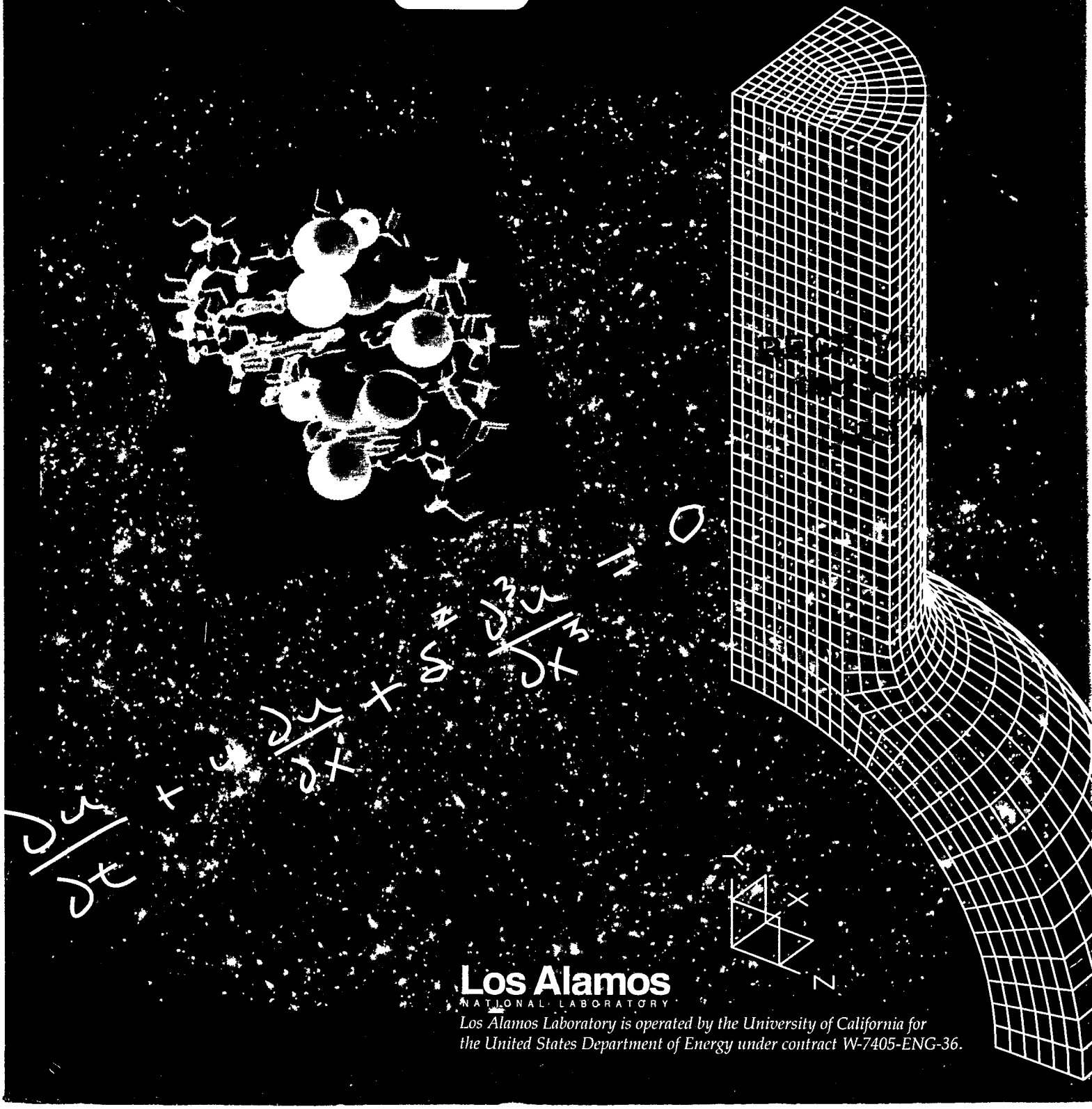
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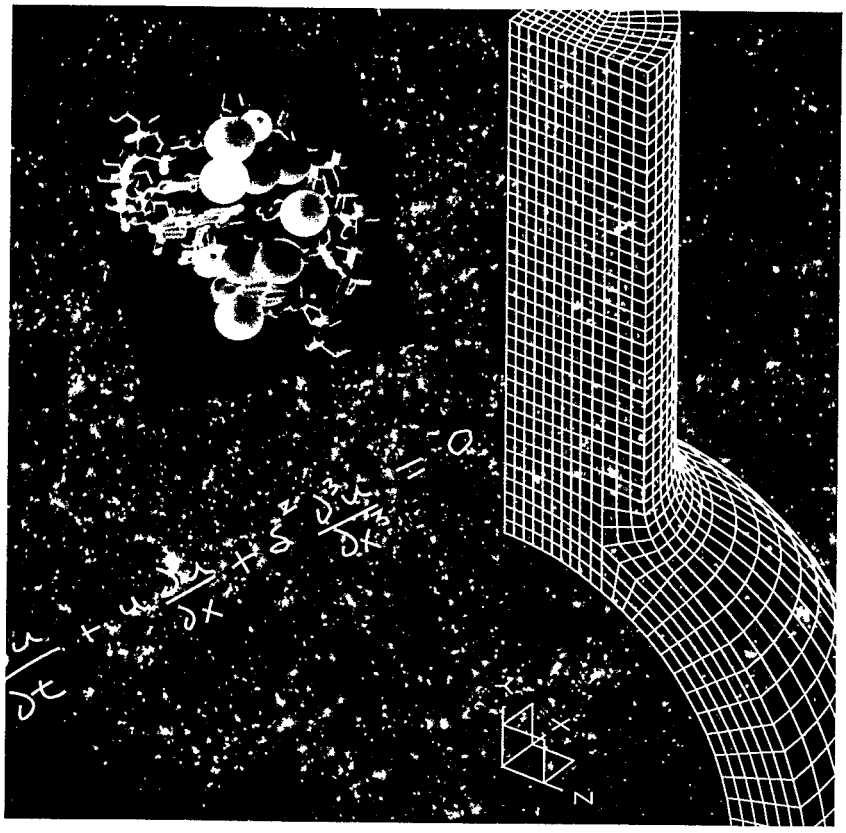


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ABSTRACT

This is the FY 1997 Progress Report for the Laboratory Directed Research and Development (LDRD) program at Los Alamos National Laboratory. It gives an overview of the LDRD program, summarizes work done on individual research projects, relates the projects to major Laboratory program sponsors, and provides an index to the principal investigators. Project summaries are grouped by their LDRD component: Competency Development, Program Development, and Individual Projects. Within each component, they are further grouped into nine technical categories: (1) materials science, (2) chemistry, (3) mathematics and computational science, (4) atomic and molecular physics and plasmas, fluids, and particle beams, (5) engineering science, (6) instrumentation and diagnostics, (7) geoscience, space science, and astrophysics, (8) nuclear and particle physics, and (9) bioscience.



FRONT COVER:

The background graphic simulates an intermediate stage in the evolution of the universe; the simulation tracks 322 million particles (M. Warren, "Determining the Mass of the Universe"). The upper-left inset shows the structural patterns for protein recognition (G. Goupta, "Predictive Models for Transcriptional Enhancers"). The superimposed image on the right is a computational mesh used to simulate the filling of a copper chalice mold (W. Rider, "Advanced Three-Dimensional Eulerian Hydrodynamic Algorithm Development"). The equation describes various nonlinear waves, such as acoustic waves in a crystal.

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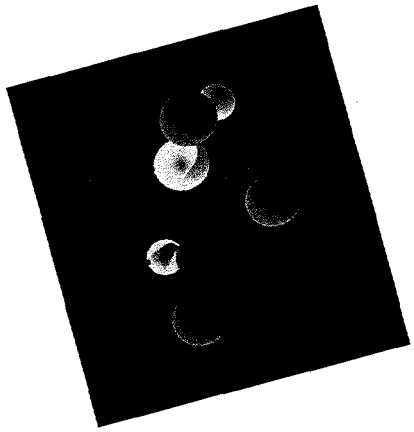
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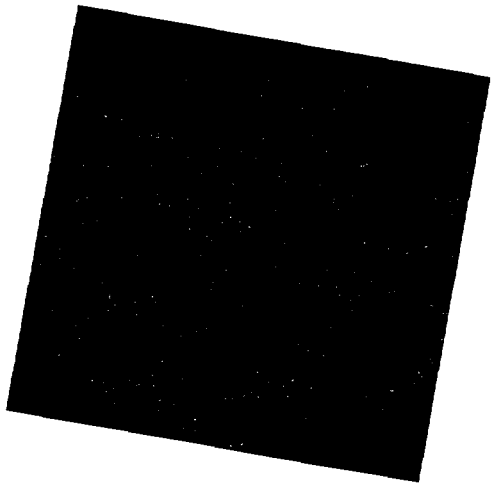
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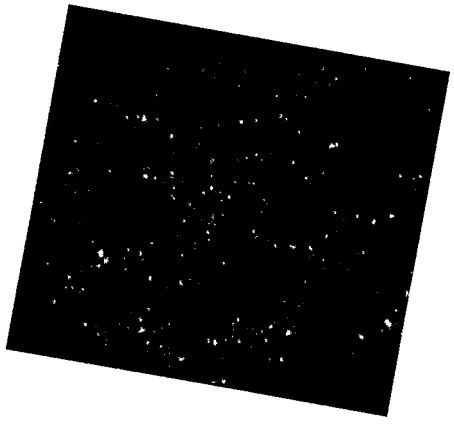
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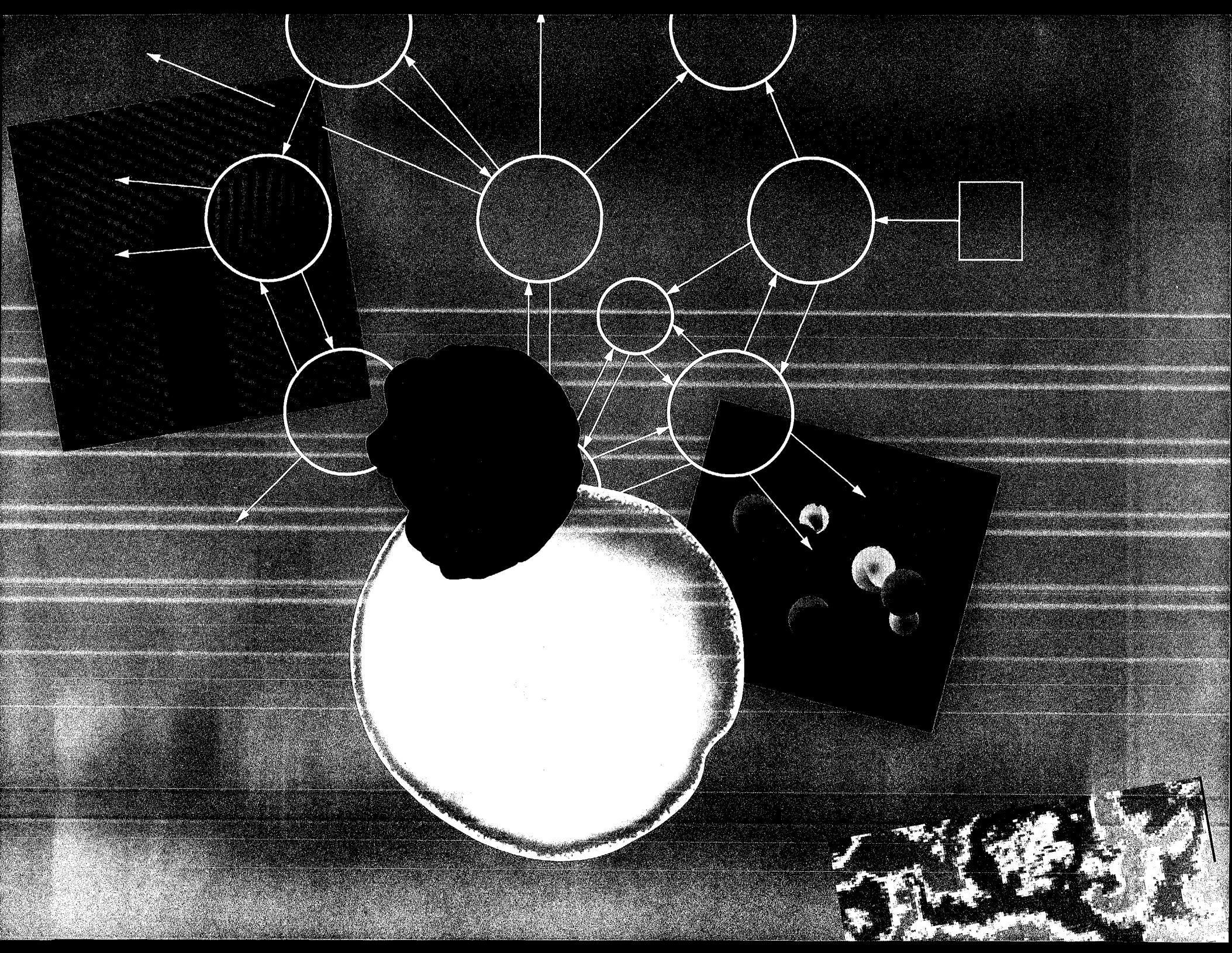
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OVERVIEW





Overview

LDRD Program Overview—FY 1997

Ed Heighway and John Vigil

The Laboratory Directed Research and Development (LDRD) Program is authorized by Congress as a means for the Laboratory to fund internally proposed, innovative research and development that extends the Laboratory's science and technology capabilities in support of its mission. There are three ways in which LDRD extends those capabilities. First, the research may explore new ways of tackling mission problems, thereby identifying opportunities to execute mission objectives in a cheaper, faster, or better way. Second, it may develop new capabilities in a Laboratory competency (area of expertise) that are needed to fulfill the mission, perhaps adding a multidisciplinary approach that provides new insight. Third, it may broaden the fundamental science and technology base that underpins the Laboratory's ability to execute its mission.

The LDRD program at Los Alamos National Laboratory sponsors all three types of research projects. In all cases, projects are chosen based on their innovation and scientific merit in a mission context, and as a result, their work leads to large numbers of publications and patents and to external recognition through awards. These, in turn, result in high visibility for the Laboratory's science and technology, making LDRD an important vehicle for attracting the best scientists and engineers to Los Alamos.

The primary mission of the Laboratory is "to reduce the nuclear danger," and there are four major components of that mission: (1) ensuring that the nation's nuclear stockpile is reliable and safe, (2) managing the production and use of nuclear materials, (3) ensuring that the environment is both restored from past nuclear activities and minimally impacted by future activities, and (4) developing technology to help eliminate proliferation of nuclear materials and weapons. To show how these mission activities are supported, Fig. 1 gives a breakdown of the Laboratory's FY 1997 funding by major program. A tabular listing of all LDRD projects as they relate to major program sponsors is provided at the end of this report (see page 275).

LDRD Program Structure

LDRD is a formally managed program that imposes accountability in the processes for selecting, executing, and reporting on projects. All projects are selected through competition and review by peers or Laboratory managers. Innovation and scientific excellence are key funding criteria. All projects must be in science and technology areas that support the Laboratory's mission.

The LDRD program at Los Alamos has three components—Competency Development (CD), Program Development (PD), and Individual Projects (IP)—each with differing goals. The CD component invests in large multidisciplinary projects that tackle significant mission-related problems from several viewpoints, often combining theory and experiment and drawing on scientists from several technical

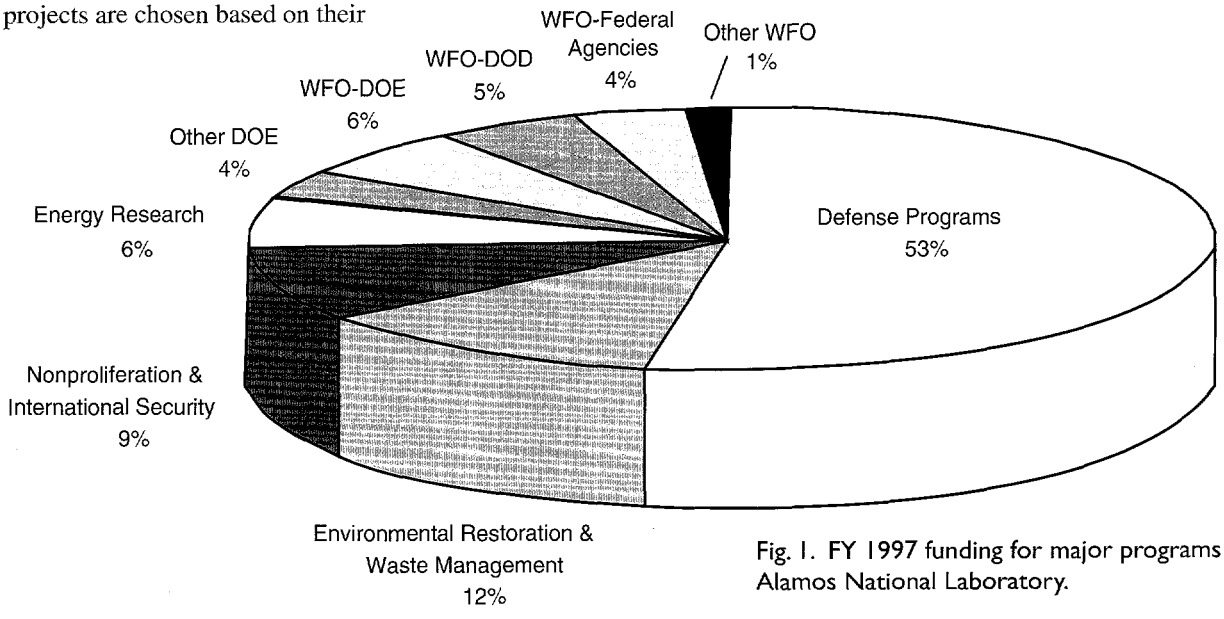


Fig. 1. FY 1997 funding for major programs at Los Alamos National Laboratory.

divisions. The PD component is intended to invest in nearer-term research and development that requires proof-of-principle, feasibility, or other confidence-building demonstrations for sponsors. The IP component is intended to fund the most far-reaching and basic research proposed by the technical staff; its goal is to extend the Laboratory's science and technology knowledge base.

In FY 1997, the LDRD program funded 309 projects for a total expenditure of $62.6 million. This sum represented 5.7% of the Laboratory operating budget. The funding distribution among the three LDRD components is shown in Fig. 2.

Competency Development. This component funds research related to the technical competencies that underpin the Laboratory's ability to execute its missions and respond to new Department of Energy (DOE) and national initiatives. The Laboratory has eight core competencies:

- Theory, Modeling and High-Performance Computing
- Complex Experimentation and Measurements
- Analysis and Assessment
- Nuclear and Advanced Materials
- Nuclear Weapons Science and Technology
- Earth and Environmental Systems
- Nuclear Science, Plasmas, and Beams
- Bioscience and Biotechnology

CD projects are intended to be cross-cutting; that is, they draw on the resources of several Laboratory divisions and have clear institutional value beyond the strategic interest of any single division. CD projects are proposed by the technical staff through their division and program offices, reviewed both for technical content and strategic value by program managers and scientific peers, and ultimately selected by the Laboratory director. Most projects are funded for three years. In FY 1997, 98 CD projects were funded.

Program Development. This component funds innovative R&D that allows the Laboratory to examine ways of meeting extended or future needs of program sponsors. The research is generally demonstrational, assessing the feasibility of new scientific approaches or technologies and establishing a technical path to advanced solutions of existing problems. PD projects are generally of shorter duration than those in the other two components, typically being funded for only one or two years. Proposals are solicited, reviewed, and ultimately selected by managers and technical staff within the Laboratory's seven program offices. These offices are as follows:

- Civilian and Industrial Technology Program
- Department of Defense Programs
- Environmental Management Program
- Nonproliferation and International Security Program
- Nuclear Materials and Stockpile Management Program
- Nuclear Weapons Technology Program
- Science & Technology Base and Energy Research Programs

In FY 1997, 64 PD projects were funded.

Individual Projects. This component funds basic research whose potential is high but whose payoff may be distant. These projects tend to be exploratory—mapping uncharted territory—and therefore involve higher risk. Proposals are evaluated and ranked by peer-review teams (one team for each of nine technical categories), with the highest ranked and most relevant ones being funded. In alphabetical order, the nine technical categories are as follows:

- Atomic and Molecular Physics and Plasmas, Fluids, and Particle Beams
- Bioscience
- Chemistry
- Engineering Science
- Geoscience, Space Science, and Astrophysics
- Instrumentation and Diagnostics
- Materials Science
- Mathematics and Computational Science
- Nuclear and Particle Physics

In FY 1997, 147 IP projects were funded.

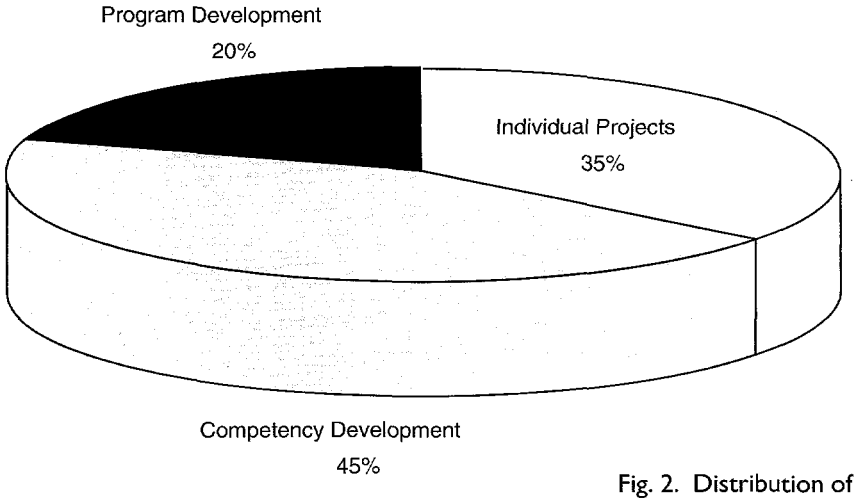


Fig. 2. Distribution of FY 1997 funding among the three LDRD components.

Quality of LDRD Research

The high quality of research supported by all components of the LDRD program is attested to by the significant number of related awards, scientific publications, and patents. In 1997, LDRD scientists continued to garner national and international recognition for their work, which included recognition by the National Academy of Sciences, the IEEE Computer Society, the Optical Society of America, the Guggenheim Foundation, the Packard Foundation, the Bomem Corporation, the von Humboldt Foundation, the American Geophysical Union, and the Biophysical Society, among others.

The number of publications continues to remain very high; this year LDRD scientists cite some 1200 publications arising from their work, a number that approaches 30% of all Laboratory publications. In addition, LDRD research contributed to 49% of the Laboratory's patent awards and 67% of the R&D 100 Awards. These achievements attest to the far-reaching and ground-breaking nature of LDRD research, which in FY 1997 was funded with only 5.7% of the Laboratory's budget.

Finally, the LDRD program is a significant proving ground for new talent attracted to the Laboratory through our postdoctoral program. In 1997, some 190 postdoctoral staff members participated at significant levels in LDRD projects. Of these, approximately one in four will become permanent Laboratory employees, rejuvenating our workforce and contributing to our central mission.

Highlights of LDRD in Support of National Security

LDRD projects contribute broadly to the science and technology that support the Laboratory's national security mission. In particular, they contribute to nuclear weapons science and technology, to nonproliferation, counterproliferation, and arms control, to nuclear materials characterization and waste disposal, and to other national security areas such as materials, detectors, and computational tools for military applications. Without nuclear testing, our fundamental understanding of the science of nuclear weapons performance must be greater, and LDRD is increasingly needed to provide innovative approaches to achieving that understanding. LDRD researchers are helping to develop new ways to make the measurements needed for stockpile certification and for improving the modeling capability of our nuclear weapons codes. Highlights of this important national security work follow.

Dynamic Radiography. LDRD researchers are exploring two new ways to make dynamic radiographs of imploding dense objects as part of an advanced hydrotest facility for stockpile stewardship. The first project explores the use of high-energy protons to probe these objects. During FY 1997, a new proton radiography experiment was installed at Los Alamos that included beam transport, a containment vessel, a magnetic lens system, and a detector system able to image small-scale contained explosions. Together with simulations and analyses, the experiment provided the first dynamic proton radiograph of a shock wave in high explosives, demonstrating proton radiography as a potentially useful technology for science-based stockpile stewardship.

This exploratory work by a multidisciplinary team of scientists convincingly demonstrated the technology's utility to a previously skeptical community—a perfect example of the value of LDRD. The experimental data provided the proof of principle for an extensive series of shots that will help researchers better understand the performance of high explosives.

The second radiography project is exploring advances in the traditional x-ray approach to hydrodynamic testing. The team is working on how to algebraically reconstruct three-dimensional images from multiview x-ray data. Particular issues under study are the limit to resolution imposed by scattering, the optimal number of views needed, and the precision with which test object edges can be located and densities estimated. Other supporting tasks are to develop photocathode technology, to simulate low-energy beam transport, particularly high-current beam-cavity interactions, to model target bremsstrahlung converters and target-beam interaction, and to develop solid-state detectors capable of high frame rates with very large frame storage capability.

Both radiography initiatives are already influencing the way the core program staff think about addressing the Laboratory's stockpile stewardship mission.

Understanding Mix. Another key initiative is applying multiscale science to the examination of dynamic mix processes in high-density matter. The term *multiscale* refers to simultaneously modeling physics phenomena from the atomic to the bulk-material scale. Mix is a process in which distinct substances (solids, fluids, and gases) are combined into some aggregate material.

The goals of this work are to develop new approaches to creating validated, predictive models for fluid and materials mix based on microphysical descriptions of key processes. Once the models are understood and verified by experiment, their features can be incorporated into design and assessment codes. The project combines such diverse elements as adaptive mesh simulation, multifluid turbulent mixing, experiments on and modeling of solid interactions, statistical representations of mixing, quantum molecular dynamics studies, shock-wave modeling, and spall experiments. This fundamental theoretical and experimental study will impact basic systems modeling for stockpile stewardship.

Improving the Monte Carlo Method. Researchers are making rapid advances in the Monte Carlo method for calculating particle transport, a key component in modeling complex nuclear systems. A major goal is to find ways to reach

exponential convergence in moderately nontrivial problems. The ongoing goal is to generalize new exponential convergence methods and apply them to realistic problems that currently cannot be solved.

Researchers are applying advanced statistical methods to Monte Carlo estimates of density distribution functions in order to improve both the analysis of Monte Carlo data and the efficiency of Monte Carlo algorithms. They are also using learning algorithms to improve importance sampling. All of these developments may radically increase the speed with which Monte Carlo calculations can be applied to complex problems while maintaining fidelity and accuracy in the representation. The results can be applied to both defense and civilian problems.

Understanding Actinides. One research project is focused on determining the electronic structure of highly correlated 5f elements and compounds, with particular emphasis on plutonium. Being very complex, plutonium is one of the few materials that are not yet well understood at a fundamental level and whose behavior is inadequately predicted by current models. Scientists are using high magnetic fields to produce special states of actinide compounds for which an oscillatory magnetization, known as the de Haas van Alphen effect, yields fundamental information about their electronic structure. This work on static properties is complemented by investigation of microscopic dynamical phenomena that occur on time scales of 10^{-11} to 10^{-14} seconds. The purpose of these experiments is to reveal the role of many-body interactions in f-electron systems.

These investigations into the physics of actinides are paralleled by a separate investigation into their chemistry, i.e., how they react with other materials. Actinide chemistry is critically important, providing the technical basis for plutonium process and separation chemistry and metallurgy. This project closely integrates chemical synthesis and characterization, spectroscopy, and theory and modeling to understand the chemical properties of actinides. The research specifically investigates actinide speciation, structure, and chemical equilibrium in aqueous/nonaqueous systems, on oxide surfaces, and of small molecules on actinide surfaces. The results will help us address complex chemical problems associated with the actinides.

These varied investigations into the nature of complex actinides will add to our knowledge base for stockpile stewardship.

Aging of Polymeric Materials. Researchers are developing a science-based methodology for predicting the aging processes of polymeric materials, materials that are integral to national defense. The approach being taken is based on characterizing molecular response to mechanisms that cause aging and extending those responses through various length scales, i.e., from atomic to bulk material. The work includes both modeling and experiments.

At the atomic and molecular level, the effects of water and hydrogen bonding on polymers are being modeled in a three-dimensional lattice, Monte Carlo polymer code based on bond fluctuation models. At the mesoscale level, a model describing microphase island structure has been developed. Preliminary evaluation has been completed of network models and the viscoelastic response of networks to constant strain experiments. At the bulk-material scale, continuum models based on microstructure considerations have been developed and the pressure/volume response and high-shear-rate response evaluated. This work is already influencing thinking about how to monitor the stockpile.

Metal Deformation under High Strain. Stockpile stewardship requires accurate predictions of the implosion dynamics when materials are subjected to extreme conditions (large strain, high strain rates, and high temperatures); precise knowledge of these conditions is critical. Through coordinated experiments, mathematical modeling, and code implementation, researchers are improving the calibration of strength models using data at elevated temperatures and at very large strains, extending these models to include texture evolution, and then using molecular dynamics to improve the models at intermediate strain rates.

Among other achievements, this work has successfully studied the intersection process for dislocations and applied a ductile damage model to spallation data. The texture and constitutive behavior evolution of steel has been characterized and incorporated into physics models. These successes add to our fundamental knowledge of the properties and behavior of key materials in the nation's stockpile.

Remote-Sensing Technology. A significant part of the Laboratory's national defense mission is to help detect and then counter the proliferation of weapons of mass destruction. A key to such work is developing the means to remotely detect proliferation. One LDRD project encompasses a threefold development of remote-sensing capabilities: hyperspectral remote sensing, active remote sensing (lidar), and a broad exploration of new remote-sensing concepts. The specific goal is to detect gaseous plumes from proliferation activities and to understand the atmospheric boundary layer in which gases may be transported.

This work has achieved "first-of-a-kind" measurements of microstructural convection cells in the equatorial-marine atmospheric boundary layer. These measurements are being modeled by three-dimensional computations with unprecedented meter-size spatial resolution in a fully coupled radiation-mass transfer code. Several detectors have been developed and are now making new and exciting measurements. A microwave interferometer has provided unique measurements of water vapor heterogeneities in the atmospheric boundary layer. A modified Fourier transform infrared spectrometer has produced high-resolution emission spectra of gaseous components in volcanic plumes at large stand-off distances. These and

other developments are significantly advancing a remote-sensing capability that has both civilian and defense applications.

Fundamental Studies of High Explosives. The science and modeling of high explosives lie at the heart of the Laboratory's national security mission. Improved approaches to modeling high-explosive reactions are critical to success in both stockpile stewardship and the Accelerated Strategic Computing Initiative. A new LDRD study is investigating condensed-phase chemistry, widely recognized as the weakest link in our understanding of the decomposition of energetic materials. New diagnostic methods, combined with a sophisticated modeling effort and an array of benchmark experiments over a broad range of pressures and temperatures, will enable predictive modeling of high-explosive response. The final products of this work will be an understanding of the fundamental chemistry and a predictive kinetics modeling that are validated by coupled (chemistry and transport) experiments.

Separations of Nuclear Materials. Another new LDRD project seeks to understand the molecular mechanisms governing the transport of ions and molecules through membrane pores and to develop a new class of molecular recognition membranes. This work exploits recent advances in nanotechnology to fabricate thin membrane assemblies that incorporate specific receptors for molecular recognition in their pore structure. Using organic and inorganic supports with well-defined structures, researchers can control the arrangement of ionic and molecular receptors in the pores in order to gain a molecular-level understanding of the fundamental principles that govern transport.

A new class of membranes with unprecedented selectivity and throughput is expected. These synthetic membranes will mimic the extraordinary specificity of biological membranes but will operate under a broader range of conditions, including the harsh conditions of nuclear processing operations. The work will include development of a theoretical model that can predict the detailed molecular functioning of the membranes. The knowledge gained will allow the design of molecular recognition membranes in which the relationships between selectivity, flux, and stability are well-defined and the application of such membranes to myriad defense and civilian applications.

Understanding Nonequilibrium Systems. The goal of another new LDRD project is to understand the physical principles underlying the complex real-time dynamics of nonequilibrium phase transformations and multiscale phenomena. Such phenomena are important to many facets of the Laboratory's core mission.

The plan is to develop a microscopic theory of nonequilibrium processes and corresponding numerical methods at the finest spatiotemporal scales. This work will be augmented by identifying coherent structures, significant variables, and the spectrum of fluctuations to apply coarse-graining techniques in space and time. Numerical methods will be developed to solve the resulting mesoscale, stochastic partial-differential equations. Specific tasks include looking at fluctuation hierarchy in microtheory, carrying out high-resolution numerical simulations of microtheory, numerically characterizing the non-Gaussian noise spectrum, and identifying nonlinear coherent structures or variables. This highly mathematical approach will bring new insight into the complex processes prevalent in the nuclear environment.

New Nuclear Measurement Systems. A research project was started that focuses on the science of nuclear material measurements, a science that is integral to the Laboratory's nonproliferation work. The technology being developed can be applied to the disposal of nuclear waste, to residue stabilization, and to arms control.

The challenges in this work include managing the rapidly growing inventories of nuclear materials from civilian programs, safeguarding materials recovered from dismantling nuclear weapons in both the United States and the former Soviet Union, and removing the residues and wastes left over from weapons production. Project goals include developing the means of assaying difficult-to-measure nuclear material, which in turn will require developing fast-neutron detectors and new methods of active interrogation as well as integrating large volumes of data. These developments will support the Laboratory's primary mission of reducing the global nuclear danger.

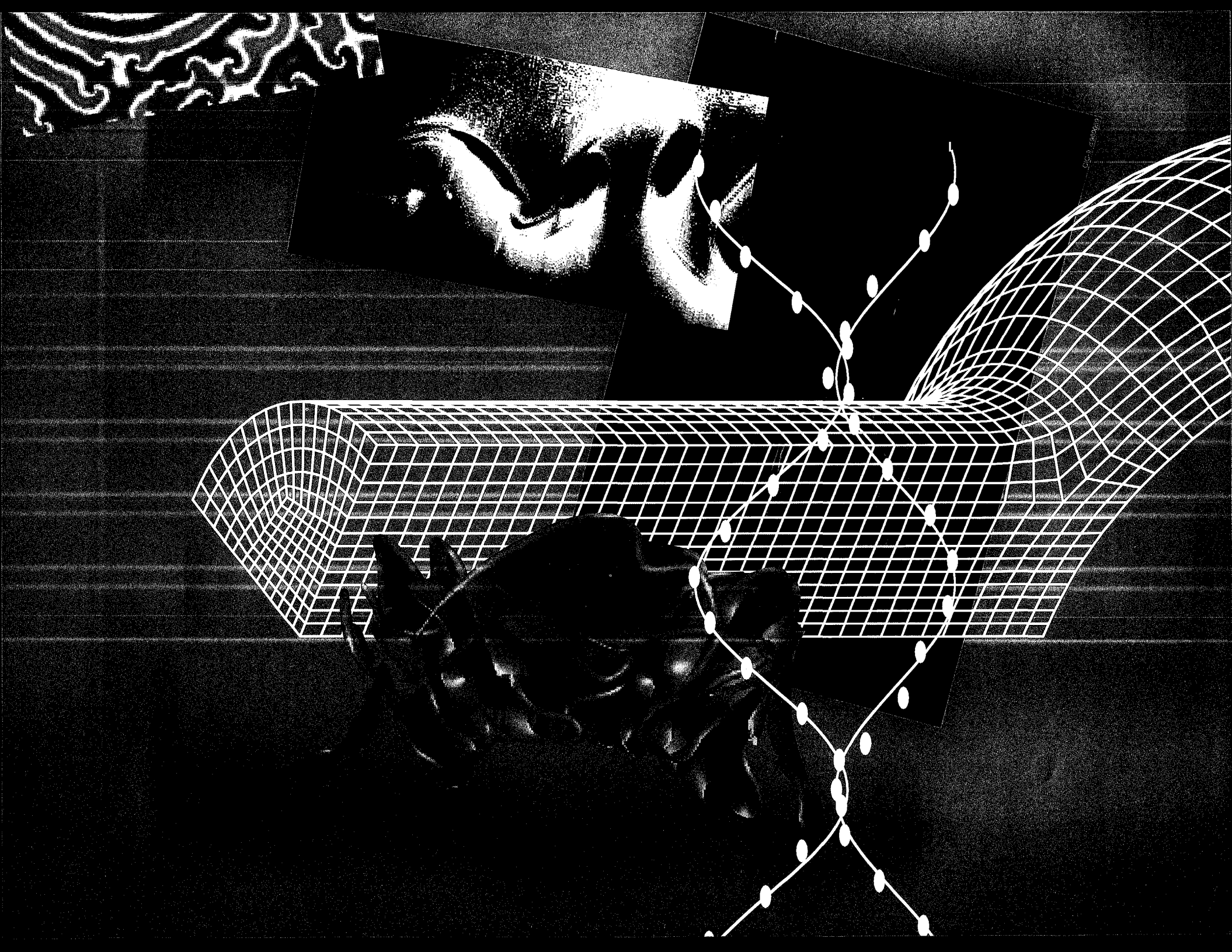
LDRD Project Summaries

The above highlights exemplify but a few of the ways in which LDRD nurtures the science and technology required to fulfill the Laboratory's national security mission. These examples primarily explore new ways of tackling specific mission problems. Other projects invest in expanding the Laboratory's core competencies. Still others seek to broaden the fundamental science and technology base that underpins our ability to carry out the Laboratory mission. All projects contribute to the long-term vitality of the Laboratory's national security work.

In the sections of our annual progress report that follow, project summaries are grouped by their LDRD component—CD, PD, and IP. Within each component, they are further grouped into the nine technical categories for projects, ordered as follows:

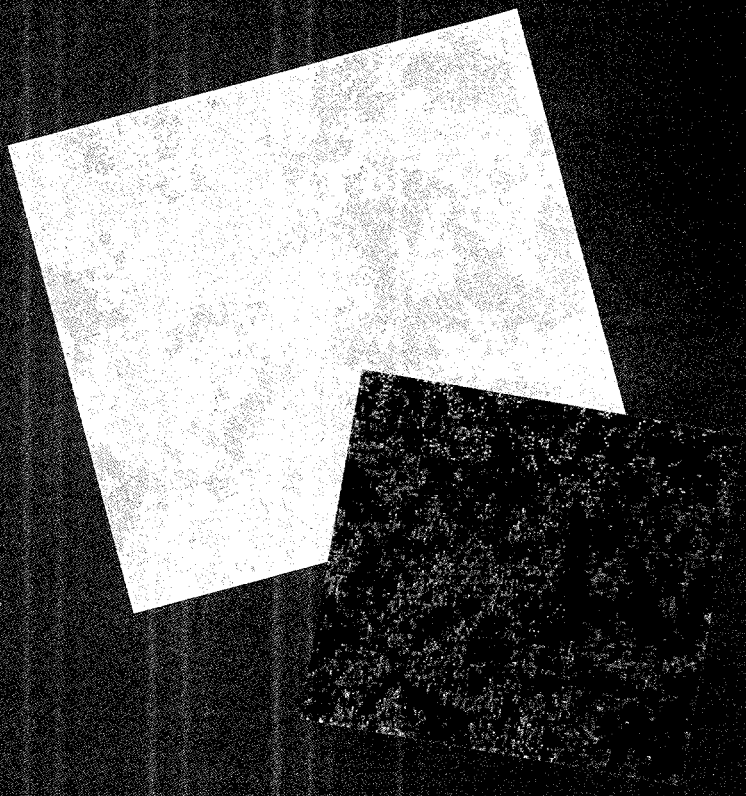
- Materials Science
- Chemistry
- Mathematics and Computational Science
- Atomic and Molecular Physics and Plasmas, Fluids, and Particle Beams
- Engineering Science
- Instrumentation and Diagnostics
- Geoscience, Space Science, and Astrophysics
- Nuclear and Particle Physics
- Bioscience

An index to the projects' principal investigators is included at the end of this report.



1997 LDRD

Competency Development Projects



A Neutron Diffraction and Computational Micromechanics Study of Deformation in Advanced Materials

Mark Bourke

This project focuses on the use of neutron diffraction to study deformation in polycrystalline materials. Our goal is to demonstrate and establish synergies between neutrons, used as a unique experimental tool, and recently developed models of polycrystalline behavior (including both self-consistent and finite-element elastic-plastic formulations). The effort includes making measurements of residual and in situ loading strains, as well as thermal relaxation effects.

During the past year, we made many significant achievements. In collaboration with the Illinois Institute of Technology, we used a high-temperature (1300°C) furnace on the neutron powder diffractometer to measure room-temperature residual strains and high-temperature relaxation of dispersion-strengthened NiAl reinforced with alumina particulates. Using a finite-element code (ABAQUS™), we successfully compared these measurements with creep-relaxation predictions.

Following a series of in situ deformation measurements on stainless steel, we demonstrated the use of Rietveld refinement both for macrostrain interpretation of neutron data and for semiquantitative inference of plastic strain history (see accompanying figure). We measured strain and texture evolution in shape-memory-alloy composites (NiTi-TiC) under varying loads. We performed routine measurements on both Ti-SiC and W-Kanthal metal-matrix composites and performed complementary finite-element calculations in which we demonstrated the effect of thermomechanical processing on residual strain evolution. In a two-pronged effort, polycrystalline deformation of both high- and low-symmetry materials was explored numerically and experimentally at Los Alamos and the University of California, San Diego, respectively. The focus of this effort was on simple austenitic steel and on TiAl, respectively.

Publications

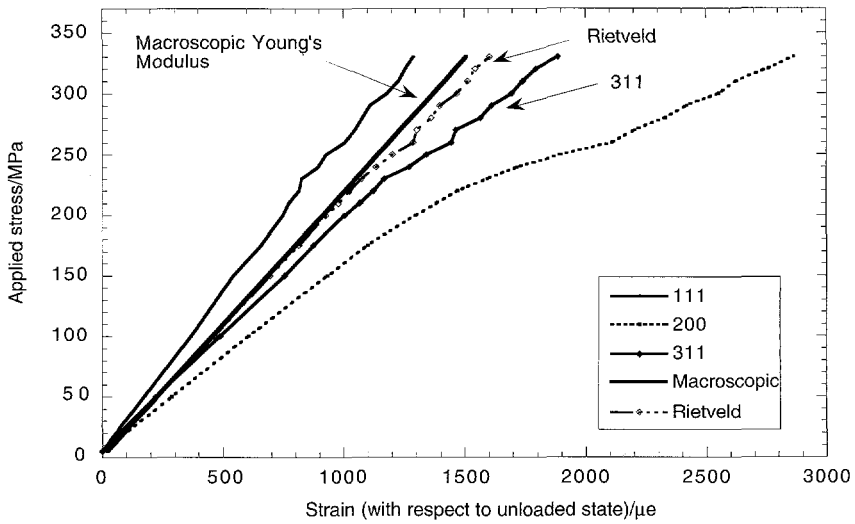
Daymond, M.R., et al, "Effect of Texture on hkl Dependent Intergranular Strains Measured In Situ at a Pulsed Neutron Source" (to be published in *Proc. Fifth Int. Conf. Residual Stress*).

Daymond, M.R., et al., "Elastic Strain Distribution in Copper Reinforced with Molybdenum Particulates during Deformation at Low and High Temperature" (submitted to *Met. Trans.*).

Daymond, M.R., et al., "Use of Rietveld Refinement for Elastic Macrostrain Determination and for Evaluation of Plastic Strain History from Diffraction Data," *J. Appl. Phys.* **82** (4), 1554 (1997).

Dunand, D.C., et al., "Mechanical Properties of NiTi-TiC Shape Memory Alloys," *Mat. Res. Soc. Symp. Proc.* **459**, 131 (1997).

Vaidya, R., et al., "Measurement of Bulk Residual Stresses in Molybdenum Disilicide/Stainless Steel Joints Using Neutron Scattering" (to be published in *Acta. Met.*).



Elastic strains were measured using neutron diffraction during an in situ load test of austenitic stainless steel. The graph shows three lattice reflections (111, 200, 311), the Rietveld "average" response, and the macroscopic response.

Microstructure and Microanalysis of Materials at Atomic Resolution

Terence Mitchell

We are studying structure-property relationships in a wide variety of materials by determining their microstructure and microchemistry at atomic resolution. We are able to image with 1-Å structural resolution by using the field-emission high-resolution transmission electron microscope (HRTEM) and to perform chemical analysis and imaging at the 5- to 10-Å level with the field-emission scanning transmission electron microscope. Examples of applications include studying (1) the atomic structure of domain boundaries in ferroelectrics and their interaction with defects that control mobility, (2) the structure and chemistry of interfaces in nanolayered materials, (3) the structure and chemistry of grain boundaries in structural ceramics, and (4) the atomic structure of dislocations in intermetallics, which controls their mechanical properties.

We have tackled a wide range of materials science problems using electron microscopy. A few recent results are the following:

- We found dislocations in molybdenum disilicide, imaged at the atomic level, to be dissociated on various planes, leading to an understanding of its deformation behavior.
- X-ray energy dispersive spectrometry and HRTEM show that little intermixing occurs at the interfaces of copper/niobium multilayers.
- We used HRTEM to study the atomic structure of fine twins and stacking faults formed in conductive $SrRuO_3$ thin films, which are important in various electrode applications.
- We studied the atomic structure of various defects in chemical-vapor-deposition silicon nitride, in particular showing that 60° rotation boundaries are quite common.
- We discovered the existence of body-centered cubic (bcc) copper in copper/niobium multilayers with spacings of 12 Å or less; parallel electron energy loss spectrometry also shows that the Fermi energy of bcc copper is lower than that of face-centered cubic copper.
- We imaged 180° ferroelectric domain walls in $LiNbO_3$ by both scanning and transmission electron microscope techniques.

Publications

Chu, F., et al., "Elastic Properties of the O Phase in Ti-Al-Nb Alloys," *Intermetallics* **5**, 147 (1996).

Han, K., and G.C. Weatherly, "Transmission Electron Microscopy Investigation of Twinning in Titanium Carbonitride," *Philos. Mag. Lett.* **76**, 247 (1997).

Jia, Q.X., et al., "Heteroepitaxial Growth of $Ba_{0.5}Sr_{0.5}TiO_3$ Thin Films on Si with Conductive Metallic $SrRuO_3$ as a Bottom Electrode," *Thin Solid Films* **299**, 115 (1997).

Jia, Q.X., et al., "High Temperature Superconductor Edge-Geometry SNS Junctions with Engineered Normal-Metal Layers," *Supercond. Sci. Technol.* **9**, 985 (1996).

Kotula, P.G., et al., "Orientation Relationships in the System Nb-$NbCr_2$," *Microsc. Microanal.* **3**, **Suppl. 2**, 707 (1997).

Kotula, P.G., et al., "Site Occupancies in Ternary C15 Ordered Laves Phases," *Mater. Res. Soc. Symp. Proc.* **460**, 617 (1997).

Liu, Y.M., et al., "Anisotropic Damage Evolution in Unidirectional Fiber Reinforced Ceramics," *Acta. Mater.* **45**, 3981 (1997).

Liu, Y.M., et al., "Elastic Properties of Laminated CAS/SiC Composites Determined by Resonant Ultrasound Spectroscopy," *J. Am. Ceram. Soc.* **80**, 147 (1997).

Liu, Y.M., et al., "The Influence of Fiber/Matrix Interface on the Mechanical Behavior of Nicalon SiC Fiber-Reinforced Glass-Ceramic Composites," *Mater. Res. Soc. Symp. Proc.* **348**, 161 (1997).

Lu, Y.-C., et al., "Effect of 690 keV Xe Ion Irradiation on the Microstructure of Amorphous $MoSi_2$/SiC Nanolayer Composites," *Nucl. Instrum. Methods. Phys. Res., Sect. B* **127**, 648 (1997).

Lu, Y.-C., et al., "Microstructural Instability in $MoSi_2$/SiC Nanolayers," *Microsc. Microanal.* **3**, **Suppl. 2**, 399 (1997).

Lu, Y.-C., et al., "Observations of Dislocations in Cu/Nb Nanolayer Composites after Deformation," *J. Mater. Res.* **12**, 1939 (1997).

Mitchell, T.E., et al., "Structure and Mechanical Properties of Copper/Niobium Multilayers," *J. Am. Ceram. Soc.* **80**, 1673 (1997).

Ormeci, A., et al., "Elastic Constants of a Laves Phase Compound: C15 $NbCr_2$," *Mater. Res. Soc. Symp. Proc.* **460**, 623 (1997).

Suematsu, H., et al., "Plastic Deformation of Silicon Nitride Single Crystals," *Mater. Sci. Eng. A* **209**, 97 (1996).

Suematsu, H., et al., "The a-b Transformation in Silicon Nitride Single Crystals," *J. Am. Ceram. Soc.* **80**, 615 (1997).

Thoma, D.J., et al., "Comparison of $NbCr_2$ and HfV_2 C15 Laves Phases," *Mater. Res. Soc. Symp. Proc.* **460**, 689 (1997).

Manipulation of Residual Stresses to Improve Material Properties

Mark Bourke

This project takes an innovative approach to control material properties by manipulating residual stresses generated during phase transformations. The focus is on a reduction reaction; however, the capabilities and methodology will be applicable to other transformations in ceramic and metallic materials. Our initial approach has been to study residual stresses generated due to the volume shrinkage under constraint that occurs when a ceramic oxide, $NiAl_2O_4$, is reduced to a metal-ceramic composite consisting of nickel and Al_2O_3.

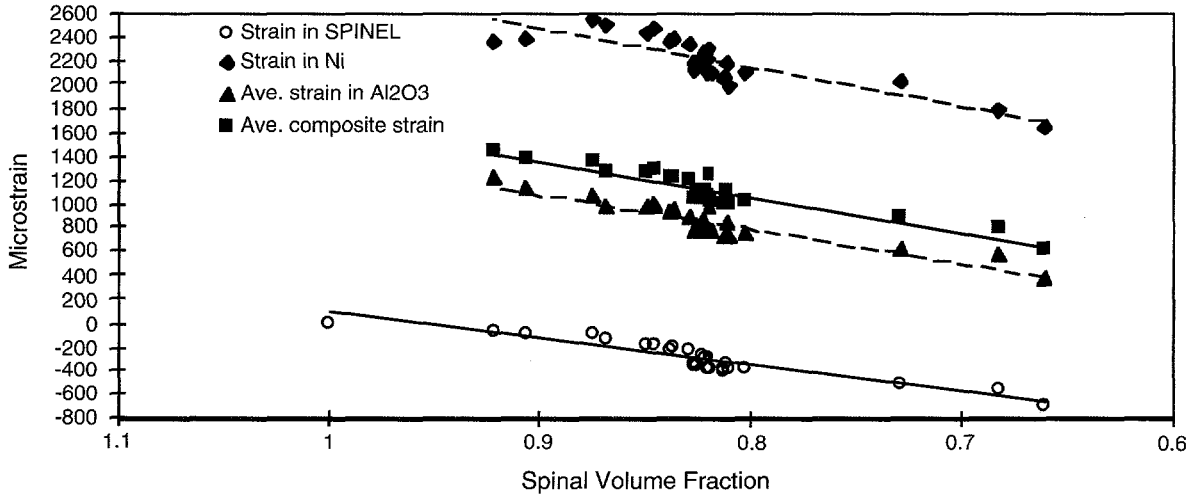
Results obtained during the past year derive from four areas: x-ray diffraction (XRD) stress measurements on an application prototype, neutron diffraction kinetics measurements, finite-element simulations, and theoretical calculations of vacancy formation and migration energies for the spinel reduction of interest.

Significant results are listed below.

- A controlled-atmosphere furnace was used to perform in situ reductions while simultaneously collecting neutron diffraction data (see accompanying figure).
- The effect of temperature and sample density on the nickel-spinel reduction reaction was studied. Compressive residual stresses (up to 300 MPa) in the core of a cylindrical specimen were observed during reduction.
- Compressive residual stresses in model specimens consisting of alumina disks surrounded by reduced spinel were measured by XRD.
- Finite-element modeling studies were initiated on both the model specimens and on those used in neutron diffraction experiments.
- XRD measurements on magnetic thin films produced by the same reduction reaction demonstrated the presence of in-plane tensile residual stresses (up to 300 MPa), which was correlated to the perpendicular magnetization.

Publications

Ustundag, E., et al., "Influence of Residual Stresses on the Evolution of the Microstructure during the Partial Reduction of $NiAl_2O_4$," *Mater. Sci. Eng. A* **238**, 50 (1997).



Change in spinel volume fraction plotted against the evolution of strain in the Al_2O_3 core. These measurements were recorded over approximately 36 hours at a temperature of 1220°C.

Structure/Property Relations in Elasticity and Plasticity

U. Fred Kocks

This project addresses a comprehensive modeling of the mechanical response of materials, including elasticity and thermal expansion, anelasticity and viscous behavior, creep and plasticity, and eventually recovery and recrystallization. Such modeling is based on microstructural observations that we will carry to a macroscopic scale. Our end result will be "average constitutive relations" at the large scale, derived from those at the small scale. Inasmuch as the basis of the macroscopic modeling is physical fundamentals, it should be applicable beyond the range of experimental verification. We can also predict mesoscopic internal stresses, which influence macroscopic behavior (e.g., that occurring upon stress reversals).

In the past year, the thrust of our work has been on internal stresses. We organized an international workshop on the subject, in conjunction with the Manuel Lujan Jr. Neutron Scattering Center. Our internal stress modeling has focused on interpreting neutron diffraction measurements done in nickel-based alloys and steels and on developing new polycrystal models capable of predicting residual strains after large plastic deformation. We collaborated with experimentalists in the Materials Science and Technology Division and at Argonne National Laboratory to develop a comprehensive analysis of the residual stresses and fracture of beryllium.

Applications of the Los Alamos polycrystal plasticity and self-consistent computer codes to various Laboratory problems provided new insight. Specifically, the incorporation of recrystallization effects into the viscoplastic self-consistent code permitted us to explain deformation textures of calcite rocks under low- and high-temperature conditions. In addition, we initiated a study of rate effects upon the relative activity of slip and twinning systems.

Publications

Kocks, U.F., C.N. Tomé, and R.-H. Wenk, Eds., *Texture and Anisotropy* (Cambridge University Press, New York, NY, in press).

Lebensohn, R.A., et al., "Self-Consistent Calculation of Intergranular Stresses Based on a Large Strain Visco-Plastic Model" (5th Int. Conf. on Residual Stresses, Linkoping, Sweden, June 16–18, 1997).

Tomé, C.N., "Tensor Properties of Textured Polycrystals," in *Texture and Anisotropy*, U.F. Kocks et al., Eds. (Cambridge University Press, New York, NY, in press), Chap. 7.

Tomé, C.N., and G.R. Canova, "Self-Consistent Modeling of Heterogeneous Plasticity," in *Texture and Anisotropy*, U.F. Kocks et al., Eds. (Cambridge University Press, New York, NY, in press), Chap. 11.

Tomé, C.N., et al., "Interpretation of Intergranular Stress Measurements in Monel-400 Using Polycrystal Models" (5th Int. Conf. on Residual Stresses, Linkoping, Sweden, June 16–18, 1997).

Advanced Research Capabilities for Neutron Science and Technology—New Polarizers

Robert A. Robinson

The goal of this project is to develop laser-driven helium-3 polarizers suitable for condensed-matter research at the Los Alamos Neutron Science Center (LANSCE). Helium-3 polarizers are the technology of choice for broadband applications at wavelengths below 2 Å and for large-divergence applications at all wavelengths. This project involves developing the design, including the laser, glass cell, magnetic shielding, and neutronics, and testing the cell both on the surface profile analysis reflectometer (SPEAR) and on a diffractometer.

This year we improved the system diagnostics. The laser wavelength is defined by the operating temperature. We purchased a spectrometer system that allows accurate tuning of the lasers and measuring the degree of light absorption.

We directed most of our effort toward maximizing the helium-3 polarization. We developed nuclear magnetic resonance techniques that allow accurate measuring of the helium-3 polarization. Helium-3 is polarized in a cell made from special glass; the degree of polarization depends on physical and chemical properties of the glass. We manufactured and tested small quantities of special low-iron, low-boron-10 glass and performed polarization tests on SPEAR with thermal neutrons and epithermal neutrons on flight path 2 at LANSCE. We achieved helium-3 polarizations of 43% and 55% in the beam and in the laboratory, respectively. In addition, we performed an experiment at the Manuel Lujan Neutron Scattering Center using two polarized helium-3 cells, one as a polarizer and one as an analyzer. Data analysis of this experiment has not yet started.

Structure/Property Relationships in Metal Oxides and Magnetic Materials Studied by Scanning Probe

Marilyn Hawley

Our work during the past year had three goals. The first goal was to probe the relationship between growth parameters, structure, local and bulk properties, stress, and defects in strontium-doped, pulsed-laser-deposited colossal-magnetoresistive (CMR) films (ferromagnetic technetium ~370°C) with potential application in the next-generation magnetic read heads. The second goal was to use atomic- and magnetic-force microscopies (AFM and MFM) to study plastic-deformation-induced residual stress in amorphous FeSiB ribbons (see accompanying figure). Our third goal was to demonstrate the utility of MFM to study austentite to martensite stress-induced transformations in iron.

For these objectives, MFM was fully implemented. For the CMR films, primary interest was in the role of lattice-mismatch, growth processes, and post-deposition anneal in generating magnetic domains and defects that impact on performance.

Films were grown on $LaAlO_3$ (compressive) and $SrTiO_3$ (tensile) at 500°C–800°C, half annealed (950°C, 10 hours in O_2). Substrate-dependent differences in transport and magnetization were observed despite indistinguishable structural differences. Magnetic structures were associated with stress around pinholes, pits caused by incomplete coalescence on annealing, and domains induced by residual stress. The work was extended to other oxide films and development of electric-force microscopy.

The combined work resulted in over 10 papers. Work on iron-based ribbons continued with measurements relating inhomogeneous plastic-deformation shear-band structure to residual stress as observed by MFM. Last, dimpled burst valves were used to map ferrous martensitic regions as a function of post-stress annealing temperature.

Publications

Brown, G.W., et al., "Growth and Magnetic Structure of $La_{0.67}Sr_{0.33}MnO_3$ Films," *Mater. Res. Soc. Symp. Proc.—Epitaxial Oxide Thin Films III* **474**, 179 (1997).

Hawley, M.E., "Scanning Tunneling Microscopy," in *Characterisation of High-Tc Materials and Devices by Electron Microscopy* (Cambridge Press, Cambridge, U.K., 1997).

Hawley, M.E., et al., "CMR Films' Structure as a Function of Growth and Processing," *J. Cryst. Growth* **174**, 455 (1997).

Hawley, M.E., et al., "Growth-Related Magnetic and Physical Structures in CMR Films" (to be published in *Trans. Am. Ceram. Soc. 99th Annu. Symp.*).

Hawley, M.E., et al., "STM and X-Ray Diffraction Temperature-Dependent Growth Study of $SrRuO_3$ PLD Thin Films," *Mater. Res. Soc. Symp. Proc.—Thin Films: Surface and Morphology* **441**, 57 (1997).

Jia, Q.X., et al., "Pulsed Laser Deposition of Conductive $SrRuO_3$ Thin Films," *J. Vac. Sci. Technol. A* **15** (3), 1080 (1996).

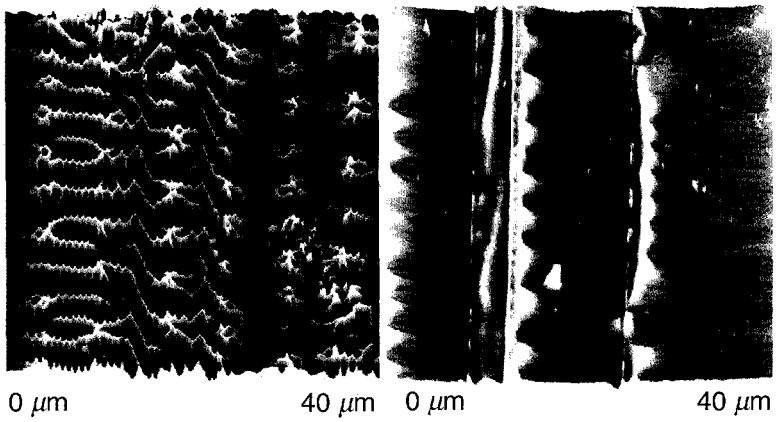
Reagor, D., et al., "Noise and Operational Characteristics of Magnetometers Made From Superconducting-Normal-Superconducting Josephson Junctions," *Mater. Res. Soc. Symp. Proc.—Epitaxial Oxide Thin Films III* **474**, 113 (1997).

Schlom, D.G., et al., "The Controlled Growth of Perovskite Thin Films: Opportunities, Challenges, and Synthesis" (to be published in *Trans. Am. Ceram. Soc. 99th Annu. Symp.*).

Schlom, D.G., et al., "Epitaxial $YBa_2Cu_3O_{7-\delta}/SrRuO_3$ Heterostructures," *Mater. Res. Soc. Symp. Proc.—Epitaxial Oxide Thin Films III* **474**, 85 (1997).

Theis, C.D., et al., "Adsorption-Controlled Growth of $Bi_4Ti_3O_{12}$ by Reactive Molecular Beam Epitaxy" (to be published in *Appl. Phys. Lett.*).

Theis, C.D., et al., "Adsorption-Controlled Growth of Ferroelectric $PbTiO_3$ and $Bi_4Ti_3O_{12}$ Films for Nonvolatile Memory Applications by MBE," *Mater. Res. Soc. Symp. Proc.—Epitaxial Oxide Thin Films III* **474**, 297 (1997).



Compressive (normal: left panel) versus tensile (in-plane: right panel) stress-induced magnetic structure in amorphous FeSiB ribbons studied by MFM.

Vesicle and Lamellar Phase Stability: An Experimental Approach to a Problem Central to the Theory of Complex Fluids

Rex Hjelm

On theoretical grounds, membrane stability in aqueous media is due to spontaneous curvature and bending rigidity—whether the membrane comprises stacked, infinite bilayer sheets (lamellae) or closed, finite bilayer sheets (vesicles). In other theoretical work, the effects of surface tension are thought to come into play through the work required to change the volume and surface area. These ideas have never been subjected to rigorous experimental tests. Our objective is to design such experimental tests, using the predictions of the theories on changes in stability with composition, temperature, and pressure.

During the past year, we designed and built a pressure system to study the morphological behavior of membrane systems. We made significant advances in a fundamental problem in complex fluids, understanding the thermodynamic and molecular aspects of vesicle and lamellar phase stability, through small-angle neutron scattering studies of pressure- and temperature-induced changes in vesicle formation in mixed colloids.

We demonstrated that there are two transitions, one from vesicles to stacked lamellar sheets at low pressure and another from stacked lamellae to a stacked ribbon phase at higher pressure. The low-pressure transition was in a pressure range predicted by the Young-LaPlace equation, though the results lead us to question the standard interpretation of this effect at the molecular level. We also demonstrated that the pressure- and temperature-induced transitions are reversible, opening the way to thermodynamic analysis of vesicle stability.

Publications

Mang, J., and R.P. Hjelm, "SANS Investigation of the Pressure and Temperature-Dependent Structure of the Bile Salt Lecithin System," *Mol. Cryst. Liq. Cryst.* **299**, 439 (1997).

Mang, J., and R.P. Hjelm, "Small-Angle Neutron Scattering Studies of Vesicle Stability" (to be published in *Int. J. Thermophys.*).

Nonequilibrium Phase Transitions

Emil Mottola

Nonequilibrium phase transitions play a central role in a very broad range of scientific areas, ranging from nuclear, particle, and astrophysics to condensed matter physics and the materials and biological sciences. The aim of this project is to begin creating a deeper and more fundamental understanding of the common physical principles underlying the complex real-time dynamics of phase transitions. To achieve this goal, we have assembled a multidisciplinary team with integrated theoretical and experimental expertise in statistical mechanics, many-body theory, field theory, and materials science.

This past year, our main accomplishment was to develop general theoretical tools to deal with nonequilibrium processes and numerical methods robust enough to capture the time-evolving structures that occur in actual experimental situations. We have begun applying these tools to Laboratory-wide, multidivisional efforts in relativistic heavy-ion physics (transition to a new phase of nuclear matter consisting of a quark-gluon plasma) and layered high-temperature superconductors (critical currents and flux flow at the National High Magnetic Field Laboratory).

Publications

Cooper, F., et al., "Anomalous Transverse Distribution of Pions as a Signal for the Production of DCCs," *Phys. Rev. C* **54**, 3298 (1996).

Cooper, F., et al., "Non-Equilibrium Dynamics of Symmetry Breaking $\lambda\phi^4$ Field Theory," *Phys. Rev. D* **55**, 6471 (1997).

Dominquez, D., et al., "First-Order Melting of a Moving Vortex Lattice: Effects of Disorder" (to be published in *Phys. Rev. Lett.*).

Habib, S., et al., "Winding Transitions at Finite Energy and Temperature: An 0(3) Model," *Phys. Rev. D* **54**, 7774 (1996).

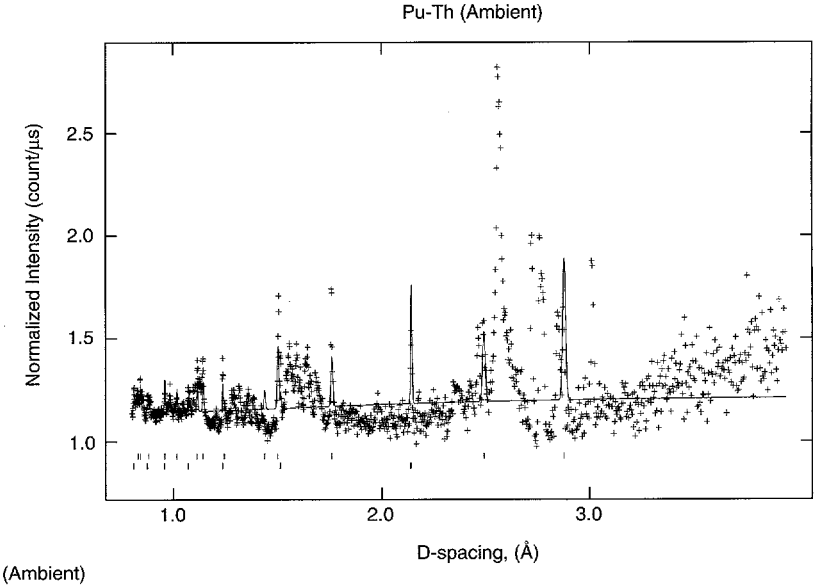
Zang, J., et al., "Nonequilibrium Transport and Population Inversion in Double Quantum Dot Systems" (to be published in *Mod. Phys. B*).

Actinide Crystal Structures with an Emphasis on Plutonium Alloys

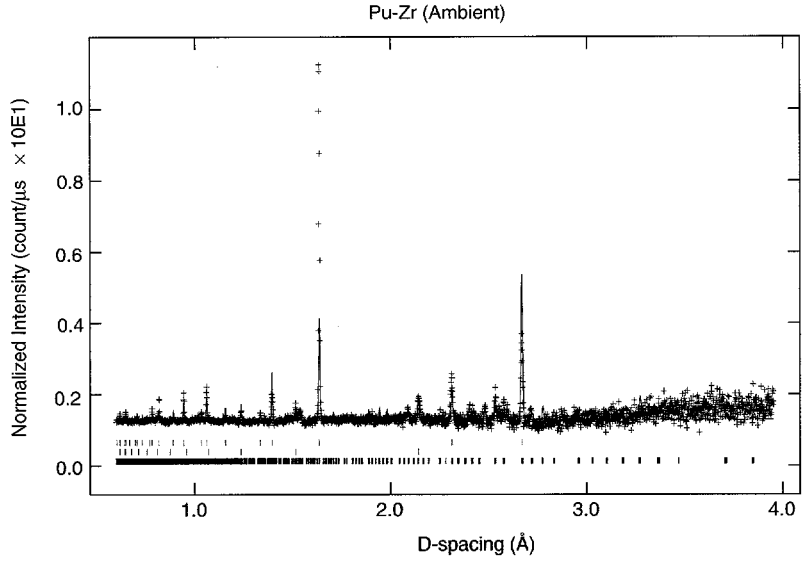
Joyce Roberts

The complexity of the crystal structure is the outstanding scientific problem of actinide crystal chemistry. We do not know why the idiosyncratic structures of plutonium metal occur, and we do not know how they transform among themselves. The electronic instability of the 5f electronic shell that is just beginning to be filled in the light actinides causes the formation of "self-intermetallic" compounds (in which the same element is in more than one valence state or crystallographic site) in both plutonium metal and its alloys, and the formation of these compounds is responsible for the observed complexity of the structure and the phase behavior. Our objective is to contribute to this understanding by determining the crystal structures of several actinide alloys: Np-U, Pu-Th, and Pu-Zr.

During the past year we prepared two plutonium-alloy samples and obtained diffraction patterns for them on the neutron powder diffractometer at the Manuel Lujan Jr. Neutron Scattering Center. One sample was a Pu-Th alloy, prepared at the composition appropriate for zeta-phase Pu_2Th. The diffraction pattern (see first plot) does not support the orthorhombic structure described in the literature, but the presence of pure thorium in the pattern suggests that we should anneal the sample longer. The second sample was a Pu-Zr alloy corresponding to the composition Pu_4Zr for the theta phase in this alloy system. The phase diagram suggests that this alloy is a superstructure of delta-phase plutonium, and this notion is supported by the diffraction pattern (see second plot). However, the diffraction pattern is so complex that we have not yet been able to provide a satisfactory indexing.



Neutron diffraction pattern for a Pu-Th alloy. The pattern shows the presence of pure thorium; contamination from pure thorium prevents us from solving the structure of the Pu-Th alloy. The plus signs correspond to the data, and the solid line represents the calculated profile. Reflection markers, the vertical bars just below the data, are shown for vanadium (lower) and thorium metal (upper). The reflection markers indicate the allowed diffraction peaks for the atomic structure of the material.



Neutron diffraction pattern for a Pu-Zr alloy. This alloy is a superstructure of the face-centered-cubic delta-phase plutonium. The plus signs correspond to the data, and the solid line represents the calculated profile. Reflection markers are shown for the Pu-Zr alloy (lower), vanadium (middle), and delta-phase plutonium (upper).

Science of Polymer-Based Materials Aging

Douglas Hemphill

The key focus for this project is to develop a science-based methodology predicting aging processes of polymeric materials. The approach being taken is based on characterizing molecular response to aging drivers and extending those responses through various length scales, i.e., molecular to mesoscale to bulk material. Project activities are divided into two areas, modeling and experiment.

Modeling work this year has progressed through different length scales. At the atomic and molecular level, we have been modeling the effect of water and hydrogen bonding on polydimethyl siloxanes and Estane (see first figure) and have constructed a three-dimensional lattice, Monte Carlo polymer code based on bond fluctuation models. At the mesoscale, we have developed a model that describes Estane microphase island structure. We have completed preliminary evaluation of existing siloxane network models and viscoelastic response of networks to constant strain experiments. At the bulk-material scale, we have developed continuum models based on polyurethane microstructure considerations and evaluated the pressure-volume response and high shear rate response.

Experimental efforts this year have focused on evaluating appropriate experimental techniques and establishing base-line Estane structure. Characterization techniques evaluated include gel permeation chromatography, differential scanning calorimetry, thermal gravimetric analysis, dielectric relaxation analysis, Fourier transform infrared (FTIR) Raman spectroscopy, solution and solid-state nuclear magnetic resonance, small-angle x-ray diffraction, dynamic mechanical analysis, stress/strain response, small-angle neutron scattering, thermally stimulated luminesence, two-dimensional FTIR (see second figure), and plate impact. Additionally, we are exposing samples to accelerated aging environments including various temperatures, humidities, and atmospheres.

Publications

Bardenhagen, S.G., et al., "Viscoelastic Models for Explosive Binder Materials," in *Shock Compression of Condensed Matter* (American Physical Society, Amherst, MA, in press).

Chitanvis, S., "A Gauge Theory of Mesoscopic Structure in Diblock Copolymers" (submitted to *Phys. Lett. A*).

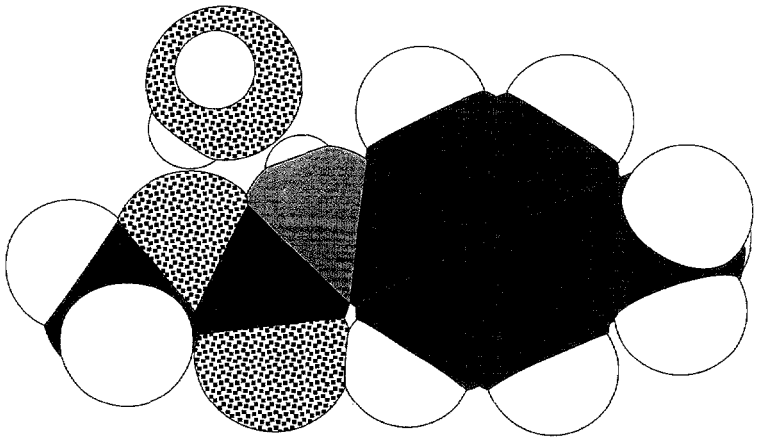
Chitanvis, S., "The Onset of Self Assembly" (submitted to *Phys. Rev. Lett.*).

Marsh, A., and J.R. Schoonover, "Infrared Spectroscopy of a Polyurethane Elastomer Under thermal Stress" (Eleventh International Conference on Fourier Transform Spectroscopy, Athens, GA, August 7–11, 1997).

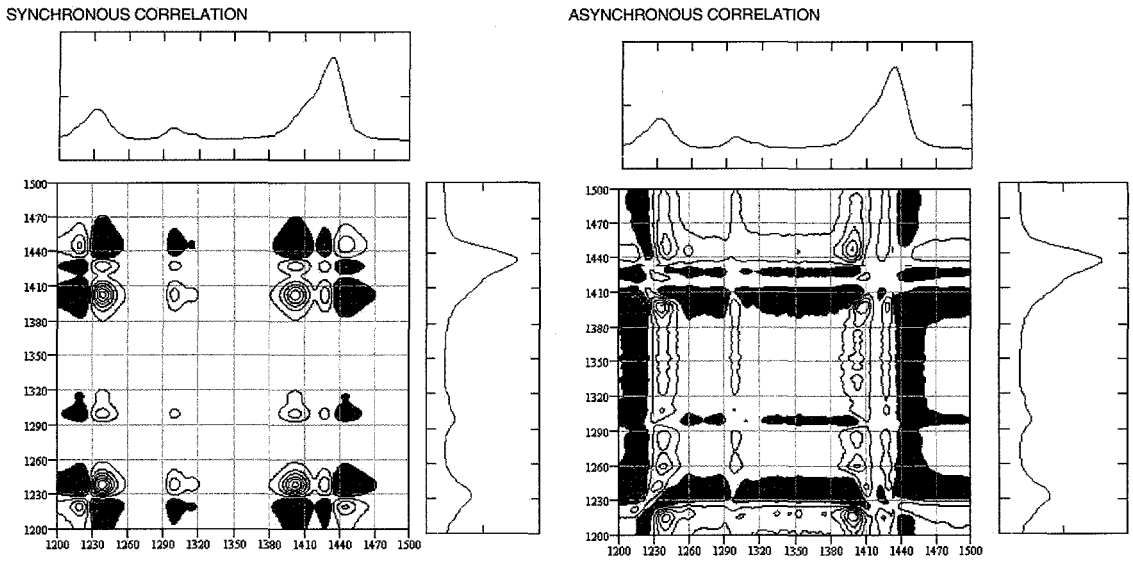
Palmer, R.A., et al., "Step-Scan FTIR Dynamic Polymer Opto-Rheology: Time vs. Frequency Domain" (Twenty-Fourth Annual Conference of the Federation of Analytical Chemistry and Spectroscopy Societies, Providence, RI, October 26–30, 1997).

Patterson, C.W., et al., "Conformational Analysis of the Crystal Structure for MDI/BDO Hard Segments of Polyurethane Elastomer" (submitted to *J. Polym. Sci., Part B: Polym. Phys.*).

Subia, S.R., et al., "Modeling of Concentrated Suspensions using a Continuum Constituitive Equation" (submitted to *J. Fluid Mech.*).



Molecular structure of water interaction with (N-p-tolyl)(O-methyl)urethane, a model compound for the Estane hard segment: C = dark gray, N = light gray, O = dotted, H = white, obtained with high-level ab initio calculations (DFT BLYP 6-311G**). The molecular vibrational spectra, and changes in the intensities and positions of bands that result from such interactions, are calculated and used in comparison with experimental spectra (such as the two-dimensional IR results) to deduce the molecular structure and interactions present in the pristine and aged polymer. These calculations also quantitate the potential energy functions that are used in the molecular dynamics and "coarse-grained" simulations that will model the changes in macroscopic properties.



The synchronous (real) and asynchronous (imaginary) components of the two-dimensional FTIR cross section of a thermally perturbed sample of Estane 5703. The dark contours are in the positive direction. The 1500 to 1800 wavenumber region is shown, and the correlation includes spectra taken at 16 temperatures between 30°C and 80°C. The two-dimensional correlation deconvolutes each of the broad peaks (shown above and on the right of each graph) into multiple spectral components as a function of their differing behaviors to the thermal perturbation.

Studies of Ultrahigh-Strength Materials

J. David Embury

The objective of this project is to explore both experimentally and theoretically the behavior of materials at stresses close to their theoretical strength. This effort involves the preparation of ultrafine-scale structures by a variety of fabrication methods.

In this past year our work has concentrated on wire drawing of in situ composites such as copper-silver and copper-niobium. We also fabricated materials by melting alloys in glass and drawing them into filaments at high temperatures using a method known as the Taylor wire technique. With this technique, we have drawn copper-silver microwires 10 μm in diameter that consist of nanoscale grains of a supersaturated solid solution.

To complement the experimental work, we have initiated theoretical studies to explore how the interfaces contribute to the production of dislocations in ultrafine-scale materials and how they control the distribution and scale of plastic flow events. In addition, we are studying the properties of structure with very high dislocation densities ($10^{17}/m^3$).

In the drawn in situ composites, we are able to achieve strengths of 1 to 1.5 GPa in materials that exhibit high ductilities, as evidenced by their reduction in area of 50% at failure. Also, by deforming copper single crystals and polycrystals at 4.2 K, we are able to achieve strength levels of about one-third of the theoretical strength because of the storage of very high densities of dislocations. In the two-phase materials, strengthening occurs by the production of very large areas of interphase interfaces and by the storage of dislocations at these interfaces.

Publications

Embury, J.D., "The Plasticity of Embedded Phases" (6th Int. Symp. on Plasticity, Juneau, AL, June 6, 1997).

Han, K., et al., "The Fabrication and Properties of Composite Conductors" (Conf. of Composites, Lake Louise, Canada, Oct. 8, 1997).

Wood, J.T., et al., "An Approach to Materials Processing and Selection for High-Field Magnet Design," *Acta Mater.* **45**, 1099 (1997).

Electrons in High Magnetic Fields—Ground-State Electronic Structure of 5f Materials

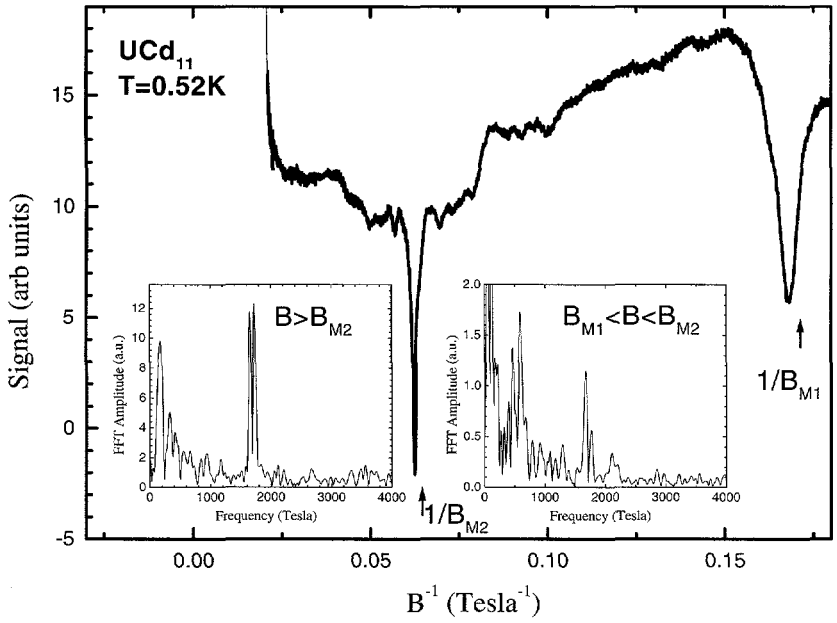
Aloysius Arko

The goal of this project is to determine the electronic structures of 5f elements and compounds having highly correlated electrons. We are particularly interested in measurements on delta-phase plutonium (δ-Pu, a crystalline form of plutonium) using de Haas-van Alphen (dHvA) and specific heat (C_p) measurements in the 50- and 60-T pulsed magnets at the Los Alamos National High Magnetic Field Laboratory.

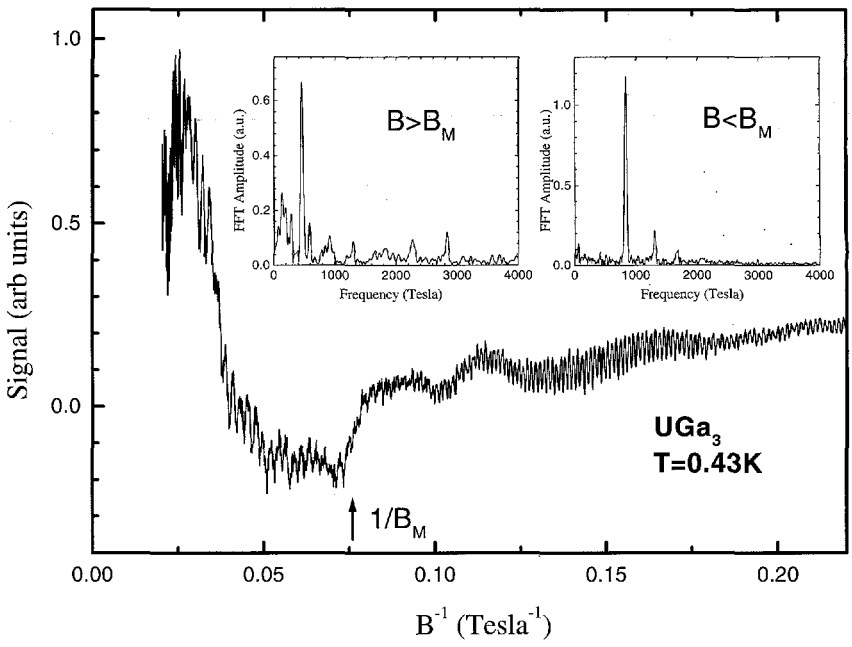
Measuring oscillatory magnetization, i.e., the dHvA effect, yields both the Fermi surface of the material as well as the orbital effective mass of the electrons. These two items provide extensive information on the electronic structure of the material. We compare the effective mass in high fields obtained from a dHvA measurement, which is related to the strength of the correlations, with the mass obtained by measuring the specific heat in high fields.

To date we have obtained dHvA data on several crystals of uranium compounds whose C_p γ-values range from ~20 (for USb_2) to more than 800 (for UCd_{11}). (The γ-value is a linear term in C_p that is related to the electron mass.) The data shown in the figures display a richness of magnetic behavior and suggest that dHvA data can readily be obtained for δ-Pu, whose g-value is only ~50. The highest effective mass seen thus far is 15 m_0 in USn_3, where m_0 is the rest mass of the electron, about 3 times higher than what we expect for δ-Pu. Metamagnetic transitions are common in highly correlated compounds and often completely restructure the Fermi surface (e.g., as seen in UGa_3).

We have a nearly completed system for making dHvA measurements in plutonium. The system consists of a rotating dHvA coil, a He^3 pumping station to reach low specimen temperatures, and a plastic cryostat which contains the sample and the He^3. Compliance with environmental, safety, and health requirements is also nearly complete and appears to present no problem. The experimental design for C_p measurements in pulsed fields is complete, and we have already tested the temperature-measurement apparatus in pulsed fields. In these tests we saw no heating or magnetoresistive effects that could affect the temperature measurements.



The output voltage of a dHvA pickup coil containing the heavy fermion crystal UCd_{11} as the magnet was pulsed to 50 T. (A heavy fermion material is one whose conduction electrons behave as though they have extremely high effective mass.) The data are plotted in inverse magnetic field and clearly show small dHvA magnetization oscillations as well as large spikes corresponding to two metamagnetic transitions (B_{M1} and B_{M2}). The insets are Fourier transforms of the data above and below B_{M2} and show no change in the large frequency spike at 1700 T. (Since dHvA oscillations are periodic in inverse magnetic field, the cyclotron frequency is expressed in units of magnetic field.) The transitions (arrows) likely correspond only to spin reorientation.



The output of a pickup coil containing UGa_3 during a magnet pulse. The smaller effective masses in UGa_3 yield much stronger dHvA oscillations than the those of the heavy fermion UCd_{11}. The arrow points to the metamagnetic transition at B_M, with oscillation frequencies clearly changing at B_M. The insets are Fourier transforms of the data above and below B_M and show a complete change in the frequency spectrum. In all likelihood the magnetic state changes from antiferromagnetic to paramagnetic at B_M, thus completely restructuring the Fermi surface.

Dynamic Fracture of Heterogeneous Materials

Michael Stout

The objective of this project was to investigate the fundamental aspects of the process of dynamic fracture propagation in heterogeneous materials. Our work focused on three important, but poorly understood, aspects of dynamic fracture in materials with a heterogeneous microstructure: (1) the appropriateness of using a single-parameter asymptotic analysis to describe dynamic crack-tip deformation fields; (2) temperature rises at the tip and on the flanks of a running crack; (3) and the constitutive modeling of damage initiation and accumulation.

Last year, we characterized experimentally the mechanical properties of the PBS-9501 mock-simulant sugar material and the PBX-9501 high explosive. We measured the fracture behavior and toughness of the PBS-9501 mock-simulant sugar material. We used the speckle photograph technique to measure directly the strain field evolution during dynamic impact of the PBX-9501 high-explosive material. We developed a physically based, micromechanical constitutive model that describes the thermomechanical behavior of the heterogeneous materials. We implemented the theoretical model into an explicit finite-element code to simulate the experiments. We will also use this code as a design tool to simulate the process of dynamic fracture.

Publications

Asay, B.W., et al., "Speckle Photography During Dynamic Impact of an Energetic Material Using Laser Induced Fluorescence," *J. Appl. Phys.* **82**, 1093 (1997).

Bennett, J.G., and K.S. Haberman, "A Constitutive Model for the Non-Shock Ignition and Mechanical Response of High Explosives (PBX-9501)" (to be published in *J. Mech. Phys. Solids*).

Haberman, K.S., et al., "Modeling, Simulation, and Experimental Verification of Constitutive Models for Energetic Materials," in *Shock Compression of Condensed Matter* (American Physical Society, Amherst, MA, in press).

Henson, B.F., et al., "Surface Temperature Measurements of Heterogeneous Explosives by IR Emission," in *Shock Compression of Condensed Matter* (American Physical Society, Amherst, MA, in press).

Williams, T.O., and J. Aboudi, "A Generalized Micromechanical Model With Shear-Coupling" (submitted to *Int. J. Solids Struc.*)

Tailoring the Interfacial Electronic Structure of Organic Electronic Materials and Devices

Ian Campbell

We are using self-assembled molecular coupling layers to tailor the electronic structure of metal/organic interfaces and organic/organic heterojunctions in order to control the electrical transport properties of these interfaces. Metal/organic interfaces are the essential active element in every organic electronic device, and organic/organic heterojunctions are the key element in efficient, organic light-emitting diodes. Because of their processing advantages, the tunability of their electronic properties, and their flexibility in both materials and device design, organic electronic materials and devices are poised to revolutionize major technological areas such as information display and optical communication. Learning to manipulate the electrical transport properties of these interfaces is the key to realizing the potential of organic electronics.

This year we used self-assembly techniques to demonstrate control and improvement of charge injection in organic electronic devices by manipulating the Schottky energy barrier between a metal electrode and the organic electronic material. We varied hole injection from copper electrodes into the electroluminescent conjugated polymer MEH-PPV (a derivative of poly[phenylene vinylene]) by using two conjugated-thiol-based self-assembled monolayers. We incorporated the chemically modified electrodes in organic diode structures and measured changes in the metal/polymer Schottky energy barriers and current-voltage characteristics. We developed a theoretical model that describes tunneling through disordered organic materials with strong electron-lattice coupling.

Publications

Campbell, I.H., et al., "Controlling Charge Injection in Organic Electronic Devices Using Self-Assembled Monolayers" (to be published in *Appl. Phys. Lett.*).

Campbell, I.H., et al., "Controlling Schottky Energy Barriers in Organic Electronic Devices Using Chemically Tailored Electrodes," *Phys. Rev. B* **54**, 14321 (1996).

Davids, P.S., et al., "Charge Injection in Organic Light-Emitting Diodes: Tunneling into Low Mobility Materials," *Appl. Phys. Lett.* **69**, 2270 (1996).

Heller, C.M., et al., "Chemical Potential Pinning Due to Equilibrium Electron Transfer at Metal/C-60-Doped Polymer Interfaces," *J. Appl. Phys.* **81**, 3227 (1997).

Smith, D.L., "Steady State Model for Polymer Light-Emitting Electrochemical Cells," *J. Appl. Phys.* **81**, 2869 (1997).

Smith, D.L., et al., "Theory of Ballistic Electron Emission Microscopy for Nonepitaxial Metal/Semiconductor Interfaces" (submitted to *Phys. Rev. Lett.*).

Yu, Z.G., et al., "Effects of Lattice Fluctuations on Electronic Transmission in Metal/Conjugated-Oligomer/Metal Structures," *Phys. Rev. B* **56**, 6494 (1997).

Yu, Z.G., et al., "Electronic Transmission in Conjugated-Oligomer Tunnel Structures: Effects of Lattice Fluctuations" (submitted to *J. Phys.: Condens. Matter*).

Science and Technology of Reduced-Dimensional Magnetic Materials

Robert Heffner

Our work involved synthesizing single-crystal and thin-film samples of magnetoresistive manganites ($LaMnO_3$ doped with calcium and strontium) and characterizing their electronic transport properties. Our objective is to understand the underlying physical mechanisms responsible for the colossal magnetoresistance (CMR) of these materials. We supplemented the experimental work with an effort to develop microscopic mathematical models of the observed phenomena.

In the experimental work, we succeeded in finding an important relation between the magnetization and resistivity in the materials, helping to explain the importance, in the CMR phenomena, of lattice distortions that are accompanied by clusters of ferromagnetic spins (called spin-lattice polarons). In addition, we developed materials with rudimentary tunnel junctions consisting of multilayers of an insulator sandwiched between CMR materials. These materials will have possible applications as magnetic sensors.

In the theoretical realm, using a combination of exact-diagonalization and series-expansion analytical techniques, we predicted novel spin textures that should induce anomalous, persistent currents in applied magnetic fields. We also found a strong tendency for small polaron

formation, which is consistent with a large body of experimental data, including local structural and magnetic probes, very large isotope shifts, and thermodynamic, transport, and susceptibility measurements. Finally, we have begun modeling polaron and spin-coupling crossover effects in layered magnetoresistive perovskites, as well as the mechanisms for ferromagnetic coupling in the magnetoresistive pyrochlores.

Publications

Hawley, M.E., et al., "CMR Films Structure as a Function of Growth and Processing," *J. Cryst. Growth* **174**, 455 (1997).

Hundley, M.F., et al., "Interplay between Electronic Transport and Magnetic Order in Ferromagnetic Magnetic Manganite Thin Films," *Mat. Res. Soc. Symp. Proc.* **474**, 167 (1997).

Hundley, M.F., et al., "Thermoelectric Power of $La_{1-x}Ca_xMnO_{3+d}$: Inadequacy of the Nominal $Mn^{3+/4+}$ Valence Approach," *Phys. Rev. B* **55**, 11511 (1997).

Kwon, C., et al., "Large Magnetoresistance in $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3/La_{0.7}Sr_{0.3}MnO_3$ Ramp-Edge Junctions" (to be published in *Appl. Phys. Lett.*).

Li, Q., et al., "Charge Localization in Disordered Colossal-Magnetoresistance Manganites" (to be published in *Phys. Rev. B*).

Neumeier, J.J., et al., "Volume-Based Thermodynamic Considerations for the Magnetoresistance Effect and Metal-Insulator Transition of CMR Manganese Oxide Perovskites" (submitted to *Phys. Rev. B*).

Zang, J., et al., "Magnetic Properties of the Double-Exchange Model," *J. Phys. Condens. Matt.* **9**, 157 (1997).

Zang, J., et al., "Persistent Current in the Ferromagnetic Kondo Lattice Model" (to be published in *Phys. Rev. B*).

Chemistry and Microstructure of High-Temperature Superconductor Interfaces

Quanxi Jia

Superconductor /normal-metal/ superconductor (SNS) junctions are a very promising technology for the fabrication of high-performance superconducting quantum interference devices (SQUIDs). This technology provides a desirable alternative to bicrystals because it costs less, it is highly flexible, and it is easily integrated into SQUID systems. Most significant, when SNS junctions are used, external magnetic fields of up to about 1G do not influence the noise of the SQUIDs. It is speculated that the junctions are intrinsically shielded from external magnetic fields by the Meissner effect in the top electrode. During this year, our research objectives have been to investigate the interface chemistry, physics, and microstructure of superconductor and normal-metal composites for the development of ultrasensitive SQUID magnetometers.

We have successfully demonstrated, for the first time, directly coupled SQUID magnetometers based on a ramp-edge SNS technology. Processing controllability and reproducibility improved substantially when silver-doped YBCO was used for the electrodes. The device routinely displayed I_cR_n products of over 120 μV and a transfer function of over 100 $\mu V/\Phi_0$ (where $\Phi_0 = 2.07 \times 10^{-15}$ Wb). We have fabricated directly coupled SQUID magnetometers with an I_c of 17 μA and an R_n of 4.4 Ω at 75 K for each junction. The field sensitivity of the magnetometers increased with increasing pickup loop area. We demonstrated directly coupled SQUID magnetometers on $LaAlO_3$ substrates. The magnetometers had a field sensitivity of around 400 $fTHz^{-1/2}$ at kilohertz frequencies for a pickup loop area of 8.5 mm × 7.5 mm. Our experimental results also show that edge-geometry SNS junctions using silver-doped YBCO as electrodes are stable in air and have potential applications in fields such as low-frequency radio waves.

Publications

Jia, Q.X., et al., "Characterization of Ramp Edge Geometry SNS Junctions and dc SQUIDs," *IEEE Trans. Appl. Supercond.* **7**, 3005 (1997).

Jia, Q.X., et al., "Stability of dc SQUIDs Fabricated Using Ramp Edge SNS Technology," *Appl. Phys. Lett.* **71**, 1721 (1997).

Zhou, J., et al., "Improved N-Layer Materials for High-T_c SNS Junctions and SQUID Sensors" (to be published in *Appl. Phys. Lett.*).

X-Ray and Neutron Characterization of Transition Metal Oxides

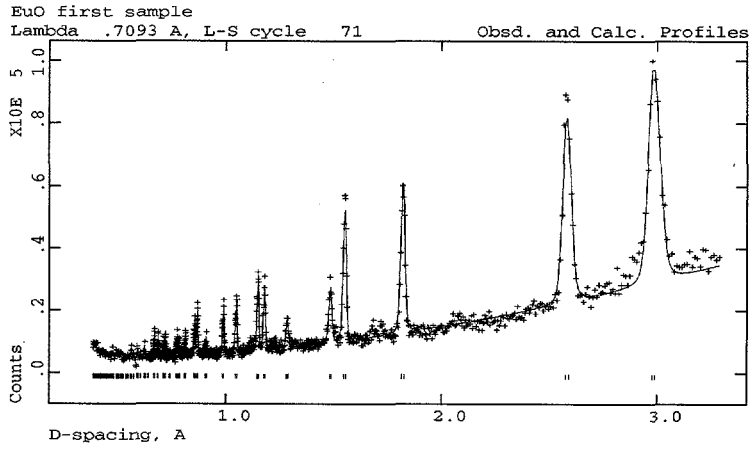
Michael Fitzsimmons

A key issue in the colossal magnetoresistance (CMR) of compounds containing manganese is the relative importance of the electronic exchange versus local lattice distortions in determining the coupled magnetic and transport properties. For example, charge exchange between Mn^{+3} and Mn^{+4} in some magnetite compounds is affected by the magnetic environment of the material. This behavior is a consequence of the multivalent electronic structure of manganese. Other elements, in particular europium, are also multivalent. We explore the magnetic and transport properties and the detailed atomic structures of europium-containing materials which might also exhibit CMR behavior or induce magnetism in otherwise nonmagnetic materials, for example, lead.

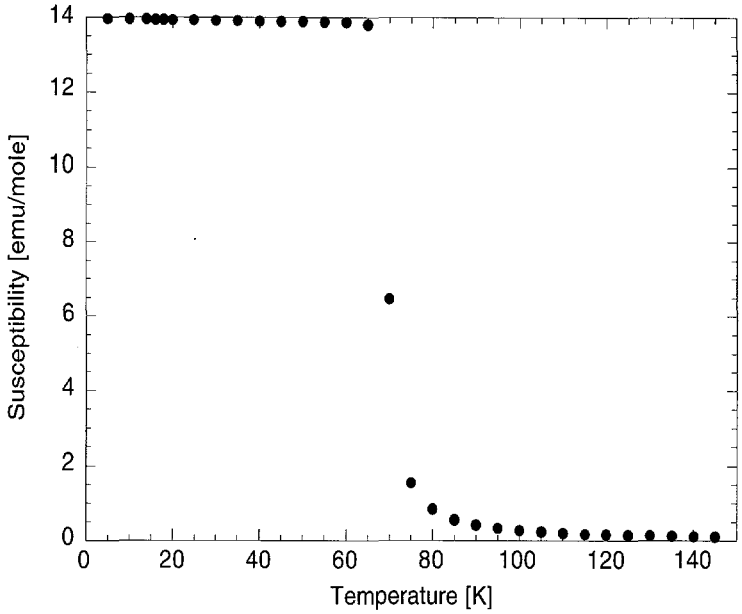
This year, we fabricated about 8 g of europium mono-oxide (EuO) by heating a mixture of Eu_2O_3 and europium to 1800 K inside an argon-filled tantalum casing. X-ray diffraction (first graphic) of the material confirmed it to be virtually single-phase EuO. Magnetometry showed the material to have magnetization and a Curie temperature consistent with EuO (second graphic).

In addition, we made films of NiMnSb sputtered from a composite target onto heated (150°C to 300°C) Si(100) and MgO(100) substrates. The crystal structure was characterized by high-angle x-ray diffraction, and polycrystalline samples with the structure C1b were found for the higher deposition temperatures. Magnetization measurements confirmed ferromagnetic layers with a strong dependence between the coercive field strength and deposition temperature. The saturation moment was also affected by deposition temperature, and in all cases was less than 2.5 μB per manganese site.

Recently, we developed a deposition procedure that produced NiMnSb films with coercivity of less than 3 Oe (third graphic)—one requirement for use in a magnetic recording read head.



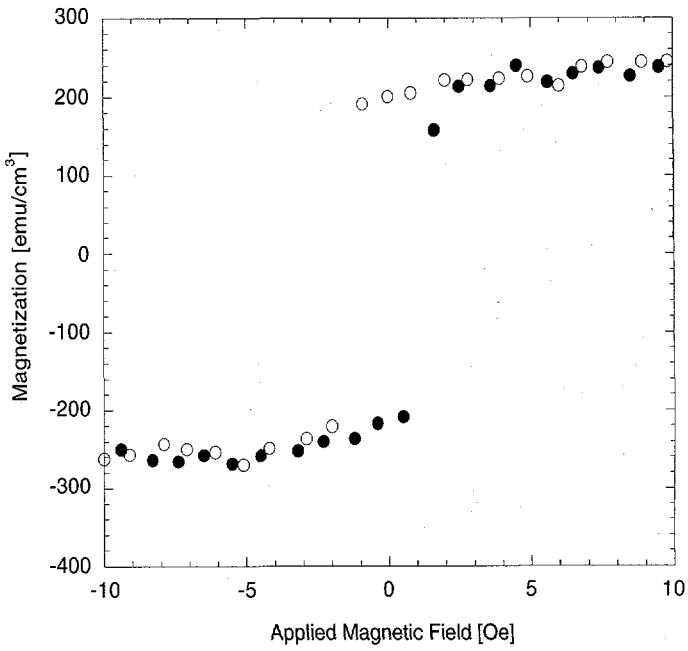
X-ray diffraction pattern of EuO fabricated for this project.



Magnetization of the EuO sample.

Publications

Schlomka, J-P. F., et al., "Structural and Magnetic Properties of Ion Beam Sputtered NiMnSb Films" (to be published in *Physica B*).



Magnetization of a thin NiMnSb film exhibiting low coercivity.

High-Performance Computing of Electron Microstructures

Alan Bishop

The purpose of this project, a research collaboration with scientists at the University of California at Santa Barbara (UCSB), is to model novel quantum properties of optically driven nanostructured (quantum-well) electronic materials. These materials are fabricated and measured at UCSB by using dedicated molecular-beam epitaxy and free-electron laser facilities. We developed a new time-dependent density-functional model that is being implemented with large-scale parallel-processing computation and that can systematically handle all the relevant physical interactions controlling nonequilibrium properties.

This project takes excellent advantage of the Laboratory's core competency in theory, modeling, and high-performance computing. It is relevant to programs in nanotechnology; semiconductor modeling; information and sensor technology; and multiscale, nonlinear, and nonequilibrium processes.

This year we extended the time-dependent density-functional approach to include a density matrix formalism that allows for energy relaxation and multiple quantum-level processes. This nonperturbative, dynamic quantum theory for periodically driven systems can be used to accurately predict dynamic energy spectra (including sidebands), the distribution of wave function amplitudes for composite electron and photon systems, and both resonant and off-resonant processes.

Several predictions for experimental investigations are clear and are ongoing with UCSB colleagues. Those investigations include a photoluminescence study in which a free-electron laser drives a two-dimensional sample. A pumping laser populates the levels, and measurements of the quasi-energy peaks with predicted sideband structures are made. Another involves an excitonic effect below the band edge in a superlattice. USCB experiments confirm the predicted excitonic resonance peaks with sidebands.

Publications

Birnir, B., and H.A. Hauksson, "A Finite Dimensional Attractor of the Moore-Greitzer PDE Model" (to be published in *SIAM J. Appl. Math.*).

Birnir, B., and J. Xin, "The Global Attractor of the Transverse Maxwell-Bloch Equations" (to be published in *Physica D*).

Salkola, M., et al., "Coupled Spin-Boson Systems Far from Equilibrium," *Phys. Rev. B* **R54**, 12645 (1996).

Wang, L., et al., "Time-Periodic Quantum Theory: Interpretation of Quasi-Energies" (to be printed in *Phys. Rev. B*).

Zang, J., et al., "Nonequilibrium Transport and Population Inversion in Double Quantum Dot Systems," *Int. J. Mod. Phys.* **11**, 1463 (1997).

Dynamic, High-Strain Deformation of Metals: Experiments, Advanced Constitutive Modeling, and Computational Implementation

Dean Preston

Science-based stockpile stewardship requires the capability to predict the implosion dynamics of system components in which the materials are under extreme conditions (large strain, high strain rates, and high temperatures) and for which a precise knowledge of those conditions is critically important. This project is a coordinated experimental, mathematical-modeling, and code-implementation effort aimed at (1) improving the calibration of the mechanical threshold stress (MTS) and Preston-Tonks-Wallace (PTW) strength models using new Hopkinson bar data at elevated temperatures and at very large strains, (2) extending the models to include texture evolution, and (3) employing the methods of molecular dynamics (MD) to improve the models at intermediate strain rates.

Our major accomplishments reflect the coordinated experimental, mathematical-modeling, and code-implementation nature of the project. We performed MD simulations to study short-distance dislocation dynamics; to date we have investigated the intersection process for dislocations in face-centered cubic metals (see accompanying three-dimensional representation). We also successfully applied the Tonks ductile damage model to spallation data of two grades of tantalum. We measured and analyzed the texture and constitutive-behavior evolution resulting from upset forging in 304L stainless-steel bar stock (see plot), and we used these data as the basis for fitting the large-strain response to the PTW and MTS models.

In addition, we studied the transition of the body-centered cubic lattice to a hexagonal closest-packed lattice in iron. We are also applying a three-dimensional, physically based material model of constitutive strength to various high-rate deformations of spherical shells. This model, which is appropriate for metallic anisotropic elastoplasticity, accounts for the directional propagation of stress waves, the shape and evolution of the yield surface, and the hardening. We are validating the modeling by geometrically comparing the model to Taylor impact specimens or quasi-static compression specimens.

Publications

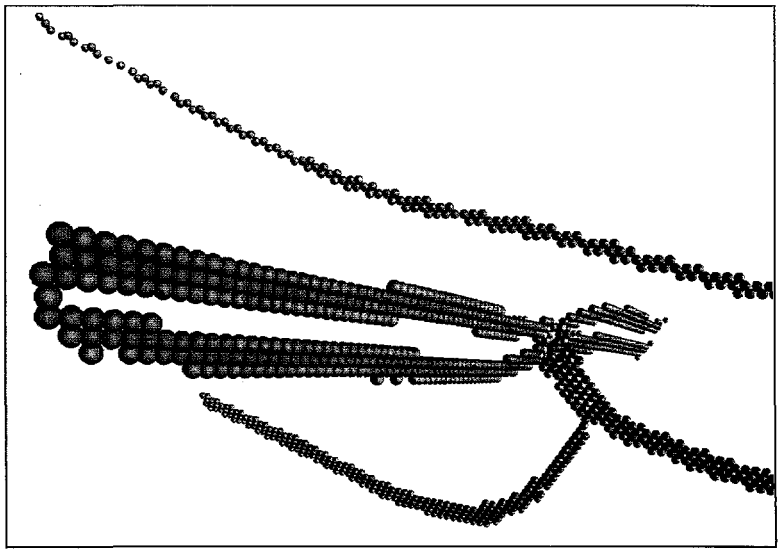
Chen, S.R., and G.T. Gray III, "Constitutive Behavior of Tantalum and Tantalum-Tungsten Alloys," *Metall. Mater. Trans. A* **27**, 2994 (1996).

Chen, S.R., and G.T. Gray III, "Influence of Twinning on the Constitutive Response of Zr: Experiments and Modeling," *J. Phys. IV* **7**, 523 (1997).

Gray, G.T. III, "Influence of Strain Rate and Temperature on the Structure/Property Behavior of High-Purity Titanium," *J. Phys. IV* **7**, 423 (1997).

Maudlin, P.J., et al., "On the Modeling of the Taylor Cylinder Impact Test for Orthotropic Textured Materials," in *Proceedings of Conference Plasticity '97: The 6th International Symposium on Plasticity and Its Current Applications*, A.S. Khan, Ed. (Neat Press, Fulton, MD, 1997), p. 125.

Maudlin, P.J., et al., "On the Modeling of the Taylor Cylinder Impact Test for Orthotropic Textured Materials: Calculations and Experiments" (submitted to *Int. J. Plasticity*).

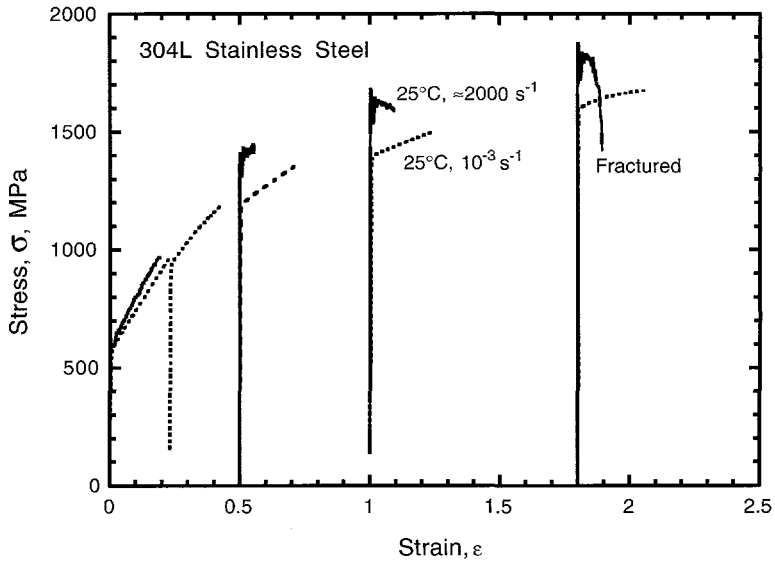


A three-dimensional MD simulation of the perpendicular intersection of extended dislocations in copper. The only atoms displayed are those that are in the dislocation cores and that have potential energies higher than the energy of the bulk. The edge partial of the 60° extended dislocation has moved down and passed the stationary screw dislocation, and the 30° partial is about to pass. The cubic computational cell contains 3.5 million atoms interacting via an embedded-atom-model potential.

Tonks, D.L., et al., "Quantitative Analysis of Damage Clustering and Void Linking for Spallation in Tantalum," *J. Phys. IV* **7**, 841 (1997).

Tonks, D.L., et al., "Spallation Studies on Shock-Loaded Uranium," in *Proceedings of Shock Compression of Condensed Matter 1997* (American Institute of Physics Press, Woodbury, NY, in press).

Zhou, S.J., et al., "Large-Scale Molecular Dynamics Simulations of Dislocation Intersections in Copper" (submitted to *Science*).



Shown here are our measurements, as a function of temperature and strain rate, of the stress-strain response of 304L stainless steel following upset forging to true strains of approximately 2. We are reloading the prestrained samples over a wide range of temperatures and strain rates in order to fit the large-strain response to the PTW and MTS constitutive models.

Neutron Scattering Studies on Shear-Induced Structure in Polymers

Rex Hjelm

Shear is introduced in industrial processing when a polymer melt or concentrated solution is passed though an orifice such as a spinner used in fiber formation, when it is injected into a form for molded parts, or when it is spread for a surface coating. This naturally introduced flow shear affects the conformation and orientation of polymer chains. We need a detailed understanding of the physics of polymers that are under shear strain in order to connect the structure of polymers with their unique rheological properties; currently, we have little basis from which to predict the effect of a processing strategy on the material properties. Our objective is to study polymer-surface interactions and shear-induced polymer structure in bulk solutions. These aspects include self-lubrication effects in polymers under flow- and shear-induced disentanglement and conformational and phase-transition effects in bulk solutions.

This year we finished the construction of thermostatically controlled Couette-geometry shear cells for the study of high-temperature polymer melts, completed a cone and plate rheometer for neutron reflectometry experiments, and successfully performed proof-of-principle experiments for both the shear apparatuses. We performed the experiments on concentrated solutions of polystyrene of different molecular weights in good solvents, using small-angle neutron scattering to study the bulk-solution structure and using neutron reflectometry to study the near-surface structure under shear strain. In addition, we successfully demonstrated that triblock copolymers, which form micelles in water, undergo significant changes in structure under shear strain. We are analyzing the results of these measurements.

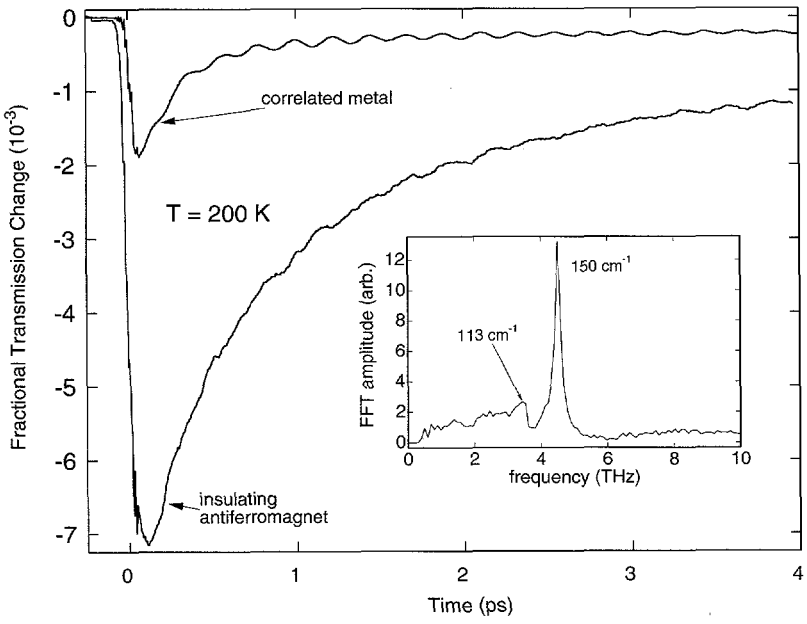
Electrons in High Magnetic Fields—Femtosecond Dynamics

Timothy R. Gosnell

The objective of this project is to apply ultrafast optical techniques to the measurement of nonequilibrium properties of correlated d and f electrons in condensed matter. Such experiments will allow the investigation of microscopic dynamical phenomena that occur on time scales of 10^{-11} to 10^{-14} s. The purpose of these experiments is to reveal in unique ways the role of many-body interactions in d- and f-electron systems. A magnetic field will be used as an independent control variable to tune the energy scale of the many-body phenomena under study. These ultrafast dynamics measurements will be combined with original theoretical research on ultrafast many-body physics that directly supports the experiments.

This year the following results were obtained in zero magnetic field:

- Using time-resolved far-infrared transmission measurements, we discovered oscillatory ultrafast dynamics revealed in transient carrier conductivity of a photo-excited insulating antiferromagnet.
- Using transient optical transmission measurements, we observed systematic variations in charge carrier cooling dynamics attributable to varying many-body interactions in an insulating antiferromagnet, a correlated metal, and a superconductor (see accompanying figures). All samples exhibited electron-phonon coupling as revealed in the modulation of the sample dielectric constant by optic phonons.
- The theory of dynamic excitation of coherent phonons was developed. We discovered the first example of transient far-infrared emission from correlated electron materials and identified second-order, electric-quadrupole optical nonlinearity as the emission mechanism.



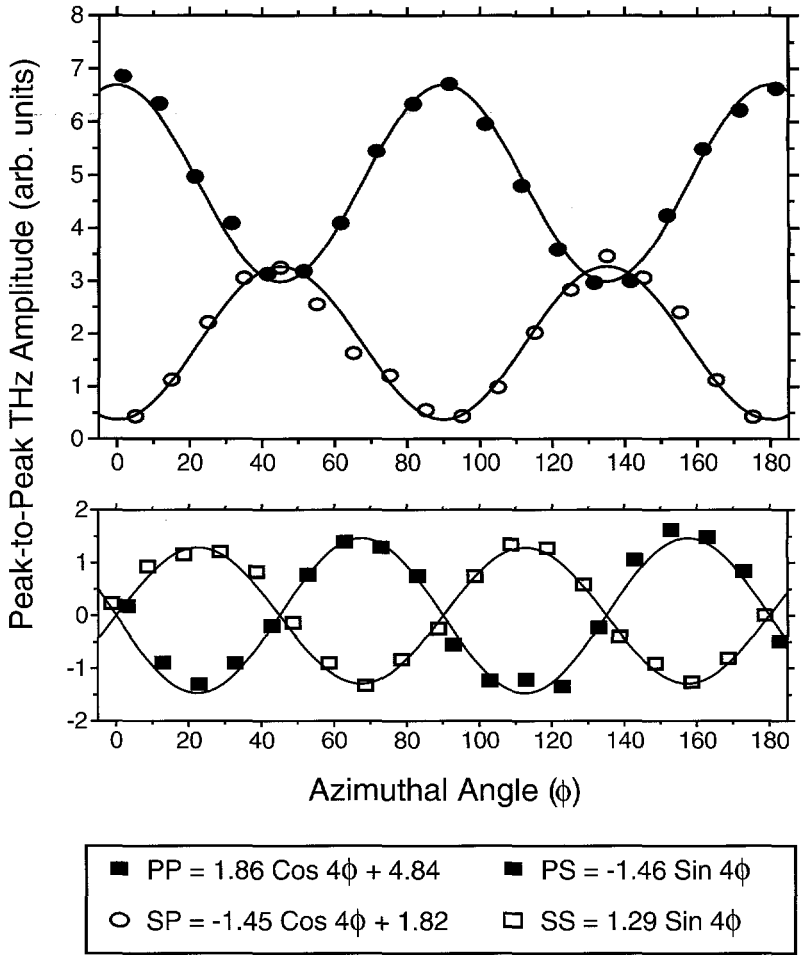
Absorption of a 30-fs pulse by a correlated metal and an insulating antiferromagnet creates hot carriers that cool by carrier-phonon and carrier-carrier interactions, as revealed in this transient transmission data. Differences in many-body interactions yield qualitatively different cooling dynamics for the two materials. For both, strong electron-phonon coupling gives rise to coherent phonon oscillations as reflected in the signal modulation shown in the data. In the inset, a Fourier transform of the data reveals frequencies of the excited optical phonons.

Publications

Bonca, J., and S.A. Trugman, "Inelastic Quantum Transport" (submitted to *Phys. Rev. Lett.*).

Siders, J.L.W., et al., "Terahertz Emission From YBa2Cu3O(7-x)" (submitted to *Phys. Rev. Lett.*).

Siders, J.L.W., et al., "Terahertz Emission From YBa2Cu3O(7-x) Thin Films," *OSA Trends in Optics and Photonics: Ultrafast Electronics and Optoelectronics* **14**, 49 (1997).



Amplitude of single-cycle terahertz emission from a thin film of $YBa_2Cu_3O_7$ excited by a 100-fs, 800-nm pulse as a function of the azimuthal angle j in the a-b plane. The fourfold symmetry of the emission is indicative of a second-order, electric-quadrupole optical nonlinearity.

Development of Ion-Beam Techniques for the Study of Special Nuclear Materials–Related Problems

Carl Maggiore

The scientific objective of this project was to develop the ion-beam techniques for the characterization of actinides and their effects on other materials. It was designed to enhance our ability to quantitatively understand the oxidation, corrosion, diffusion, stability, and radiation damage of actinides and the materials with which they are in contact.

We developed and applied several low-energy nuclear techniques (resonant and nonresonant backscattering, nuclear reaction analysis, and particle-induced x-ray emission) to the quantitative study of the near surfaces of actinide and tritide materials and determined the absolute accuracy and precision of ion-beam measurements on these materials. We also demonstrated the use of variable-energy alpha beams for the study of accelerated aging of polymeric materials in contact with actinide materials.

Publications

Doyle, B.L., et al., Eds., "Nuclear Microprobe Technology and Applications," in *Proc. Fifth Int. Conf. Nucl. Microprobe Tech. Appl.* (North-Holland, New York, NY, 1997).

Lucca, D.A., and C.J. Maggiore, "Subsurface Lattice Disorder in Polished II–VI Semiconductors," *Ann. CIRP* **46**, 485 (1997).

Lucca, D.A., et al., "Subsurface Damage Distribution in Ultraprecision Machined CdS," *Ann. CIRP* **45**, 1 (1996).

McIntyre, P.C., et al., "Oxidation Kinetics of TiN Layers: Exposed and Beneath Pt Thin Films," *Appl. Phys. Lett.* **70**, 6 (1997).

Synthesis and Characterization of Correlated-Electron Materials

Joe D. Thompson

With discoveries—heavy fermions, conducting polymers, fullerenes, and colossal magnetoresistive materials—beyond traditional solid-state physics, we are at the beginning of a golden age for complex electronic materials. These new materials are characterized by strong electronic correlations that arise from competitions between microscopic electronic, magnetic, and structural degrees of freedom.

Our objective in this project is to understand the competition and interplay between these three degrees of freedom in correlated-electron materials in order to provide the underpinning science for key next-generation civilian and defense technologies. Our attention focuses on three important science issues: (1) How does this competition control the metal-insulator transition in correlated-electron materials? (2) What conditions induce non-Fermi-liquid behavior in these systems? (3) How can the local structure be probed?

This past year we established that inhomogeneous spin fluctuations are intrinsic to hole-doped lanthanum-manganite materials and are not due to chemical clustering. From a pair-distribution-function analysis of neutron diffraction data, we showed that local structural changes in manganites are identical, irrespective of whether the metal-insulator boundary is crossed by changing temperature or by cation doping. We performed a comprehensive neutron diffraction study of the entire calcium-doped manganite series. Using resonant ultrasound spectroscopy, we observed a signature of local-structure/polaron formation.

We also demonstrated the importance of "hybridization disorder" for producing non-Fermi-liquid behavior in some correlated-electron metals. This work was closely coordinated with a companion theoretical project to close the loop on synthesis characterization modeling. Lastly, we hosted an international workshop titled "Spin-Charge-Lattice Coupling in Complex Electronic Materials."

Publications

Bernal, O.O., et al., "μSR Studies of Kondo Disorder in $UCu_{5-x}Pd_x$ and $CeCu_{5.9}Au_{0.1}$," *Physica B* **230**, 606 (1997).

Booth, C.H., et al., "Direct Relationship between Magnetism and MnO_6 Distortions in $La_{1-x}Ca_xMnO_3$" (submitted to *Phys. Rev. Lett.*).

Booth, C.H., et al., "Lattice Effects in in $La_{1-x}Ca_xMnO_3$ (x = 0–1): Relationship between Distortions, Charge Distribution, and Magnetism" (submitted to *Phys. Rev. B*).

Darling, T.W., et al., "Measurement of the Elastic Tensor of a Single Crystal of $La_{0.83}Sr_{0.17}MnO_3$ and Its Response to Magnetic Fields" (submitted to *Phys. Rev. B*).

DiFrancesco, R.G., et al., "Local Structure and Polaron Formation in $La_{1-x}Ca_xMnO_3$" (to be published in *Physica B*).

Feyerherm, R., et al., "Competition between Magnetism and Superconductivity in $CeCu_2Si_2$," *Phys. Rev. B* **56**, 699 (1997).

Graf, T., et al., "Comparison of $CeRh_2Si_2$ and $CeRh_{2-x}Ru_xSi_2$ near Their T = 0 Magnetic-Non-Magnetic Boundaries," *Phys. Rev. Lett.* **78**, 3769 (1997).

Hawley, M.E., et al., "CMR Films Structure as a Function of Growth and Processing," *J. Cryst. Growth* **174**, 455 (1997).

Heffner, R.H., et al., "Unusual Relaxational Dynamics in $La_{1-x}Ca_xMnO_3$ (x = 0.33 and 0.67)," *Physica B* **230**, 759 (1997).

Heffner, R.H., et al., "Zero-Field μSR Study of the Colossal Magneto-resistance Material $La_{0.67}Ca_{0.33}MnO_3$," *Hyperfine Interact.* **104**, 29 (1997).

Hundley, M.F., and J.J. Neumeier, "Thermoelectric Power of $La_{1-x}Ca_xMnO_{3+d}$: Inadequacy of the Nominal $Mn^{3+/4+}$ Valence Approach," *Phys. Rev. B* **55**, 1016 (1997).

Hundley, M.F., et al., "Interplay between Electronic Transport and Magnetic Order in Ferromagnetic Manganite Thin Films" (submitted to *J. Appl. Phys.*).

Kwei, G.H., et al., "Lattice Effects in Perovskite and Phyochlore CMR Materials" (to be published in *Physica B*).

Neumeier, J.J., et al., "Volume-based Thermodynamic Considerations for the Magnetoresistance Effect and Metal-Insulator Transition of CMR Manganese Oxide Perovskites" (submitted to *Phys. Rev. B*).

Nieuwenhuys, G.J., et al., "μSR in the Random Bond Spin Glass URh_2Ge_2" (submitted to *Physica B*).

Thompson, J.D., et al., "Cerium Heavy Fermion Compounds near Their T = 0 Magnetic-Non-Magnetic Boundary" (to be published in *High Pressure Science and Technology*).

Thompson, J.D., et al., "Correlated Electron Materials Near Their T = 0 Magnetic-Non-Magnetic Boundary: Implications for delta-Pu" (submitted to *J. Alloys Compd.*).

Role of Charge Localization in the Basic High-Temperature Superconductivity Mechanism

P. Chris Hammel

It is clear that local atomic structure profoundly affects the superconducting, normal electronic, mechanical (for example, elastic), and other response functions of materials. In this project, we are examining this difficult, important, and as yet barely studied problem. In particular, we are focusing on the study of the oxide superconductors and related nonsuperconducting compounds by using an integrated array of experimental and theoretical techniques. Our goal is to create a new theoretical paradigm and to develop the associated computational tools necessary to understand the properties of the high-critical-temperature (high-T_c) superconductors.

This year, we have modeled both the coupled spin-charge-lattice distortions in CuO_2 planes and the stripe pattern formation. We have developed a model of quantum paraelectric superconductors based on oxygen-buckled CuO_2 planes. We have modeled photoexcited local structural distortions in yttrium-barium-copper oxide, as probed by x-ray absorption fine spectrum. We have analyzed a high-magnetic-field mechanism for driving non-Fermi-liquid behavior in coupled Luttinger chains. We have discovered a phase transition at $T = 0.2$ K in the superconducting phase of nickel-doped Bi-2212, providing direct evidence that the superconductivity in high-temperature superconductors is unconventional, and we have developed a preliminary theory for this new phase transition.

In addition, we have calculated the effect of impurities on the superconducting density of states; developed a new, hybrid density functional and applied it to cluster calculations of the coupling constant and local moment in high-T_c and related compounds; measured the static and dynamic properties of lightly lithium-doped La_2CuO_4, providing evidence for collective hole effects; observed the microscopically inhomogeneous magnetic environment arising from stripe formation in $(La, Sr)_2NiO_4$; and obtained experimental evidence that the orthorhombic to tetragonal transition in $(La, Sr)_2CuO_4$ is better described as an order-disorder transition rather than a displacive one.

Publications

Balatsky, A.V., and S.A. Trugman, "On the Lifshitz Tail in the Density of States of a Superconductor with Magnetic Impurities" (to be published in *Phys. Rev. Lett.*).

Balatsky, A., et al., "On Spontaneous Violation of Time Reversal and Parity in d-Wave Superconductor with Magnetic Impurities" (to be published in *J. Phys. Chem. Solids*).

Bozin, E., et al., "Reexamination of the Second Order Structural Phase Transition in $La_{2-x}A_x CuO_4$ (A = Ba, Sr)" (submitted to *Physica B*).

Bussmann-Holder, A., and A.R. Bishop, "Doping Dependence of T_c and Its Related Isotope Effect in High-Temperature Superconductivity," *Phys. Rev. B* **55**, 11751 (1997).

Bussmann-Holder, A., et al., "The Role of Apical Oxygens in Superconducting Cuprates," *Phil. Mag. B* **75**, 463 (1997).

Haas, S., et al., "Extended Gapless Regions in Disordered d-Wave Superconductors" (to be published in *Phys. Rev. B*).

Moshopoulou, E.G., et al., "An Electron Diffraction Study of $La_2Li_{0.5}Cu_{0.5}O_4$" (to be published in *J. Phys. Chem. Solids*).

Movshovich, R., et al., "Low Temperature Phase Transition in $Bi_2Sr_2Ca(Ni_xCu_{1-x})_2O_8$: Evidence for Unconventional Superconductivity" (submitted to *Science*).

Movshovich, R., et al., "Low Temperature Phase Transition in $Bi_2Sr_2Ca(Ni_xCu_{1-x})_2O_8$: Second Superconducting Phase?" (to be published in *J. Phys. Chem. Solids*).

Salkola, M., et al., "Spectral Properties of Quasiparticle Excitations Induced by Magnetic Moments in Superconductors," *Phys. Rev. B* **55**, 12648 (1997).

Shenoy, S., et al., "Quantum Paraelectric Model for Layered Superconductors" (to be published in *Phys. Rev. Lett.*).

Siders, J.L.W., et al., "Terahertz Emission from $YBa_2Cu_3O_7$ Thin Films," in *Trends in Optics and Photonics, Ultrafast Electronics and Optoelectronics*, M. Nuss and J. Bowers, Eds. (Optical Society of America, Washington, DC, 1997), Vol. 13, p. 252.

Suh, B.J., et al., "Suppression of Antiferromagnetic Order by Light Hole Doping in $La_2Cu_{(1-x)}Li_xO_4$: A ^{139}La NQR Study" (submitted to *Phys. Rev. Lett.*).

Trugman, S.A., and J. Bonca, "Inelastic Quantum Transport" (submitted to *Phys. Rev. Lett.*).

Tyson, T.A., and A.R. Bishop, "Observation of Local Structural and Electronic Changes Accompanying Photodoping in $YBa_2Cu_3O_{6+x}$" (to be published in *Physica C*).

Yu, Z.G., et al., "Charge Localization and Stripes in a Two-Dimensional Three-Band Peierls-Hubbard Model" (to be published in *Phys. Rev. B*).

Yu, Z.G., et al., "Low-Energy Magnetic Excitations in Doped $La_2Cu_{0.5}Li_{0.5}O_4$" (submitted to *Phys. Rev. Lett.*).

Neutron Scattering from Correlated-Electron Systems

Robert A. Robinson

The purpose of this project is to study correlated-electron behavior in f-electron systems (e.g., heavy fermions) as well as in transition-metal oxides (e.g., high-temperature superconducting cuprates). There are two main directions: (1) the use of neutron inelastic scattering for the study of magnetic excitations in such systems and (2) the use of pair-distribution-function structural methods to study local disorder, particularly in the cuprates. The pair-distribution-function method consists of analyzing the data in real (rather than reciprocal) space, much as one does in measuring pair-distribution functions in glasses or liquids; it complements more conventional crystallographic methods like Rietveld refinement.

Our inelastic scattering studies performed on the high-resolution chopper spectrometer PHAROS continued to be highly successful this year. These included discovery of new localized magnetic excitations in UNiGe; an observation that confirms and complements our previous work on UPdSn; observation and assignment of crystal-field levels in the quadrupolar Kondo system $PrAg_2In$ (see the figure); observation of quasi-elastic magnetic scattering in both ordered and paramagnetic phases of the two-layer colossal magnetoresistance material $La_{1.4}Sr_{1.6}Mn_2O_7$; and observation of magnetic excitations in the molecular system manganese-12 acetate.

We have also determined the crystal structure of several correlated-electron materials, both in polycrystal and single-crystal form. The focus has been on $La_2Cu_{0.5}Li_{0.5}O_4$, in which the lithium atoms form an ordered two-dimensional superstructure within the CuO_4 plane but are disordered along the direction perpendicular to the plane. This structure is important for interpreting nuclear quadrupole resonance data that show the development of a spin gap.

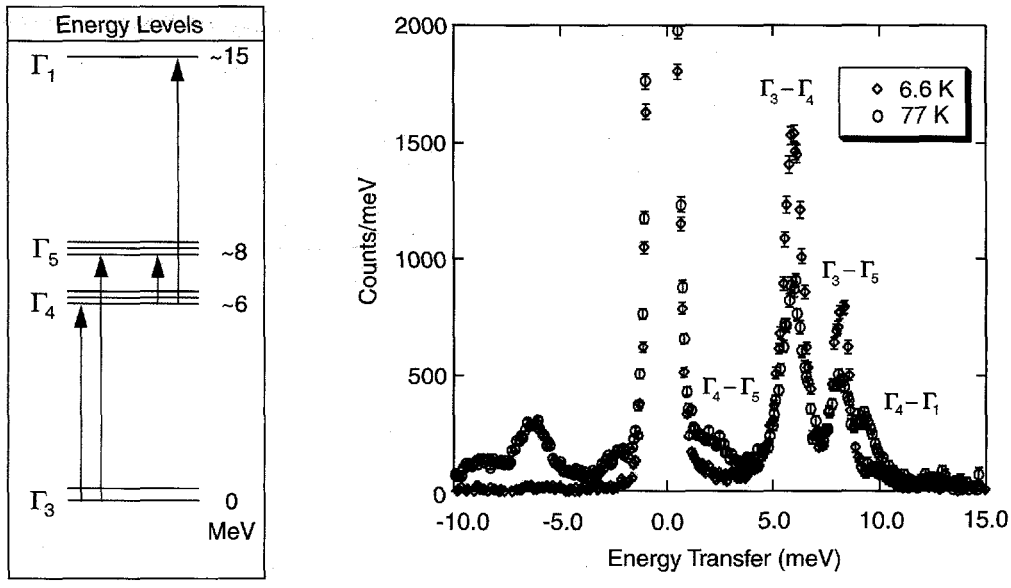
Another system studied is SrB_6, which is a very unusual metal at low temperatures. Detailed single-crystal structure parameters have been critical for input into band-structure calculations that depend sensitively on the boron-boron and strontium-boron interatomic distances.

Publications

Javorsky, P., et al., "Crystal Fields in ErNiAl and ErCuAl Studied by Inelastic Neutron Scattering" (submitted to *J. Magn. Magn. Mater.*).

Kelley, T.M., et al., "Short-Range Spin Correlations in the CMR Material $La_{1.4}Sr_{1.6}Mn_2O_7$" [to be published in *Physica B (Amsterdam)*].

Nakotte, H., et al., "Localized Excitations in UPdSn" [to be published in *Physica B (Amsterdam)*].



Crystal-field excitations in $PrAg_2In$ (for an incident energy of 25 meV) measured in both neutron energy loss and gain at two different temperatures, along with the level diagram for the Pr^{3+} ion in a cubic environment. Note that the ground state is a nonmagnetic doublet.

Understanding and Controlling Self-Assembly

Basil Swanson

In this project, synthesis, characterization, and modeling are integrated to help develop a better understanding of how self-assembly works. Our goal is to lay a foundation for the preparation of useful self-assembled films. The target of this work is the synthesis of functionalized self-assembled monolayers (SAM), in which the tail groups at the surface of the film are chemically modified to optimize chemical reactivity and/or materials properties.

We prepare and characterize single- and multi-component SAMs on both gold and silica surfaces by using experimental techniques (such as Fourier transform infrared spectrometry, ellipsometry, surface-enhanced Raman spectroscopy, and others) to develop a database for use in developing models of the structure and dynamics of self-assembled films.

Another target is the development of new synthetic approaches to self-assembled films that will allow us to control the structure, composition, and component distribution of the films. Our goal is to develop a predictive model that will guide the preparation of self-assembled hierarchical structures that can be used in novel materials applications.

Some of our significant achievements in the past year are the following: (1) establishing new experimental facilities that include surface-enhanced Raman spectroscopy and a new Langmuir-Bloddgett (LB) trough, (2) initiating experimental studies of the role that defects play in controlling the subsequent chemistry of SAMs as well as studies of domain formation for SAMs with mixed components that have different chain lengths and/or different tail groups, (3) using LB films to template the formation of covalently bonded SAMs in order to control structure, defect distribution, domain size, and incorporation of macromolecules, (4) using glancing-angle x-ray diffraction to structurally characterize SAMs formed by microcontact printing that would lead to understanding images of patterned SAMs taken with an atomic force microscope, (5) completing full quantum mechanical calculations of the gold-sulfur interaction at the organic-inorganic interface and thereby completing the first step in hierarchical modeling that bridges quantum and classical mechanics, and (6) developing statistical potentials between alkane chains modeled as rods and a liquid-crystal Ginzburg-Landau model for macroscopic identification of orientational order in self-assemblies.

Publications

Bar, G., et al., "Dendrimer-Modified Silicon Oxide Surfaces as Platforms for the Deposition of Gold and Silver Colloid Monolayers: Preparation Method, Characterization and Correlation between Microstructure and Optica Properties," *Langmuir* **12**, 1172 (1996).

Bar, G., et al., "Scanning Force Microscopy Study of Patterned Monolayers of Alkanethiols on Gold: Importance of Tip-Sample Contact Area in Interpreting Force Modulation and Friction Force Microscopy Images," *Langmuir* **13**, 373 (1997).

Bishop, A.R., et al., "Determination of Headgroup-Gold Potential Surface for Alkanethiol Self-Assembled Monolayers by *ab initio* Calculation" (submitted to *Phys. Rev. Lett.*).

Jensen, N., et al., "Lekner Summation of Coulomb Interactions in a Partially Periodic System" (to be published in *Mol. Phys.*).

Kress, J., et al., "*Ab initio* Calculations of the Gold-Sulphur Interaction for Alkanethiol Monolayers," *Synth. Met.* **84**, 317 (1997).

Parikh, A.N., et al., "Langmuir-Phase Assisted Assembly of Bifunctional Monolayers" (submitted to *Langmuir*).

Pinning Vortices and Enhancing High-Temperature Superconductor Critical Currents

Martin Maley

This project addresses a problem of fundamental technological importance for high-temperature superconductivity (HTS): the stabilization of high current densities at high temperatures in high magnetic fields. Stabilization can only be accomplished if a dense array of microstructural defects is introduced that "pin" superconducting vortices. Our work is to study such flux-pinning mechanisms and develop methods for enhancing critical current density (J_c) at high temperatures and high magnetic fields.

In the past year, we have achieved a greatly enhanced flux pinning by introducing nanoscale amorphous columnar defects (CDs) into HTS material by irradiation with fast uranium ions. We concentrated on high-J_c thick films of the HTS compound YBaCuO prepared by ion-beam-assisted deposition. We performed a transport study of the dependence of J_c on the density of CDs and found a strong correlation between the magnetic field range in which large J_c enhancement is observed and the matching field in which the density of the defects equals that of the vortices. A distinct "kink" at this same matching field is observed in the "glass line" that defines the disappearance of J_c above a line in the magnetic field vs temperature plane.

We also continued theoretical work on the Josephson plasma resonance (JPR). We have discovered that the JPR frequency measures the correlation of vortices normal to the CuO layers. We have now modeled this phenomenon into a powerful tool for studying changes in vortex structure with increasing temperature and field. JPR is now being investigated by experimental groups worldwide. A new direction entails the use of JPR to investigate vortex structure in the presence of CDs. Recent JPR results suggest the presence of a new phase of vortex matter caused by interaction with CDs.

Publications

Bulaevskii, L.N., et al., "Josephson Plasma Mode in the Mixed State of Long-Junction and Layered Superconductors," *Phys. Rev. B* **55**, 8482 (1997).

Bulaevskii, L.N., et al., "Vortex Phases in Irradiated Highly Anisotropic Superconductors" (submitted to *Phys. Rev. Lett.*).

Dominguez, D., et al., "First Order Melting of a Moving Vortex Lattice: Effects of Disorder," *Phys. Rev. Lett.* **78**, 2644 (1997).

Dominguez, D., et al., "Simulations of Current-Driven Three-Dimensional Josephson Junction Arrays," *Physica B* **222**, 396 (1997).

Findikoglu, A.T., et al., "Power-Dependent Microwave Properties of Superconducting YBaCuO Films on Buffered Polycrystalline Substrates," *Appl. Phys. Lett.* **70**, 3293 (1997).

Jensen, N.G., et al., "Metastable Filamentary Vortex Flow in Thin Film Superconductors," *Phys. Rev. Lett.* **76**, 2985 (1997).

Sasik, R., et al., "Vortices and Plasmons in Inductive Periodic Josephson-Junction Arrays," *Phys. Rev. B* **55**, 688 (1997).

Willis, J.O., et al., "Magnetic Field Orientation Dependence of J_c in Bi-2212 Round Wires," *IEEE Trans. Appl. Supercon.* **7**, 2022 (1997).

Advancing X-Ray Hydrodynamic Radiography: Advanced Cathodes

Daniel Prono

Science-based stockpile stewardship requires that x-ray hydrodynamic radiography be able to distinguish fine-scale feature sizes and shapes to <0.25-mm resolution, which is about 10 times better than present capabilities. Systematic effects can blur radiography resolution, but a fundamental limit is the spot size of the x-ray illumination source (the more it is like a point source, the better the image resolution). System requirements for the desired resolution are x-ray spot sizes of <1 mm, with a 4:1 magnification ratio (object-to-image distance divided by source-to-object distance). X-ray spot size results from an electron beam striking a high-Z metalic converter that produces x-rays; hence the x-ray spot can be no smaller than the electron beam spot, which is intrinsically limited by the transverse random motion (temperature) of the beam's electrons. At present, electron beam temperatures are ~100 eV for the Dual-Axis Radiographic Hydrodynamics Test (DARHT) Facility.

We are developing a large-area (~200 cm^2), polycrystalline diamond-film photocathode with high quantum efficiency (QE > 1%), low electron temperature (~1 eV), and the capability of operating in the modest vacuum environments characteristic of hydrodynamic firing sites. Such a photocathode will be a significant improvement over existing metal-oxide photocathodes that have QE of <0.1% and require ultrahigh vacuum environments.

This year we worked with MIT's Lincoln Laboratory to produce a small (4-cm^2), boron-doped, polycrystalline diamond-film photocathode, demonstrating a QE of 0.5% to 1.0%; developed the experimental capability to terminate the boron-doped polycrystalline diamond with hydrogen atoms so that the diamond cathode could function without degradation due to surface oxidation in modest vacuum; performed analyses to determine diamond surface properties (temperature and crystalline configuration) for successful hydrogen bonding and termination; and began to design an experimental apparatus for producing large-area (200 cm^2) polycrystalline surfaces of proper surface uniformity and mechanical integrity.

Publications

Shurter, R.P., et al., "Development and Characterization of High QE Diamond Film, High Current Photocathodes" (Particle Accelerator Conference, Vancouver, British Columbia, May 1997).

Shurter, R.P., et al., "Large Area Polycrystalline Diamond Films as High Current Photocathodes for Linear Induction Accelerators" (8th European Conference on Diamond, Diamond-like, and Related Materials, Herriot-Watt University, Edinburgh, Scotland, August 1997).

Advanced Biomolecular Materials Based on Membrane-Protein/Polymer Complexation

Gregory Smith

The overall goal of our work is to apply neutron reflectivity and atomic force microscopy to the characterization of the structure and phase behavior, the interlayer and intermacromolecular interactions, and the protein shape stability of a fundamentally new class of bioactive molecular materials. The new materials are composed of reconstituted and native, self-assembled membrane proteins complexed with associating and surfactant polymers. The ultimate goal of our research is to create synthetic materials in which the natural function of the proteins is preserved in harsh environments. Such materials are extremely important in developing novel sensors, particularly gas sensors.

We found that the key to these studies is proper sample preparation. Flat, uniform films that are only a few hundred angstroms thick are required for these experiments. We have prepared oriented films of the lipid dimyristoyl phosphatidylcholine (DMPC) on silicon substrates. However, these films have not proved to be ideal for our studies. Although the films initially formed well-oriented samples, they delaminated from the substrates upon hydration. To solve this problem, we developed a new sample chamber for drawing substrate-free, freely suspended films of the lipids and lipid-protein mixtures. This chamber is capable of maintaining an environment with tunable, yet stable, temperature and humidity.

In the past year, we drew the first lipid films, which were 2.5 cm × 2.5 cm. The molecules within these films spontaneously orient perpendicular to the plane of the film. This particular orientation gives an enhanced signal for neutron scattering experiments, which enable understanding of the DMPC structure and of the effect of adding proteins to the DMPC structure.

In July 1997, we conducted the first neutron reflection experiments on these films at the Manuel Lujan Jr. Neutron Scattering Center. The results showed that the scattering was dominated by the excess lipid powder surrounding the film and not by the film itself. We therefore changed the design of the oven used in the experiments, and that change corrected the original problem. The oven is operating well and is ready for experiments on the lipid-protein materials.

Structural and Magnetic Characterization of Actinide Materials

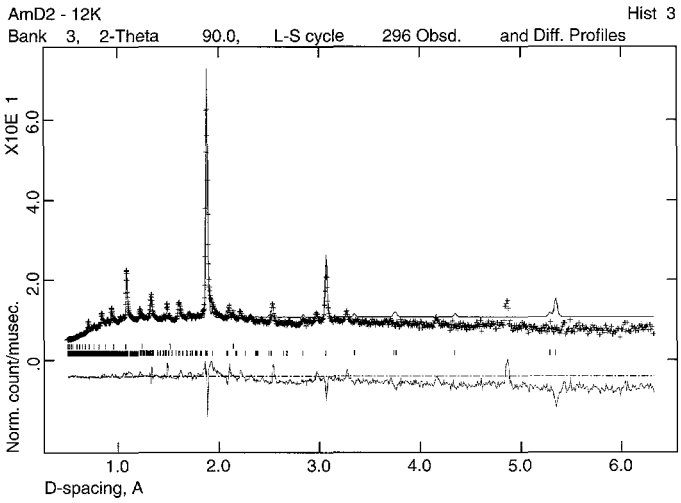
Barbara Cort

The focus of this work is to extend our fundamental research capability in neutron scattering, equilibrium studies, kinetics, and x-ray diffraction to address questions of practical importance to stockpile integrity and long-term storage of nuclear material. Our major accomplishment this year is the use of small-sample encapsulation techniques we developed in the previous two years to perform neutron diffraction studies of $AmD_{2.4}$ (see first figure).

The best solution is a tetragonal structure with twice the fluorite volume. The structure is different from $NpD_{2.1}$; the basal plane area is doubled for $AmD_{2.4}$, while the c-axis length is doubled for $NpD_{2.1}$. The tetrahedral and octahedral occupancies are 1.9 and 0.5, respectively, and are not significantly temperature dependent. Atomic positions are given in the table. The lattice constants vary in a normal way with temperature (see second figure). No structural correlates are found for the reported anomalies in the electrical resistivity. However, the true structure may be more complicated.

Crystal data for $AmD_{2.4}$ at 300 K. Space group is P4/mmm with lattice parameters of a = 7.490 Å, and c = 5.352 Å.

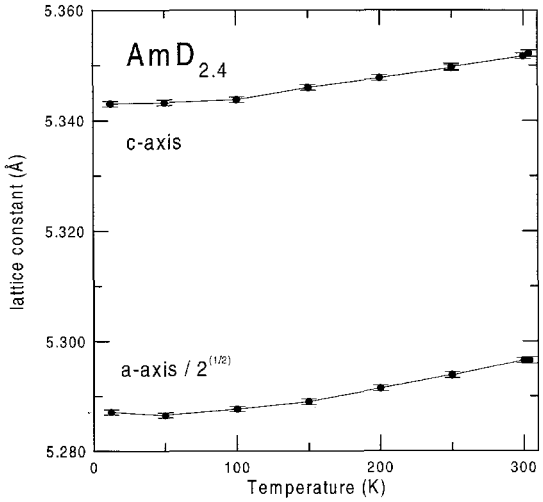
atom	x	y	z	frac
Am(1)	0	0	0	1
Am(2)	–	–	0	1
Am(3)	.261	.261	–	1
Am(4)	–	0	0	1
D(5)	0	0	–	0
D(6)	–	–	–	.98
D(7)	.225	.225	0	.70
D(8)	–	0	0	0
D(9)	.233	0	.259	1.06
D(10)	–	.233	.220	.80



Neutron diffraction pattern from a 200-mg sample of $AmD_{2.4}$. The vertical lines indicate peak positions, and the lower curve indicates deviations from the fit to the data.

We also conducted neutron diffraction studies to determine the structure of the reaction product between PuO_2 and water. We synthesized hyperstoichiometric oxide, $^{242}PuO_{2+x}$, by reacting 400 mg of $^{242}PuO_2$ with water vapor at 300°C for 30 days. The diffraction pattern (third figure) features a high background from the vanadium encapsulation and from the hydrogenous water known to be present in the sample. We used a simple fluorite structure model for PuO_2. The refined stoichiometry is $PuO_{2.30(8)}$, with a lattice constant of 5.395 Å.

We plan to repeat this experiment with the small-sample encapsulation assembly used for $AmD_{2.4}$.

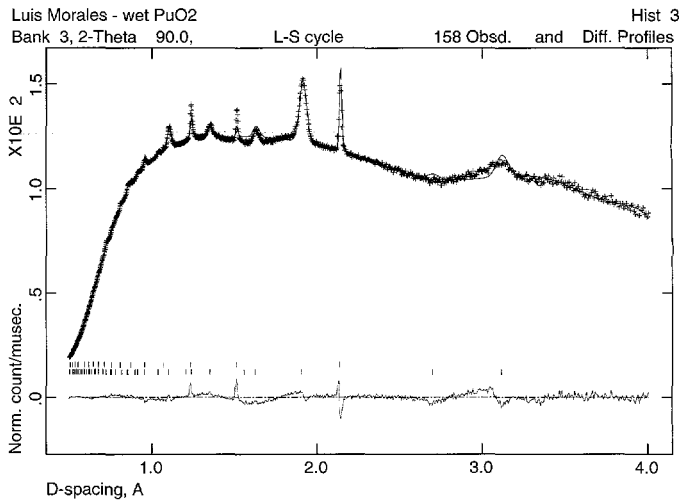


Lattice constants versus temperature for $AmD_{2.4}$.

Publications

Haschke, J.M., and T.E. Ricketts, "Adsorption of Water on Plutonium Dioxide," *J. Alloys Compd.* **252**, 148 (1997).

Klemens, P.G., and B. Cort, "Thermodynamic Properties of Helium Bubbles in Aged Plutonium," *J. Alloys Compd.* **252**, 157 (1997).



Neutron diffraction data for PuO_2, including peaks for vanadium. The vertical lines indicate peak positions, and the lower curve indicates deviations from the fit to the data.

Texture Science and Technology

Michael Stevens

The goals of this project are (1) to increase the utilization of texture and anisotropy both within and without the Laboratory's programmatic, basic, and industrial-related efforts; (2) to improve our capabilities for measuring and modeling texture; and (3) to maintain the Laboratory's recognition as an international leader in the analysis and understanding of crystalline texture effects through basic research in the field.

During the past year, we had many accomplishments. We brought the electron backscatter diffraction technique for measuring microtexture on-line in conjunction with a new field-emission-gun scanning electron microscope. We investigated deformation and reaction-induced texture development in bismuth-based superconductors at both the bulk and local scales. We researched texture evolution in refractory metals and alloys and furnished experimental validation for plasticity modeling of these materials. We provided an x-ray texture measurement and analysis resource for various research programs and collaborations and in support of students and visiting scientists.

We used neutron diffraction to determine quantitatively the texture of Bi-2212 and Bi-2223 crystallites in tapes and wires. We developed Rietveld texture analysis to deconvolute measured time-of-flight spectra in order to obtain information about texture, crystal structure, microstructure, and internal strains. We analyzed the influence of different states of precipitation/supersaturation on the recrystallization behavior in a ternary Al-Fe-Si model alloy. We developed and validated a model to simulate recrystallization textures of aluminum alloys. We developed a semiquantitative model to simulate the impact of orientation pinning on the evolution of recrystallization textures in aluminum alloys.

Publications

Bingert, J.F., et al., "Texture Evolution in Upset-Forged P/M and Wrought Tantalum: Experimentation and Modeling" (International Conference on Tungsten, Refractory Metals, and Alloys, Orlando, FL, November 16–19, 1997).

Engler, O., "On the Influence of Orientation Pinning on Growth Selection of Recrystallization" (to be published in *Acta Mater.*).

Engler, O., "A Simulation of Recrystallization Textures of Al-Alloys which Considers the Probabilities of Nucleation and Growth" (submitted to *Textures and Microstructures*).

Engler, O., et al., "Influence of Particle Stimulated Nucleation on the Recrystallization Textures in Cold Deformed Al-Alloys, Part I—Experimental Observations" (to be published in *Scr. Mater.*).

Maudlin, P.J., et al., "On the Modeling of the Taylor Cylinder Impact Test for Orthotropic Textured Materials: Calculations and Experiments" (International Symposium on Plasticity, Juneau, AK, July 14–17, 1997).

Schoenfeld, S.E., et al., "The Bulk Processing of 2223-BSCCO Powders; Part II, Tape Rolling," *Philos. Mag. A* **73**, 1565 (1996).

Vaudin, M.D., et al. "A Method for Crystallographic Texture Investigation Using Standard X-Ray Equipment" (submitted to *J. Mater. Res.*).

Wenk, H.-R., et al., "Texture Analysis of Bi-2212 and 2223 Tapes and Wires by Neutron Diffraction," *Physica C* **227**, 1 (1996).

Willis, J.O., et al., "Processing Factors for High Critical Current Density in Bi-2223/Ag Tapes," *Physica C* **78**, 1 (1997).

Bulk Amorphous Materials

Ricardo Schwarz

Metallic glasses have many technologically important properties that are not found when the alloy is in its crystalline form. These include high strength (in excess of 2 GPa), ductility in tension, and high corrosion resistance (especially for metal-metalloid glasses containing phosphorus). In addition, some ferromagnetic metallic glasses have extremely low hysteretic losses when they are used as cores of transformers and motors operating at 60 Hz. Other applications of ferromagnetic metallic glasses include toroidal magnetic cores for high-frequency transformers, targets in article-surveillance systems, sensors, transducers, and high-energy pulsed-power devices.

The objective of this project was to develop the competency to synthesize novel bulk amorphous alloys and radiation-resistant alloys. We researched synthesis methods and alloy properties, including thermal stability, mechanical properties, and transport properties. This year, we discovered a new system that forms bulk glass (Pd-Ni-Fe-P), and we determined the compositions for glass formation. In this system, bulk glasses can be formed only if there is less than 20 at.% iron, an amount that is below the level needed for a glass to be ferromagnetic.

These Pd-Ni-Fe-P glasses show spin-glass behavior, which we investigated. Its associated magnetocaloric effect suggests that these glasses may potentially be used in magnetic refrigeration. The difference between the crystallization and glass-transition temperatures is in the range of 60 to 100 K, opening the possibility for us to fabricate net-shape components with techniques such as injection molding. The temperature dependence of the magnetic susceptibility of amorphous $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$ alloy is shown in the accompanying figure.

In addition, we used mechanical alloying techniques to prepare a novel vanadium-spinel alloy that is quite resistant to degradation (such as swelling and bubble formation) in radiation environments. The composite has a hardness in excess of 12 GPa. With only 20 vol.% vanadium, the toughness of this composite is about five times that of pure spinel but is still too low for structural applications. We found that the toughness was limited by the formation of an oxygen-stabilized, tetragonal vanadium phase. We prepared new alloys by adding titanium and chromium. In the new alloys the oxygen content is below 1 at.% and shows no tetragonal vanadium phase. Currently, we are measuring the mechanical properties of these alloys.

Publications

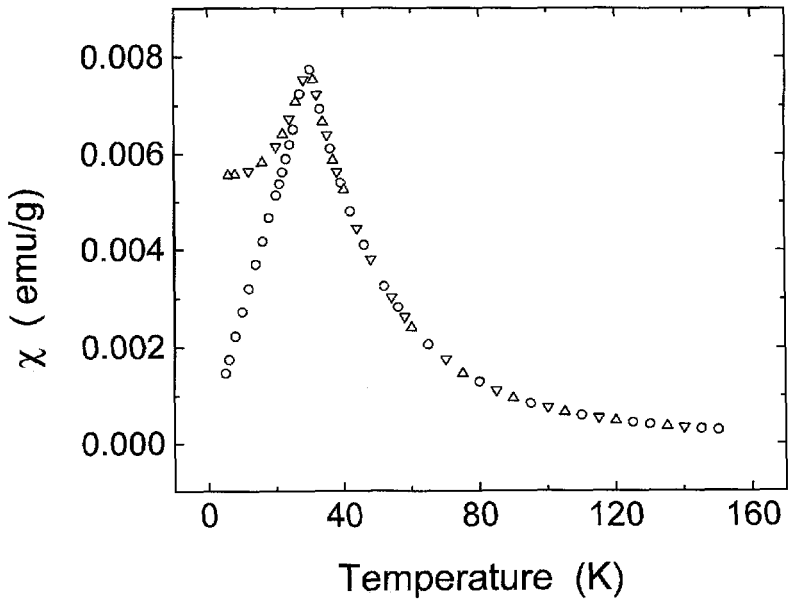
Egami, T., et al., "Structure of Bulk Amorphous Pd-Ni-P Alloys Determined by Synchrotron Radiation" (to be published in *Metall. Trans.*).

He, Y., et al., "Bulk Amorphous Metallic Alloys: Synthesis and Fluxing Techniques" (to be published in *Metall. Trans.*).

Miller, M.K., et al., "Decomposition in $Pd_{40}Ni_{40}P_{20}$ Metallic Glass" (to be published in *Mater. Sci. Eng. A*).

Schwarz, R.B., and Y. He, "Bulk Metallic Glass Formation in the Pd-Ni-P and Pd-Cu-P Systems," in *Properties of Complex Inorganic Solids*, A. Gonis, Ed. (Plenum Press, New York, 1997), p. 287.

Schwarz, R.B., et al., "Vanadium Spinel Composites for Structural Applications in Hostile Environments," *Mater. Sci. Eng. A* **234–236**, 1091 (1997).



Temperature dependence of the magnetic susceptibility of amorphous $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$ alloy. The triangles are measurements taken while we cooled and warmed the sample under an applied field of 10 Oe, and the circles denote the measurements we took while we heated the sample after it was cooled in zero applied field. The magnetic susceptibility (χ) is measured in electromagnetic units per gram.

Controlling Function of Polar Organic Multilayers

Duncan McBranch

We are investigating photo-induced charge and energy transfer in organic polar superlattices for applications in optoelectronic devices. We are studying self-assembled multilayers from two materials classes: (1) light-absorbing chromophores linked to fullerenes for ultrafast, photo-induced, intermolecular charge transfer, and (2) one- and two-dimensional oligomers of porphyrin molecules used for energy and electron transport. Our focus is on ultrafast excited-state spectroscopy, with the following objectives: to study the relationship between photo-excited charge transfer and molecular nonlinearities; to develop nonadiabatic dynamical models that describe electronic states following photo-excitation; and to understand the effects of polar multilayer structures.

This year we demonstrated rapid multilayer formation by using ionic bonding between several molecular species, including conducting polymers, fullerenes, metalloporphyrins, and metallophthalocyanines. We used ultrafast spectroscopy to characterize the photoexcitation dynamics and photo-induced charge and energy transfer. Our studies of energy transfer within weakly coupled arrays of porphyrins demonstrated energy transfer efficiencies in excess of 95% between porphyrin pairs. We have developed the first microscopic model of charge transfer in conducting polymers and fullerenes.

Our characterization studies of self-assembled, "solid-state" films have revealed contributions to the linear absorption features and excited-state absorption dynamics from both aggregation in a single monolayer and interactions among adjacent molecular layers. We developed an exciton theory for quasi-one-dimensional organic materials with strong electron-electron correlation. In addition, we used semiempirical molecular orbital theory to perform configuration interaction calculations on the excited states of the biphenyl molecule.

Publications

Brazovskii, S., et al., "Stability of Bipolarons in Conjugated Polymers" (to be published in *Opt. Mater.*).

Hsiao, J.S., et al., "Soluble Synthetic Multiporphyrin Arrays. 2. Photodynamics of Energy-Transfer Processes," *J. Am. Chem. Soc.* **118**, 11181 (1996).

Li, D., et al., "Self-Assembled, Molecular and Macromolecular p-Conjugated Optical Materials and Their Nonlinear Optical Properties," *Synth. Met.* **86**, 1849 (1997).

Luett, M., et al., "X-ray Reflectivity Study of Self-Assembled Multilayers of Macrocycles and Macromolecules" (submitted to *J. Phys. Chem.*).

Maniloff, E.S., "Charge Transfer Polymers: A New Class of Nonlinear Optical Materials," *SPIE Proc., Fullerenes and Photonics III* **2854**, 208 (1997).

Maniloff, E.S., et al., "Ultrafast Holography in Charge Transfer Polymers," *Opt. Comm.* **141**, 243 (1997).

McBranch, D.W., et al., "Ultrafast Holography and Transient Absorption Spectroscopy in Charge-Transfer Polymers" (to be published in *SPIE Proc., Fullerenes and Photonics IV*).

McBranch, D., et al., "Ultrafast Nonlinear Optical Properties of Charge-Transfer Polymers" (to be published in *J. Nonlinear Opt. Phys. Mat.*).

Vacar, D., et al., "Charge Transfer Range for Photoexcitations in Conjugated Polymer/Fullerene Bilayers and Blends," *Phys. Rev. B* **56**, 4573 (1997).

Yu, Z.G., et al., "Excitons in Quasi-One-Dimensional Organics: Strong Correlation Approximation," *Phys. Rev. B* **56**, 3697 (1997).

Yu, Z.G., et al., "Variational Studies of Charge Transfer in a Molecularly Doped Polymer" (to be published in *SPIE Proc., Fullerenes and Photonics IV*).

New Initiatives in Materials Characterization, Modeling, and Synthesis

Don M. Parkin

This project explores new techniques in characterization, constitutive modeling, and synthesis of materials. Advances in these three central elements of materials science drive the frontiers of materials research. This past year we discovered a new phenomenon in which abrupt changes in the hydrogen capacities of $GdFe_2$ hydride occur during hydrogen-induced amorphization. In the temperature range 22°C to 230°C, all the absorption isotherms show a break in the continuity at a critical amorphization pressure where the hydrogen capacity decreases. Above 230°C, the effect is reversed, and a positive jump in hydrogen capacity is observed during amorphization.

We developed a new technique for actinide and f-electron systems that enables a comparison of total energies between completely delocalized states, completely localized ones, and a somewhat more exotic electronic configuration that has a component from localized states and a component from delocalized ones. The latter configuration is consistent with most of the observed data (equilibrium volume, cohesive energy, and elastic properties) of delta-phase plutonium. With our previous work and that from the literature, these calculations give a rather complete description of the different allotropes of plutonium and of the actinide metals in general.

To further our understanding of characterization, modeling, and synthesis, we conducted workshops on "Defects in Crystals," "Residual Stress," "Spin-Charge-Lattice Coupling in Complex Electronic Materials," "Many-Body Physics," "Many-Body Theory of Actinides," and "Electronic Structure and Experimentation in Plutonium."

Publications

Brooks, M.S.S., et al., "Density Functional Theory of Crystal Field Quasi-Particle Excitations and the Ab Initio Calculation of Spin Hamiltonian Parameters," *Phys. Rev. Lett.* **79**, 2546 (1997).

Delin, A., et al., "Full-Potential Optical Calculations of Lead Chalcogenides" (submitted to the *Int. J. Quantum Chem.*).

Delin, A., et al., "A Method for Calculating Valence Stability in Lanthanide Systems" (to be published in *Phys. Rev. Lett.*).

Eriksson, O., et al., "Theoretical Aspects of the 4f Localization at the Surface of Alpha-Ce," *Surf. Sci.* **382**, 93 (1997).

Eriksson, O., et al., "Theoretical Confirmation of Near Degenerate States in Delta-Pu" (submitted to *Phys. Rev. Lett.*).

Grechnev, G.E., et al., "Pressure Effect on Magnetic Properties of UGa_3," *Acta Phys. Pol. A* **92**, 331 (1997).

Hjortstam, O., et al., "Accurate LSDA Calculations of the Magneto Crystalline Anisotropic Energy for bcc Fe, hcp Co, and fcc Ni" (submitted to *Phys. Rev. B*).

Hjortstam, O., et al., "Calculated Bond Properties of K Adsorbed on Graphite" (submitted to *Phys. Rev. B*).

Hjortstam, O., et al., "Calculated Surface Layer Relaxation and Surface Energies for Y, Zr, and Ru at hcp (0001) and (1010) Surfaces" (submitted to *Phys. Rev. B*).

Lundin, U., et al., "Transition Metal Dioxides with a Bulk Modulus Comparable to Diamond" (submitted to *Phys. Rev. B*).

Lutterotti, L., et al., "Combined Texture and Structure Analysis of Deformed Limestone from Time-of-Flight Neutron Diffraction Spectra," *J. Appl. Phys.* **81**, 2 (1997).

Matthies, S., et al., "Advances in Textures Analysis from Diffraction Spectra," *J. Appl. Cryst.* **30**, 31 (1997).

Movshovich, R., et al., "Low-Temperature Phase Transition in $Bi_2Sr_2Ca(Ni_xCu_{1-x})_2O_8$: Evidence for Unconventional Superconductivity" (submitted to *Science*).

Ravindran, P., et al., "Electronic Structure, Chemical Bonding and Ground State Properties of Superconducting and Non-Superconducting $YNi_{2-x}(Co/Cu)_xB_2C$" (submitted to *Phys. Rev. B*).

Ravindran, P., et al., "Optical Properties of Monoclinic SnI_2 from Relativistic First Principles Theory," *Phys. Rev. B* **56**, 6851 (1997).

Ravindran, P., et al., "Theoretical Investigation of the High Pressure Phasers of Ce" (submitted to *Phys. Rev. B*).

Soderlind, P., and O. Eriksson, "Pressure Induced Phase Transitions in Pa Metal from First Principles Theory" (to be published in *Phys. Rev. B*).

Soderlind, P., et al., "A Simple Model for Complex Structures" (to be published in *Phys. Rev. B*).

Svane, A., et al., "Electronic-Structure Calculations of Praseodymium Metal by Means of Modified Density-Functional Theory," *Phys. Rev. B* **56**, 7143 (1997).

Many-Body Theory Research

Alexander Balatsky

There is a broad spectrum of materials that include high-temperature superconductors, heavy fermions, and low-dimensional compounds that exhibit a number of common physical phenomena. These include unconventional superconductivity, metal-insulator transitions, antiferromagnetism, and unusual normal-state characteristics, to name a few. We are investigating the common trends of these materials to increase our understanding of quantum many-body aspects of these phenomena.

This year we obtained the following results:

Serious progress was made in our understanding of impurity states in unconventional superconductors, where anisotropic gap leads to the anisotropic impurity states.

Theory of ferromagnetic metals and superconducting instability in the presence of ferromagnetism was considered.

Experimental work on characterization of heavy fermion materials continued.

A large amount of work focused upon the properties of CuO ladders. Recently, with Rich Martin, we have been looking at the relationship of the leg and rung exchange couplings in $SrCu_2O_3$.

The novel model of volume collapse in alpha-delta plutonium phase, taking into account the electron-phonon interaction as well as f-d hybridization, has been proposed by J. R. Schrieffer (Laboratory Fellow, Florida State University) and by E. Abrahams (Rutgers University). J. Wilkins (Oklahoma State University) is working on both electronic structure and many-body computational schemes that have yielded insight into the formation of magnetic states in quasi-one-dimensional systems.

Publications

Fisk, Z., and J.L. Sarrao, "The New Generation of High Temperature Superconductors," *Annu. Rev. Mater. Sci.* **27**, 35 (1997).

Fisk, Z., et al., "The 5f Band Structure of Antiferromagnetic USb_2 from Angle-Resolved Photoemission Spectroscopy: Application to Heavy Fermions," *Phil. Mag. B* **75**, 603 (1997).

Fisk, Z., et al., "Effect of Pressure on the Electrical Resistivity and Magnetization of CeScGe," *Physica B* **237**, 207 (1997).

Fisk, Z., et al., "The Electrodynamic Response of Heavy-Electron Materials with Magnetic Transitions," *Z. Phys. B* **102,** 367 (1997).

Fisk, Z., et al., "Electronic Structure of Heavy Fermions: Narrow Temperature-Independent Bands," *Physica B* **16**, 230 (1997).

Fisk, Z., et al., "ESR of Gd^{+3} in the Intermediate-Valence $YbInCu_4$ and Its Reference Compound $YInCu_4$," *Phys. Rev. B* **55**, 1016 (1997).

Fisk, Z., et al., "Further Evidence for Time Reversal Symmetry Breaking in the Heavy Electron Superconductor UPt_3," *Physica B* **338**, 230 (1997).

Fisk, Z., et al., "High Magnetic Field Ground State in the Molecular Conductor eta-Mo_4O_{11}," *Synth. Met.* **86**, 1963 (1997).

Fisk, Z., et al., "Inelastic Neutron Scattering in Single Crystal $YbInCu_4$," *Phys. Rev. B* **55**, 14465 (1997).

Fisk, Z., et al., "Interplanar Coupling between CuO_2 Planes in $Eu_{1.9}Y_{.1}CuO_4$ Single Crystals," *Physica B* **233**, 241 (1997).

Fisk, Z., et al., "Is FeSi a Kondo Insulator?" *Physica B* **237**, 240 (1997).

Fisk, Z., et al., "Low-Energy Excitations of the Correlation-Gap Insulator SmB_6: A Light Scattering Study," *Phys. Rev. B* **55**, 12488 (1997).

Fisk, Z., et al., "Low-Temperature Magnetic and Thermodynamic Properties of $Fe_{1-x}Co_xSi$," *Physica B* **230**, 790 (1997).

Fisk, Z., et al., "Low-Temperature Transport, Optical, Magnetic and Thermodynamic Properties of $Fe_{1-x}Co_xSi$," *Phys. Rev. B* **56**, 1366 (1997).

Fisk, Z., et al., "Magnetic-Field, Pressure and Temperature Scaling of the First-Order Valence Transition in Pure and Doped $YbInCu_4$," *Phys. Rev. B* **56**, 71 (1997).

Fisk, Z., et al., "Mu-SR Studies of Li-Doped La_2CuO_4," *Hyperfine Interactions* **104**, 91 (1997).

Fisk, Z., et al., "Quantum Limit and Anomalous Field-Induced Insulating Behavior in eta-Mo_4O_{11}," *Phys. Rev. B* **55**, 2018 (1997).

Fisk, Z., et al, "Spectroscopic Study of Bound Magnetic Polarons Formation and the Metal-Semiconductor Transition in EuB_6," *Phys. Rev. B* **56**, 2717 (1997).

Fisk, Z., et al., "Structure and Low Temperature Properties of SrB_6," *Z. Phys. B* **102**, 337 (1997).

Fisk, Z., et al., "Structural, Magnetic and Transport Properties of $La_2Cu_{1-x}Li_xO_4$," *Phys. Rev. B* **54**, 12014 (1996).

Fisk, Z., et al., "Thermal Expansion of CeRhSb under High Pressure," *Physica B* **239**, 104 (1997).

Fisk, Z., et al., "Transport Properties of FeSi," *Physica B* **230**, 784 (1997).

Schrieffer, J.R., "ARPES: Novel Effects in the Energy and Momentum Distributions of Nearly Antiferromagnetic Metals," in *Proceedings of the Physical Phenomena at High Magnetic Fields—I, May 1995* (World Scientific, River Edge, NJ, 1996).

Schrieffer, J.R., "Exotic Condensates and Excitations in Condensed Matter Physics" (to be published in *Korean J. Phys.*).

Schrieffer, J.R., "The Physics of High Temperature Superconductivity," *J. Korean Phys. Soc.* **31** (1), 1 (1997).

Schrieffer, J.R., and A.S. Alexandrov, "Radiative Corrections to the Long-Wavelength Optical-Mode Spectrum of the Electron-Phonon System" (to be published in *Phys. Rev. B*).

Schrieffer, J.R., and Z.X. Shen, "Momentum, Temperature and Doping Dependence of Photoemission Lineshape and Implications for the Nature of the Pairing Potential in HTS Materials," *Phys. Rev. Lett.* **78**, 1771 (1997).

Schrieffer, J.R., et al., "Collective Modes, Pseudogaps, and Pairing Polaron Effects in HTS Materials," in *Systematic Numerical Study of Spin-Charge Separation in One Dimension*, J.R. Schrieffer, Ed. (World Scientific, River Edge, NJ, in press).

Schrieffer, J.R., et al., "Spectral Properties of Quasiparticle Excitations Induced by Magnetic Moments in Superconductors," *Phys. Rev. B* **55**, 12648 (1997).

Theory and Modeling of Correlated-Electron Materials

Alan Bishop

We are entering a golden age for exploiting complex electronic materials based on discoveries totally beyond traditional solid state—heavy fermions, conducting polymers, high-temperature superconductors, fullerenes, and quantum-Hall-effect and quantum-well systems. The new materials are characterized by strong competitions between coupled electronic, structural, and magnetic degrees of freedom and by sensitivities to dimensionality and geometry. Tuning these competitions by chemistry, pressure, magnetic fields, temperature, etc., ultimately controls macroscopic functionalities that will enable next-generation technologies.

To gain control of the properties of these novel materials, we are conducting theoretical and modeling studies related to three major tasks: (1) isolation of novel phenomena, (2) integration of synthesis and characterization with theoretical modeling of phenomena in specific classes of materials, and (3) development of scientific control of synthesis-microstructure relationships. These tasks are focused on answering three critical questions that are at the cutting edge of research in correlated-electron materials: (1) What are the roles of electron-electron and electron-lattice interactions, disorder, chemical doping, magnetic field, pressure, and temperature in controlling metal-insulator transitions in correlated-electron materials? (2) What conditions induce non-Fermi-liquid metallic behavior and what are their experimental signatures? (3) How can the local structure of these materials be probed? Answering these questions will provide the scientific underpinning for key next-generation civilian and defense technologies.

This year we continued joint experimental-theoretical meetings, hosted an international workshop on "Spin-Charge-Lattice Coupling" that was attended by Nobel laureate K. A. Mueller, and prompted extensive Laboratory discussion of state-of-the-art multidisciplinary approaches to structure-function relations in modern materials. We developed modeling techniques for polarons and charge ordering in transition metal perovskite oxides, and developed new methods for calculating transport in strongly driven polaron systems, retaining full quantum coherence. We analyzed impurity-induced and localized quasiparticle gap states in novel superconductors. We calculated metal-insulator/magnetic transitions in colossal magnetoresistance materials, including the effects of Jahn-Teller coupling and spin-disorder. We estimated the effects of interchain coupling in spin-Peierls systems. Finally, we initiated data analysis techniques based on information entropy and analyzed the signatures of local structure in neutron diffuse scattering and pair-distribution functions in direct space.

Publications

Aronov, I., et al., "Collective Electromagnetic Excitations in a Double-Layer Two-Dimensional Electron System in a High Magnetic Field" (to be published in *Phys. Rev. B*).

Balatsky, A.V., and S.A. Trugman, "On the Lifshitz Tail in the Density of a Superconductor with Magnetic Impurities" (to be published in *Phys. Rev. Lett.*).

Bishop, A.R., and H. Roeder, "Theory of Colossal Magnetoresistance," *Curr. Opin. Solid State Mater. Sci.* **2**, 244 (1997).

Bussmann-Holder, A., and A.R. Bishop, "Competing Length Scales in Anharmonic Lattices: Domains, Stripes and Discommensurations" (to be published in *Phys. Rev. B*).

Dimitrov, D., et al., "The Crystallography of a Two-Dimensional Binary Crystal with Local Distortions," *Phys. Rev. B* **56**, 2969 (1997).

Gulacsi, M., and A.R. Bishop, "Excitonic States in the One-Dimensional Hubbard Model" (to be published in *Phil. Mag. Lett.*).

Haas, S., et al., "Extended Gapless Regions in Disordered d-Wave Superconductors" (to be published in *Phys. Rev. B*).

Louca, D., et al., "Local Jahn-Teller Distortion in $La_{1-x}Sr_xMnO_3$ Observed by Pulsed Neutron Diffraction" (to be published in *Phys. Rev. B*).

Matveenko, S., et al., "Superconductivity Fluctuations in a One-Dimensional Two-Band Electron-Phonon Model with Strong Repulsive Electron-Electron Interactions," *Zh. Eksp. Teor. Fiz.* **111**, 1 (1997).

Qi, S., et al., "Localization Effects in Colossal-Magnetoresistance Manganites" (to be published in *Phys. Rev. B*).

Raghavan, S., et al., "Relation between Dynamic Localization in Crystals and Trapping in Two-Level Atoms," *Phys. Rev. B* **54**, 1781 (1996).

Salkola, M., et al., "Spectral Properties of Quasiparticle Excitations Induced by Magnetic Moments in Superconductors," *Phys. Rev. B* **55**, 12648 (1997).

Salkola, M., et al., "Coupled Spin-Boson Systems Far from Equilibrium," *Phys. Rev. B* **R54**, 12645 (1996).

Scalapino, D.J., and S.A. Trugman, "Local Antiferromagnetic Correlations and d(x2-y2) Pairing," *Phil. Mag. B* **74**, 607 (1996).

Schmeltzer, D., and A.R. Bishop, "Using a Magnetic Field to Probe the Normal State in High-Temperature Superconductors" (to be published in *Phys. Rev. B*).

Siders, J.L.W., et al., "Terahertz Emission from $YBa_2Cu_3O_7$ Thin Films via Bulk Electric Quadrupole Optical Rectification" (CLEO/IQEC 1998, San Francisco, CA, May 3–8, 1998).

Silver, R.N., "Bose Condensation in ^{4}He and Neutron Scattering," in *Condensed Matter Theories,* J.W. Clark and P.V. Pariat, Eds. (Nova Science Publishers, Inc., 1997), Vol. 12, p. 77.

Silver, R.N., "Neutron Scattering Evidence for Bose Condensation in Superfluid ^{4}He" (International Workshop on High Temperature Superconductivity: 10 years After Its Discovery, Jaipur, India, December 1996).

Silver, R.N., and H. Roder, "Calculation of Densities of States and Spectral Functions by Chebyshev Recursion and Maximum Entropy" (to be published in *Phys. Rev. B*).

Silver, R.N., M. Suzuki, N. Kawashima, Eds., "Chebyshev Recursion Methods: Kernel Polynomials and Maximum Entropy," in *Coherent Approaches to Fluctuations,*" M. Suzuki and N. Kawashima, Ed. (World Scientific, Singapore, 1996), p. 119.

Silver, R.N., et al., "Kernel Polynomial Approximations for Densities of States and Spectral Functions," *J. Comput. Phys.* **124**, 115 (1996).

Silver, R.N., et al., "The Kernel Polynomial Method Applied to the Non-Orthogonal Electronic Structure Calculation of Amorphous Diamond," *Phys. Rev. B* **55**, 15382 (1996).

Silver, R.N., et al., "Spectral Properties of the 2D Holstein t-J Model" (to be published in *Physica C*).

Trugman, S.A., and J. Bonca, "Inelastic Quantum Transport" (to be published in *Phys. Rev. Lett.*).

Tyson, T.A., and A.R. Bishop, "Observation of Local Structural and Electronic Changes Accompanying Photodoping in $YBa_2Cu_3O_{6+x}$," *Physica C* **292**, 163 (1997).

Yu, Z.G., et al., "Charge Localization and Stripes in Two-Dimensional Three-Band Peierls-Hubbard Model" (to be published in *Phys. Rev. B*).

Yu, Z.G., et al., "Low-Energy Magnetic Excitations in Doped $La_2Cu_{0.5}Li_{0.5}O_4$" (submitted to *Phys. Rev. Lett.*).

Zang, J., et al., "Interchain Coupling Effects in Spin-Peierls Systems," *Phys. Rev. B* **55**, 14705 (1997).

Zang, J., et al., "Low Temperature Magnetic Properties of the Double Exchange Model," *J. Phys. Cond. Matter* **9**, 157 (1997).

Zang, J., et al., "Persistent Current in the Ferromagnetic Kondo Lattice Model," *Phys. Rev. B* **56**, 11839 (1997).

Dynamics of Polymers at Interfaces

Gregory Smith

This project addresses fundamental questions concerning the behavior of polymers at interfaces and poses the following questions: What processes control the formation of an adsorbed layer on a clean surface? What processes control the desorption or displacement of preadsorbed polymers? Can we accurately predict the structure of nonequilibrium layers?

To study these systems, we performed two new sets of experiments during the past year. In the first experiments, we continued our studies of the swelling of a (poly)methyl methacrylate (PMMA) layer on a solid surface. We placed the surface in contact with a reservoir of benzene and used neutron reflectometry to determine the structure. The molecular weight of the PMMA varied between 68 and 520 g/mol. We are presently analyzing the data to see if it agrees with the theoretical prediction that such films should have a density profile that follows a well-defined mathematical function (a power law).

In the second set of experiments, we continued our studies of the exchange of a polymer at a surface. The experiment consisted of depositing a deuterated layer of PMMA with 188-g/mol molecular weight onto a quartz substrate. Then, we spun a second layer of hydrogenated PMMA on top of the first. The molecular weights of the second layers for the three experiments performed were 22, 68, and 174 g/mol. We then annealed the sample at 140°C in situ on the neutron reflectometer and measured the density profiles as a function of time. Our preliminary analysis of the data shows that the smaller molecular weights exchange with the surface-bound polymer at higher rates. We are analyzing the data further to understand this effect in detail.

Multiscale Phenomena in Materials

Alan Bishop

This project develops and supports a technology base in nonlinear, nonequilibrium phenomena underpinning fundamental issues in condensed matter and materials science, and applies this technology to selected problems. In this way the precision of synthesis and characterization available for classes of complex electronic and structural materials provides a test bed for nonlinear science, while nonlinear and nonequilibrium techniques help advance our understanding of the scientific principles underlying the control of materials microstructure, textures, and their evolution, which are fundamental to macroscopic functionalities.

Our project focuses on overlapping areas of emerging interests in the materials community for which nonlinear and nonequilibrium approaches will have decisive roles, and in which productive teamwork among elements of modeling, synthesis, characterization, and applications can be anticipated. Principal topics are (1) materials microstructure and evolution in multiscale and hierarchical materials; (2) complex organic and inorganic electronic materials, including soft condensed matter, self-assembly, and photophysics; (3) dynamic friction and fracture; and (4) equilibrium and nonequilibrium phases and phase transformations.

During the past year, we held a "Nonlinearity in Materials" seminar series and recruited senior visitors, postdoctoral fellows, and graduate research students. We developed novel nonlinear Ginzburg-Landau elasticity and complex Ginzburg-Landau surface growth theories (see the figure) and implemented large-scale simulations to study pattern formation and evolution. Our qualitative modeling of new effects of lattice discreteness in nonlinear systems allowed us to exploit these effects for modeling stick-slip dynamics and energy localization (fracture, friction, filamentary flux flow in superconductors, and multiphonon bound states). We have developed new exciton and polaron models of conjugated polymers for time-resolved photophysics and charge injection at metal/polymer interfaces.

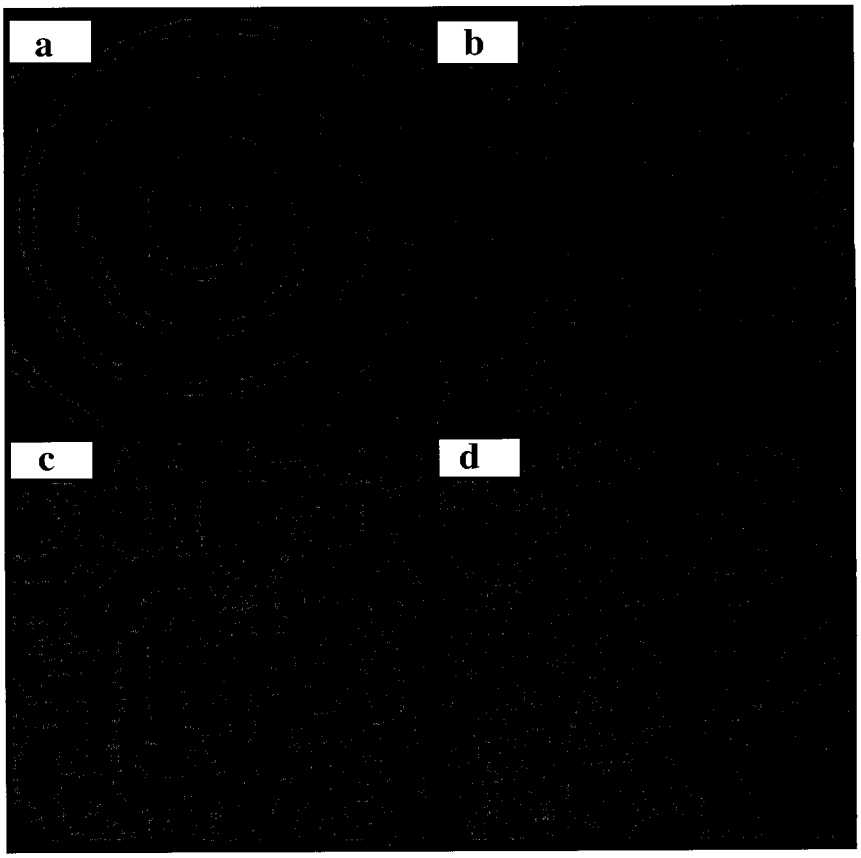
Publications

Alekseev, K.N., et al., "Dissipative Chaos in Semiconductor Superlattices," *Phys. Rev. B* **54**, 10625 (1996).

Aronov, I.E., et al., "Collective Electromagnetic Excitations in a Double-Layer Two-Dimensional Electron System in a High Magnetic Field" (to be published in *Phys. Rev. B*).

Aronov, I.E., et al., "On the Crossover of the Surface Plasmon Spectrum from Two-Dimensional to Quasi One-Dimensional in a Quantum Point Contact" [submitted to *Physica B (Amsterdam)*].

Aronov, I.E., et al., "Wigner Function Description of the A.C.-Transport through Two-Dimensional Quantum Point Contact," *J. Phys.: Condens. Matter* **9**, 5089 (1997).



A complex Ginzburg-Landau model of spiral surface growth. Figure shows gray-scale images of the surface height: (a) square-shaped spirals on a square lattice surface; (b) nucleation of new spirals at low temperatures; (c) nucleation of spirals at higher temperatures; (d) rough growth at high temperature.

Aronson, I., and A.R. Bishop, "Stretching of Vortex Lines and Generation of Vorticity in the 3-Dimensional Complex Ginzburg-Landau Equation," *Phys. Rev. Lett.* **79**, 4174 (1997).

Balakrishnan, R., and R. Blumenfeld, "Transformation of General Curve Evolution to a Modified Belavin-Polyakov Equation" [to be published in *J. Math. Phys. (N.Y.)*].

Balakrishnan, R., and R. Blumenfeld, "On the Twist Excitations in a Classical Anisotropic Antiferromagnetic Chain" (to be published in *Phys. Lett. A*).

Ben-Naim, E., and P.L. Krapivsky, "Domain Number Distribution in the Nonequilibrium Ising Model" (submitted to *Phys. Rev. Lett.*).

Ben-Naim, E., and P.L. Krapivsky, "Multiscaling in Fragmentation" [to be published in *Physica D (Amsterdam)*].

Ben-Naim, E., and P.L. Krapivsky, "Stationary Velocity Distributions in Traffic Flows" (to be published in *Phys. Rev. E*).

Ben-Naim, E., et al., "Theoretical Model of Granular Compaction" [submitted to *Physica D (Amsterdam)*].

Berman, G.P., et al., "Electron Dynamics in Intentionally Disordered Semiconductor Superlattices," *Phys. Rev. B* **54**, 14550 (1996).

Berman, G.P., et al., "Error Correction for a Spin Quantum Computer," *Phys. Rev. B* **55**, 5929 (1997).

Berman, G.P., et al., "Implementation of a Control-Not Gate Using a Quantum System of Two Interacting Spins" [to be published in *Physica B (Amsterdam)*].

Berman, G.P., et al., "Miniband Landscape of Disordered Dimer Superlattice" [to be published in *Physica D (Amsterdam)*].

Berman, G.P., et al., "On the Phase Transition in One-Dimensional Quantum Discrete ϕ^4 Model" (to be published in *Phys. Rev. B*).

Berman, G.P., et al., "Quantum Nonlinear Resonance and Quantum Chaos in Aharonov-Bohm Oscillations" (to be published in *Phys. Rev. B*).

Berman, G.P., et al., "Rugged Landscape and the 'Quantori' Ground State of 1-D Quantum System of Adatoms on Substrates" [to be published in *Physica D (Amsterdam)*].

Beardmore, K.M. et al., "Ab-Initio Calculations of the Gold-Sulfur Interaction for Alkanethiol Monolayers," *Synth. Met.* **84**, 317 (1997).

Beardmore, K.M., et al., "Determination of the Headgroup-Gold (III) Potential Surface for Alkanethiol Self-Assembled Monolayers by Ab Initio Calculation" (to be published in *Chem. Phys. Lett.*).

Bronold, F.X., and A.R. Bishop, "Nonlinear Optics of Conjugated Polymers: A Coupled Exciton-Phonon Gas Approach," *Phys. Rev. B* **53**, 13456 (1996).

Dominguez, D., et al., "First-Order Melting of a Moving Vortex Lattice: Effects of Disorder," *Phys. Rev. Lett.* **78**, 2644 (1997).

Dominguez, D., et al., "Interaction of Vortices with Ultrasound and the Acoustic Faraday Effect in Type-II Superconductors," *Phys. Rev. B* **53**, 6682 (1996).

Frachebourg, L., et al., "Spatial Organization in Cyclic Lotka-Volterra Systems," *Phys. Rev. E* **54**, 6186 (1996).

Gronbech-Jensen, N, et al., "Lekner Summation of Coulomb Interactions in Partially Periodic Systems" (to be published in *Mol. Phys.*).

Grossman, E.L., et al., "Towards Granular Hydrodynamics in Two-Dimensions," *Phys. Rev. E* **55**, 4200 (1997).

Jensen, N.G., et al., "Metastable Filamentary Vortex Flow in Thin Film Superconductors," *Phys. Rev. Lett.* **76**, 2985 (1996).

Jensen, N.G., et al., "Simulations of Current-Driven Three-Dimensional Josephson Junction Arrays," *Physica B (Amsterdam)* **222**, 396 (1996).

Kenkre, V.M., et al., "Memory Function Approach to Interacting Quasi Particle-Boson Systems," *Phys. Rev. B* **53**, 5407 (1996).

Kirova, N., et al., "Understanding the Excitations and Optical Properties of Phenylene-Based Conjugated Polymers: A Hybrid Molecular and Band Model" (submitted to *Phys. Rev. Lett.*).

Krapivsky, P.L., and E. Ben-Naim, "Domain Statistics in Coarsening Systems" (to be published in *Phys. Rev. E*).

Li, Q., et al., "Localization Effects in Colossal-Magnetoresistance Manganites" (to be published in *Phys. Rev. B*).

Nowak, E.R., et al., "Density Fluctuations in Vibrated Granular Material" (submitted to *Phys. Rev. E*).

Raghavan, S., et al., "Validity of the Discrete Nonlinear Schrodinger Equation in the Context of the Fluorescence Depolarization of a Spin-Boson System," *Phys. Rev. B* **53**, 8457 (1996).

Roeder, J., et al., "A Multichain Frenkel-Kontorova Model for Interfacial Slip" (to be published in *Phys. Rev. B*).

Sasik, R., et al., "Vortices and Plasmons in Inductive Periodic Josephson Junction Arrays," *Phys. Rev. B* **55**, 11688 (1997).

Schnitzer, H.J., and A.R. Bishop, "Hierarchy of Equations of Motion for Nonlinear Coherent Excitations Applied to Magnetic Vortices," *Phys. Rev. B* **56**, 2510 (1997).

Shuai, Z., et al., "Electro-Absorption of Poly(paraphenylene vinylene) and PtI: Exciton Versus Band Descriptions," *Solid State Commun.* **97**, 1063 (1996).

Yu, Z.G., et al., "Charge Localization and Stripes in a Two-Dimensional Three-Band Peierls-Hubbard Model" (to be published in *Phys. Rev. B*).

Yu, Z.G., et al., "Effects of Lattice Fluctuations on Electronic Transmission in Metal/Conjugated-Oligomer/Metal Structures," *Phys. Rev. B* **56**, 6494 (1997).

Yu, Z.G., et al., "Electronic Transmission in Conjugated Oligomer Tunnel Structures: Effects of Lattice Fluctuations" (to be published in *J. Phys.: Condens. Matter*).

Yu, Z.G., et al., "Excitons in Quasi-One-Dimensional Organics: Strong-Coupling Approximation" (to be published in *Phys. Rev. B*).

Yu, Z.G., et al., "Excitons in Two Coupled Conjugated Polymer Chains," *J. Phys.: Condens. Matter* **8**, 8847 (1997).

Yu, Z.G., et al., "Low-Energy Magnetic Excitations in $La_2Cu_{(0.5)}Li_{(0.5)}O_4$" (submitted to *Phys. Rev. Lett.*).

Zang, J., et al., "Interchain Coupling Effects in Spin-Peierls Systems," *Phys. Rev. B* **55**, 14705 (1997).

Zang, J., et al., "Low Temperature Magnetic Properties of the Double Exchange Model," *J. Phys.: Condens. Matter* **9**, 157 (1997).

Zang, J., et al., "Nonequilibrium Transport and Population Inversion in Double Quantum Dot Systems," *J. Mod. Phys.* **11**, 1463 (1997).

Zang, J., et al., "Persistent Current in the Ferromagnetic Kondo Lattice Model," *Phys. Rev. B* **56**, 11839 (1997).

Zhou, S., et al., "Nonlinear-Discrete Model of Dynamic Fracture Instability," *Phys. Lett. A* **232**, 183 (1997).

Chemistry

Energy Transfer in Molecular Solids

Juergen Eckert

Our goal is to significantly advance our understanding of energy transfer processes and vibrational dynamics in molecular crystals, particularly those closely related to high explosives (HEs). The question of how mechanical energy from a shock wave is transferred to chemical energy in individual molecules remains essentially unsolved. Virtually all microscopic models of the vibrational energy transfer from shock-wave (acoustic) energy to molecular vibrations require detailed knowledge of both external and internal vibrational densities of state. We are combining inelastic neutron scattering (INS) vibrational spectroscopy with theoretical modeling to derive a reliable, transferable interatomic force field for this class of compounds. This combination will enable us to perform reliable molecular dynamics simulations of HEs and to calculate properties such as vibrational heating rates.

We have collected high-quality INS spectra on a myriad of HE-related molecular crystals (such as nitroanilines and nitrotoluenes, including some isotopically substituted compounds) and on three HEs as well (TATB, RDX, and HMX). These data will serve as the basis of our molecular dynamics simulations and other theoretical studies, which we will use to produce a transferable intermolecular force field for HE systems in order to model energy transfer rates. We have also obtained an accurate single-crystal neutron structure of 4-nitroaniline, which will also serve to refine the force field.

Actinide Molecular Science: f-Electronic Structure in Synthesis, Spectroscopy, and Computation

David L. Clark

Actinide chemistry is critically important to many Laboratory and DOE core missions, providing the technical basis for plutonium process and separation chemistry and metallurgy that underpin the weapons and environmental missions. This interdisciplinary project closely integrates chemical synthesis and characterization, spectroscopy, theory, and modeling to understand the manifestation of actinide electronic structure in the physical and chemical properties of actinide materials. The research specifically investigates actinide speciation, structure, and chemical equilibrium in aqueous/nonaqueous systems, on oxide surfaces, and of small molecules on actinide surfaces. The result will be enhanced scientific competency to rationally address complex chemical problems in a number of important Laboratory and DOE missions.

This year we put into place the theoretical, spectroscopic, and chemical framework that will be applied throughout the project. Initial scientific work was carried out on uranium, neptunium, and plutonium systems in the following areas: (1) Investigation of emission branching ratios and radiative lifetimes of uranyl hydrolysis products. (2) Investigation of luminesence/Raman spectra of 20 uranyl complexes. (3) Investigation of voltammetric aqueous uranyl and neptunyl systems. (4) Determination of the solid-state and solution structures of hexavalent uranium, neptunium, and plutonium aquo ions for input to theoretical models. (5) Preparation and characterization of the first fully encapsulated actinide crown ether complex. (6) Synthesis of nonaqueous aryloxide complexes of uranium, neptunium, and plutonium for characterization by structural, chemical, electrochemical, and theoretical methods. (7) Application of density functional theory toward prediction of the molecular structures of actinyl compounds, and comparison to experimentally determined structures. (8) Development of theory capability for geometry/vibrational optimization in complexes. (9) Development of theory for calculating actinide solvation effects. (10) Initiation of solid-state calculations of actinides on silica surfaces. (11) Discovery of possible low-lying *cis*-uranyl states.

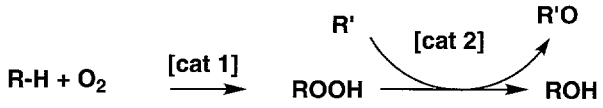
Catalysis Science and Technology

Kevin Ott

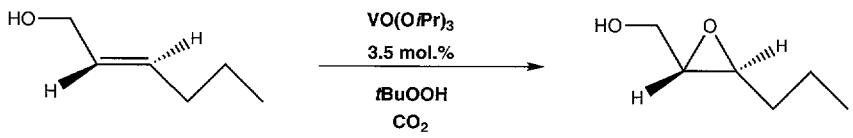
We are investigating heterogeneous and homogeneous catalytic processes for the chemical synthesis of a variety of compounds. These compounds range from simple organic compounds that are the building blocks of the chemical industry to complex agrochemical intermediates and compounds. We are focusing on two general classes of catalytic reactions: selective oxidation and catalytic diboration.

In the area of selective oxidation, we have constructed a reactor system to rapidly screen catalysts. We have screened over 50 materials and have run over 250 reactions for the oxidation of cyclohexane. We have identified useful catalysts for the formation of cyclohexylhydroperoxide (ROOH, where R is the cyclohexyl group), which we intend to use in the peroxidation of alkenes (producing R′O, where R′ is an alkyl group) in the next phase of this research. These two steps are shown in the first reaction sequence. We have also investigated the kinetics of the catalytic epoxidation of alkenes with vanadium catalysts in supercritical carbon dioxide (see second reaction), and we found supercritical carbon dioxide to be a useful solvent for these reactions.

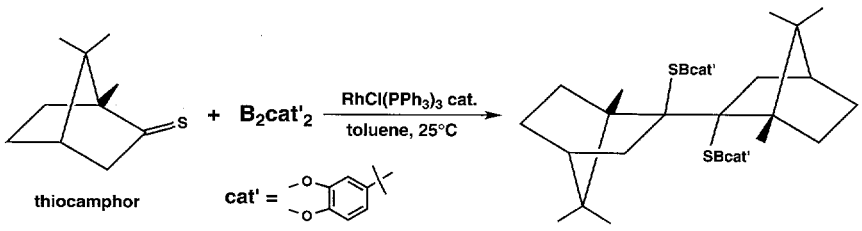
Sulfur-containing boronic acids show herbicidal activity. We sought direct routes to this class of compounds via metal-catalyzed diboration of the C=S moiety. Instead of the expected reaction products, we observed reductive carbon-carbon couplings that formed new C_2-bridged disulfur compounds (see third reaction); these new compounds may be useful ligands in transition-metal complexes with applications to homogeneous catalysis. Analogous metal-catalyzed diborations of sulfoxides (see fourth reaction) produce the corresponding sulfides in high yields. This is one of the mildest sulfoxide reductions known and can be used to control the stereochemistry at the alpha carbon either through chiral dialkoxy substituents on boron or chiral ligands on the metal-complex catalyst.



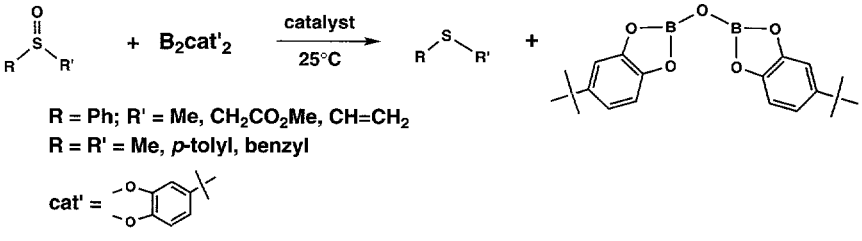
Two-step oxidation of a substrate R′ with dioxygen via an intermediate alkylhydroperoxide. In this reaction sequence, R and R′ are alkyl groups, and cat 1 and cat 2 are catalysts.



Vanadium-catalyzed epoxidation of alkenes with *t*-butylhydroperoxide (*t*BuOOH) in supercritical carbon dioxide.



Reductive diboration of thiocamphor (where Ph is a phenyl group).



Reductive deoxygenation of sulfoxides, in which R is a phenyl group and R′ is a methyl, $CH_2CO_2CH_3$, or $CH{=}CH_2$ group; also, both R and R′ can be methyl, *p*-tolyl, or benzyl groups.

Publications

Pesiri, D., and W. Tumas, "Selective Epoxidation in Dense Phase Carbon Dioxide" (submitted to *Angew. Chem.*).

Foreign Nuclear Test Radiochemical Diagnostics

Michael Macinnes

During the time of atmospheric nuclear testing, the United States and its allies maintained a capability to sample, analyze, and interpret material released by the detonation of nuclear devices. The goal of this project is to conduct training and document the methodology while the necessary knowledge still exists at the Laboratory. An additional goal is to provide additional evaluated radiochemical data for future efforts in nonproliferation and nuclear test characterization.

In the first two years, we brought computational codes back on-line and trained two analysts in their use in the evaluation of simple fission devices. In addition, technical staff members were involved in modeling one of these devices to illustrate the iterative nature of nuclear device interpretation to our trainees.

This year we expanded our interpretive training to include evaluation of thermonuclear devices. We revitalized an additional computer code and instructed our two trainees in its use. They evaluated two events, using all the tools they had mastered over the last three years. We compiled a working handbook for Laboratory radiochemists from three years of techniques that were used to evaluate several classes of nuclear devices. In addition, we evaluated and archived over 40 benchmark tests, forming a database for use in interpretation. Because we were successful in maintaining this important capability, the US Air Force has selected Los Alamos to take over work in this area of expertise.

Molten Salt and Separations Technologies Evaluation

Francesco Venneri

Nonaqueous, molten-salt- and liquid-metal-based processes are the preferred options for process chemistry in the accelerator-driven transmutation of waste (ATW) concept. We are developing waste treatment flow sheets that include front-end preparation from spent fuel, back-end cleanup, fission-product partitioning, and final waste disposition. The building blocks of the ATW treatment, electrorefining and reductive extraction, require work to extend the current state of the technology. Issues of fission-product separation chemistry and waste-form/repository interaction are also specific to the ATW concept and need to be studied. All work is directed toward developing proliferation-resistant processes.

The objective of this project is to develop the pyrochemical expertise for ATW treatment applications. This research leverages the pyrochemistry developed at Argonne National Laboratory and Los Alamos, extending it to waste transmutation applications. Our focus is on the areas of electrorefining, reductive extraction, fission-product chemistry, and waste-form chemistry, with special emphasis on the first two.

We selected and scoped out pyrochemical processes to be employed in the treatment of spent fuel and preparation of residual waste for ultimate disposal in the new liquid-lead ATW concept.

In addition, we initiated thermodynamic modeling of the actinide-bismuth alloy systems. The data will be used to model and understand the electrorefining process.

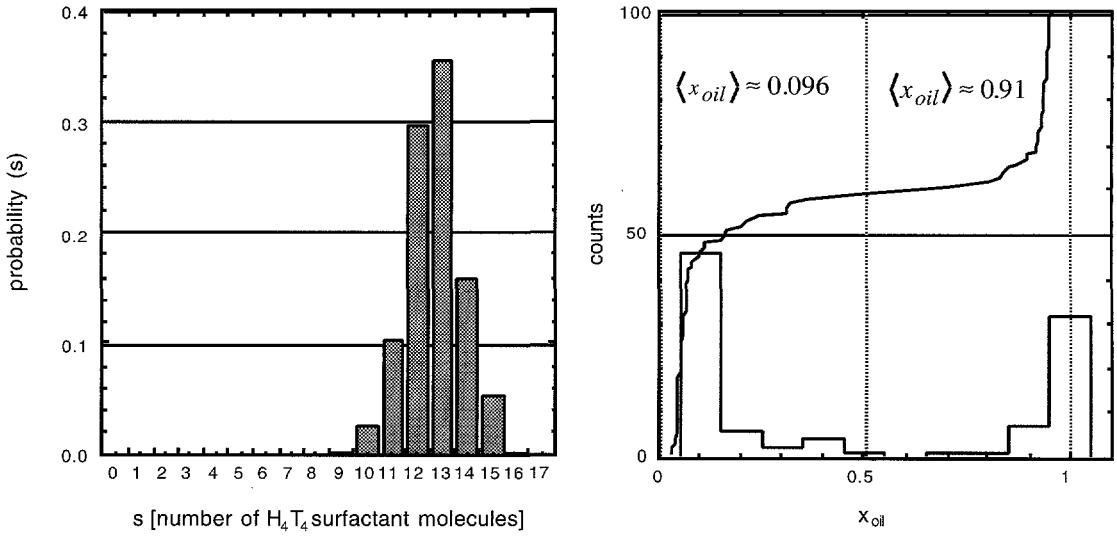
Mathematics and Computational Science

Novel Monte Carlo Algorithms for Statistical Mechanics

James Gubernatis

Monte Carlo simulations of many interesting and important physical systems often require sampling from multimodal distributions in the phase space of the key measurable quantities like the energy and magnetization. We are seeking significantly more efficient Monte Carlo algorithms for these types of problems. This year we emphasized problems near first-order phase transitions, where the distributions are bimodal, and adopted several different model systems. For a three-dimensional hard-sphere system near its solid-liquid phase transition, we devised a technique that combines Monte Carlo configurations for the same particle number into a new "super" configuration that maintains the correct partitioning of the system into a solid and liquid phase. This combination should improve the efficiency of the simulation in the phase coexistence region.

We also developed a Monte Carlo procedure that, for a given phase, samples a large region of configuration space by adding unweighted moves in temperature and volume to the standard moves in energy. For a Lennard-Jones model, we can now obtain in one Monte Carlo simulation the equation of state for a given phase over a wide range of densities and temperatures. We began investigating similar techniques across phase boundaries. For an Ising model realization of a micelle solution, we combined the inverse-restricted sampling method with the Swendsen-Wang cluster method and produced a technique that allows the direct estimation of the free energy in addition to the model's equation of state (see accompanying figure).



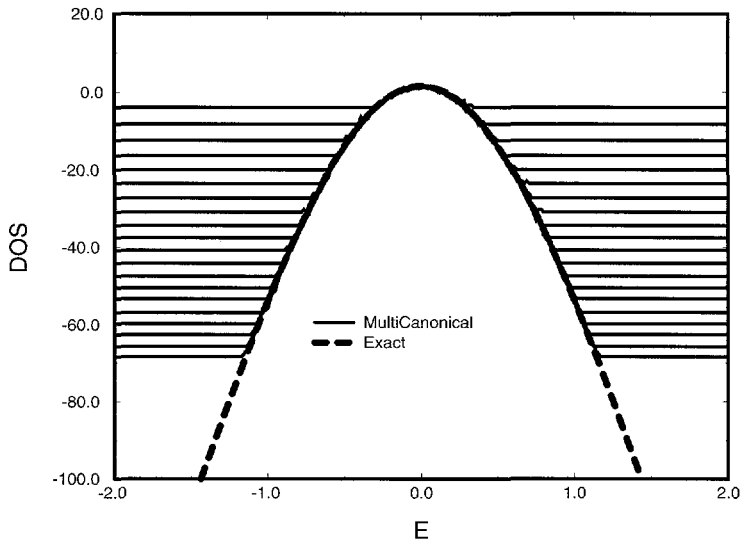
Monte Carlo simulation of a micellizing solution of surfactant chain molecules in a mixture of oil and water (left panel). The most stable phase is water-rich, with oil and surfactant concentrations each approximately 10%. The right panel shows observations of two-phase solubilization phenomena, as illustrated by the two peaks in the histogram. The horizontal and vertical lines are guides to the eye. The remaining curve is the accumulated number of counts.

Density Function Estimation for Monte Carlo Simulations

James Gubernatis

We are applying advanced statistical methods to the Monte Carlo estimation of density distribution functions to improve the analysis of Monte Carlo data and the efficiency of Monte Carlo algorithms. This past year we emphasized the improvement of algorithms. In particular, we examined several ways to improve the adaptive density estimation step in the multicanonical method and applied them to the Ising model near its order-disorder transition. Comparing test results with exact results (see accompanying figure), we found that while they were highly accurate, we were unable to reduce the autocorrelation times significantly. We determined that the multicanonical algorithm is more limited by the proposal than by the density estimation.

We tested a new proposal in which we estimate the transition matrix probability function (a conditional probability), use it to bias the move, and then use it with the detailed balance condition to estimate the density function. This procedure reduced the autocorrelation times. We are testing this procedure on the three-dimensional random-bond Ising model near its glass transition temperature, where the efficiency of the standard multicanonical method is greatly reduced. Below the glass transition temperature, we also developed a novel extension of the multicanonical method that biases the Monte Carlo moves by an adaptively determined bivariate distribution in the energy and order parameter instead of the standard method, which determines a distribution in just the energy. This new approach appears effective for small systems and is being tested for larger systems.



Calculation of the density of states (DOS) of the two-dimensional Ising model by the multicanonical method. The solid lines are the DOS learned by sucessive iterations of this adaptive Monte Carlo method. The dashed line is the exact DOS.

Geometrically Compatible, 3-D Monte Carlo and Discrete-Ordinates Methods

Jim E. Morel

The first objective of this project was to develop a discrete-ordinates (S_n) radiation transport code capable of modeling complex three-dimensional (3-D) geometries. The second objective was to modify the general-geometry Monte Carlo transport code, MCNP, to use adjoint or importance solutions from the 3-D, tetrahedral-mesh S_n code for purposes of variance reduction. The S_n code that we developed was the first 3-D, unstructured-mesh S_n code ever developed. As such, it provides us with a unique capability for efficiently and accurately performing calculations in complex 3-D geometries. In addition, this S_n code can generate adjoint or importance solutions that can be used by the MCNP code to dramatically reduce the statistical variance obtained in difficult Monte Carlo calculations.

In the past year, we developed and implemented several new techniques for improving the accuracy of our S_n code for difficult problems having collimated sources and detectors. In addition, we modified the MCNP code so that it could accept importance functions from our tetrahedral-mesh S_n code. Numerical testing on problems related to the design of neutron-based, oil-well logging tools indicated that the scheme works extremely well. The run time for the Monte Carlo computations was decreased by a factor of 30 or more on all of the test problems.

Theoretical Foundation for Adaptive Monte Carlo

Richard R. Picard

The Monte Carlo method is an important analysis tool for many complex problems, especially in the nuclear arena, in which computer simulation is more cost-effective than conducting physical experiments. This project is one of several that are aimed at improving the effectiveness of such Monte Carlo simulations.

The standard central-limit theorem, applicable to Monte Carlo work where N simulation runs are statistically independent, provides for the standard deviation to converge to zero at the rate of the square root of N. Under restrictive conditions, the replacement of independent sampling with adaptive learning algorithms has recently been shown to provide exponential convergence. The goal of this project is to obtain exponential convergence under less restrictive conditions.

In this second year of the effort, we successfully extended the major result of the first year (theoretical proof of exponential convergence for discrete-state problems) to the more general context of continuous-state spaces. To illustrate the method, we applied it to an idealized one-dimensional particle transport setting ("the rod problem").

In addition, we made progress on obtaining valid error estimates for Monte Carlo estimates from low-discrepancy sequences (i.e., quasi-random numbers). These sequences can be quickly generated and often lead to convergence at rates better than the square root of N.

Our efforts related to statistical mechanics focused on the use of proposed transitions in the Metropolis algorithm. As applied to multi-canonical methods, this approach allows for the accumulation of information from iteration to iteration, thereby improving efficiency over existing methods.

Publications

Kollman, C., et al., "Adaptive Importance Sampling on Discrete Markov Chains" (submitted to *Ann. Appl. Probability*).

Evolutionary Computation

Christian Reidys

The objective of this project is to extend the mathematical basis for a theory of computer simulation of dynamical systems. This theory will lead to a practical engineering science of simulation applicable to socio-technical and physical systems simulated on a computer. It will refine and extend the approach to achieve the encompassing mathematical theory of computer simulation by defining how the properties of dynamical systems generated depend on underlying rules together with their structural and logical constraints and the order in which the rules are iterated.

In our first year's work, we have made considerable progress in establishing credibility for what we call the "Theory of Simulation." We believe we have established general properties of such a formal simulation and engendered wide community discussion of the subject. We have related formal simulations to molecular properties to study computationally relevant properties of molecules and explored the possibility of mutation-based molecular computation. Using the genotype-phenotype paradigms, we worked on the development of mathematical foundations for a theory of computer simulation. For this theory we employ sequentially updated cellular automata over arbitrary graphs as a paradigmatic framework. In the development of the theory, we focus on the properties of causal dependencies among local mappings in a simulation.

We have completed a series of investigations of random components in n-cubes. One central aspect for the thorough understanding of evolutionary computation is the mapping between genotypes and phenotypes and aspects of the modeling of such maps. We worked on maps defined on n-cubes and achieved relevant structure theorems on connectivity and components of induced subgraphs of generalized n-cubes. We have also looked at connectivity in random Cayley graphs with a view to prove some very general and useful results on "search-spaces" in the context of update schedules.

Publications

Barrett, C.L., and C. Reidys, "Elements of a Theory of Computer Simulation I: Sequential CA Over Random Graphs" (to be published in *Appl. Math. Comput.*).

Reidys, C.M., "Random Induced Subgraphs of Generalized N-Cubes," *Adv. Appl. Math.* **19**, 360 (1997).

Reidys, C.M., "Random Subgraphs of Cayley Graphs Over p-Groups" (submitted to *Eur. J. Combinatorics*).

Adaptive Monte Carlo Methods for Radiation Transport

Thomas Booth

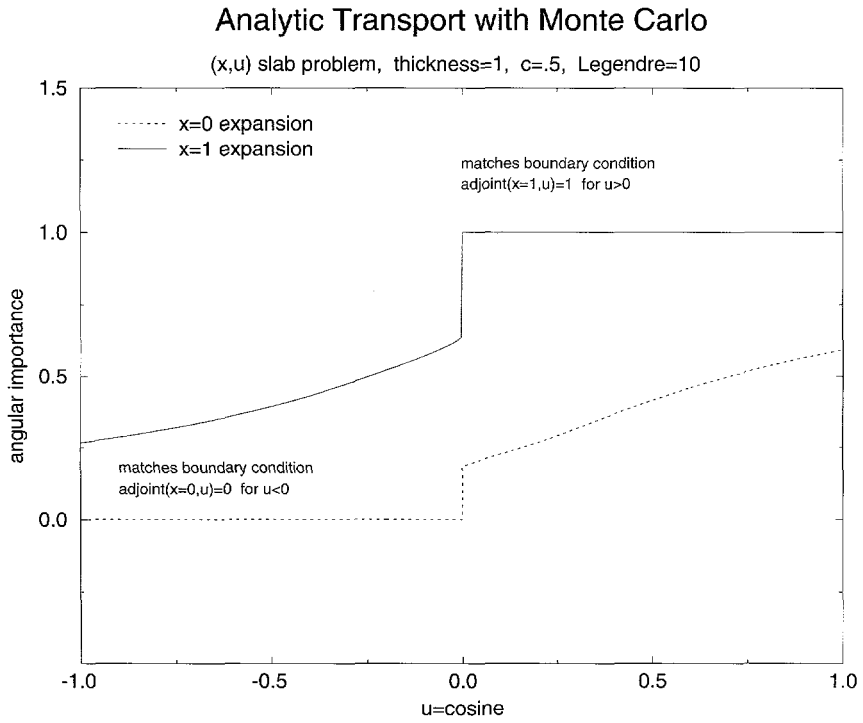
During the first two years of this project, we have sought to determine the feasibility of exponential convergence for simple one- and two-dimensional (1- and 2-D) continuous transport problems. The ongoing goal will be to generalize these developed exponential convergence methods and apply them to problems that currently cannot be solved. In this project we are working with external collaborators from the Claremont group.

The work this year was aimed at an extension of earlier results, when we obtained the first exponential convergence for the one-speed, 1-D continuous space problem. We sought to extend the results to more general 1-D continuous space problems and to extend the results to 2-D continuity in both space and angle (the "slab problem"). Additionally, the project sought to investigate the apparent false-convergence protection procedure discovered earlier.

We have successfuly derived a new analytic transport result for the slab problem and written a Monte Carlo method coded to estimate the analytic transport solution coefficients (see first figure). All expansion coefficients of the importance function can now be estimated simultaneously with zero variance. This multiple zero-variance technique has widespread applicability.

We have implemented a method for false-convergence protection that has been empirically demonstrated on many bidirectional transport problems over a wide range of parameters. Additionally, it empirically obviates Kollman's artificial delta. The protection method has been given to the statisticians for possible mathematical proof.

Our Claremont collaborators showed exponential convergence with the reduced-source method on the continuous bidirectional problem. Also, they obtained exponential convergence on a special slab problem with the reduced-source method (second figure). Studies of discrete transport problems indicate that the exponential convergence rate is faster for zero-variance methods than for reduced-source methods.



An analytic transport method was used to specify the form of the exact solution to the slab problem, and the Monte Carlo method was used to supply the coefficient functions for the analytic form. This extremely tight coupling between an analytic method and Monte Carlo allows Monte Carlo to match the exact solution to within 0.0001 with only 10 Legendre terms.

Publications

Booth, T.E., "Exponential Convergence with Adaptive Monte Carlo," *Trans. Am. Nucl. Soc.* **77**, 178 (1997).

Booth, T.E., "Exponential Convergence on a Continuous Monte Carlo Transport Problem," *Nucl. Sci. Engin.* **127**, 3 (1997).

Booth, T.E., "Monte Carlo Estimates of Transport Solutions to the Isotropic Slab Problem" (submitted to *Nucl. Sci. Engin.*).

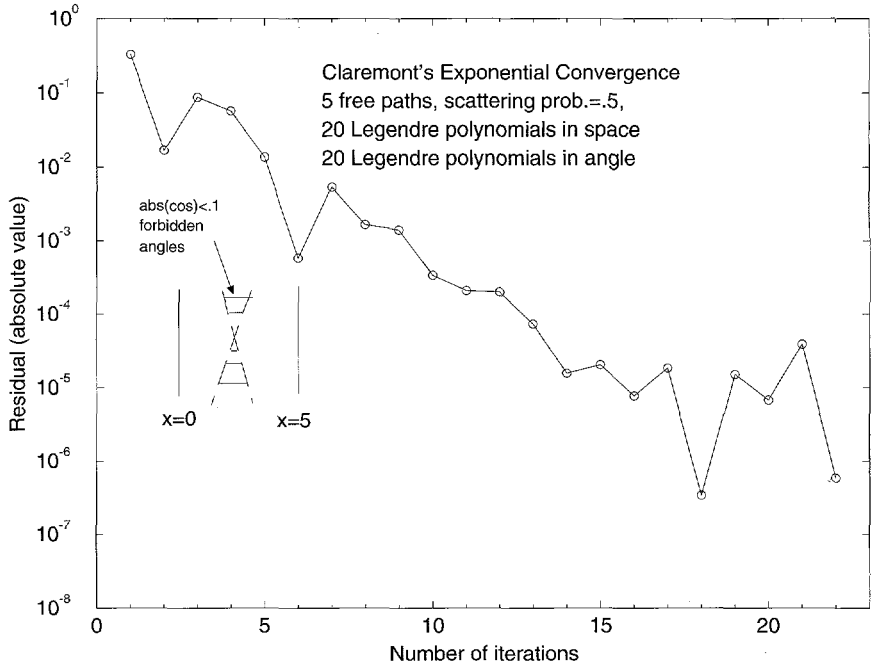
Booth, T.E., "Progress in the Quest for Exponential Convergence with Adaptive Monte Carlo" (Code Developers Conference, San Diego, CA, October 22–25, 1996).

Booth, T.E., "Simultaneous Monte Carlo Zero-Variance Estimates of Several Correlated Means," *Trans. Am. Nucl. Soc.* **77**, 185 (1997).

Booth, T.E., "Simultaneous Monte Carlo Zero-Variance Estimates of Several Correlated Means" (to be published in *Nucl. Sci. Engin.*).

Lichtenstein, H., "Generalization of Booth's Code for Adaptive Monte Carlo Transport" (available online, *http://www-xdiv.lanl.gov/XTM/projects/mc21/docs/*).

Lichtenstein, H., "Parametric Investigation of Potential False Learning in Adaptive Monte Carlo Transport" (submitted to *Nucl. Sci. Engin.*).



The reduced-source method with a double Legendre flux expansion in space and angle leads to exponential convergence for a special slab problem having continuous space and angle dependence.

Advancing X-Ray Hydrodynamic Radiography: Radiography Chain Model

Daniel Prono

A major component of science-based stockpile stewardship is advanced x-ray radiography, which must furnish two-dimensional (2-D) images from multiple lines of sight to create time-dependent, three-dimensional [3-D(t)] reconstructions of primary systems undergoing hydrodynamic tests. These reconstructions must have sufficient spatial and temporal resolution of the primary's internal structure and accurate enough density distributions to validate hydrocodes and infer nuclear criticality. What makes this task daunting is that we must develop analytical and computational techniques that can deal with "real-life" data and its imperfections: noise, scattered signals, motion-, source-, and detector-blur, detector sensitivity, and other systematic errors. Also, analysis must address the fundamental issue of how many experimental images and different lines of sight are needed to generate 3-D(t) reconstructions with enough accuracy and structural detail.

Accomplishments during the year include the following:

- In experiments at the microtron facility, we quantified systematic errors of source and detector blur, cone-beam illumination of a test object, detector sensitivity, edge resolution, and 3-D shape identification. Sources and magnitude of scattered x-ray signals were also quantified and used to validate transport-code simulations performed on the Accelerated Strategic Computing Initiative (ASCI) Blue Machine. Our results were presented at two conferences and were used in the performance specifications for the second-axis project of the Dual-Axis Radiographic Hydrodynamic Test (DARHT) Facility that is being built at Los Alamos.
- We created new analysis and optimization algorithms for studying the dependence of 3-D(t) reconstruction accuracy on the number of lines of sight. These results were presented at review meetings for the Advanced Hydrodynamic Facility (AHF) and its external advisory committee and form the basis for defining AHF system parameters.
- We created 3-D synthetic reconstructions based on hydrocode predictions and compared them with 2-D radiographic data from tests on weapon configurations. We used the results to identify the resolution needed to distinguish 3-D features and density distributions. Our results were presented at international weapons meetings.
- We started, using the ASCI Blue Machine, to develop code linkages between weapons-design hydrocodes, neutronics codes that infer criticality, and 3-D(t) reconstruction codes that build 3-D images from synthetic radiographs of primary systems undergoing hydrotests. The synthetic radiographs are based on real 2-D radiography data taken from single lines of sight.

Self-Organization and Pattern Formation

Robert Ecke

The formation of regular structures is important in science and technology, and recently there has been much attention focused on self-organizing and self-assembling systems. Instead of building structures by hand, as in microelectronic lithography, one would design or search for systems that put themselves together. If successfully developed, self-organizing and self-assembling systems could have a major impact on a wide range of technologically useful applications.

One variety of such systems is pattern-forming systems that can produce simple regular and irregular structures. Two examples of pattern formation are thermal convection and reaction-diffusion systems, in which two-dimensional patterns of stripes, spirals, targets, hexagons, squares, and "self-replicating" spots have been observed and modeled. The elucidation of the types of patterns, their dynamics, and the possible application of such mechanisms to problems of technological interest is a large part of our work. Specific systems we have studied are spatiotemporal patterns in cellular automata, vortex structures in rotating thermal convection, replicating spot dynamics in reaction-diffusion systems, instability and turbulence in flowing soap films, and self-organized criticality.

Our important accomplishments include deriving a new set of kinematic equations for two-dimensional bistable media that allows the study of front dynamics. The resulting equations describe naturally the core structure of a spiral wave without a special parameterization at the spiral tip. We have also discovered a new scaling regime in self-organized criticality. We have further constructed an experimental facility for flowing soap films and measured the properties of two-dimensional vortex streets and turbulence (see accompanying figure). In the area of vortex structures, we have analyzed mushroom formation in impulsively accelerated gas curtains, and we have characterized the formation of vortices from the unstable boundary layer in thermal convection.

Publications

Boettcher, S., "Aging Random Walks" (submitted to *Am. J. Phys.*).

Boettcher, S., and M. Paczuski, "Aging in a Model of Self-Organized Criticality," *Phys. Rev. Lett.* **79**, 889 (1997).



The coalescence of vortices behind a turbulent bluff body in a two-dimensional flowing-soap-film experiment. The intensity indicates film thickness (lighter areas are thicker). These measurements are critical in determining the correlation of thickness and velocity fluctuations in the real films.

Hagberg, A., and E. Meron, "The Dynamics of Curved Fronts: Beyond Geometry," *Phys. Rev. Lett.* **78**, 1166 (1997).

Hagberg, A., and E. Meron, "A Mechanism for Spatio-Temporal Disorder in Bistable Reaction-Diffusion Systems," *Nonlinear Science Today* (available online, *http://www.springer-ny.com/nst/nstarticles.html*).

Hagberg, A., et al., "Order Parameter Equations for Front Transitions: Planar and Circular Fronts," *Phys. Rev. E* **55**, 4450 (1997).

Liu, Y., and R.E. Ecke, "Heat Transport Scaling in Turbulent Rayleigh-Benard Convection: Effects of Rotation and Prandtl Numbers," *Phys. Rev. Lett.* **79**, 2257 (1997).

Nowak, E., et al., "Density Fluctuations in Vibrated Granular Materials" (submitted to *Phys. Rev. E).*

Paczuski, M., and S. Boettcher, "Avalanches and Waves in the Abelian Sandpile Model" (submitted to *Phys. Rev. E).*

Rightley, P.M., et al., "Evolution of a Shock-Accelerated Thin Fluid Layer," *Phys. Fluids* **9**, 1770 (1997).

Vorobieff, P., and R.E. Ecke, "Flow Structure in a Rayleigh-Benard Cell upon Impulsive Spin-up" (to be published in *Proc. 1997 ASME Meet.*).

Vorobieff, P., and R.E. Ecke, "Regular and Chaotic Flow Patterns upon Spin-up of a Rayleigh-Benard Convection Cell" (to be published in *Proc. 4th Experimental Chaos Conference, August 1997*).

Vorobieff, P., and R.E. Ecke, "Vortex Structure in Rotating Rayleigh-Benard Convection" (to be published in *Physica D*).

Wang, X., and C. Doering, "Attractor Dimension Estimates for Two-Dimensional Sheer Flows" (to be published in *Physica D*).

Simulation Methods for Advanced Scientific Computing

James Gubernatis

The rapid increase in raw computational power has created exciting opportunities for scientific computation. Accordingly, scientific computing is enjoying considerable worldwide popularity. Yet many challenging scientific problems remain unsolved because of inadequate algorithms, especially algorithms that are suitable for massively parallel architectures.

In this project our objective is to create effective new algorithms for solving these problems, giving the Laboratory the initiative in key technical areas by combining the talents and efforts of our physical scientists and applied mathematicians. We concentrate on developing advanced n-body simulation techniques, including classical and quantum Monte Carlo techniques, molecular dynamics techniques, and Langevin methods for stochastic differential equations. We are also improving current algorithms used to simulate problems with disparate time and/or space scales.

This year we focused on three problems. We sought to develop more efficient ways to perform quantum Monte Carlo simulations in real time. Here, we performed preliminary tests on an extension of the stationary-phase Monte Carlo method. For "toy" problems the new methods worked very well.

We also sought to develop ways to compute excited states in the diffusion quantum Monte Carlo method. Our approach was to compute concurrently many excited states in a natural-orbital-like basis. Preliminary results on quantum oscillators appear promising. More complicated problems are under study.

Finally, we sought ways to compute imaginary-time correlation functions in the constrained-path quantum Monte Carlo method. The issue here is one of numerical stability. In some cases we can compute even superconducting pairing-correlation functions for the Hubbard model for large imaginary times. We are presently examining advanced matrix decomposition methods to handle the other cases.

Publications

Zhang, S., "Pairing Correlations in the Two-Dimensional Hubbard Model," *Phys. Rev. Lett.* **78**, 4486 (1997).

Zhang, S., et al., "A Constrained Path Monte Carlo Method for Fermion Ground States," *Phys. Rev. B* **55**, 7464 (1997).

Multiscale Science for Science-Based Stockpile Stewardship

Len G. Margolin

Our overall goal is to develop and apply the methods of multiscale science to the problems of materials characterization and of fluids and materials mixing. The specific focus is on the science-based stockpile stewardship issue of assessing performance of a weapon with off-design, aged, or remanufactured components in the absence of full-scale testing. Our product will be physics models that are based on microphysical principles and parameters, and that are suitable for implementation in the large-scale design and assessment codes used in the weapons program.

We have organized our research into three general areas. The first is the evolution of interfacial fluid instabilities and the resulting interpenetration mix of materials. We are developing a multiphase description of Rayleigh-Taylor instability growth including the evolution of region boundaries and are doing direct numerical simulations to construct and validate closures. Also, we are combining turbulence and multiphase models, using the turbulent estimate of length scales to construct the drag coefficients between fluid phases. We are looking at crenulation, which is a bulk instability, in a converging nonhomogeneous material, based on a turbulence transport theory approach.

The second research area is the prediction of material properties as a function of microstructure. Our studies of the spallation process in ductile metals are based on void growth and include the effects of damage clustering and void linking. In addition, we are modeling interfacial slip and using large molecular dynamics calculations to validate our theories. We are using large-scale quantum molecular dynamics simulations to simulate the dynamical properties of hot dense mixtures of materials of greatly differing mass ratios.

The third research area is the study of the formation of surface ejecta from a shocked metal/gas interface and the subsequent ejecta transport. We have begun developing a discrete model for metal surfaces by adding contact forces representing grain bonding to a smooth-particle hydrodynamics code. We are also developing new formulations of macroscopic multiphase equations, based on a microscopic model of suspended particles. The new formulation aims to include the effects of particle-size

distributions and particle/particle interactions, both short-range (through wakes) and long-range effects. In addition, we are studying the mass and energy transfer (interfacial) processes between the phases, including the growth/evaporation rate of spherical particles immersed in a thermal plasma.

To foster meaningful interactions and to tie the research more closely to the needs of the nuclear design community, we sponsor biweekly seminars. We also schedule informal, but more deeply focused, discussions on current research topics.

Publications

Clark, T., "Two-Point Closures and Statistical Equilibrium: Implications for Engineering Turbulence Models," in *Proc. NASA/ICASE/LaRC/AFSOR Symposium on Modeling Complex Turbulent Flows* (Kluwer Academic Publishers, Dordrecht, Netherlands, 1997).

Clark, T., and C. Zemach, "Symmetries and the Approach to Spectral Equilibrium in Isotropic Turbulence" (submitted to *Phys. Fluids*).

Glimm, J., and D.H. Sharp, "Stochastic Partial Differential Equations: Selected Applications in Continuum Physics," in *Stochastic Partial Differential Equations: Six Perspectives, Mathematical Surveys and Monographs*, R.A.Carmona and B.L. Rozovskii, Eds. (American Mathematical Society, Providence, RI, 1997).

Glimm, J., et al., "The Statistical Evolution of Chaotic Fluid Mixing" (submitted to *Phys. Rev. Lett.*).

Hammerberg J.E., and J. Pepin, "An Analytic Solution to a Driven Interface Problem" (APS Topical Conf. on the Shock Compression of Condensed Matter, Amherst, MA, July 27–Aug. 1, 1997).

Hammerberg, J.E., et al., "Non-linear Dynamics and the Problem of Slip at Material Interfaces" [to be published in *Physica D (Amsterdam)*].

Lenosky, T., et al., "Molecular Dynamics Modeling of Shock-Compressed Liquid Hydrogen," *Phys. Rev. B* **55,** 11907 (1997).

Nadiga, B.T., et al., "On Simulating Flows with Multiple Time Scales Using a Method of Averages" (to be published in *Theor. Comput. Fluid Dyn.*).

Plohr, B.J., and D.H. Sharp, "Instability of Accelerated Metal Plates" (submitted to *Z. Angew. Math. Phys.*).

Roder, J., et al., "A Multichain Frenkel-Kontorova Model for Interfacial Slip" (to be published in *Phys. Rev. B*).

Speziale, C.G., et al., "On the Origin and Structure of Self-Preserving Solutions in Homogeneous Turbulence" (submitted to *J. Fluid Mech.*).

Tonks, D., et al., "Quantitative Analysis of Damage Clustering and Void Linking for Spallation Modeling in Tantalum," *J. Phys. IV* **C3**, 841 (1997).

Tonks, D., et al., "Spallation Modeling in Tantalum" (International Workshop on New Models and Numerical Codes for Shock Wave Processes in Condensed Media, Oxford, UK, Sept. 1997).

Tonks, D., et al., "Spallation Studies on Shock Loaded Uranium" (APS Topical Conf. on Shock Compression of Condensed Matter, Amherst, MA, July 27–Aug. 1, 1997).

Advanced Three-Dimensional Eulerian Hydrodynamic Algorithm Development

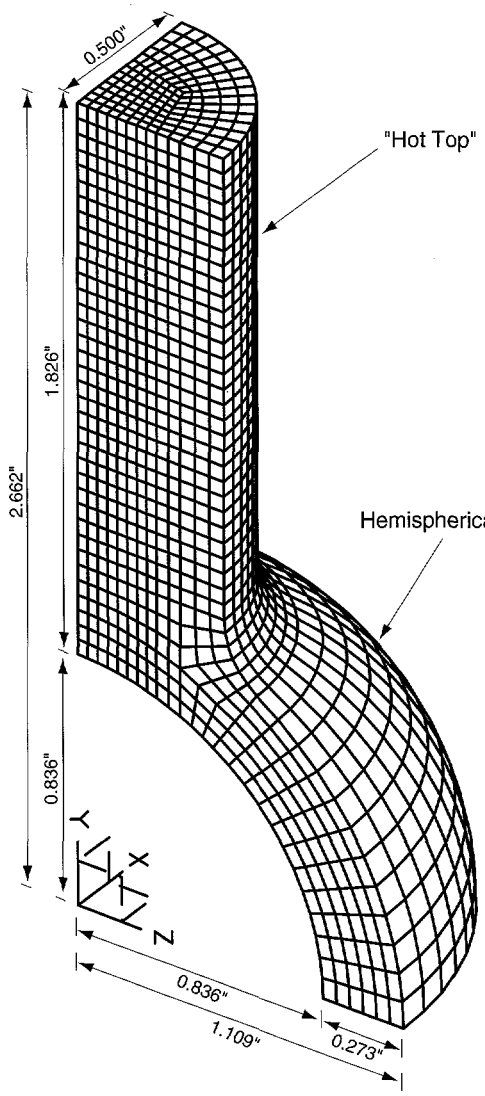
William J. Rider

The purpose of this project is to investigate, implement, and evaluate algorithms that have high potential for improving the robustness, fidelity, and accuracy of three-dimensional (3-D) Eulerian hydrodynamic simulations. Eulerian computations are necessary to simulate a number of important physical phenomena, including the molding process for metal parts, nuclear weapons' safety issues, and astrophysical phenomena. This research explores a number of algorithmic issues: interface/volume tracking, surface physics integration, multilevel iterative methods, multimaterial hydrodynamics, and coupling radiation with hydrodynamics.

Several of our efforts from the last two years significantly changed the basis for determining the fidelity of interface tracking. The tests we proposed are becoming new standards in testing (and improving) interface tracking. Our efforts also produced a new high-fidelity standard for integrating low-speed flow hydrodynamics with interfaces.

During this year we continued (1) developing unsplit advection algorithms for improved accuracy and robustness, with a focus on multimaterial shock hydrodynamics and low-speed hydrodynamics in conjunction with the TELLURIDE code; (2) developing unsplit volume-of-fluid methods for multimaterial shock hydrodynamics and 3-D low-speed flow; (3) implementing the surface tension model in TELLURIDE and improving its ability to track subgrid material property change fronts, such as melting and solidification fronts (see the figure); (4) improving the basis for the implementation of multimaterial

integrators, including mixed-cell thermodynamics and "stiff" equation-of-state Riemann solvers; (5) "field" testing and disseminating the parallel communication libraries in a wider realm of simulation codes; and (6) developing the basic technology for implementing dynamic load balancing on parallel computer architectures of the future. Further details on progress in some of these areas are given in the publications.



Computational mesh (6480 cells) used for TELLURIDE simulation of copper chalice mold filling. Note that the computational model is one-fourth of the full physical model.

Publications

Knoll, D.A., and W.J. Rider, "A Multigrid Preconditioned Newton-Krylov Method" (submitted to *SIAM J. Sci. Comput.*).

Knoll, D.A., and W.J. Rider, "Multilevel Newton-Krylov Methods for Nonsymmetric, Nonlinear Boundary Value Problems" (10th International Conference on Domain Decomposition, Boulder, CO, Aug. 10–14, 1997).

Kothe, D.B., "Perspective on Eulerian Finite Volume Methods for Incompressible Interfacial Flows" (Free Surface Flow Workshop, Udine, Italy, September 1–5, 1997).

Kothe, D.B., et al., "A High Resolution Finite Volume Method for Efficient Parallel Simulation of Casting Processes on Unstructured Meshes" (8th SIAM Conference on Parallel Processing for Scientific Computing, Minneapolis, MN, March 14–17, 1997).

Puckett, E.G., et al., "A Second-Order Projection Method for Tracking Fluid Interfaces in Variable Density Incompressible Flows," *J. Comput. Phys.* **130**, 269 (1997).

Reddy, A.V., et al., "High Resolution Finite Volume Parallel Simulations of Mould Filling and Binary Alloy Solidification on Unstructured 3-D Meshes" (SP97: The Fourth Decennial International Conference on Solidification Processing, The University of Sheffield, UK, July 7–10, 1997).

Rider, W.J., "An Adaptive Riemann Solver Using a Two-Shock Approximation" (submitted to *Comput. Fluids*).

Rider, W.J., "Filtering Nonsolenoidal Modes in Numerical Solutions of Incompressible Flows" (to be published in *Int. J. Numer. Methods Fluids*).

Rider, W.J., and D.A. Knoll, "Solving Nonlinear Heat Conduction Problems with Multigrid Preconditioned Newton-Krylov Methods" (to be published in *Lect. Notes Comput. Appl. Math.*).

Rider, W.J., and D.B. Kothe, "Constrained Minimization for Monotonic Reconstruction" (13th AIAA Computational Fluid Dynamics Conference, Snowmass, CO, June 29–July 2, 1997).

Rider, W.J., and D.B. Kothe, "Reconstructing Volume Tracking" (submitted to *J. Comput. Phys.*).

Rider, W.J., et al., "Accurate and Robust Methods for Variable Density Incompressible Flows with Discontinuities," in *Proceedings of the Workshop on Barriers and Challenges in CFD* (Kluwer Academic Publishers, Norwell, MA, in press).

Crisis Forecasting

Andrew White

This year we focused on two problems relevant to crisis forecasting: (1) fire weather prediction and (2) epidemiological modeling and analysis. We demonstrated the advantages of combining a hydrodynamic model with a traditional fire-behavior model by simulating the South Canyon fire near Glenwood Springs, Colorado, and the Calabasas fire near Malibu, California. In both of these simulations, the combination of the heat produced by the fire and the air flow over complex terrain produced blow-up phases, as observed in the fires themselves. The fires were driven up steep slopes by fire-induced winds that were much stronger than ambient winds.

For the South Canyon fire, the simulations produced results that agree well with the observed fire behavior (see first figure). The total length of time required for the simulated fire to race from the bottom of South Canyon to the ridge line was approximately 18 minutes, which agrees well with the published accounts of the actual fire spread.

For the Calabasas fire, the simulated fire moved up the steep slope of Malibu Bowl in several minutes. For this simulation, the heat released by the fire extended several hundred feet above the active fire line. The simulation clearly reveals how an intense wildfire can be carried by fire-induced winds that are much stronger than the ambient winds (see second figure).

Although the traditional fire models are capable of describing significant aspects of wildfire behavior, they are empirically based and thus of restricted value for a wide scope of predictions. Accordingly, we have also developed a physics-based model for wildfires that has proved capable of accounting for complex terrain, variable types of fuels, and unsteady variable winds in the prediction of the most probable wind-fire behavior.

Publications

Ahrens, J., et al., "Wildfire Visualization" (IEEE Vizualization Conference, Phoenix, AZ, October 19–24, 1997).

Bossert, J., et al., "Coupled Weather and Wildfire Behavior Modeling at Los Alamos: An Overview" (Second Symposium on Fire and Forest Meteorology, Phoenix, AZ, January 11–16, 1998).

Linn, R., and F. Harlow, "FIRETEC: A Transport Description of Wildfire Behavior" (Second Symposium on Fire and Forest Meteorology, Phoenix, AZ, January 11–16, 1998).

Reisner, J., et al., "Numerical Simulations of Two Wildfire Events" (Second Symposium on Fire and Forest Meteorology, Phoenix, AZ, January 11–16, 1998).



Image of the South Canyon fire simulation after 10 minutes of simulated time. The black areas have already burned. The active fire line is midway up the slope, with light shading showing the high-temperature air moving upslope ahead of the fire line.

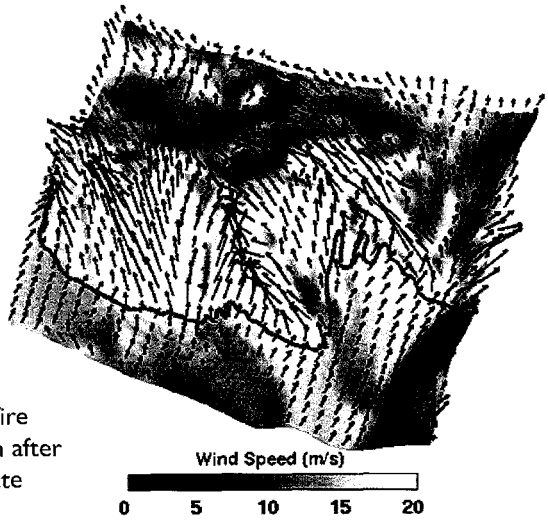


Image of wind vectors, wind speed, and fire perimeter from the Calabasas simulation after 30 minutes. Lighter shades of grey indicate higher wind speeds.

Applications of Nonlinear Stochastic Dynamics

Darryl Holm

Fundamental research in the dynamics of nonlinear and noisy systems, the aim of this project, has various applications in theoretical physics, chemistry, and biology. These applications typically involve interactions among nonlinearity, noise, and disorder in the dynamics of both simple and complex systems. The underlying theoretical problems have much in common within the paradigms of nonlinear science.

Last year, we (1) developed and applied cycle-expansion methods for nonlinear ray equations resulting from asymptotic expansions of linear partial differential equations, including the Schrödinger equation; (2) developed and analyzed models of molecular motors using nonlinear stochastic differential equations; (3) used a new variational formulation of global transport bounds to derive and analyze low-dimensional dynamical system models for turbulent, incompressible Couette-Taylor flows; (4) calculated nonequilibrium quantum dynamics of coupled-spin boson systems; (5) demonstrated stability properties of multiphonon bound states in classical and quantum discrete nonlinear systems; (6) analytically studied the interactions of mean fluid circulation with noise and disorder, which are applied to assessing predictability in large-scale ocean models; and (7) analytically and numerically studied new power-enhancement schemes for fiber-optic telecommunications, including use of the sliding-frequency guiding filter technique and dispersion-managed solitons. This last aspect of our research follows on our earlier work with nonlinear amplifying loop mirrors for fiber-optic telecommunications, whose patent application was completed this year.

Publications

Aceves, A., et al., "Homoclinic Orbits and Chaos in a Second-Harmonic Generating Optical Cavity," *Phys. Lett. A* **233**, 203 (1997).

Allen, J.S., et al., "A Note on Kelvin Waves in Balance Models," *J. Phys. Ocean* **27**, 2060 (1997).

Aronson, I., et al., "Ginzburg-Landau Theory of Spiral Surface Growth" (submitted to *Phys. Rev. Lett.*).

Ben-Naim, E., and P.L. Krapivsky, "Domain Number Distribution in the Nonequilibrium Ising Model" (submitted to *Phys. Rev. Lett.*).

Ben-Naim, E., and P.L. Krapivsky, "Domain Statistics in Coarsening Systems" (to be published in *Phys. Rev. E*).

Ben-Naim, E., and P.L. Krapivsky, "Stationary Velocity Distributions in Traffic Flows" (to be published in *Phys. Rev. E*).

Braun, O.M., et al., "Hysteresis in the Underdamped Driven Frenkel-Kontorova Model" (to be published in *Phys. Rev. Lett.*).

Cai, D., et al., "Low-Loss Nonlinear Transmission Lines in a Technologically Interesting Regime" (to be published in *Physica D*).

Cai, D., et al., "Resonance in the Collision of Two Discrete Intrinsic Localized Excitations" (to be published in *Phys. Rev. E*).

Cendra, H., et al., "The Maxwell-Vlasov Equations in Euler-Poincare Form" (to be published in *J. Math. Phys.*).

Clague, D.S., and R.J. Phillips, "Hindered Diffusion of Spherical Macromolecules through Dilute Fibrous Media," *Phys. Fluids* **8**, 1720 (1996).

Clague, D.S., and R.J. Phillips, "A Numerical Calculation of the Hydraulic Permeability of 3-Dimensional Disordered Fibrous Media," *Phys. Fluids* **9**, 1562 (1997).

Fabijonas, B., et al., "Secondary Instabilities of Flows with Elliptic Streamlines," *Phys. Rev. Lett.* **78**, 1900 (1997).

Gabitov, I., et al., "Generation of Soliton Pulse Trains from Sinusoidally Modulated cw Beams with Nonlinear Amplifying Loop Mirrors" (submitted to *Opt. Lett.*).

Gabitov, I., et al., "Low-Noise Picosecond Soliton Transmission Using Concatenated Nonlinear Amplifying Loop Mirrors," *J. Opt. Soc. Am. B* **14**, 1850 (1997).

Holm, D.D., and V. Zeitlin, "Hamilton's Principle for Quasigeostrophic Motion" (to be published in *Phys. Fluids*).

Holm, D.D., et al., "The Euler-Poincare Equations and Semidirect Products with Applications to Continuum Theories" (submitted to *Adv. Math.*).

Holm, D.D., et al., "Euler-Poincare Models of Ideal Fluids with Nonlinear Dispersion" (submitted to *Phys. Rev. Lett.*).

Konotop, V., et al., "Interaction of a Soliton with Point Impurities in an Inhomogeneous Discrete Nonlinear Schrödinger System" (to be published in *Phys. Rev. E*).

Raghavan, S., et al., "Relation between Dynamic Localization in Crystals and Trapping in Two-Level Atoms," *Phys. Rev. A* **54**, 1781 (1996).

Rasmussen, K.O., et al., "Dynamics of Nonlinear Localized States on Finite Discrete Chains," *Phys. Rev. E* **55**, 6151 (1997).

Sanchez, A., and A.R. Bishop, "Collective Coordinates, Lengthscale Competition and Complex Behavior in Soliton-Bearing Equations" (submitted to *SIAM Rev.*).

Atomic and Molecular Physics and Plasmas, Fluids, and Particle Beams

Short-Pulsed, Electric-Discharge Degradation of Toxic and Sludge Wastes

Louis Rosocha, Los Alamos National Laboratory
Frank J. Wessel and Vitaly M. Bystritskii, University of California, Irvine

The project is a collaborative effort with the University of California at Irvine (UCI). The main objective is to degrade aqueous-based pollutants, particularly organic compounds (e.g., chlorinated solvents and biphenyls), to harmless or more easily managed compounds, using a pulsed, electric-discharge chemical reactor (see accompanying diagram). The reactor parameters are 50- to 90-kV voltage amplitude, 100- to 150-ns pulse duration, 100- to 1000-Hz repetition rate, and 40-L volume. Electric field enhancement near 10- to 100-μm aerosol drops, combined with large specific water-to-air surface, results in the copious generation of energetic and hydrated electrons and other radicals (O, H, OH), thereby providing efficient degradation and partial dechlorination of the primary organic pollutant molecules.

Last year we modified the aerosol reactor for improved operation; this year we used it to process additional compounds. Estimated wall-plug specific energy in these tests was in the range of 100 to 200 eV per degraded molecule for paranitrophenol and 200 to 1500 eV per moleclule for dichlorophenol (DCP) and perchloroethylene; the amount of energy depends on the discharge parameters and the pollutant concentration. These values are many times lower than those for other aqueous-pollution treatment technologies (sonohydraulic, electrohydraulic, high-energy electron beam).

In additional experiments, we innoculated the aerosol-processed DCP water samples (having residual chlorination degree of 65% to 70%) with bacteria Pseudomonas mendocina and achieved up to 90% degree of dechlorination in the first 40 hours after inoculation, demonstrating the potential for a novel, two-stage treatment technology.

Publications

Bystritskii, V.M., et al., "Aerosol Plasma for Aqueous Waste Treatment" (24th IEEE International Conference on Plasma Science, Newport Beach, CA, May 19–22, 1997).

Bystritskii, V.M., et al., "Aerosol Pulsed Corona Discharge for Degradation of Water Borne Chlorinated Pollutants" (1997 High Voltage Workshop, San Diego, CA, May 6–8, 1997).

Bystritskii, V.M., et al., "Pulsed Discharge in Aerosol for Waste Water Clean-Up," *Bull. Am. Phys. Soc.* **41**, 1641 (1996).

Bystritskii, V.M., et al., "Pulsed Power for Advanced Waste Water Remediation Technology" (to be published in *Proceedings of the 11th IEEE International Pulsed Power Conference*).

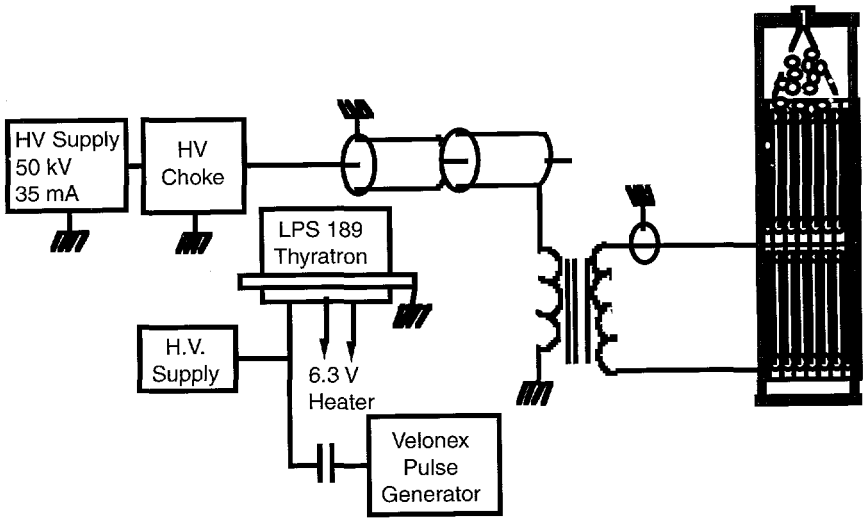


Diagram of the pulsed, electric-discharge aerosol reactor.

Advancing X-Ray Hydrodynamic Radiography: Multipulse Converter Development

Daniel Prono

To be able to resolve the features and material density distributions necessary for accurate three-dimensional (3-D) reconstructions, the next generation of x-ray radiography will need x-ray imaging flashes ~4 times more intense in a spot-size diameter 3 to 4 times smaller; therefore, energy densities will be >40 times higher than now possible. In addition, these flashes must be time-sequenced with ~250 to 500 ns between them in order to produce a "movie" of the temporal evolution of a hydrodynamic event.

The x-ray flashes are made by impacting a 50-ns, 4-kA, 20-MeV, 1-mm-spot electron beam onto a high-atomic-number metal plate. Because of the high energy densities involved, the converter targets will become plasmas and violently disassemble, ejecting a plasma plume in the ~500 ns between sequential incoming electron beams. Target-plasma interaction with, and coupling to, the incident electron beam can disrupt the beam spot size, thereby degrading image resolution. The interaction of primary concern is the electron beam's extraction of ions from the target plasma, causing an imbalance of space charge and altering beam trajectories. Our understanding of these interactions, gained over the past year, now leads us to identify them as a serious limitation on how far we can ultimately develop x-ray radiography.

Using computational modeling and simulations, we investigated the production of ejecta and plasma plumes when the converter targets are struck by high-energy-density electron beams. Depending upon target material (density, melt temperature, and specific heat are the most important properties) and target composition, ejecta speeds of 0.2 to 1.5 cm/μs can be achieved. We found that placing inertially confining cylinders on the target surface effectively inhibits the ejecta's radial expansion, thereby minimizing the expansion volume. We also performed experiments on beam/target interactions using the Laboratory's integrated test stand (ITS), which produces a 50-ns, 4-kA, 6-MeV electron beam focused to a 1.3-mm spot. The observed ejecta speeds and their dependence on material properties and geometries validated computational predictions.

Finally, using computational modeling and simulations, we investigated beam interaction with target plasmas, concentrating on the issue of time-dependent beam-spot degradation. Both two- and three-dimensional models indicate the seriousness of how ions extracted by the electron beam's self-potential will lead to an imbalance of space charge and to spot-size degradation. Experiments using ITS verified strong time-dependent, spot-size degradation under conditions of very high beam-energy density. We are computationally testing ways to control and mitigate ion extraction.

All of the above results were presented at the technical reviews and planning meetings for the Advanced Hydrodynamics Facility, at the technical reviews for the Dual-Axis Radiographic Hydrodynamics Test Facility, and at technical workshops convened by DOE.

Laser-Sheet Imaging of HE-Driven Interfaces

Robert Benjamin

Our primary goal has been the development of a new diagnostic tool called multiple imaging of laser-sheet illumination (MILSI) for experiments at high-explosive (HE) firing sites. In MILSI, a sheet of laser light illuminates a two-dimensional slice of a complex flow, and then a high-speed camera records multiple images of the event. This technique improves the understanding of shock-induced interfacial instabilities, which is a fundamental research problem in fluid dynamics and weapons physics. Experimental data produced by MILSI are valuable for the validation of fluid simulations developed under the Accelerated Strategic Computing Initiative.

We successfully fielded MILSI systems at two Los Alamos firing sites this year. We tested several combinations of lasers and cameras and found the best system to be an array of frequency-doubled yttrium-aluminum-garnet lasers for illumination and an eight-frame, intensified charge-coupled-device camera for recording. Tests with argon lasers and image-converter cameras produced less satisfactory results.

MILSI systems were fielded on two fluid instability experiments: (1) a "liquid curtain" experiment consisting of an initially frozen layer of a high-density fluid embedded in water, and (2) a single-interface instability experiment. In both experiments, fluorescent dye was added to the higher-density fluid (e.g., bromoform) so the images showed optical emission from this fluid. We observed the emitted fluorescence signal and the onset of the instability, but experimental difficulties involving the container prevented a measurement of the instability growth rate. Development of better shot containers will enable MILSI to measure instability growth.

Advanced Modeling of High-Intensity Accelerators

Robert Ryne

The primary goal of this project has been to advance the state of the art in modeling high-intensity accelerators through the use of high-performance computers and the development of software and algorithms targeted to these platforms. Such software and codes are essential for modeling the next generation of accelerators that will be used for waste transmutation, tritium production, and future spallation sources.

Last year we succeeded in developing and using fully three-dimensional (3-D), parallel beam dynamics codes on the SGI/CRI Origin2000 and Cray T3E parallel computers. We used these codes to perform the largest simulations to date using the Accelerator Production of Tritium (APT) accelerator as a test case. We also performed detailed comparisons of 2-D and 3-D space charge algorithms. We used our 3-D code to analyze halo formation in a new class of 3-D equilibria. This analysis showed that for elongated beam bunches, the longitudinal beam halo develops more quickly than the transverse halo (for comparable longitudinal and transverse mismatches). We also developed parallel Langevin and direct Fokker-Planck codes, and we used the Langevin code as another means to produce equilibria used as the starting point in beam halo studies.

Our accomplishments in this project and our track record in high-performance modeling of accelerators helped lead to approval in 1997 of a DOE Grand Challenge in Computational Accelerator Physics.

Publications

Ryne, R., "Beam Dynamics Simulations Using a Parallel Version of PARMILA," *Proc. XVIII Int. Linear Accel. Conf.* **1**, 234 (1997).

Ryne, R., and S. Habib, "Parallel Beam Dynamics Calculations on High Performance Computers," *Comput. Accel. Phys., AIP Conf. Proc.* **391**, 377 (1997).

Wangler, T., et al., "Dynamics of Beam Halo in Mismatched Beams," *Proc. XVIII Int. Linear Accel. Conf.* **1**, 372 (1997).

Fundamental Studies of Radiation Damage in Two-Phase Oxide Composites

Kurt Sickafus

Ceramic composites have great potential for use in high-radiation environments, although few studies have addressed radiation effects in such systems. For this research, we have refined the composite concept to address radiation effects in systems that allow for precipitation of single-crystal secondary phases within a single-crystal matrix. In particular, we are focusing on the systems $MgO{\cdot}TiO_2$, $MnO{\cdot}TiO_2$, and $FeO{\cdot}TiO_2$.

We have developed the capability to produce single crystals and composites in the system $MgO{\cdot}TiO_2$. We are characterizing these materials by using x-ray diffraction, optical and scanning electron microscopy, and transmission electron microscopy (TEM). Bulk and TEM samples of $MgTiO_3$ with TiO_2 and Mg_2TiO_4 precipitates were neutron-irradiated at the facility for the accelerator production of tritium at the Los Alamos Neutron Science Center (LANSCE). Oak Ridge National Laboratory has prepared TEM samples of pure $MgTi_2O_5$ and the $MgTiO_3$ composite for neutron irradiation. Characterization of these materials will begin once the irradiated samples are available.

In addition, we irradiated $MgTi_2O_5$ thin foils with electrons and ions while directly observing structural changes using TEM. During irradiation with 1-MeV electrons, we observed no changes up to 0.75 displacements per atom (dpa) between 300 and 1000 K. When irradiated with 1-MeV krypton, we found that the material amorphizes at 30 K after 0.1 dpa but remains crystalline at 310 K and 1.5 dpa. We performed time-of-flight neutron-scattering analyses on Fe_2TiO_5, $MgTiO_3$, $MnTiO_3$, and $FeTiO_3$ powders during the last beam cycle at LANSCE. We will use these data as base lines for comparison with neutron-irradiated samples.

Publications

Mitchell, J.N., et al., "Radiation Effects in Corundum Structure Derivatives" (to be published in *Nucl. Instrum. Methods Phys. Res., Sect. B*).

Generation and Compression of a Target Plasma for Magnetized Target Fusion

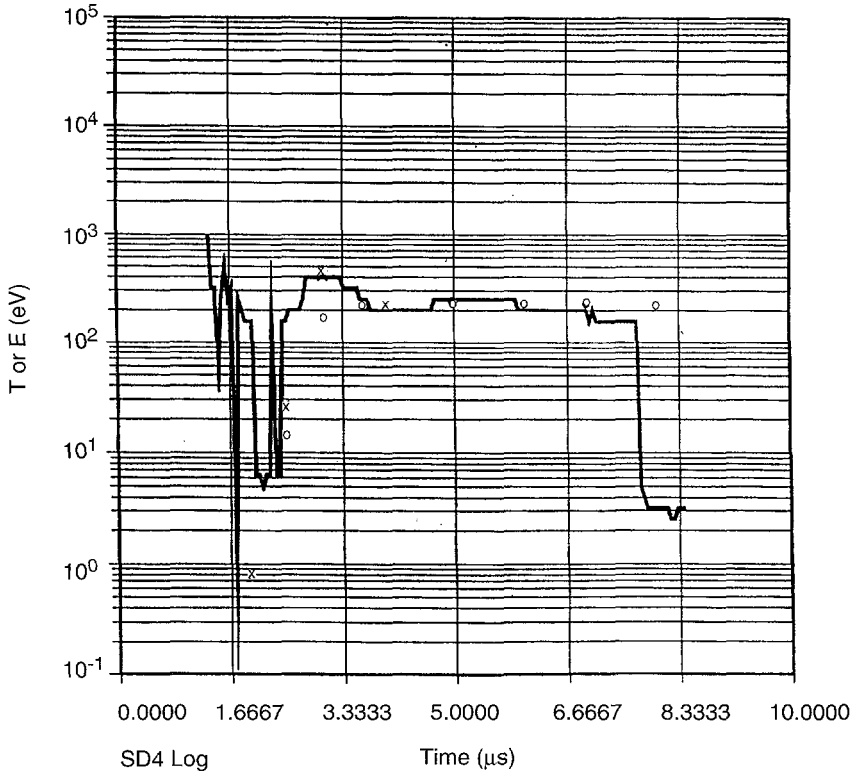
Ronald Kirkpatrick

Magnetized target fusion (MTF) is an approach to controlled thermonuclear fusion that uses a magnetic field to reduce electron thermal conduction and enhance the heating by the fusion reaction products in a fusion plasma. It involves two steps: first, creation of a warm target plasma and, second, compression of that plasma by an imploding shell, or "wall." The magnetic field and lower operation density reduce the energy loss from the plasma so that a slow compression is adequate for heating it to fusion temperatures. The slow compression and larger target size reduce both the power and the intensity that must be delivered to the target by the driver to achieve ignition, so efficient pulsed-power machines become viable candidates for igniting fusion reactions.

This year, a major goal of our research in MTF was to understand the filtered silicon diode signals for MAGO II and MAGO III. These two MTF target-plasma-generation experiments were performed jointly by Los Alamos and the All-Russian Scientific Institute for Experimental Physics. We were able to refine our analysis and present the results at two conferences. The results indicate that the MAGO plasma attains a temperature of about 200 eV and lasts for at least 5 μs, with a thermal relaxation time of about 10 μs. The temperature and density histories compare very favorably with the magnetohydrodynamic (MHD) code calculations (see accompanying graph). This good comparison increases our confidence that we are correctly modeling the atomic physics as well as plasma dynamics in this complex experiment. In addition, the analysis indicated that shocks disrupt the filtered silicon diode measurements at late times. We have devised ways to avoid this problem on future experiments.

A second goal was to better understand the plasma-wall interaction problem. As the target plasma is compressed, it transfers heat to the wall both through convection and conduction; this heat transfer cools the plasma near the wall. This reduces its electrical conductivity and may allow the magnetic field to diffuse through the plasma, possibly leading to some loss of magnetic insulation. We have begun MHD calculations and theoretical analysis of the plasma near a conducting wall. In addition, a consultant from New Zealand during the summer provided us with his extensive expertise of the evolution of material from a cathode in the presence of a plasma. We now have a good start on understanding the problem and plan to use our knowledge to develop a computer model that can augment our MHD codes.

Another goal was to improve our computational capabilities for alpha-particle energy deposition in a magnetized deuterium-tritium (DT) plasma. This year we were able to develop new analytic results that should enable both Monte Carlo simulations and a transport-matrix approach.



MAGO II temperature history. The curve shows the analysis of the filtered silicon diode data, and the points show the results from the MHD calculations. This good comparison increases our confidence that we are correctly modeling the atomic physics as well as plasma dynamics in this MAGO experiment.

Publications

Kirkpatrick, R.C., "Energetic Alpha Particle Transport Method EATM" (Fifth Joint Conference on Computational Mathematics, Albuquerque, NM, September 2–5, 1997).

Kirkpatrick, R.C., "Magnetized Target Fusion (MTF): Relaxing the Driver Requirements," in *Proceedings of the Second Symposium on Current Trends in International Fusion Research* (Plenum Press, New York, in press).

Kirkpatrick, R.C., "Report on Innovative Confinement Concepts Workshop," in *Proceedings of the Second Symposium on Current Trends in International Fusion Research* (Plenum Press, New York, in press).

Kirkpatrick, R.C., "Transport Method for Energetic DT Alpha Particles in a Magnetized Plasma" (Innovative Confinement Concepts Workshop, Los Angeles, CA, March 3–8, 1997).

Kirkpatrick, R.C., "VNIIEF Magnetic Bubble Approach to an X-Ray Source" (International Pulsed Power Conference, Baltimore, MD, June 1997).

Kirkpatrick, R.C., and G. Idzorek, "Analysis of Filtered Silicon Diode Data from MAGO/MTF Experiments" (International Conference on Plasma Science, San Diego, CA, May 1997).

Kirkpatrick, R.C., and G. Idzorek, "Analysis of the Filtered Silicon Diode Data from the MAGO II and MAGO III Experiments" (Innovative Confinement Concepts Workshop, Los Angeles, CA, March 3–8, 1997).

Sheehey, P.T., et al., "Computational Modeling of Joint US-Russian Experiments Relevant to Magnetic Compression/Magnetized Target Fusion" (Fifth Joint Conference on Computational Mathematics, Albuquerque, NM, September 2–5, 1997).

Sheehey P.T., et al., "Computational Modeling of Wall-Supported Dense Z-Pinches" (Innovative Confinement Concepts Workshop, Los Angeles, CA, March 3–8, 1997).

Sheehey, P.T., et al., "Computational Modeling of Wall-Supported Dense Z-Pinches" (Dense Z-Pinch Conference, Vancouver, Canada, June 1997).

Wysocki, F.J., et al., "Progress with Developing a Target for Magnetized Target Fusion" (International Pulsed Power Conference, Baltimore, MD, June 6–8, 1997).

Engineering Science

Development of an Automated Core Model for Nuclear Reactors

Russell Mosteller

The objective of this project is to develop an automated package of computer codes that can model the steady-state behavior of nuclear-reactor cores of various designs. As an added benefit, the data produced for the steady-state analysis also can be used as input to TRAC (the Transient Reactor Analysis Code) for subsequent safety analysis of the reactor at any point in its operating lifetime.

The basic capability to perform steady-state reactor-core analysis already existed in the combination of the HELIOS lattice-physics code and the NESTLE core-simulation code. Our project has completed the automated package by (1) obtaining cross-section libraries for HELIOS, (2) validating HELIOS by comparing its predictions with results from critical experiments and from the MCNP Monte Carlo code, (3) validating NESTLE by comparing its predictions to results from numerical benchmarks and to measured data from operating reactors, and (4) developing a linkage code to transform HELIOS output into NESTLE input.

During this past year, we completed the validation of HELIOS and wrote and documented a linkage code, PHONICS, to automate the transformation of HELIOS output into NESTLE input. We compared HELIOS predictions with results from established benchmarks for two different series of critical experiments, one with lattices of conventional uranium oxide fuel rods and the other with lattices of mixed oxide (MOX) fuel rods. HELIOS produced good agreement with the benchmark results for the lattices of the uranium oxide fuel rods, as shown by the comparison with results from the MCNP code in the accompanying table. However, its results for the MOX lattices were inconsistent. We believe that the observed inconsistencies are due to computational limitations (such as file size) rather than to any fundamental deficiency in the code's treatment of neutron transport.

Publications

Mosteller, R.D., "Static Benchmarking of the NESTLE Advanced Nodal Code," in *Proceedings of the Joint International Conference on Mathematical Methods and Supercomputing for Nuclear Applications* (American Nuclear Society, La Grange Park, IL, 1997), Vol. 2, p. 1596.

Mosteller, R.D., "Two-Dimensional Benchmark Calculations for PNL-30 through PNL-35" (to be published in *Trans. Am. Nucl. Soc.*).

Comparison of Results from MCNP and HELIOS

Case	Fuel Pins	Water Holes	Poison Pins	Soluble Boron (ppm)	k_{eff} MCNP	k_{eff} HELIOS
A	4961	0	0	1511	0.9956 ± 0.0003	0.9956
B	4808	153	0	1335.5	0.9957 ± 0.0003	0.9971
C	4808	9	144	794	0.9940 ± 0.0003	0.9917

Development of an Integrated System for Estimating Human Error Probabilities and Modeling Their Effects

Jack Auflick

Human reliability analysis (HRA) is the probabilistic evaluation of work-oriented human performance. Analysts examine human-machine relationships, identify error-likely situations, and provide probabilistic estimates for human errors on critical tasks. Comprising at least 40 methodologies, HRA presents the practitioner a bewildering set of philosophical and applied choices. In addition, unskilled analysts, the improper application of methodology, and the lack of human performance data can produce invalid human error probabilities (HEPs). Invalid HEPs in turn affect the derived probabilities related to the estimated occurrence of accidents that cause injury, death, or property damage.

Our objective is to address the difficulties by developing an effective, HRA-centered expert system to provide probabilistic estimates for potential human error actions. The system combines the HRA problem-solving skills and techniques of several experts and disseminates their expertise to minimally trained users. This system thus includes several of the well-known HRA core techniques and a newly developed Bayesian update method. Its expertise is contained in 1500 if/then rules and numerous hypertext links, enabling users to access HRA definitions, assumptions, and specific rules used to derive the HEP.

This system will help improve the validity of and standardize the conclusions for a given set of data, help codify HRA quantification techniques, and act as an effective training tool. Use of the system is appropriate for DOE safety and hazard analyses, Nuclear Regulatory Commission probabilistic risk assessments, and evaluations of various industrial applications regulated by the Occupational Safety and Health Administration.

Publications

Auflick, J.L., et al., "Development of a Prototype Knowledge-Based System for Estimating Human Error Probability" (to be presented at the 1998 American Society of Mechanical Engineers International Pressure, Vessel, and Piping Symposium, San Diego, CA, July 1998).

Auflick, J.L., et al., "HuRa! The HRA Knowledge-Based System for Quantifying Human Errors" (submitted to the 1998 International Probabilistic Safety Assessment Meeting, New York, NY, September 1998).

Auflick, J.L., et al., "On-Going Development of an Expert System for Estimating Human Error Probabilities" (submitted to ESREL '98, the International European Safety Analysis Conference, Trondheim, Norway, June 1998).

Neural-Network-Based System for Damage Identification and Location in Structural and Mechanical Systems

Charles Farrar

Our objective is to integrate new damage identification algorithms that examine changes in the vibration characteristics of structures and mechanical systems with adaptive computing algorithms. The coupling of these technologies is the first step in developing a computer-based system that can remotely monitor and detect damage in structural and mechanical systems.

The achievements this year include the development of a toolbox of time-frequency signal analysis algorithms that enables us to identify the temporally varying frequency of a signal. With this technology we can identify the onset of nonlinear response in a dynamic system, which is often indicative of damage. These algorithms are now incorporated into a larger general-purpose damage identification code. The algorithms that we developed as part of the project are currently aiding the vibration testing of nuclear weapons systems.

Publications

Doebling, S.W., and C.R. Farrar, "A Monte Carlo Based Technique for Determination of Statistical Uncertainty on Identified Modal Parameters" (submitted to *J. Vib. Control*).

Binding Carbon Dioxide in Mineral Form: A Critical Step toward a Zero-Emission Power Plant

Klaus Lackner

We have successfully developed the scientific basis for sequestering CO_2 in mineral form. Our goal is to maintain the competitiveness of coal energy even when environmental and political pressures require drastic reductions in CO_2 emissions. In contrast to most other sequestration methods, ours does not aim at buying time to phase out fossil energy; instead, it offers a complete and economic solution that maintains access to the vast fossil energy reservoir. Even if world economic growth exceeds the most optimistic estimates, our method will ensure energy availability for centuries. In contrast to other researchers, we are developing an industrial process for transforming CO_2 into a stable compound rather than injecting the gas into a reservoir that for safety reasons needs to be monitored indefinitely.

In an exothermic reaction, we chemically bind CO_2 to form a thermodynamically stable and environmentally benign carbonate from readily available mineral rock. We focus on magnesium silicates, because ores rich in magnesium are readily accessible in quantities that far exceed even the most optimistic estimate of coal reserves. We have developed a process that utilizes serpentinite or peridotite rock. The most promising implementation is based on an HCl extraction in which magnesium is obtained as $MgCl_2$. HCl is quantitatively recovered, while $MgCl_2$ is transformed into $Mg(OH)_2$. The hydroxide is carbonated in a gas-solid reaction that provides the energy consumed in recovering HCl.

The overall process is exothermic, and our goal is a process design that does not require an external supply of energy. Economically, the process appears viable: the mining, crushing, and milling operation is estimated at about \$8/ton of CO_2. Overall, we consider \$15/ton of CO_2 (0.8¢ to 1.5¢/kW h) a reasonable goal.

Publications

Butt, D.P., et al., "Kinetics of Thermal Dehydroxylation and Carbonation of Magnesium Hydroxide," *J. Am. Ceram. Soc.* **79**, 1892 (1996).

Butt, D.P., et al., "A Method for Permanent Disposal of Carbon Dioxide" (to be published in *World Resource Rev.*).

Lackner, K.S., et al., "Magnesite Disposal of Carbon Dixoide," *Proc. 22nd Int. Tech. Conf. Coal Util. Fuel Syst.* **22**, 419 (1997).

Lackner, K.S., et al., "Progress on Binding CO_2 in Mineral Substrates," *Energy Convers. Manage.* **38** Suppl., 259 (1997).

Pile, D., et al., "A Simple Method for Determining the Carbon Dioxide Content of Carbonated Minerals" (submitted to *J. Chem. Educ.*).

Nuclear Futures Analysis and Scenario Building

Edward Arthur

In this effort, we are examining future nuclear energy roles, the resulting global inventories of plutonium, and technological approaches to, and impact on, the management of such inventories. We are also focusing on establishing quantitative metrics that can be used to assess proliferation risks resulting from such materials accumulations. A major goal of our analyses is to identify strategies for technology selections and nuclear material management that will contribute positively to future security, nonproliferation, and robust energy scenarios.

During the past year, we presented and published a major invited paper at the International Atomic Energy Agency's Symposium on Fuel Cycle and Reactor Strategy: Adjusting to New Realities. In this paper, we describe our results from major efforts this past year in the areas of scenario development for nuclear energy and materials futures, and we describe our initial assessment of the impact of technologies on global plutonium management.

In addition, we completed technology assessment and impact studies that analyze results from plutonium inventory management worldwide. Technology areas of initial focus included multiple recycle of mixed oxide fuel in thermal reactors and the use of specialized fast-neutron-spectrum burner systems. We also investigated the economic impact resulting from use of such technological strategies to examine projected changes in future nuclear energy market shares and to set criteria required for the operation of a future plutonium management system.

Publications

Krakowski, R.A., et al., "Nuclear Energy and Materials in the 21st Century" (International Symposium on Fuel Cycle and Reactor Strategy: Adjusting to New Realities, Vienna, Austria, June 2–6, 1997).

Instrumentation and Diagnostics

Ultrasensitive Sensors for Weak Electromagnetic Fields Using SQUIDs for Biomagnetism, Nondestructive Evaluation, and Corrosion Currents

Robert Kraus

This project integrates various Los Alamos advances in the development of exceptional sensors for measuring weak magnetic fields for the nondestructive evaluation of materials, underground sensing, nonproliferation experiments, and biomedical applications. This year we made significant progress in several efforts, including (1) background noise rejection, (2) a superconducting quantum interference device (SQUID) microscope, which is a sensor for nondestructively detecting, localizing, and imaging microscopic flaws in materials, and (3) a small, hand-held biomagnetic sensor system.

The background rejection method was the result of a close collaboration with Andrei Matlashov of Conductus, Inc. The method allows the use of ultrasensitive SQUID sensors in unshielded environments. The applications for background rejection range from biomagnetic sensors for field use to sensors for nondestructive evaluation of material flaws or corrosion.

We made significant strides in the development of the SQUID microscope in the last half of this year. We developed this sensor to localize hidden material flaws, often located below intervening layers of material. This effort has brought us significant follow-on programmatic support as part of the DOE's Enhanced Surveillance Program. In late September we obtained the preliminary results of this novel system.

Concurrent with the development of the SQUID microscope, we also fabricated a small, hand-held sensor capable of detecting magnetic fields from the human heart and brain. This sensor system is designed for hand-held operation in unshielded environments to provide the first method for high-temporal-resolution sensing of human heart and brain signals. The applications for this sensor include emergency and battlefield medical needs. There is a reasonable probability of follow-on DoD funding for this work in FY 1998.

Publications

Espy, M.A., et al., "Two Methods for a First Order Hardware Gradiometer Using Two HTS SQUIDs" (to be published in *Rev. Sci. Instrum.*).

Flynn, E., et al., "A Digitial-Signal-Processor Flux-Locked Loop for Biomagnetic Applications of SQUIDs," in *Proceedings of the 1996 International Conference on Biomagnetism* (Springer Verlag, Berlin, in press).

Kraus, R.H. Jr., et al., "High-Temperature SQUID Magnetometer and Gradiometer Systems with Digital Signal Processor Flux-Locked Loop for Biomagnetic Applications," in *Proceedings of the 1996 International Conference on Biomagnetism* (Springer Verlag, Berlin, in press).

Neutron Metrology for Science-Based Stockpile Stewardship

Chris Morris

Our overall goal is to improve the detector technology available at the Los Alamos Neutron Scattering Center (LANSCE).

One objective emphasizes an imaging detector for LANSCE research. Previous work concentrated on building one layer of an active readout imaging ion chamber. An earlier version of such a detector was reported previously. This year we performed experiments at the Weapons Neutron Research Facility to demonstrate a new technology using stacked image plates and neutron converters. We were able to demonstrate a detector with an efficiency of about 20% and submillimeter-position resolution. This compares with 2% and 2.5 mm obtained previously.

A second objective focuses on a delay-line-readout area detector for cold neutrons. We completed and tested a 5-cm by 40-cm active area, fast delay-line-readout, two-dimensional imaging detector for cold neutrons and used it in the focal plane of the Bragg spectrometer on the ultracold neutron (UCN) rotor on flight path (FP) 11B at LANSCE. The data from this detector helped us to verify that dynamic distortions of the Bragg reflector were not large enough to influence the UCN production rates. The advantage of this detector over more conventional resistive-capacitive-network encoding detectors is that it is much faster and essentially immune to dead time caused by the gamma flash.

In addition, we designed and built UCN detectors for FP 11B. These are stable detectors with very good signal-to-noise ratios. The background rates were on the order of 0.01/s. This enabled us to detect UCN at a rate of less than 1/s in our first year of running with the rotor source. We have since improved many aspects of the source and are now up to a production rate of 800/s.

We also designed, built, and used a helium-3–filled ion-chamber detector for monitoring neutron fluxes from LANSCE beam lines. We developed a method for obtaining absolute flux measurements by analyzing the fluctuation levels from the detector.

A third objective targets large-area, inexpensive neutron detectors. We have begun a development project to design and construct inexpensive amplifiers and readout electronics for large-area-pad, readout ion-chamber detectors. They should provide a low-cost alternative to the area detectors currently used for neutron work.

Advanced Dynamic Radiography with Protons

Chris Morris

Proton radiography is being developed as a potential tool for providing data of interest to stockpile stewardship in the near term and as a candidate technology for a possible next-generation advanced hydrotest facility for science-based stockpile stewardship.

This year we installed a new proton radiographic experiment in Line B at the Los Alamos Neutron Science Center (LANSCE). It includes a beam transport system, containment vessel, magnetic lens system, and detector system able to image small-scale contained explosions. Together with a simulation and analysis effort, this setup provided the first dynamic proton radiograph of a shock wave in high explosives, demonstrating proton radiography as a potentially important technology to support science-based stockpile stewardship. This initial set of experiments provided the proof-of-principle basis for an extensive series of shots whose goal is to better understand the performance of high explosives.

We have studied the potential capability at LANSCE to include demonstration of material identification through a second lens system and to include investigation of the effects of beam motion on the quality of the radiographs through the use of an imaging system in front of the object to be radiographed. To have the space required to install these additional components, we will move the experiment to Line C at LANSCE in FY98. We will also install a larger containment vessel so we can investigate radiographic performance on larger high-explosive systems.

Geoscience, Space Science, and Astrophysics

Algorithm Development for Ocean Models

Len G. Margolin

We are applying technology developed in various Laboratory programs, ranging from weapons physics to mesoscale atmospherics and general turbulence modeling, toward improvements of numerical models for global ocean simulation. These improvements include increased numerical stability and computational efficiency, and better physical realization, which we are focusing on the two ocean models currently supported at Los Alamos—the Parallel Ocean Program and the Miami Isopycnic Coordinate Ocean Model. The models are used for studying ocean circulations and long-term climate changes. Our project involves university personnel in a variety of disciplines including applied mathematics, oceanography, and engineering turbulence. The involvement of personnel from the DOE CHAMMP (Computer Hardware, Advanced Mathematics, and Model Physics) program will help to ensure the strong scientific tie of this work to ocean modeling.

This year we developed a "method of averages" that accurately and efficiently treats the numerical splitting of fast and slow modes in the ocean. We implemented and tested the method in a basin-scale shallow-water model of the ocean; generalized the method to a fully three-dimensional nonhydrostatic model of the atmosphere; and derived a linear stability analysis for the method. Currently, we are extending the analysis to include nonlinearity.

We improved our generalized conjugate-residual elliptic solver by using the hydrostatic solution as an initial guess and by developing a matrix-free, alternate-direction-implicit preconditioner. We developed and tested subgrid-scale turbulence parameters for representing the planetary boundary layer and performed numerous large eddy simulations. In addition, we developed and tested a volume-of-fluid algorithm that couples to the stiff thermodynamic processes of condensation and evaporation and reduces the resolution required for following the dynamical evolution of clouds in the atmosphere.

We developed a composite algorithm for multidimensional hydrodynamics that combines two schemes sequentially—a high-order oscillatory method and a low-order method that is nonoscillatory but diffusive. For optimal combinations of the two schemes, the new composite scheme is also nonoscillatory, yet it exhibits much less diffusion. Currently, we are studying how to predict this optimal mixture.

Publications

Liska, R., and B. Wendroff, "Analysis and Computation with Multi-Layer Fluid Models" (to be published in *J. Comput. Phys.*).

Liska, R., and B. Wendroff, "Composite Schemes for Conservation Laws" (to be published in *SIAM J. Numer. Anal.*).

Margolin, L.G., et al., "Application of the Volume-of-Fluid Method to the Advection-Condensation Problem, *Mon. Weather Rev.* **125**, 2265 (1997).

Smolarkiewicz, P.K., and L.G. Margolin, "MPDATA: A Positive Definite Solver for Geophysical Flows" (submitted to *J. Comput. Phys.*).

Smolarkiewicz, P.K., et al., "On Forward-in-Time Differencing for Fluids: Stopping Criteria for Iterative Solutions of Anelastic Pressure Equations," *Mon. Weather Rev.* **125**, 647 (1997).

Lithospheric Processes

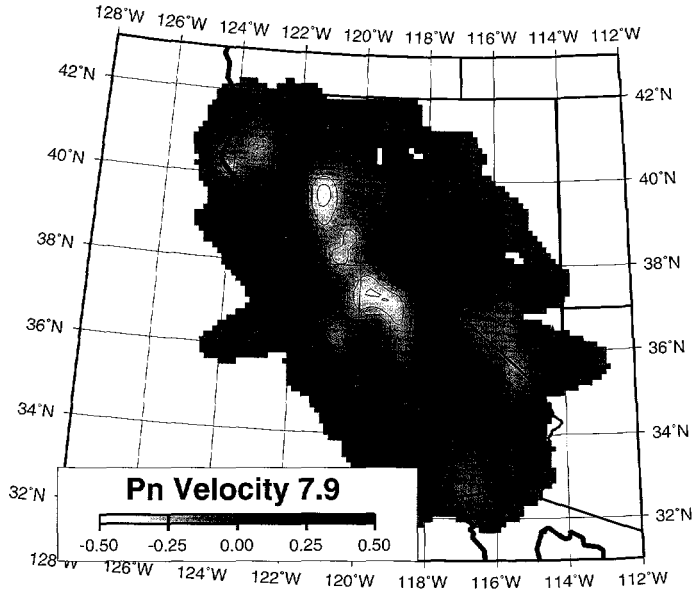
W. Scott Baldridge

This project focuses on the investigation of key problems associated with the evolution of the oceanic and continental lithosphere. Problems include the thermal structure of the lithosphere, thickness of the crust and lithosphere, melt formation within and beneath the lithosphere, residence time of magmas in the crust and mantle, and processes by which the lithosphere is thinned and ruptured during rifting. We use a variety of seismic and geochemical techniques, each providing different information. We expect to achieve a better model of upper-mantle velocity structure in different regions of the earth, an enhanced understanding of magma generation and extraction in different tectonic environments, and improved understanding and ability to model fluid transport processes on a range of scales in the lithosphere.

Using seismic techniques, we made progress in understanding the structure and thermal state of the lithosphere in the western United States and China. We constructed a database of Pn (upper-mantle compressional wave velocity) travel times for California and Nevada and inverted 75,000 Pn arrivals to image velocity variations (see the accompanying figure). Anisotropy is oriented with fast-velocity direction parallel to the San Andreas Fault, implying simple shear-strain-oriented olivine crystals within the mantle. We imaged low velocity beneath the Sierra Nevada and dramatic crustal thinning across the San Andreas Fault in central California. We reconnoitered for a teleseismic experiment across the southern Rio Grande rift to determine lithospheric structure and thickness.

We assembled surface-wave data for China and merged them with global data. Inversion of these data will enhance understanding of the heterogeneous lithospheric structure and wave propagation characteristics in this important region. We use geochemistry to investigate processes of melt formation and crustal interaction. Primarily, we measured uranium-series disequilibrium in Hawaiian rocks to determine fractionation and/or time since melting and the residence time and contamination in crustal magma chambers. We made mineral separates, measured uranium and thorium, and completed chemical separations on selected samples.

We studied cooling in pyroclastic flows, using the finite-element code FEHM to investigate multiphase hydrothermal systems, and compared the results with field observations. FEHM models the effects of water infiltrating an ignimbrite flow on heat and mass transport processes. Finally, investigations on volcanism, rifting, and paleoenvironmental studies continued in the Ethiopian rift system. New geochemical, geochronological, and paleomagnetic data refined chronostratigraphic sequences in the field.



Variations in Pn velocity in California and southwestern Nevada (preliminary results).

Publications

Baldridge, W.S., et al., "Quartz-Bearing Basalts: Oxygen Isotopic Evidence for Crustal Contamination of Continental Mafic Rocks," *Geochim. Cosmochim. Acta* **60**, 4765 (1996).

Fan, G., and T. Lay, "Multivariate Analysis of Waveguide Effects on Regional Seismic Phases in Western China" (to be published in *Bull. Seismol. Soc. Am.*).

Fan, G., and T. Lay, "Statistical Analysis of Irregular Waveguide Influences on Regional Seismic Discriminants in China" (to be published in *Bull. Seismol. Soc. Am.*).

WoldeGabriel, G., et al., "Volcanism, Tectonism, Sedimentation, and the Paleontological Record in the Ethiopian Rift System," in *Volcanic Hazards and Disasters in Human History* (Geological Society of America, Boulder, CO, in press).

Zhang, Y.-S., "Three Dimensional Velocity Structure beneath East Asia and Its Tectonic Implications," in *Mantle Dynamics and Plate Interactions in East Asia, AGU/GSA Geodynamics Series* (American Geophysical Union, Washington, DC, in press).

Zhang, Y.-S., and T. Lay, "Global Surface Wave Phase Velocity Variations," *J. Geophys. Res.* **101**, 8415 (1996).

Earth Materials and Earth Dynamics

Thomas Shankland

The objective of this research is to use laboratory methods and associated theory to describe processes responsible for Earth's structure, composition, and internal activity. In providing the "ground truth" linking different geophysical measurements, we are approaching these problems with a program of complementary experiments and theoretical models. Because fluids are important in nearly every geological process and of great concern for waste isolation and water quality, we are also examining rock-fluid interactions. Complex feedback relationships affect fluid flow in the presence of temperature and compositional gradients. Owing mainly to the cracks and pores through which fluids are transported, rocks display a surprising variety of nonlinear elastic effects that we are uncovering through our experiments and models. Nonlinear attributes of rock affect earthquake slip, reservoir subsidence, seismic-wave propagation and attenuation, stress-fatigue damage, and hydraulic fracturing.

This year, we used a powerful Los Alamos computer code to investigate water-rock feedback relationships that operate in hydrothermal systems and to evaluate the time-dependent behavior of fluid flow that is driven by thermal and chemical buoyancy, e.g., visualization of thermal evolution and rock texture above a subsurface volcanic intrusion. Related to such phenomena are the nonlinear elastic interactions that we associate with crystal-to-crystal contacts inside the pores of rock. On a global scale, we have begun to couple laboratory-derived rules to relate textural anisotropy of rocks to their strain history. For the first time, we have mapped a seismic anisotropy map onto a convection flow field in the earth. Using high-pressure methods to obtain equations of state for deep-Earth compositions, we found that our calculations for the dominant mineral phases agree surprisingly well with seismic observations.

Publications

Abramson, E.H., et al., "Elasticity of San Carlos Olivine to 17 GPa," *J. Geophys. Res.* **102**, 12253 (1997).

Bassett, W.A., et al., "The Hydrothermal Diamond Anvil Cell (HDAC) and Its Applications," *Geochem. Soc., Special Publication* **5**, 261 (1996).

Boness, D.A., and J.M. Brown, "Heat Capacity of Shocked Alkali Halides," *Shock Compression Condensed Matter* **1**, 85 (1996).

Chai, M., and J.M. Brown, "Effects of Static Non-Hydrostatic Stress on the R Lines of Ruby Single Crystals," *Geophys. Res. Lett.* **24**, 3539 (1997).

Chai, M., et al., "The Elastic Constants of an Aluminous Orthopyroxene to 12.5 GPa," *J. Geophys. Res.* **102**, 14779 (1996).

Chai, M., et al., "The Elastic Constants of a Pyrope-Grossular-Almandine Garnet to 20 GPa," *Geophys. Res. Lett.* **24**, 3539 (1997).

Chai, M., et al., "Thermal Diffusivity of Mantle Minerals," *Phys. Chem. Min.* **23**, 470 (1996).

Hearn, E., et al., "The Effect of Anisotropy on the Development of Oceanic Asthenosphere," *Geophys. Res.* **102**, 11943 (1996).

Pasternak, M.P., et al., "High Pressure Collapse of Magnetism in $Fe_{0.94}O$: Mössbauer Spectroscopy beyond 100 GPa" (submitted to *Phys. Rev. Lett.*).

Ragan, D., et al., "Silicone Fluid as a High Pressure Medium in Diamond-Anvil Cells," *Rev. Sci. Instrum.* **67**, 494 (1996).

Zhao, Y., et al., "Thermoelastic Equation of State of Jadeite $NaAlSi_2O_6$: An Energy-Dispersive Rietveld Refinement Study of Low Symmetry and Multiple Phase Diffractions," *Geophys. Res. Lett.* **24**, 5 (1997).

Elements of Water Resources and Urban Pollution

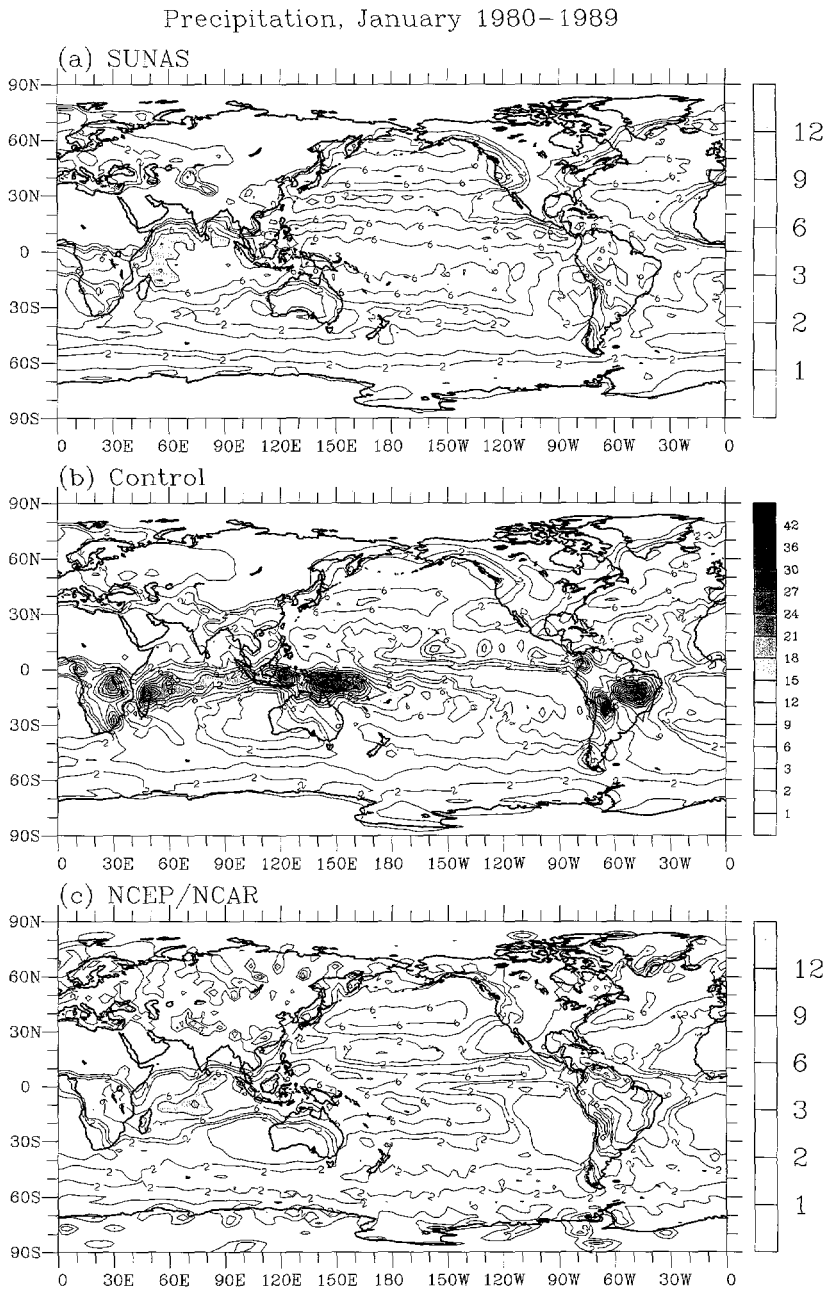
Charles Keller

The purpose of this project is to provide basic research results in support of the Laboratory's tactical goal of global environmental security as well as the Earth and Environmental Science Division's strategic plan for establishing a strong capability in water resources and air quality. Collaborations with University of California faculty and postdoctoral or graduate student interns at Los Alamos have substantially enhanced this effort.

Progress during this past year includes advances in characterization of water vapor in the atmospheric column, which have improved our ability to reproduce global precipitation primarily in the equatorial regions where convective storms dominate (see the first figure). Our analysis of American summer monsoon precipitation in the southwestern United States traced its source to the Gulf of Mexico. Similar analyses of winter storms over California showed no coherent connection to El Niño, but rather a complex connection to several weather patterns in the North Pacific. Regional coupled ocean/atmosphere modeling will study several representative cases.

We are modeling and analyzing the mean circulation and the variability of the North Pacific and their effect on the weather regime over the Pacific coast of America, particularly as it is affected by the Kuroshio Current and, in turn, by the dependence of the Kuroshio on wind-driven currents in the shallow continental shelf seas to the west.

In the Parallel Ocean Program (POP), a Los Alamos three-dimensional ocean code, we simulated introducing nitrogen oxides from Asian air pollution into the western Pacific, fertilizing phytoplankton growth. The results show surprising



Precipitation modeled by (a) the Los Alamos Cloud Physics Parameterization (SUNAS), (b) the standard Cloud Physics Parameterization in the NCAR (National Center for Atmospheric Research) atmospheric global model, and (c) the National Center for Environmental Prediction's latest data set (NCEP/NCAR). Note particularly the greatly improved simulation of equatorial precipitation in our parameterization; the control plot (b) shows more rain than is observed.

migration into the equatorial countercurrent (see the second figure).

In addition, we did the initial work to implement the University of California, Los Angeles, aerosol model, SMOG, into a Los Alamos urban-scale atmospheric circulation model. The resulting code will be used to simulate data taken in Mexico City and Los Angeles.

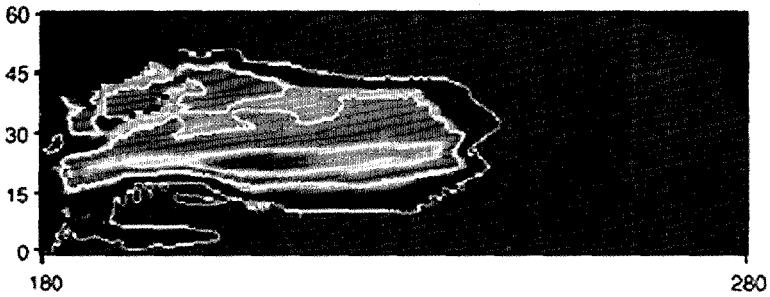
Publications

Elliot, S., et al., "Motorization of China Implies Changes in Pacific Air Chemistry and Primary Production" (to be published in *Geophys. Res. Lett.*).

Huang, Z., and C.-C.A. Lai, "Multidecadal Variability of the Southwestern U.S. Monsoon Precipitation in the Past Century" (submitted to *J. Climate*).

Kao, C.-Y.J., et al., "Sensitivity of a Physically-Based Cloud Package in the NCAR CCM" (submitted to *J. Geophys. Res.*).

Zhao, X., et al., "Aerosol-Induced Chemical Perturbations of Stratospheric Ozone" (to be published in *J. Geophys. Res.*).



POP simulation showing pollution-induced fertilization (nitrogen oxides from Asia) and drift of phytoplankton in the North Pacific Ocean. The nitrate concentration outside the contour lines is zero. Note both the northward migration of the springtime phytoplanktonic bloom, along with the advective effects of western boundary currents, and the initial drift into the equatorial countercurrent to the south from west to east.

Development of the First Nonhydrostatic, Nested-Grid, Grid-Point Global Atmospheric Modeling System on Parallel Machines

Chih-Yue J. Kao

All the existing global atmospheric models suffer from one or more of the following shortcomings: (1) lack of an interactive nested-grid capability, which makes it difficult to evaluate the regional impact, (2) spherical spectral (as opposed to grid-point finite-difference) representation of model variables, which hinders model performance on parallel machines, and (3) use of the hydrostatic approximation, which makes the models potentially ill posed. As a result, no atmospheric model is capable of simulating the full range of spatial scales of atmospheric circulations from global-scale wave disturbances to microscale plume dispersion.

The goal of this project is to develop a highly modularized, nested-grid, self-calibrating (for further physical parameterization development) global atmospheric modeling system (GAMS) with state-of-the-science physics and chemistry. This system will also provide a unique test bed for high-performance computing architecture.

Last year we incorporated and tested a global reduced-grid system into GAMS and tested the lower boundary conditions in terms of sea surface temperature and orography in the model. In addition, we studied the intrinsic difference between the hydrostatic and nonhydrostatic global models.

This year we upgraded the dynamics of our global modeling systems from the original anelastic approach to a fully compressible framework. The upgrade enables the model to predict the absolute pressure field and then to better interact with data assimilation. We handle the problem of dealing with the fast-moving sound waves in a compressible code with a temporal-averaging technique.

Publications

Kao, C.-Y.J., and W. S. Smith, "Sensitivities of a Physically Based Cloud Parameterization in CCM" (submitted to *J. Geophys. Res.*).

Reisner, J.M., and C.-Y.J. Kao, "Application of Simple Numerical Techniques for Increasing the Efficiency of a Forward-in-Time Shallow Water Code on a Sphere," in *Parallel Computation Fluid Dynamics: Algorithms and Results Using Advanced Computers*, P. Schiano et al., Eds. (Elsevier, Amsterdam, 1997).

Solar-Terrestrial Coupling through Space Plasma Processes

Joachim Birn

We continued the investigation of solar-terrestrial interaction through space plasma processes, combining efforts in space data analysis with theory plus laboratory and computer simulations.

Two phenomena observed in the magnetotail suggest that transient reconnection akin to flux transfer events at the magnetopause occur in the magnetotail. These are bursts of fast plasma flow and plasmoids, or magnetic flux ropes. We used combined Los Alamos/UCLA observations in an attempt to better understand the connection between them. We have found evidence that a substantial fraction of flux-rope core fields may form during reconnection. We have also found instances of "fossilized" flux ropes, which have not undergone lobe reconnection and are still embedded within the tail plasma sheet. The accompanying figure shows results from our modeling efforts.

In a theoretical and computational study, we investigated threshold and saturation conditions of the electromagnetic proton cyclotron ring instability. Linear Vlasov theory and one-dimensional hybrid simulations were used to examine and confirm previously derived scaling laws for the maximum instability growth rate and maximum value of the fluctuating magnetic field energy density. Simulation results showed that cessation of fluctuation growth is due to the heating of protons in the parallel direction and to the reduction of the perpendicular kinetic energy.

Laboratory simulations of magnetospheric currents showed the presence of an aurora for different orientations of the interplanetary magnetic field (IMF), with the luminosity being strongest for southward IMF. They also showed the presence of reconnection sites in the front and tail area for southward IMF.

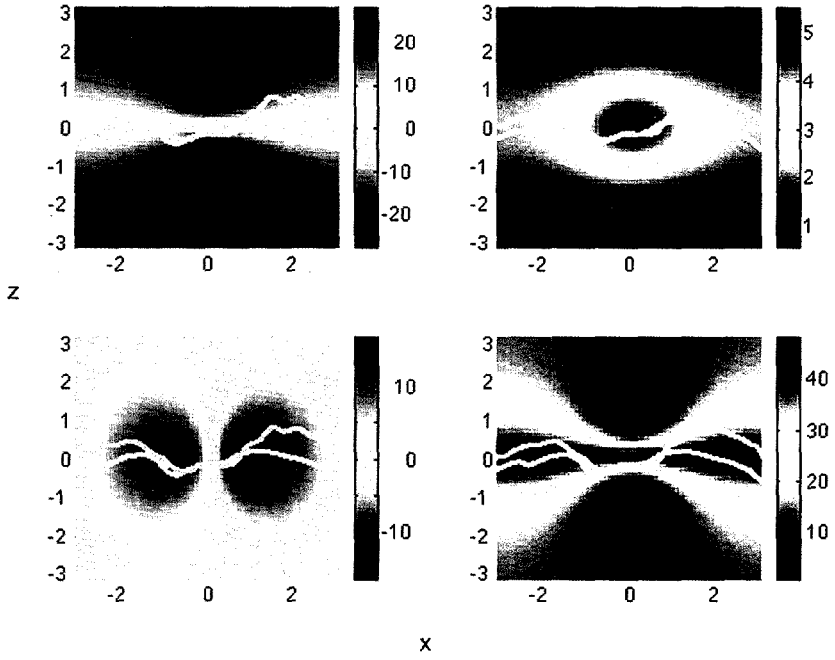
Publications

Chang, T.-F., et al., "IMF Control on the Global Structures of Laboratory Magnetospheres" (submitted to *J. Geophys. Res.*).

Convery, P.D., and S.P. Gary, "Electromagnetic Proton Cyclotron Ring Instability: Threshold and Saturation," *J. Geophys. Res.* **102**, 2351 (1997).

Hesse, M., and M.G. Kivelson, "Flux ropes in the Magnetotail" (submitted to *J. Geophys. Res.*).

Kepko, L., et al., "Observations of Flux Ropes in the Near-Earth Magnetotail" (submitted to *J. Geophys. Res.*).



Results from fitting a flux-rope model to magnetotail spacecraft data. The figures show the trajectories of two spacecraft in the *x*, *z* plane overlaid on the color-coded magnetic field intensity obtained from the model. (The coordinate *x* points sunward, *y* duskward, and *z* northward.) The four panels show the magnitude of the *x* component, *y* component, *z* component, and total magnetic field strength, respectively, in nanotesla.

Seismic Wave Propagation Modeling

Eric Jones

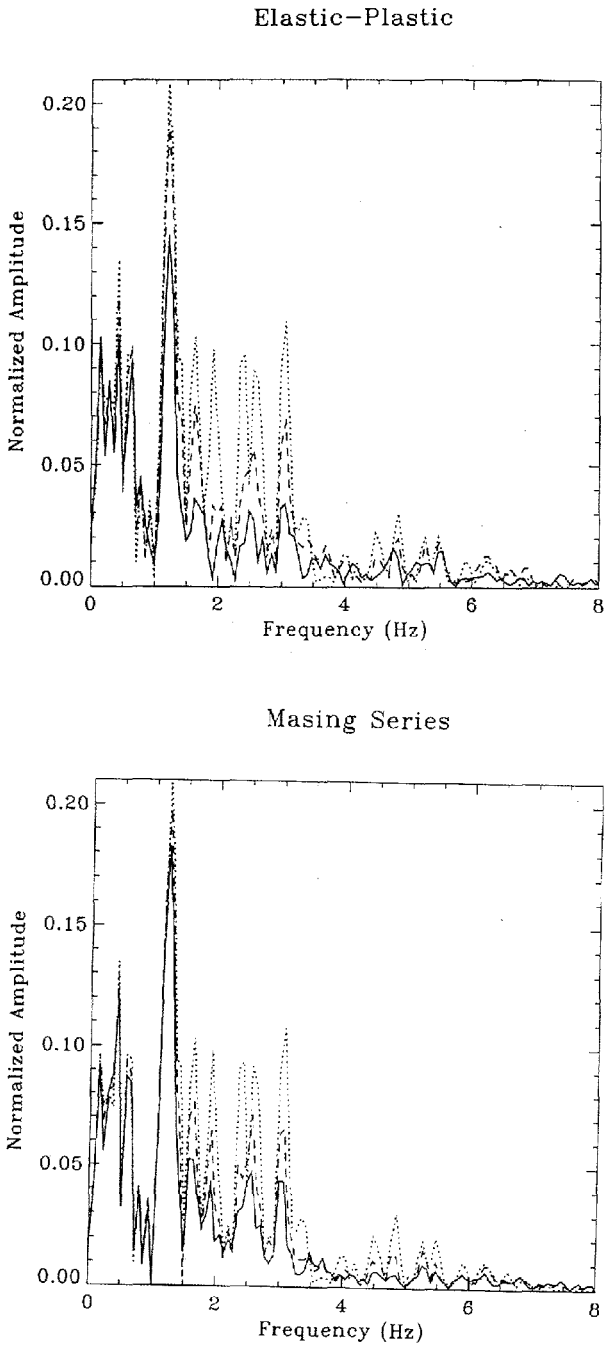
In a collaborative effort with K. B. Olsen at the University of California, Santa Barbara, we have produced realistic, high-resolution simulations of the ground motion caused by earthquakes using a new, hybrid technique. This technique couples (1) a 4th-order, finite-difference method developed by Olsen to compute linear-elastic ground motion in realistic, three-dimensional (3-D) settings such as the Los Angeles Basin and (2) Los Alamos capabilities for first-principles modeling of nonlinear ground motion in a variety of geologic materials. This was the first attempt to model earthquake ground motion by including 3-D finite-fault radiation, viscoelastic attenuation, 3-D basin response, and nonlinear effects.

We have calculated the 3-D linear-elastic ground motion at a depth of 5 km in the area of the 1995 Northridge earthquake and used those calculations as input for a one-dimensional simulation of nonlinear motion in the near-surface layers.

The accompanying figure shows the effects of nonlinearity on surface shear-velocity spectra. The dotted curve shows a linear case in which the input data was divided by a factor of 1000; the dashed curve used the realistic input data; and the solid curve had the input motion multiplied by a factor of 2. The curves are normalized so that in the absence of nonlinearity, the three cases should lie on top of each other. As can be seen in the figure, significant nonlinear effects are seen above about 1.5 Hz. At 3 Hz, for example, nonlinearity has reduced the amplitude of the realistic case (dashed curve) by a factor of almost 2.

Publications

Jones, E.M., and K.B. Olsen, "Three-Dimensional Finite-Difference Modeling of Nonlinear Ground Motion" (The Northridge Earthquake Research Conference, Los Angeles, CA, August 20–22, 1997).



Simulated shear-velocity spectra at the surface for the 1995 Northridge earthquake using either elastic-plastic material models (top) or the Masing Rule (bottom) for the three cases described in the text. The spectra are normalized to the linear (dotted) case.

Remote-Sensing Science Thrust

Siegfried Gerstl

The scope of this research encompasses three technical areas: hyperspectral remote sensing, active remote sensing (lidar), and new remote-sensing concepts. This research has application to detecting gaseous plumes and understanding the atmospheric boundary layer. These scientific tasks correlate very well with needed capabilities for future applications in nonproliferation of nuclear, biological, and chemical weapons of mass destruction and in environmental and earth sciences.

We have achieved "first-of-a-kind" measurements of Rayleigh-Benard-like microstructural convection cells in the equatorial marine atmospheric boundary layer. This discovery was published in the January 1997 issue of *Geophysical Research Letters*, and the measured data was featured on the journal's cover page. We are confirming these measurements by using a fully coupled, radiation-mass transfer code to develop three-dimensional model computations with unprecedented meter-size spatial resolution. We have upgraded the Raman lidar instrument that performed these measurements by adding a new line-narrowed Nd:YAG laser, which makes it the first scanning, boundary-layer, water-vapor/temperature lidar.

Another newly developed instrument is a microwave interferometer that has provided unique measurements of water vapor heterogeneities in the atmospheric boundary layer. We demonstrated proof of the new measurement concept using data from the Very Large Array, the microwave astronomical observatory in Socorro, New Mexico, and we have acquired additional data at the Semi-Arid Land-Surface-Atmosphere Mountain Experiment in August 1997.

We achieved another "first" by remotely sensing volcanic emissions of several passively degassing volcanoes (see the accompanying figure). We modified and calibrated a commercially available Fourier transform infrared (FTIR) spectrometer so that it allowed measurement of high-resolution emission spectra of gaseous components in the volcanic plume at large stand-off distances (2 to 10 km). Not only were we able to identify the existence of HF and HCl molecules in the plume, but we also determined the concentrations of these and other gases.

In another new task during the past year, we developed the proof of concept for an ultraspectral infrared imager based on spatial heterodyne spectroscopy. We have developed a simulation computer model that embodies the technical features of the new instrument and allows us to predict its performance. So far we have not identified any insurmountable technology hurdles.

Publications

Christenson, B., et al., "Direct Sampling and COSPEC Results Supporting FTIR and LIDAR Measurements of Volcanic Emissions at White Island and Ruapehu Volcanoes, New Zealand," *EOS Trans. AGU* **77**, 830 (1997).

Cooper, D.J., "Initial Investigations of Micro-Scale Cellular Convection in an Equatorial Marine Atmospheric Boundary Layer Revealed by LIDAR," *Geophys. Res. Lett.* **24**, 45 (1997).

Cooper, D.J., et al., "An Error Analysis and Intercomparison of Several Fast Response Eddy Covariance Sensors" (submitted to *Agricultural and Forest Meteorol.*).

Goff, F., et al. "Geochemical Surveillance of Fumarolic Gases at Popocatépetl Volcano, Mexico" (to be published in *Geol. Soc. Am. Bull.*).



Remote-sensing measurements being taken of emission-plume chemical constituents in 1997 at the active Popocatépetl Volcano near Mexico City. The FTIR and COSPEC (a correlation spectrometer operating in the solar environment) instruments are deployed side by side 10 km from the summit.

Hagelberg, C.R., et al., "Spatial Patterns in Marine Surface Layer Water Vapor Observations" (submitted to *Boundary Layer Meteorol.*).

Jacobson, A.R., and R. Sramek, "A Method for Improved Microwave-Interferometer Remote Sensing of Convective-Boundary-Layer Turbulence, Using Water Vapor as a Passive Tracer" (to be published in *Radio Sci.*).

Love, S., et al., "Integrated Infrared/UV/Lidar Remote Sensing of Volcanic Emissions at White Island and Mt. Ruapehu, New Zealand: Abst.," in *EOS Trans. AGU* **77**, 25 (1997).

Shao, X.M., et al., "Observations of Precipitable Water Vapor in the Planetary Boundary Layer via Microwave Interferometry" (submitted to *Am. Meteorol. Soc. Annu. Meet.*).

Wang, J., et al., "Comparison of Observed Marine Boundary Layer Structure with Simulations from the NCAR Community Climate Model CCM3" (to be published in *J. Climate*).

High-Performance Computing Pilot Project—Urban Security

Grant Heiken

The objective of this project is to develop a cross-divisional applied research competency to model the response and vulnerability of urban systems to malicious attacks, sociopolitical setting, the economy, and changes in physical environment.

During this year we have engaged a team representing five Laboratory divisions and a range of expertise, including atmospheric and hydrologic modeling, software design, mathematics, geographic information systems (GIS), geology, and urban planning. One major part of the pilot project was to link regional atmospheric modeling with groundwater models to assess vulnerability of urban water supplies to climate change. Another part of the pilot project was to model the transport of an airborne toxic gas, including the effects of buildings on airflow, and to link this to traffic simulations for effective emergency response.

Other activities during the year included successfully stretching existing atmospheric, hydrologic, and GIS capabilities to new regimes for urban applications; establishing design criteria for implementing coupled models on the Accelerated Strategic Computing Initiative's High-Performance Computing platform; getting appointments to the Mega-cities Committee of the International Union of Geodesy and Geophysics; collaborating with UCLA (School of Public Health, Institute of the Environment) and Arizona State University (Department of Mechanical and Aerospace Engineering); and collaborating with the DOE Center for Excellence for Sustainable Development and with academic urban simulation experts.

Publications

Brown, M., and C. Muller, "The Effect of Microscale Urban Canyon Flow on Mesoscale Puff Dispersion" (Am. Meteorol. Soc. 12th Symp. on Boundary Layers and Turbulence, Vancouver, Canada, July 1997).

Brown, M., and M. Williams, "The Effect of Urban Canopy Parameterizations on Mesoscale Meteorological Model Simulations in the Paso del Norte Area" (Air and Waste Manage. Assoc. 90th Annual Conf., Toronto, Canada, 1997).

Costigan, K.R., et al., "Atmospheric/Hydrologic Models for the Rio Grande Basin: Simulations of Precipitation Variability" (Fall Meeting, Am. Geophys. Union, San Francisco, CA, Dec. 1997).

Jones, E., and K. Olsen, "Three-Dimensional Finite-Difference Modeling of Nonlinear Ground Motion" (The Northridge Earthquake Research Conference, Los Angeles, CA, Aug. 20–22, 1997).

Soll, W.E., et al., "Atmospheric/Hydrologic Models for the Rio Grande Basin: Effects of Streamflow Variability on Aquifer Recharge" (Fall Meeting, Am. Geophys. Union, San Francisco, CA, Dec. 1997).

Turin, H.J., et al., "Albuquerque, New Mexico, USA: A Sunbelt City Rapidly Outgrowing Its Aquifer" (Int. Assoc. of Hydrologists, XXVII Congress, Nottingham, UK, Sept. 1997).

Toward a Full Three-Dimensional Model of the Earth's Mantle and Core

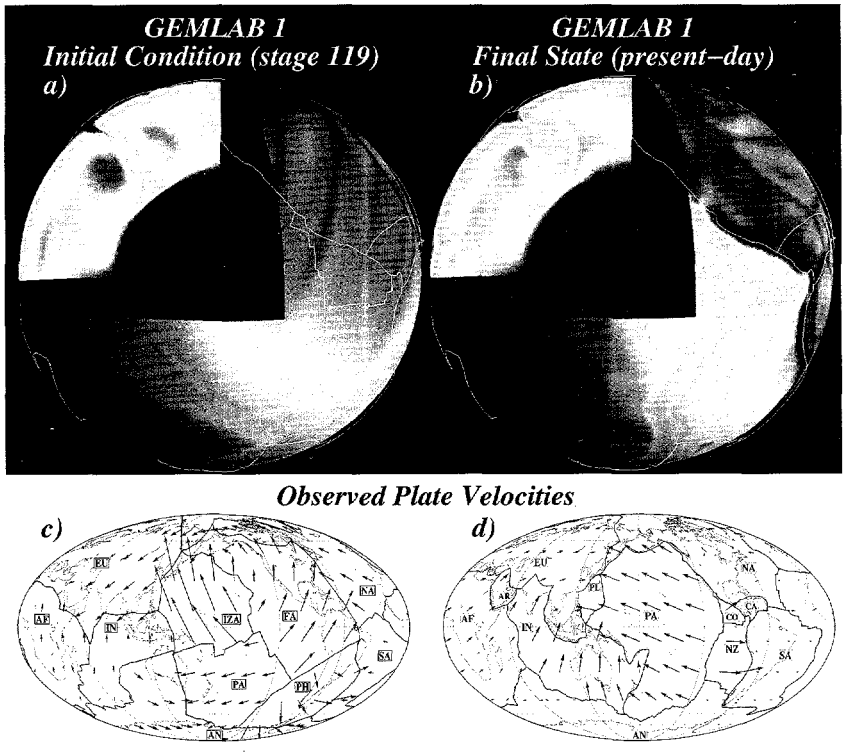
Gary Glatzmaier

Convection of both heat and composition occurs in the earth's mantle and core. It is these fluid dynamics that determine the thermal and chemical structure of the earth's deep interior and, to a major extent, the structure and dynamics of the environment at the earth's surface. Convection in the earth's mantle shapes the surface by building mountains, dispersing and aggregating continents, forming ocean basins, and controlling sea level; it determines the subsurface structure by mixing materials in some places and forming reservoirs in others; and, on short time scales, it causes earthquakes and volcanic eruptions. Convection in the earth's core maintains the geomagnetic field, which affects navigation and communication and helps to shield life on earth from cosmic radiation.

The objective of this project is to study the earth's deep interior, i.e., its mantle and core, by performing computer simulations of the three-dimensional (3-D) dynamics of these regions and comparing the results to the analysis of observational data.

Our mantle convection simulations, motivated by geochemical observations, are beginning to show how several distinct geochemical reservoirs in the earth's mantle have been able to maintain their identities over much of the age of the earth without being completely mixed by mantle convection. Our global simulations are also reconstructing the evolution of the 3-D internal structure of the mantle over the last 200 million years by using the history of the tectonic plate motions, obtained from geological and paleomagnetic observations, as a time-dependent boundary condition on mantle convection and then comparing the evolved structure with present-day seismic tomography (see figure).

Our global simulations of convection and magnetic-field generation in the earth's core are illustrating how the pattern of heat flux out of the core, which is controlled by the thermal structure of the mantle above, determines the probability of a magnetic dipole reversal. By comparing these results to the earth's paleomagnetic reversal record, we are improving our understanding of the geomagnetic field.



(a) The initial condition of the temperature structure in the mantle obtained by imposing the tectonic plate motions (for a long time) that existed 119 million years ago. (b) The present-day temperature structure obtained by running the model forward through 11 plate-motion stages. (c) The plate motions 119 million years ago. (d) The present-day plate motions.

Publications

Aurnou, J.M., et al., "Mechanics of Inner Core Super-Rotation," *Geophys. Res. Lett.* **23**, 3401 (1996).

Bunge, H.-P., and M.A. Richards, "The Origin of Long-Wavelength Structure in Mantle Convection: Effects of Plate Motions and Viscosity Stratification," *Geophys. Res. Lett.* **23**, 2987 (1996).

Bunge, H.-P., et al., "The Effect of Depth-Dependent Viscosity on the Planform of Mantle Convection," *Nature* **379**, 436 (1996).

Glatzmaier, G.A., and P.H. Roberts, "An Anelastic Evolutionary Geodynamo Simulation Driven by Compositional and Thermal Convection," *Physica D* **97**, 81 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Computer Simulations of the Earth's Magnetic Field," *Geowissenschaften* **15**, 95 (1997).

Glatzmaier, G.A., and P.H. Roberts, "On the Magnetic Sounding of Planetary Interiors," *Phys. Earth Planet. Inter.* **98**, 207 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Numerical Simulations of the Geodynamo," *Acta. Astron. Geophys.* **XIX**, 125 (1997).

Glatzmaier, G.A., and P.H. Roberts, "Rotation and Magnetism of Earth's Inner Core," *Science* **274**, 1887 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Simulating the Geodynamo," *Contemp. Phys.* **38**, 269 (1997).

Kincaid, C.R., et al., "The Dynamics of Off-Axis Plume-Ridge Interaction in the Uppermost Mantle, *Earth Planet. Sci. Lett.* **137**, 29 (1996).

Liddicoat, J.C., et al., "Transitional Palaeomagnetic Field at the Terminus of the Mammoth Reverse Subchron (3.05 MA)," *Surveys Geophys.* **17**, 183 (1996).

Olson, P., and G.A. Glatzmaier, "Magnetoconvection and Thermal Coupling of Earth's Core and Mantle," *Phil. Trans. R. Soc. Lond.* **A354**, 1413 (1996).

Ratcliff, J.T., et al., "Effects of Temperature-Dependent Viscosity on Thermal Convection in a Spherical Shell," *Physica D* **97**, 242 (1996).

Tackley, P.J., "On the Ability of Phase Transitions and Viscosity Layering to Induce Long Wavelength Heterogeneity in the Mantle," *Geophys. Res. Lett.* **23**, 1985 (1996).

Tackley, P.J., "Effects of Strongly Variable Viscosity on Three-Dimensional Compressible Convection in Planetary Mantles," *J. Geophys. Res.* **101**, 3311 (1996).

Development of Inexpensive Continuous-Emission Monitors for Feedback Control of Combustion Devices that Minimize Greenhouse Gases, Toxic Emissions, and Ozone-Damaging Products

David Funk

There is little doubt that combustion is the major cause of poor urban air quality and depletion of the ozone layer, that it is a major source of greenhouse gas and carbon dioxide, and that global combustion activity is increasing. As part of our research, we have developed sensitive, inexpensive continuous-emission monitors based on the optical properties of the expected hazardous gas-phase materials. These sensors will provide direct feedback on combustor operations, allowing the optimization of combustion while simultaneously minimizing the emission of hazardous gas-phase species.

To date we have completed two specific tasks (and worked on others):

- We have developed and demonstrated a solid-state Fourier transform spectrometer, which has led to an application for a patent. This spectrometer moves the detected signal away from 1/f noise, thereby increasing its sensitivity over traditional spectrometers for a given integration time.
- We have developed an infrared He-Ne probe for detection of hydrocarbon/oxygen ratios, which has also led to a patent. This IR He-Ne probe allows one to determine the air-to-fuel ratio (AFR) continuously. We recently examined the power spectrum of the AFR by applying the Fourier transform to the time series. We discovered several preferred frequencies in combustion chambers. This finding is most exciting to gas turbine researchers because pressure dynamics is a serious problem that limits the operational envelope of the combustor. We have evolved the IR He-Ne air-to-fuel device from gas turbines to spark engines by modifying a spark plug to accept two IR transmitting optical fibers. The IR He-Ne light passes through one fiber in the spark plug center electrode. Light exits this fiber and crosses the spark gap, where it reflects back from the polished ground electrode to a return fiber optic. The absorption of the laser beam by hydrocarbons in the gap gives the AFR in the gap.

Publications

Funk, D.J., and D.S. Moore, "Fourier Transform Spectroscopy using Liquid Crystal Technology," *Opt. Lett.* **22** (23), 1799 (1997).

Mongia, R.K., et al., "Measurement of Air-Fuel Ratio Fluctuations Caused by Combustor Driven Oscillations" (Fall Meeting of the Western States Section of the Combustion Institute, Diamond Bar, CA, October 23–24, 1997).

Mongia, R.K., et al., "Use of an Optical Probe for Time-Resolved In Situ Measurement of Local Air-to-Fuel Ratio and Extent of Fuel Mixing with Applications to Low No_x Emissions in Premixed Gas Turbines," *Twenty-Sixth Symp. (Int.) Combust.* **26**, 2749 (1996).

Low-Luminosity, Compact Stellar Objects and the Size of the Universe

Richard Epstein

We investigated the evolution of (and radiation from) neutron stars and stellar systems as well as the technologies for studying the physical properties of these objects. This investigation included a study of x-ray and gamma-ray emission from neutron stars and other compact objects. We examined the spatial and brightness distributions of gamma-ray bursts and showed that brightness-dependent anisotropy of gamma-ray bursts could arise if the sources represent a galactic distribution with a range of luminosities. We modeled stellar encounters in galactic nuclei. In particular, we simulated the high-velocity collisions in active galactic nuclei, focusing on encounters involving main-sequence stars, and calculated cross sections for mass loss and capture.

We developed critical components for a vibration-free cryocooler that will allow high-performance cryogenic detectors for astrophysical observations and remote-sensing applications.

We measured and calculated the oscillation modes of Delta Scuti stars to learn more about the internal structure and composition of these objects. We completed a comprehensive analysis of the observational data available on the Delta Scuti stars and generated a newer, more precise pulsation spectrum than was previously available. This spectrum includes a newly discovered mode that allows us to constrain theoretical models of these objects.

Publications

Berley, D. et al., "First Light from the Milagrito Air Shower Detector," *XXV Int. Cosmic Ray Conf.* **5**, 201 (1997).

Berley, D. et al., "Results from the Milagrissimo Air Shower Detector," *XXV Int. Cosmic Ray Conf.* **3**, 285 (1997).

Fajardo, J.C., et al., "Electrochemical Purification of Heavy Metal Fluoride Glasses for Laser-Induced Fluorescent Cooling Applications," *J. Non-Cryst. Solids* **213**, 95 (1997).

Link, B., and R.I. Epstein, "Are We Seeing Magnetic Axis Reorientation in the Crab and Vela Pulsars?" *Astrophys. J. (Lett.)* **478**, 91 (1997).

Mungan, C.E., et al., "Internal Laser Cooling of Yb-Doped Glass Measured between 100 and 300 K," *Appl. Phys. Lett.* **71**, 1458 (1997).

Mungan, C.E., et al., "Laser Cooling of a Solid by 16 K Starting from Room Temperature," *Phys. Rev. Lett.* **78**, 1030 (1997).

Mungan, C.E., et al., "Spectroscopic Determination of the Expected Optical Cooling of Ytterbium-Doped Glass," *Mat. Sci. Forum* **239**, 501 (1997).

Nuclear and Particle Physics

Advanced Techniques for Producing, Polarizing, and Storing Ultracold Neutrons

Susan Seestrom

This project is focused on two main areas: new techniques for producing ultracold neutrons (UCNs) using cryogenic converters and development of techniques needed to perform world-class fundamental physics experiments using those UCNs. A specific computer code was written to model UCN production in solid deuterium. That code has now been expanded to incorporate realistic source terms from the neutron transport code MCNP. Calculations indicate the possibility of constructing a UCN source with much higher flux. Measurements are needed to benchmark these calculations, and these have been planned. A cryogenic target has now been designed and is under construction.

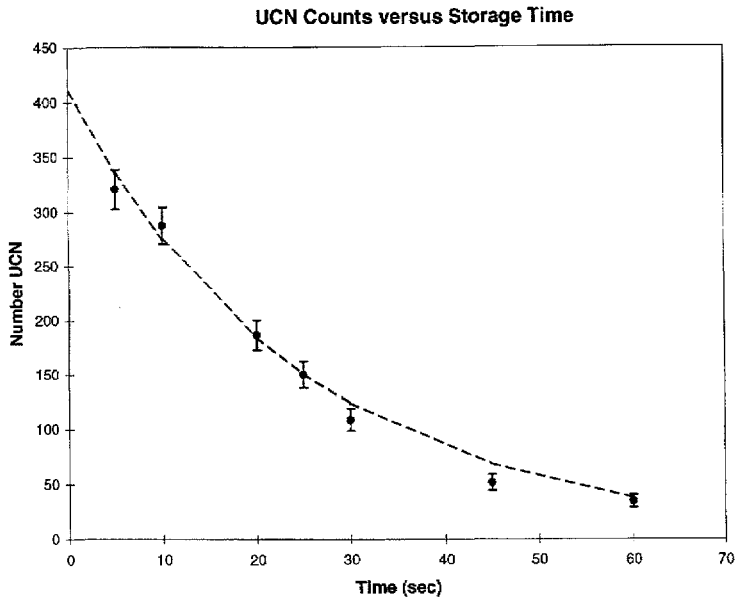
Two different types of UCN guides have been designed, built, and tested—one made from stainless steel and the other from stainless steel with a ^{58}Ni coating. Both gave similar performance results. The actual guide performance was found to be highly sensitive to the cleanliness and surface preparation. Part of our work is to build and test bottles for holding the neutrons. An improved version of the prototype UCN bottle first built in FY 1996 was built and tested in FY 1997. In initial beam tests this year, we measured a respectable bottle lifetime of 25 s (see figure). At present, after the system was taken apart and reassembled, the measured lifetime is significantly lower. At this time it is unclear whether the source of reduced lifetime is buildup of oil vapor on the surface or alignment of the bottle valves.

We have identified and obtained a 2.7-T magnet that will be used in initial tests to polarize UCNs.

Publications

Seestrom, S.J., et al., "Development of an Ultra-Cold Neutron Source at the Los Alamos Neutron Science Center" (International Conference on Nuclear Data for Science and Technology, Trieste, Italy, May 1997).

Seestrom, S.J., et al., "Initial Testing of an Ultra Cold Neutron Source at MLNSC" (Fifth International Seminar on Interaction of Neutrons with Nuclei, Duban, Russia, May 1997).



Plot of UCN counts vs storage time (circles) and the fit to a curve representing an exponential decay with a 25-s half-life.

Advancing X-Ray Hydrodynamic Radiography: Multipulse Accelerator Cores and Injectors

Daniel Prono

To be able to validate our weapon hydrodynamics codes and infer nuclear performance and criticality, we will need to collect sufficient radiography data to synthesize time-dependent, three-dimensional [3D(t)] reconstructions of weapon primary systems undergoing hydrodynamic tests. For the 3D(t) reconstructions to be accurate and have the needed resolution of shapes and density distributions requires two-dimensional (2-D) radiography data from multiple (>8) viewing lines of sight, with several time-sequential 2-D radiography images for each view (>5 images per view). The soon-to-be-operational first axis of the Dual-Axis Radiographic Hydrodynamics Test (DARHT) Facility will furnish one high-quality radiography image by generating a 50-ns, 4-kA, 20-MeV electron beam and converting it into an x-ray source of ~1-mm spot size. Future x-ray radiography systems will have to generate at least 2000 ns of comparable beam current and then subdivide and distribute this current to supply the 5 images and 8 lines of sight. This 40-fold increase in pulse duration represents a significant upgrade in induction-accelerator capability for both the injector- and accelerator-core subsystems. It will require major extensions from existing physics and engineering designs and the development of new technology.

Our objective is to extend the Labortory's capability in induction-accelerator technology to achieve the required performance enhancements. During this year, we analyzed and designed the diode subsystem for a 2000-ns (long-pulse) injector. This work specified the technology requirements and predicted performance for the DARHT second-axis architecture. We also analyzed the emittance degradation mechanisms that will plague a 2000-ns electron beam as it is transported through an induction accelerator with a 2000-ns pulse duration. In the course of this work, we uncovered limitations in the design code for electron injector optics (E-GUN) and incorporated needed corrections to the Los Alamos particle-in-cell design code (ISIS). We also modified and improved the SLICE beam transport code to analyze emittance growth in a long-pulse induction-accelerator structure. This and companion work on beam emittance was done in colloboration with Lawrence Berkeley and Lawrence Livermore National Laboratories. Finally, we designed a Long-Pulse Technology Development (LPTD) facility that will use a 1.5-μs electron beam generated by a conventional Marx-bank-driven diode to test prototype long-pulse accelerator subsystems. The LPTD facility will also contain a cathode/shroud test stand to validate our design of a long-pulse injector-diode subsystem.

Publications

Carlsten, B.E., "Centrifugal Space-Charge Force on an Electron Beam in a Focusing Element," *Phys. Rev. E* **55**, 4893 (1997).

Carlsten, B.E., "Emittance Growth Mechanisms of a Nonequilibrium Intense Electron Beam in a Transport Channel with Discrete Focusing" (to be published in *Nucl. Instrum. Meth. Phys. Res.*).

Carlsten, B.E., "Emittance Growth due to Radial Density Variations of an Emittance-Dominated Electron Beam in an Accelerating/Focusing Channel" (to be published in *Phys. Plasmas*).

Advancing X-Ray Hydrodynamic Radiography: Multipulse X-Ray Detectors

Daniel Prono

Our goal is to furnish sequential x-ray images that will enable next-generation hydrodynamic radiography to "make a movie" of a weapon system's implosion dynamics during the critical nuclear phase. A sequence of two-dimensional images, when taken from multiple views, will be used to make time-dependent, three-dimensional reconstructions on how the shape and density distributions of the primary system evolve. These reconstructions will validate the hydrodynamics code predictions and will be the basis for inferring nuclear performance.

The challenge in developing the requisite x-ray detectors is to improve individual image performance by increasing the light-gathering capability, quantum efficiency of the optics, and pixel density. Such improvements in detector performance are needed both to achieve the required resolution capability of 0.25 mm on the fine features and shapes of a primary system and to limit the required x-ray source intensity, which is a large cost-driver for any future radiography facility.

The detectors must also be able to record and store images at a rate of >5 MHz (>10 images spanning the ~2-μs duration). Electronic circuitry exists for binary data collection, storage, and transfer at such rates, but it has not been applied to converting optical signals that characteristically have 1000:1 contrast ratios and that

thus require vastly greater binary encoding per information byte. Additionally, the requirement to distinguish abutting edges, boundaries, and gradients of materials dictates that information from individual pixels be preserved and transferred. In such a system, the conventional charge-coupled device (CCD) camera that photographs an image on a scintillator would be replaced by an "all-electronic" detector: a highly pixellated scintillator would have each pixel fiber coupled to a photon-sensitive computer chip that would digitize the photon intensity and then feed it to an analogue storage device that would isolate, store, raster, and transfer information input at the 5-MHz rate.

Progress toward developing the required detectors this year includes the following:

- We performed tests with static test objects to quantify scintillator and CCD camera ability to resolve feature sizes to 1.0 mm and then used these results to specify prototype fast-camera systems. Such a prototype camera will be purchased next year from Silicon Mountain Devices of Denver, Colorado.
- We conducted a 2-day workshop on detector technology to draw on the experience and professional opinions available from other national laboratories and from outside vendors. The workshop provided us with the design information needed to begin developing an "all-electronic" detector.
- We conducted a 2-image, 500-kHz detector demonstration on an actual hydrodynamic test to gain firing-point experience with fast-detector systems. This was the first multi-image demonstration (without data superposition) of a multipulse detector on an actual hydrodynamic test.
- We developed a photo-sensitive computer chip and coupled its output to a 5-MHz data analog device; tests of this chip as the basic element for an "all-electronic" detector will begin next year.

Liquid-Lead and Lead-Bismuth Technology for Use in Subcritical Systems Applied to Nuclear Waste Destruction

Francesco Venneri

Liquid-lead technology appears to be the most suitable for accelerator transmutation of waste (ATW) applications, combining good thermohydraulics properties, excellent neutron spallation efficiency, and neutronics with adequate materials compatibility. Russian researchers have extensive experience in liquid-lead nuclear technology. The objective of this project is to develop liquid-lead and lead-bismuth eutectic (LBE) expertise for ATW applications—spallation neutron targets and nuclear coolant uses. Specifically, we will acquire the capabilities to accept and test an LBE target from the Institute of Physics and Power Engineering (IPPE) by the year 2000. We will perform thermohydraulics experiments on liquid LBE test loops with parallel analysis and simulations; we will also examine key corrosion mechanisms and prevention techniques. This research will leverage the existing Russian nuclear LBE technology and the collaborative efforts of industrial partners.

Our accomplishments this year include selecting a concept and laying out subcritical liquid-lead systems. We completely reassessed the technical and scientific basis for ATW to focus the project on the use of viable technology. We conceived the concept of a spectrum-independent, subcritical actinide burner, which could take advantage of the best characteristics of a fast spectrum system (efficient use of neutrons) and of thermal spectrum systems (low closeout inventories). This progress allowed us to develop the concept of the lead-cooled ATW actinide/fission-product burner.

In addition, we designed and built a liquid LBE loop for operation in September 1997. The loop can accommodate up to 3 tons of lead and move it at velocities over 5 m/s. We expect to be able to measure fundamental thermodynamic, chemical, and heat transfer properties of lead and LBE in the region of interest for our target/blanket work next year.

A third accomplishment involved technology transfer from Russian laboratories. We completed the first phase of the evaluation of the Obninsk–Los Alamos target design. A satisfactory design was elaborated and is ready for fabrication in Russia. In a series of interactions with the IPPE, we assessed the chemical handling of liquid lead and LBE, especially the removal of polonium and other species from the lead.

Quantum Technologies

David Vieira

Recent developments in the laser cooling and trapping of atoms and ions have revolutionized our ability to control and manipulate the quantum states of atomic systems. We are developing these laser techniques to perform fundamental experiments on multiquantal systems that have previously not been possible. Of particular interest are experiments related to the development of a quantum computer. If feasible, quantum computation would undermine the security of public key encryption systems used throughout the world. Thus understanding its feasibility is important to national security interests.

We are also advancing our abilities to confine, concentrate, manipulate, and detect selected radioactive (or stable) atoms using magnetic and optical traps. This work has important applications to understanding fundamental electroweak interactions and to detecting trace amounts of nuclear proliferants relevant to treaty verification and environmental concerns.

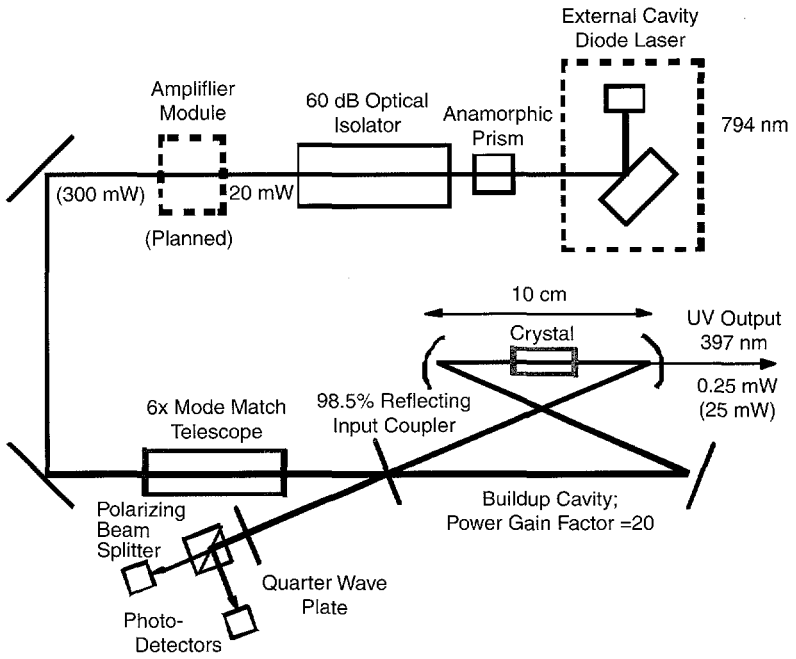
For quantum computation with calcium ions, we have developed narrow-linewidth diode lasers that operate at 794 nm (frequency-doubled to 397 nm) and 866 nm (see first figure). These lasers are required to prepare the ions' initial quantum state before computation and to read the results. We have also made important progress in the theory of quantum computation. Additionally, we are developing ultra-high-efficiency single-photon detectors (solid-state photomultipliers) for quantum information applications.

In our laser trapping work, we have made major progress in developing an atom trap for a radioactive rubidium isotope. The device consists of a magnetooptical trap coupled to a mass separator (see second figure). In parallel with this work, we have designed a time-orbiting-potential pure magnetic trap and various scintillation counters for a high-precision beta-asymmetry experiment that uses a polarized sample of the rubidium isotope.

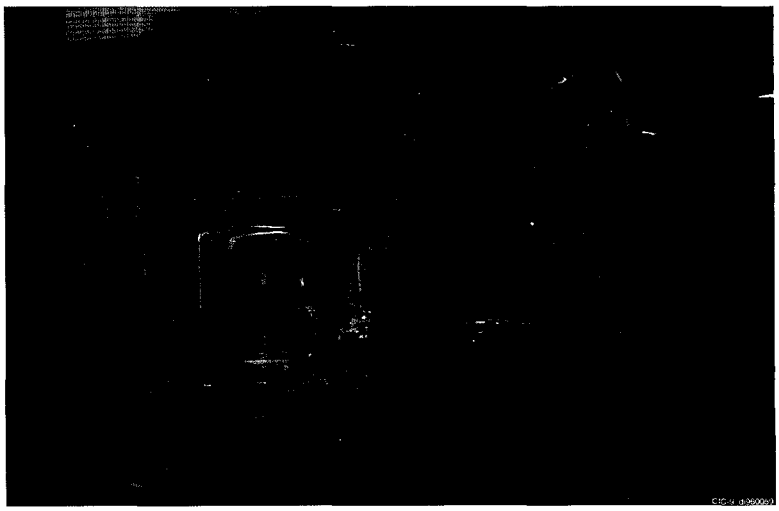
Publications

Guckert, R., et al., "Coupling an Optical Trap to a Mass Separator," *Nucl. Instrum. Methods Phys. Res., Sect. B* **126**, 383 (1997).

Hughes, R.J., et al., "The Los Alamos Trapped Ion Quantum Computer Experiment" (to be published in *Fortschr. Phys.*).



Schematic of the frequency-doubling cavity used to produce 397-nm light from a semiconductor diode laser that operates at 794 nm. A lithium iodate crystal in the buildup cavity produces the UV output. Adding an amplifier module to the setup is expected to boost the power output from 0.25 to 25 mW.



Cesium atoms (white cloud in glass cube) confined in our magnetooptical trap. When used with different lasers, the trap can also confine rubidium atoms.

Multidisciplinary Science-Based Bioremediation

James Brainard

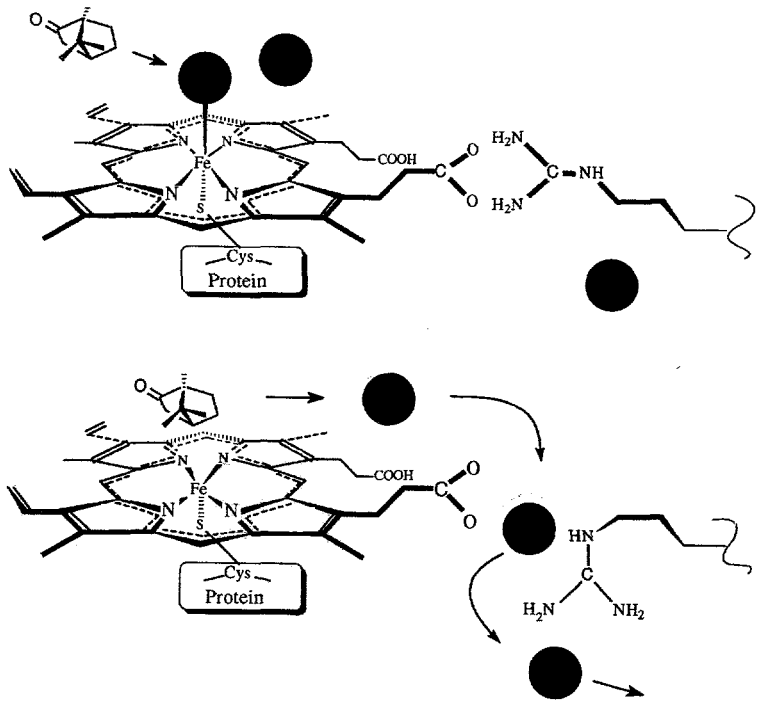
Although bioremediation approaches are potentially some of the most cost-effective solutions to the environmental cleanup problems faced by DOE and the nation, present bioremediation practices have difficulty achieving effective destruction of some contaminants, notably halogenated hydrocarbons. Many microorganisms contain halocarbon-degrading enzymes, but these enzymes are relatively poor catalysts: they have a poor affinity for the contaminant, they have slow turnover rates even when the contaminant is bound, and they sometimes generate toxic intermediates. The goal of this project is to develop the tools and understanding required to engineer and then apply enzymes with enhanced properties to the bioremediation of contaminants.

This year we performed molecular-dynamics calculations on haloalkane dehalogenase. These calculations suggested sites for site-directed mutations that will allow us to probe the reaction mechanism for this enzyme. We also obtained, by random mutation, a mutant of haloalkane dehalogenase with enhanced activity over the wild-type enzyme. We isolated dichloroethane-degrading bacteria from contaminated sites and characterized several of these bacteria. In addition, we obtained crystals and x-ray data sets from a new dehalogenase enzyme.

We proposed a new model for a functional water channel in cytochrome P450 enzymes (see figure), and we demonstrated that the cytochrome $P450_{CAM}$–oxygen intermediate produced by x-ray radiolysis is functionally competent and that the electron-density map of this intermediate is most consistent with a peroxoferryl active oxygen species. Finally, we developed and applied a new method for assessing the population heterogeneity of microbial communities in environmental samples.

Publications

Oprea, T.I., et al., "Identification of a Functional Water Channel in Cytochrome P450 Enzymes," *Proc. Natl. Acad. Sci. U.S.A.* **94**, 2133 (1997).



For cytochrome P450 enzymes to function efficiently, water in the active site must be excluded. This figure shows a two-state model for the functional water channel in cytochrome P450 enzymes that allows water to be transported from the active site to the surface of the protein and to the bulk solvent. The upper diagram shows the substrate, camphor, displacing a water molecule that is bound to the active site of the enzyme. Once the camphor is bound, the water molecules follow the transport pathway depicted in the lower diagram. The movement of the water molecules causes the arginine side chain (at the right-hand side of the diagram) to rotate, thereby opening the functional water channel.

Covariation of Mutations: A Computational Approach for Determination of Function and Structure from Sequence

Alan Lapedes

Figuring strongly in the prediction of protein structures is the understanding of protein folding—what processes transform simple linear sequences into more elaborate structures. Classical fold-recognition methods are based on the premise of distinct pairwise preferences between two given amino acids. We have studied these preferences extensively and found that, in the general case, this information is limited. Yet, by modeling pairwise interactions in the context of phylogenetic relationships and by modeling one specific type of contact, that is, the contact between interacting beta strand residues, we have recovered significant information for prediction and analysis of protein structure. At this time, when the structure of a protein is determined, its structure is more likely to belong to some existing fold than to represent a new fold. With this insight, and a number of good fold taxonomies available, fold recognition is one of the more promising subproblems in protein structure prediction.

We have developed and enhanced a set of tools for fold recognition with hidden Markov models (HMMs) and used these tools effectively in the CASP-2 protein structure prediction contest—correctly predicting the folding of a sequence before its structure was determined and verified. However, HMMs have limitations, one of which is that they do not model the long-range pairwise interactions that define the shape of a protein. Therefore, we are working on modeling pairwise interactions to incorporate them into our HMM-based framework.

Amino acid potential, the prediction of when two amino acids will be in contact, has a wide body of users as described in recent reviews. Yet we found the information in these potentials to be limited. Therefore, we have turned our attention to variants of general potentials: beta strand contacts and contacts in the context of phylogenetic relationships. Here, we have shown, is significant and useful information.

Publications

Barret, C., et al., "Scoring Hidden Markov Models," *CABIOS* **13** (2), 191 (1997).

Dalgaard, J.Z., et al., "Statistical Modeling, Phylogenetic Analysis and Structure Prediction of a Protein Splicing Domain Common to Inteins and Hedgehog Proteins," *J. Comput. Biol.* **4** (2), 193 (1997).

Grice, J.A., et al., "Reduced Space Sequence Alignment," *CABIOS* **13** (1), 45 (1997).

Karplus, K., et al., "Predicting Protein Structure Using Hidden Markov Models" (to be published in *Proteins: Struc., Funct., and Genet.*).

Lapedes, A., et al., "Correlated Mutations in Protein Sequences: Phylogenetic and Structural Effects" (to be published in *Proceedings of the AMS Conference on Statistics in Molecular Biology*).

Sjolander, K., et al., "Dirichlet Mixtures: A Method for Improving Detection of Weak but Significant Protein Sequence Homology," *CABIOS* **12** (4), 327 (1996).

Competency Development in Antibody Production for Cancer Cell Biology

Min Park

Antibodies provide humans and other animals with immune defense against a wide range of antigenic substances. Antibodies have been used as diagnostic and therapeutic tools for various diseases, including cancer. The main objective of this project is to develop a novel recombinant antibody technology that can produce therapeutically promising antibodies for immunocancer therapy. The target antigens are functionally significant molecules in cell-cycle control and DNA repair.

Our approaches to accomplish the main objective include (1) recombinant antigen production, (2) immunization and rapid isolation of splenocytes using the flow cytometry and magnetic bead-capturing process, (3) production of an immunoglobulin single-chain library, (4) production of a recombinant antibody, (5) development of a controllable expression vector for the production of intracellular antibodies, and (6) inhibition of tumor cell growth in culture.

This year we produced recombinant single-chain antibody libraries that are specific to DNA repair and cell-cycle control proteins. In addition, we designed and developed a new mammalian expression vector that can target, express, and monitor intracellular antibodies with a precise control of antibody gene expression.

Publications

Gary, R., et al., "The DNA Repair Endonuclease XPG Binds to PCNA," *J. Biol. Chem.* **272**, 24522 (1997).

Next-Generation Biological Toxin Sensors

Pat Unkefer

To develop a new approach to the detection and identification of biological toxins, we are integrating molecular recognition based on protein-receptor interactions and biomimetic materials. This effort builds on expertise in chemical and materials synthesis, surface and thin-film characterization, self-assembly, modeling, and sensor transduction that lies at the interface between the life and physical sciences. We will derivatize natural receptors that are found on the surface of neural cell membranes and incorporate the receptors into biomimetic films that are attached to transducer surfaces.

The goals of our work are to (1) develop robust biomimetic films that can be adapted to a sensor array system for the rapid identification of biological toxins, (2) modify natural receptors to provide a suite of sensing films with optimal binding affinities for the recognition of toxins, (3) develop novel optical transduction approaches and signal amplification to enhance both sensitivity and specificity, and (4) lay the foundation to apply this sensor approach to other biological sensing needs.

Highlights for work performed this year include the following actions:

- developed a new, Langmuir-phase, assisted self-assembly technique to guide self-assembly of complex, multicomponent structures on gold and oxide surfaces;
- implemented new experimental capabilities including Fourier transform infrared, Langmuir-Blodgett troughs, and surface-enhanced Raman spectroscopy;
- studied the phase behavior of GM-1, incorporated it into self-assembled monolayer (SAM)-templated hybrid bilayers, and measured the kinetics of cholera binding to GM-1 with surface plasmon resonance;
- developed a method to prepare supported bilayers containing GM-1 receptors on silica beads and applied the method to measure fluorescent cholera-toxin binding using flow cytometry;
- developed protocols to separate specific and nonspecific binding of cholera to GM-1 and a kinetic model of the binding mechanism;
- initiated surface chemistry to allow covalent attachment of glycolipid receptors to SAMs on oxide surfaces and characterized the films;
- initiated flow cytometric measurements of cholera binding on monolayer surfaces; and
- completed model development to simulate toxin-receptor binding kinetics over a wide range of parameters.

Publications

Yang, X., et al., "Covalent Attachment of Oligosaccharides to Preformed Self-Assembled Monolayers," *Abstr. Pap. Am. Chem. Soc.* **214** (2), 370 (1997).

Advanced NMR Technology for Bioscience and Biotechnology

Jill Trewhella

Nuclear magnetic resonance (NMR) plays critical roles in bioscience and biotechnology both for imaging and for structure determination. The limitations for applying NMR to many important problems are the inherently low sensitivity of the NMR experiment and the high spectral resolution required to study biological molecules.

In this past year, we focused on developing novel applications of high-field NMR with isotope labeling to overcome current limitations on the size of biomolecules whose structures can be resolved using NMR. We studied the muscle protein troponin C (TnC) complexed with segments of its regulatory target troponin I (TnI). The TnC/TnI complex is too large to be studied with conventional NMR approaches, and it also has limited solubility, thus demanding maximal resolution and sensitivity of the NMR experiment.

We measured high-field NMR data from complexes of isotopically labeled TnC complexed with the sequence segment of TnI corresponding to residues 96–115. This TnI(96–115) segment binds alternately to TnC and to actin molecules within the thin filament during the contractile cycle, thus regulating the acto-myosin ATPase activity that drives contraction. We found that when TnI(96–115) is bound to TnC, the TnI segment contains a "loose," or "nascent," helix that has been previously characterized as a peptide-folding intermediate. This loose structure appears to be key to allowing TnI(96–115) to bind alternately to TnC or actin in response to calcium signals detected by TnC. Thus, the TnC/TnI complex functions as a calcium-sensitive "switch" that regulates contraction and relaxation events in the contractile cycle. This result may be the first example of nature making use of this type of prefolding transition state for an explicit function.

Publications

Hernández, G., et al., "NMR Studies of the Troponin I Inhibitory Peptide (96–115) Bound to Skeletal Muscle Troponin C" (submitted to *J. Mol. Biol.*).

An Integrated Structural Biology Resource: Applications to the Structure and Function of DNA Repair Enzymes

L. Scott Cram, Angel E. Garcia, William Woodruff, and Joel R. Berendzen

We are developing an Integrated Structural Biology Resource (ISBR) that focuses on environmentally induced DNA damage and the assembly, structure, and function of DNA repair enzymes. Environmental effects are exerted either on genetic material or on protein expression, self-assembly, structure, or function. Many of these sites of action involve complex structures of protein and DNA. The ISBR engages the unique combination of capabilities at Los Alamos in genomics, molecular biology, and approaches to determining biomolecular structure, dynamics, and function. The resource incorporates protein expression, isotopic labeling, advanced laser spectroscopies, neutron and x-ray scattering, theory, modeling, high-resolution nuclear magnetic resonance (NMR), and x-ray crystallography.

There are five essential elements of the ISBR, which are being developed with a phased approach. This past year, we emphasized modifying the X8-C beamline at the National Synchrotron Light Source at Brookhaven National Laboratory for use in structural biology applications. This facility is now available for experimentation. Numerous experimeters from Los Alamos have already used the facility.

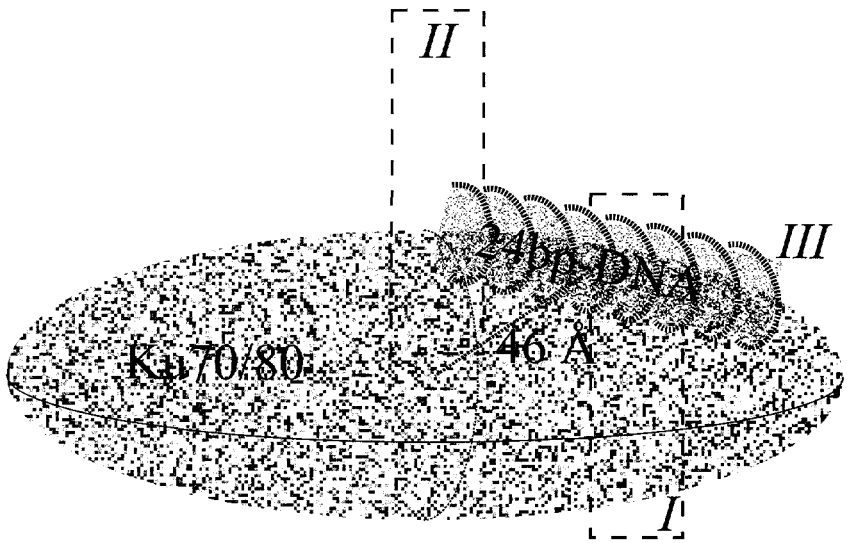
Other tasks within the ISBR have also made significant advances. We upgraded our NMR capability to be able to analyze larger biological structures. Different elements of the vibrational spectroscopy resource have been pulled together, and the resource is being optimized. We have completed an engineering design for the small-angle x-ray scattering laboratory. A theory of energetics for macromolecular complex formation is being developed using two different proteins involved in DNA damage and repair. A new state-of-the-art system for protein expression, purification, and crystallization has been designed and is under construction. Scientifically, major progress has been made in understanding the Ku/DNA complex structure through neutron scattering (see first figure), in modeling the structure of the PCNA/FEN-1 complex (second figure), and in studying the structure and function of FEN-1 by light spectroscopy (third figure).

Publications

Garde, S., et al., "Hydration of Biological Macromolecules: From Small Solutes to Proteins and Macromolecules," *Material Research Society Proceedings of the Symposium of Statistical Mechanics in Biological System* (Materials Research Society, Philadelphia, PA, in press).

Gilmanshin, R., et al., "Fast Events in Protein Folding: Relaxation Dynamics of Secondary and Tertiary Structure in Native Apomyoglobin," *Proc. Nat. Acad. Sci. U.S.A.* **94**, 3709 (1997).

Gilmanshin, R., et al., "Fast Events in Protein Folding: Relaxation Dynamics and Structure of the I Form of Apomyoglobin" (to be published in *Biochem.*).



We have used small-angle x-ray and neutron scattering to study the solution structure of Ku, a DNA double-strand break-repair protein composed of two subunits of approximately 70 and 80 kDa (Ku70/80). This figure shows two possible extreme binding positions of DNA on Ku that satisfy the experimental separation of centers of mass (*I* and *II*). Given the neutron data on both DNA oligos and the x-ray binding data, however, positions *I* and *II* are highly improbable. Position *I* will not give rise to the different separations of the centers of mass observed for the two different Ku/DNA complexes. Although position *II* could satisfy the different separations, it seem highly improbable. Since the binding of Ku to DNA is nonspecific, we would have expected to see a DNA:Ku binding stoichiometry of 1:2 instead of the 1:1 that is observed. It is more likely that DNA binds to Ku as depicted by position *III*, such that the DNA makes extensive contact with the surface of the protein.

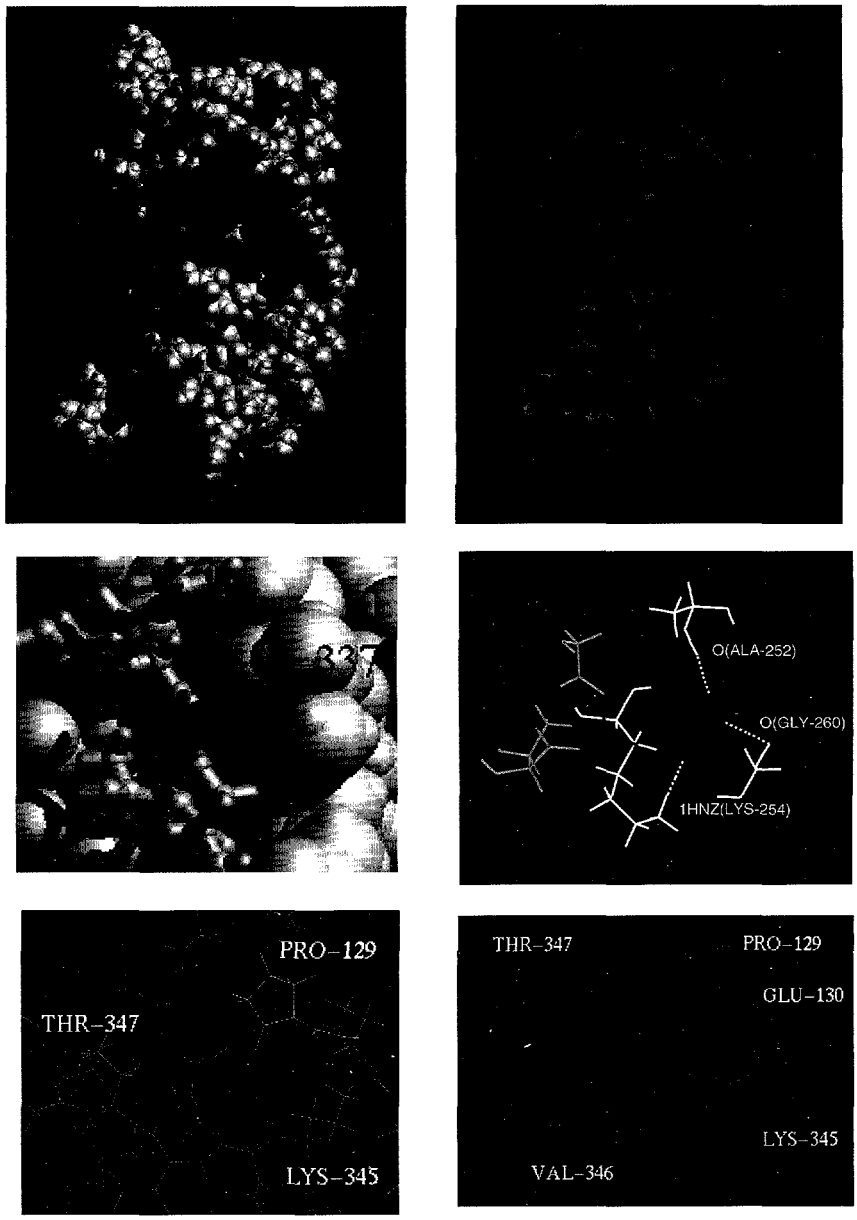
Gilmanshin, R., et al., "Structural Heterogeneity of the Various Forms of Apomyoglobin: Implications for Protein Folding," *Protein Sci.* **6**, 2134 (1997).

Hummer, G., et al., "The Pressure Dependence of Hydrophobic Interactions Is Consistent with the Observed Pressure Denaturation of Proteins" (to be published in *Proc. Natl. Acad. Sci. U.S.A.*).

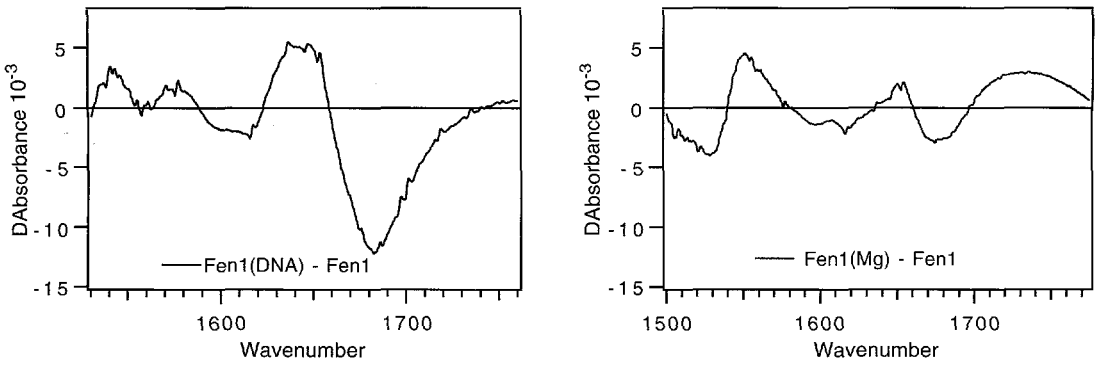
Kim, Y., et al., "Cyanide Binding and Active Site Structure in Heme-Copper Oxidases: Normal Coordinate Analysis of Iron-Cyanide Vibrations of a 32+CN-Complexes of Cytochromes ba3 and aa3" (to be published in *Biospectrosc.*).

Puustinen, A., et al., "Fourier Transform Infrared Evidence for Connectivity Between CuB and Glutamic Acid 286 in Cytochrome bo3" (to be published in *Biochem.*).

Riistama, S., et al., "Bound Water in the Proton Translocation Mechanism of the Heme-Copper Oxidases," *FEBS Lett.* **414**, 275 (1997).



PCNA (proliferating cell nuclear antigen) is involved in many important cellular functions including replication and repair mechanisms. In particular, it binds the FEN-1 and XPG repair proteins. To understand the specific binding of these DNA repair proteins to PCNA and to help in designing mutational studies, we have constructed a model of the structure of the human PCNA and FEN-1 complex. The model is based on two crystal structures: the yeast PCNA and the C-terminal region of P21 complexed with human PCNA. The modeled structure is shown in the two top panels. On the left, the FEN-1 binding peptide (residues 332–353) occupies the right-hand edge, the interdomain connecting loop (residues 119–133) is shown in dark grey, and PCNA makes up the rest of the molecule. The right panel shows the corresponding ribbonlike structure of the mainchain of the molecule. In this complex, GLN-337 of the FEN-1 binding domain is a conserved residue among several PCNA binding domains. As shown in the two middle panels, GLN-337 is buried inside a tight pocket while forming three hydrogen bonds with PCNA. Our mutation data indicates that the ILE128TRP mutation reduces the PCNA binding more drastically than does the ILE128GLU mutated sequence. The bottom two panels show the modeled structures of these two mutations. While ILE128GLU involves a nonpolar to a charged-residue mutation, the two side chains occupy a similar space. The charged group of GLU-128 is pointing toward the solvent. The larger TRP-128 requires more space than is available at ILE-128. A large structural distortion is required to accommodate the mutated TRP-128. Therefore, the PCNA binding is weaker for ILE128TRP than it is for ILE128GLU, in good agreement with the mutation data.



We have applied new Fourier transform infrared (FTIR) difference techniques to characterize the structure and function of FEN-1, a putative DNA-repair protein. FTIR measurements indicate that FEN-1 is predominantly composed of a-helix (>60%). The left panel shows the FTIR difference spectrum that is obtained when the FEN-1 apoprotein is reacted with a flap-DNA substrate. This spectrum clearly indicates that there is a substantial change in secondary structure upon binding to the DNA substrate. The amide I difference features are consistent with a significant increase (~10%) in the a-helix content in the complex. DNA binding causes the protein to self-organize, probably in the binding region. The right panel shows the reaction of the apoprotein with magnesium. In contrast with the DNA result, binding to magnesium does not cause a significant change in secondary structure. However, there is at least one carboxylate side chain that is strongly shifted upon binding. The magnitude and direction of the shift (+20 cm^{-1}) are consistent with bidentate binding of the carboxylate. These results offer important new insights into the structures involved in the DNA binding and cleavage functions of the FEN-1 enzyme.

Landscapes and Dynamics of Proteins

Hans Frauenfelder

Simple systems, such as atoms, nuclei, and many solids, can be described by energy-level schemes. In contrast, complex systems, such as glasses and proteins, must be described by an energy landscape. The goal of this project is the characterization and understanding of the energy landscape and its connection to structure and dynamics in proteins. Such an understanding will have profound impact on many fields, for instance, the production of new enzymes and of new drugs.

The work during the past year progressed in a number of different directions, as is best exemplified by the publications. Contact with experimentalists, theorists, and computer experts, both within and outside the Laboratory, stimulated new ideas and directions. Our work is recognized worldwide, and we were invited speakers at many conferences and colloquia.

An example of a recent and as yet unpublished result is the binding of small molecules such as CO to myoglobin. The process was considered to be fully understood 25 years ago. Our work last year, performed in collaboration with our various co-investigators, indicates that the mechanism is actually incredibly sophisticated and precise. If the results hold up, they indicate that the protein is built to tolerances of less than 0.1 Å! Moreover, the reaction mechanism involves very tight "collaboration" among weak and strong chemical forces. The result may be important not just to the binding process, but much more generally to all biological reactions.

Publications

Beardmore, K.M., et al., "Ab-Initio Calculations of the Gold-Sulfur Interaction for Alkanethiol Monolayers," *Synth. Met.* **84**, 317 (1997).

Beardmore, K.M., et al., "Determination of the Headgroup-Gold (111) Potential Surface for Alkanethiol Self-Assembled Monolayers by Ab Initio Calculation" (submitted to *Phys. Rev. Lett.*).

Beardmore, K.M., et al., "Molecular Dynamics Simulation of Self-Assembled Monolayers with Defects," *Abstr. Pap. Am. Chem. Soc.* **214** (2) 6 (1997).

Frauenfelder, H., "The Complexity of Proteins," in *Physics of Biological Systems*, H. Flyvbjerg et al., Eds. (Springer-Verlag, Berlin, 1997), p. 29.

Frauenfelder, H., "Proteins: A Challenging Many-Body Problem," *Philos. Mag.* **B74**, 579 (1996).

Frauenfelder, H., "Proteins and Glasses," *Struct. Dyn. Glasses and Glass Formers* **455**, 343 (1997).

Frauenfelder, H., "Proteins as Paradigms of Complex Systems," in *Roentgen Centennial*, E. Umbach, Ed. (World Scientific, Singapore, 1997), p. 227.

Frauenfelder, H., and P.G. Wolynes, "Picturing the Working Protein," in *Theoretical and Computational Genome Research,* S. Suhai, Ed. (Plenum Press, New York, 1997), p. 231.

Garcia, A.E., et al., "Variations on a Theme by Debye and Waller: From Crystal to Proteins," *Proteins* **29**, 153 (1997).

Gronbech-Jensen, N., et al., "Lekner Summation of Coulomb Interactions in Partially Periodic Systems" (to be published in *Mol. Phys.*).

Nienhaus, G.U., et al., "Exploring the Conformational Energy Landscape of Proteins," *Physica D* **107**, 297 (1997).

Similarity Landscapes: An Improved Method for Scientific Visualization of Information from Protein and DNA Database Searches

Norman Doggett

Our goal has been to answer questions about the nature of nonrandom patterns that are observed in evolutionary processes. Are they truly nonrandom? To what extent can the patterns observed be attributed to selection and other nonrandom processes, and to what extent might they have been generated by random events that sometimes mimic nonrandom ones?

This year, we used computer simulations and examination of a variety of databases to answer a wide range of evolutionary questions. We found that there is a clear distinction in the evolution of HIV-1 and HIV-2, with the former and more virulent virus evolving more rapidly at a functional level. We discovered highly nonrandom patterns in the evolution of HIV-1 which can be attributed to a variety of selective pressures.

In examining microsatellite DNA (short repeat regions) in microorganisms, we found clear differences in the distribution of microsatellites between prokaryotes and eukaryotes, differences which can be tied to different selective pressures. We developed a new method, called "topiary pruning," for enhancing the phylogenetic information contained in DNA sequences. Most recently, we discovered effects in complex rainforest ecosystems that indicate strong frequency-dependent interactions between host species and their parasites, leading to the maintenance of ecosystem variability.

Publications

Field, D., and C. Wills, "Abundant, Polymorphic Microsatellites in Yeast, Contrasted with Far Fewer Such Microsatellites in Six Prokaryotic Genomes, Show a Differential Role for Selection in Eukaryote and Prokaryote Microsatellite Evolution" (submitted to *Proc. Natl. Acad. Sci. U.S.A.*).

Gagneux, P., et al., "Resolution of Branching Order and Detection of Different Demographic Histories in Recent Hominoid Evolution" (submitted to *Science*).

Wills, C., "Directional Selection Acting on HIV-1 Genes is Correlated with Codon Mutability and Is Most Pronounced among Recently Diverged Sequences" (submitted to *Proc. Natl. Acad. Sci. U.S.A.*).

Wills, C., and R. Condit, "Strong Density- and Diversity-Related Effects Help to Maintain Tree Species Diversity in a Neotropical Rainforest" (submitted to *J. Evol.*).

Nonlinear Analysis of Biological Sequences

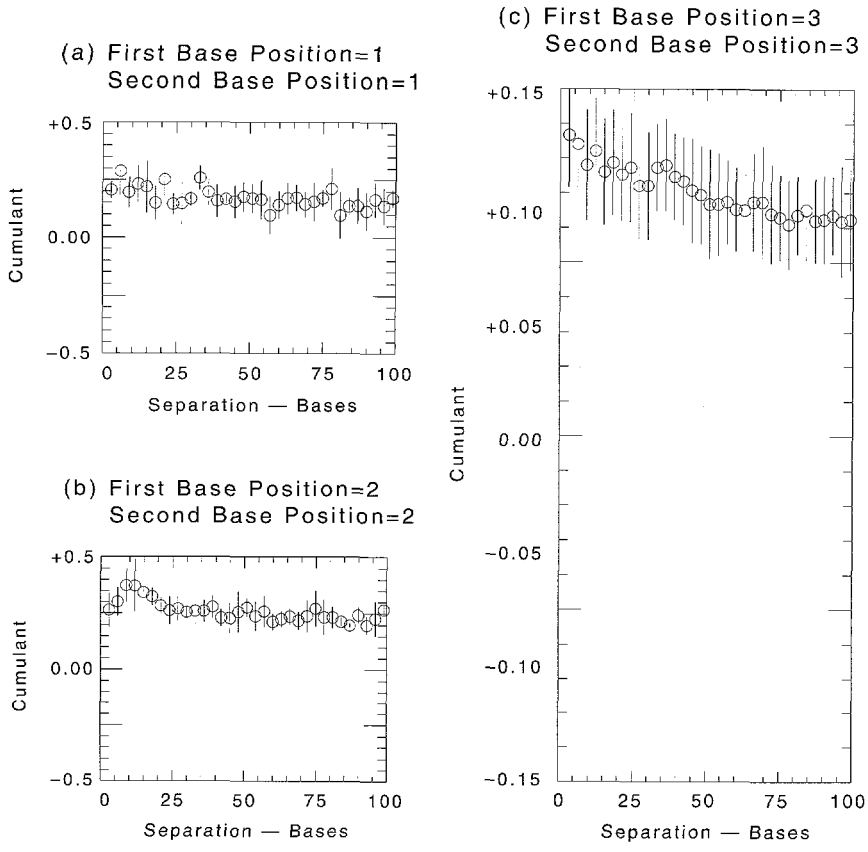
David Torney

The purpose of this project is to develop effective new computational and mathematical techniques for predicting the structure and function of biological sequences. We are currently focusing upon the detection and characterization of functional domains within biological sequences. In addition, we are modeling multigenic human diseases with the objective of designing effective experiments for discovering their genetic components.

This year we completed a full characterization of the stationary statistical properties of human DNA coding sequences. This characterization was achieved using the sample cumulants of a data set containing in-frame, nonredundant sequences totaling over 500,000 bases (see figure). Prior to our investigations, it was not known how to make the best use of a sample of sequences for classifying additional sequences. We created a tabulation of all the properties by which coding sequences might distinguish themselves and used the sample to determine the "weights" for each property.

There has been a well-justified concern that there might be additional statistical properties of coding sequences than are manifest in, say, codon frequencies, or found by neural networks, and that the identification of these properties could improve the accuracy of gene prediction. This is a natural application for our results. Our techniques are also applicable whenever the objective is to learn the statistical properties of a class of sequences from example sequences.

We have also developed a new, distance-based phylogeny reconstruction program. Molecular phylogenies (or evolutionary trees) have many uses in biology, but we are especially interested in using them in the statistical analysis of protein alignments to model protein structure and function. Existing methods are either hopelessly slow (e.g., likelihood methods) or strikingly inaccurate (e.g., neighbor-joining methods) for this application. Our new approach is similar in spirit to the neighbor-joining methods but aims to approximate the likelihood function as closely as possible without greatly sacrificing computational speed. We have found that much of the correlation structure in the problem occurs in simple weighted least-squares expressions with reduced variances, reflecting only the "non-additive noise" in the distance fluctuations.



In this figure, we display some interesting statistical properties of coding sequences. Graphs (a)–(c) are four-digit cumulants for binary encoded, DNA coding sequences with a = 1 1, c = 1 −1, g = −1 1, and t = −1 −1. These four digits are derived from two codon bases, with different spacings between the codons. In (b), there is a peak at a spacing of 10 bases, perhaps reflecting a-helices. In (c), the magnitude is the greatest because the third codon base codes degenerately.

Publications

Arvestad, L., and W.J. Bruno, "Estimation of Reversible Substitution Matrices from Multiple Pairs of Sequences" (to be published in *J. Mol. Evol.*).

Bruno, W.J., et al., "Probability Distributions on Binary Sequences" (submitted to *Proc. Nat. Acad. Sci. U.S.A.*).

Macken, C., and A. Schliep, "Equivalence of Hidden Markov Models" (submitted to *J. Appl. Probability*).

Investigations of Biomimetic Light-Energy-Harvesting Pigments

Robert Donohoe

Light-energy-harvesting pigments are of interest for energy conversion and the destruction of pollutants. The rational design of such pigments requires foreknowledge of the structure and photophysical response of the pigments—in our case porphyrins. We have succeeded in identifying and manipulating the structural features in multiporphyrin arrays that allow control of light-initiated response. The ability to absorb (or harvest) light and transmit the excited-state energy to a desired location is a process that occurs very efficiently in plants and photosynthetic bacteria. To mimic this process, we must assemble pigments in a controlled geometry so that interpigment communication is controlled by the structure. The relatively rigid diarylethyne linkers used in the multiporphyrin arrays of our study provide such control.

A second critical design requirement is the ability to predict the absorption cross section of the assembled pigments. Practically, this means that the individual pigments must retain their characteristic absorption patterns so that the arrays will manifest composite absorption data. However, such weak coupling cannot come at the expense of limited energy-transfer efficiency. Again, our diarylethyne-linked porphyrin arrays are a success: the absorption data we have obtained are a composite of those of the individual pigments, while the energy transfer rates between pigments greatly exceed the inherent relaxation dynamics of the separate porphyrin centers.

Finally, the ability to direct harvested-light energy to a desired location within an array requires a selection of multiple pigments with "donor" and "acceptor" energy-transfer characteristics. We have shown that the addition of substituents, orbital ordering, metallation, and linker geometry manipulation are all effective in this regard.

Publications

Hsiao, J.-S., et al., "Soluble Synthetic Multiporphyrin Arrays. 2. Photodynamics of Energy Transfer," *J. Am. Chem. Soc.* **118**, 11181 (1996).

Li, F., et al., "Effects of Central Metal Ion (Mg, Zn) and Solvent on Singlet Excited State Energy Flow in Porphyrin-Based Nanostructures," *J. Mater. Chem.* **7**, 1245 (1997).

Seth, J., et al., "Soluble Synthetic Multiporphyrin Arrays. 3. Static Spectroscopic and Electrochemical Probes," *J. Am. Chem. Soc.* **118**, 11194 (1996).

Strachan, J.-P., et al., "Effects of Orbital Ordering on Electronic Communication in Multiporphyrin Arrays" (to be published in *J. Am. Chem. Soc.*).

Strachan, J.-P., et al., "Synthesis and Characterization of Tetrachlorodiarylethyne-Linked Porphyrin Dimers: Effects of Linker Architecture on Intradimer Electronic Communication" (to be published in *Inorg. Chem.*).

Van Patten, P.G., et al., "Energy-Transfer Modeling for the Rational Design of Multiporphyrin Light-Harvesting Arrays" (to be published in *J. Phys. Chem.*).

Wagner, R.W., et al., "Molecular Optoelectronic Gates," *J. Am. Chem. Soc.* **118**, 3996 (1996).

Structure, Dynamics, and Function of Biomolecules

Hans Frauenfelder

We have enhanced Los Alamos's core competency in bioscience and biotechnology by building on our present strengths in experimental techniques, theory, high-performance computing, modeling, and simulation applied to biomolecular structure, dynamics, and function. Specifically, we have strengthened our neutron/x-ray scattering, x-ray crystallography, nuclear magnetic resonance, laser, and optical spectroscopies and provided a jumping-off point for the development of an integrated structural biology capability.

Initially we focused on neutron/x-ray scattering to study the structures of biomolecular complexes in solution. This year, we completed the design and implementation of new neutron scattering instrumentation for structural biology (see the first figure), continued developing new methods for analysis of scattering data from biomolecules, and developed new structural biology projects that use existing neutron and x-ray scattering instrumentation to study biomolecular interactions (see the second figure).

Building the core competency has also involved strengthening interactions between theory and experiment and between the biological and physical sciences. To foster this interaction, we continued to sponsor regular meetings of members from all interested Laboratory organizations and to support the lecture series "Issues in Modern Biology," which has attracted distinguished external speakers, including close to a dozen Nobel Prize winners over the course of this project. We also supported the formation of multidisciplinary/cross-divisional teams to develop projects in Science-Based Bioremediation and an Integrated Structural Biology Resource. Finally, we successfully worked with a multidisciplinary team to put forward the Laboratory's tactical goal of "The Genome and Beyond."

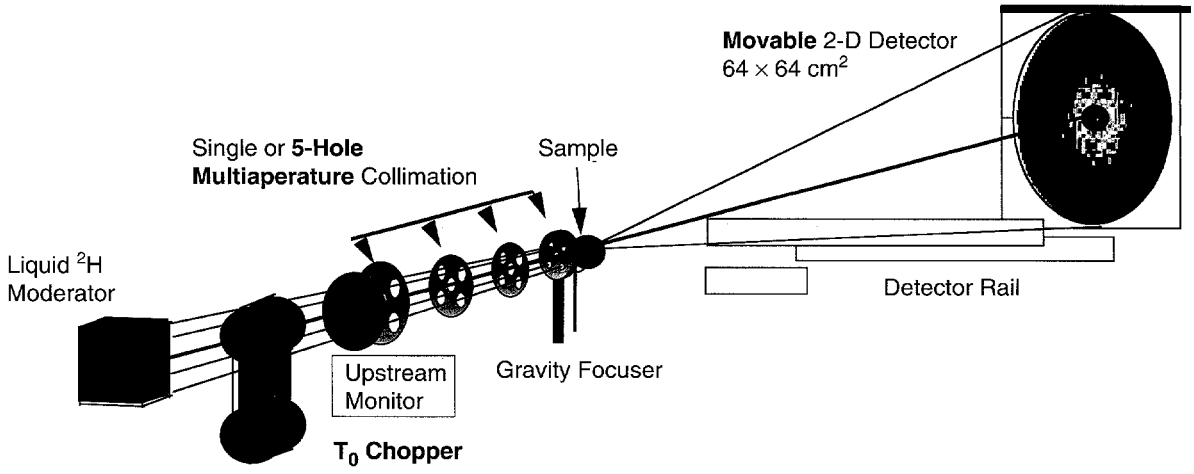
Publications

Krueger, J.K., et al., "Structures of Calmodulin and a Functional Myosin Light Chain Kinase in the Activated Complex: A Neutron Scattering Study," *Biochemistry* **36**, 6017 (1997).

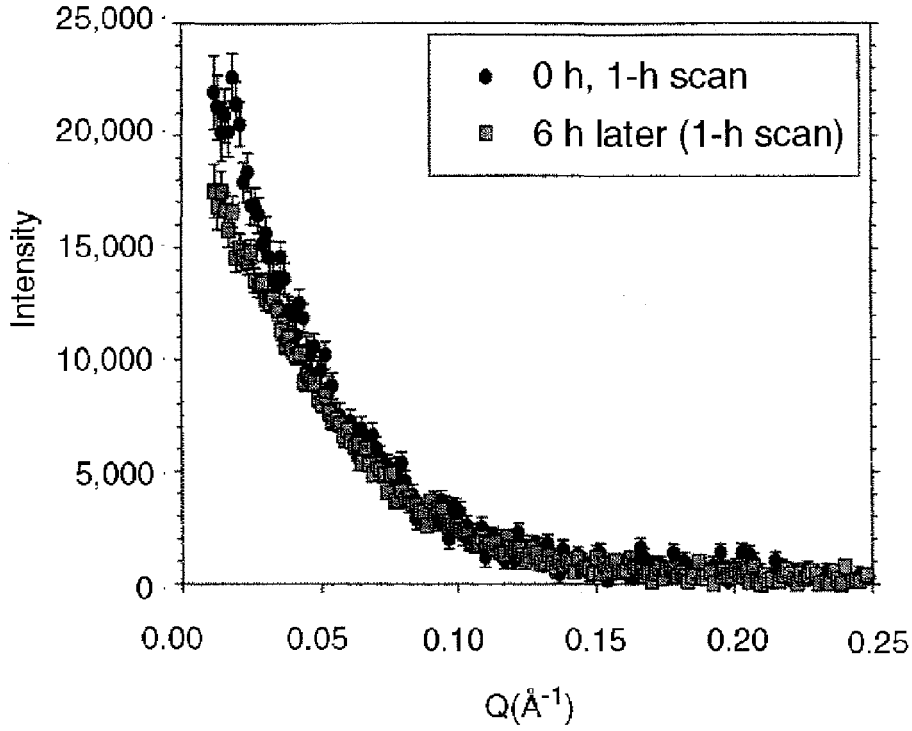
Krueger, J.K., et al., "The Structures of Calmodulin and a Functional Myosin Light Chain Kinase in Their Complex: A Study Using Small-Angle Neutron Scattering with Contrast Variation," *Biophys. J.* **72**, 129 (1997).

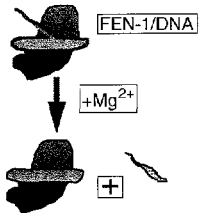
Mecklenberg, S., et al., "Tertiary Structural Changes and Iron Release from Human Serum Transferrin," *J. Mol. Biol.* **270**, 739 (1997).

Olah, G.A., and R.P. Hjelm, "Analysis and Simulation of a Small-Angle Neutron Scattering Instrument on a 1-MW Long-Pulsed Spallation Source" (to be published in *J. Neutron Res.*).



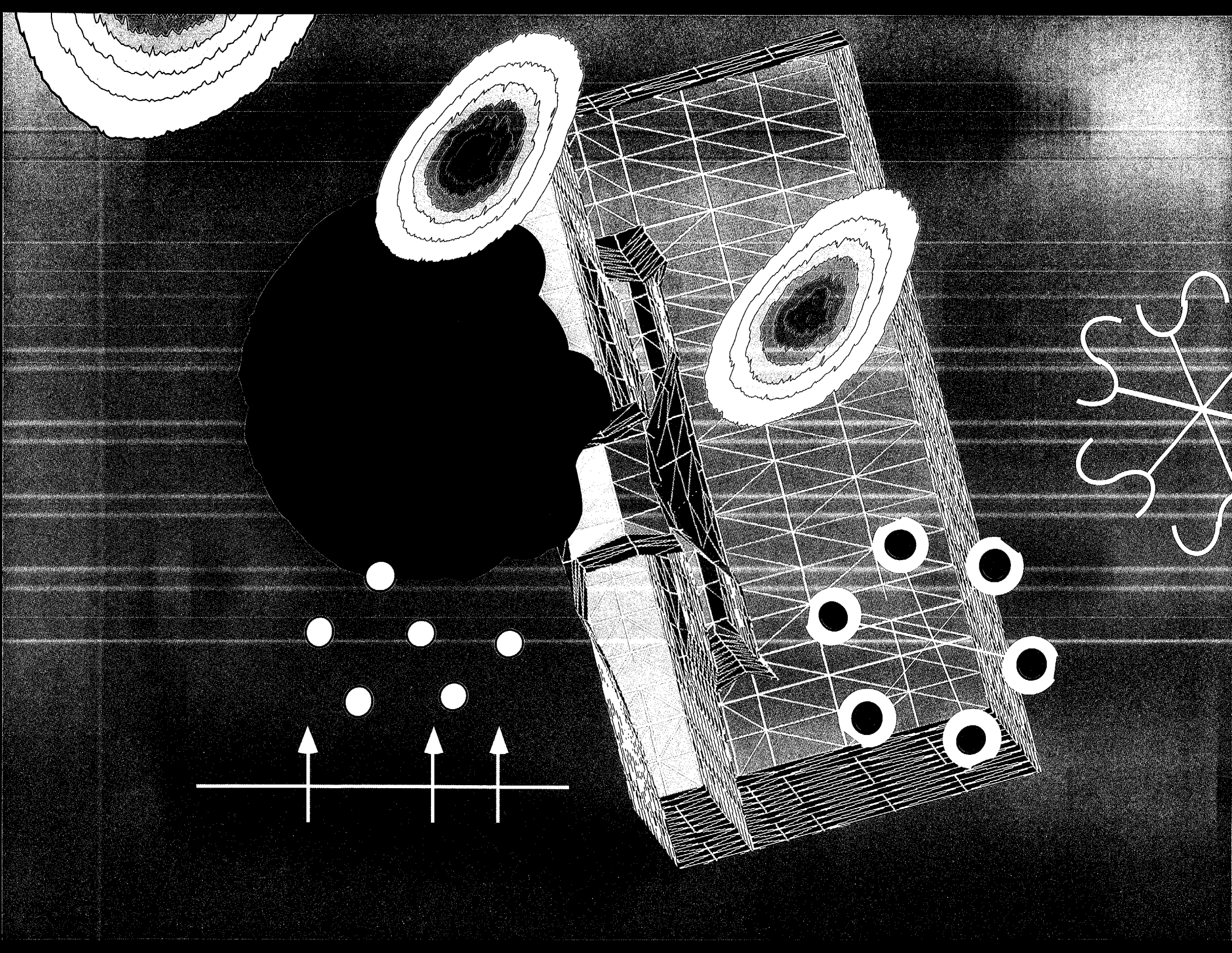
Schematic showing upgrades to the low-Q neutron diffractometer (LQD) at the Manuel Lujan Jr. Neutron Scattering Center. To date, structural biology applications using the LQD have been limited by the neutron flux from this instrument. In order to increase the flux of neutrons on a sample at high resolution, we designed and implemented a number of changes. The 5-hole multiaperture collimator allows the sample to "see" a larger area of the neutron source moderator and hence capture more neutrons. The T_0 chopper minimizes unwanted background, and the variable detector position allows for better optimization of resolution and neutron flux for individual experiments.

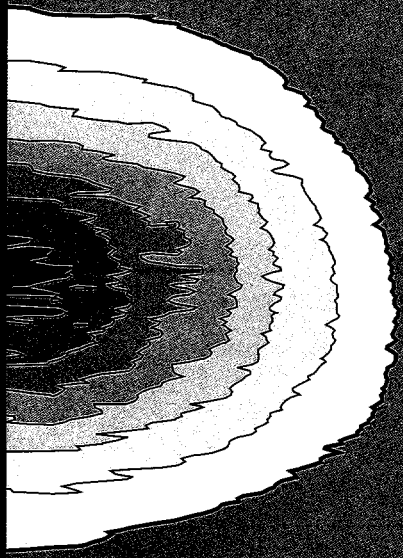




- DNA bound to FEN-1 is needed to see Mg^{2+} activation effect.
- Decrease in scattering amplitude is consistent with FEN-1 remaining bound to double-stranded DNA after cleavage.

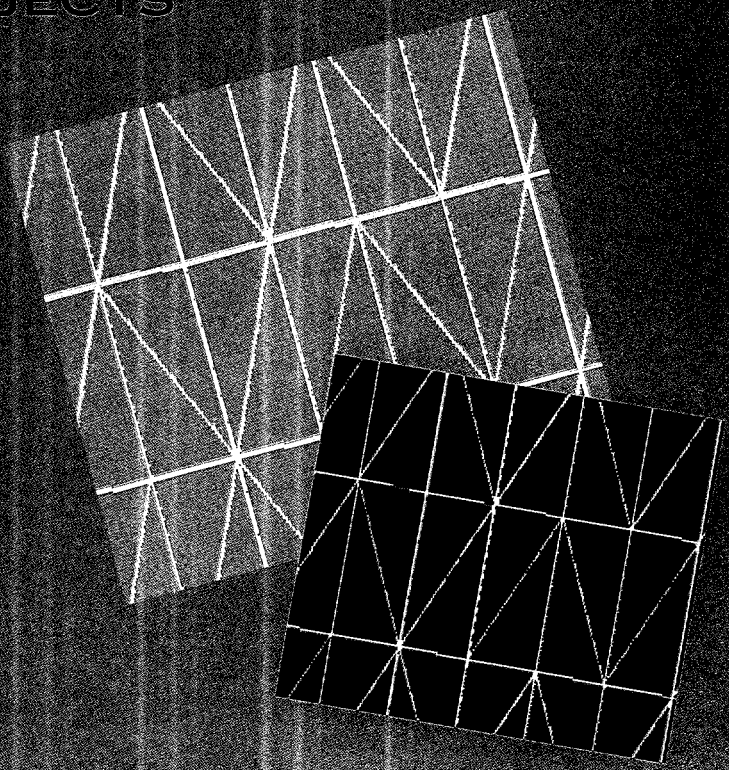
Results of an x-ray scattering experiment on a protein that repairs double-stranded DNA which has been "nicked" and the 5'-end of the nick expands to form a single-stranded "flap." The protein, FEN-1, is an endonuclease which binds to the DNA and excises the unwanted flap. This process requires a number of cofactors, including Mg^{2+}. X-ray scattering data from FEN-1/DNA complexes in which the DNA fragments are designed to readily form flap structures have shown that when Mg^{2+} is added, the flap of the DNA is cleaved and the protein remains bound to the DNA. These data provide information about the initial steps in the repair process, and future experiments will probe successive steps until a complete molecular picture is built.





1997 LDRD

Program Development Projects



Materials Science

Disposition of Plutonium as Nonfertile Fuel for Water Reactors

Kenneth Chidester

We are investigating the possibility of developing nonfertile and evolutionary mixed oxide (EMOX) fuels. Traditional nonfertile fuel does not contain uranium, which breeds plutonium, but instead uses a material such as calcia-stabilized zirconia as the fuel matrix. This fuel could be used as an option for deep-burning plutonium in that the spent-fuel standard is met or exceeded and more than 60% of the initial plutonium is destroyed through burnup, compared with approximately 20% for typical mixed oxide (MOX) fuel.

The conversion of the surplus inventory of weapons plutonium into the spent-fuel standard, however, does not address the larger, global issue of the accumulation of unseparated plutonium in spent reactor fuel. Therefore, we expanded the scope of this project to include investigations into the feasibility of developing EMOX fuel, which would be a combination of standard MOX and the nonfertile fuel. The EMOX concept has the potential for allowing existing water reactors to be used, without modification, as net consumers of plutonium instead of net breeders and could lay the groundwork for a smooth transition from conventional to nonfertile fuel cycles. This year, our project had four main objectives: develop higher-fidelity physics models, complete analysis of previous fabrication activities, incorporate results into existing Nuclear Vision models, and focus on development of follow-on programs.

We developed a four-assembly, lattice-physics, color-set model that allowed analysis of EMOX compositions and neutronic properties in more realistic flux environments. We analyzed a spectrum of nonfertile and uranium/nonfertile compositions. One-fourth core-loading results indicate two trends with increasing weight percents of plutonium compositions in the fresh EMOX: a desirable increase in the net total of plutonium destroyed per cycle but an accompanying increase in the residual plutonium discharged in the spent fuel. We used the solid-state reaction method to fabricate nonfertile and EMOX fuels containing weapons-grade plutonium. The results of microstructural and x-ray diffraction analysis indicate formation of a solid-solution, face-centered cubic structure for both fuel forms. The densities of sintered pellets ranged from 90% to 97% of theoretical calculations.

We used the EMOX analysis to feed a global energy model being developed as part of the Nuclear Visions project. The EMOX data will also serve as a reference for future systems studies on the impact of incorporating nonfertile fuel into global nuclear fuel cycles. The EMOX systems analysis and reactor physics work done this year served as the foundation for a proposal submitted through the Nuclear Visions Project to the DOE Office of Nuclear Energy for an 8-year, $44 million proliferation-resistant fuel and cycle development program scheduled to begin in FY 1999.

Publications

Beard, C.A., et al., "Development of Evolutionary Mixed Oxide Fuel," *1996 ANS/ENS Fall Int. Meet.* **75**, 75 (1996).

Buksa, J., et al., "Disposition of Plutonium as Mixed Oxide Nuclear Fuel," *Fourth Int. Conf. Nucl. Eng. (ICONE-4)* **4**, 453 (1996).

Chodak, P., and J. Buksa, "A Practical Strategy for Reducing the Future Security Risk of United States Spent Nuclear Fuel" (submitted to *Global '97 Int. Conf. Future Nucl. Systems*).

Eaton, S.L., et al., "Development of Advanced Mixed Oxide Fuels for Plutonium Management" (submitted to *Global '97 Int. Conf. Future Nucl. Systems*).

Eaton, S.L., et al., "Evolutionary Mixed Oxide Fuel Performance in Pressurized Water Reactors," *1996 ANS/ENS Fall Int. Meet.* **75**, 76 (1996).

Krakowski, R.A., et al., "Reduction of Worldwide Plutonium Inventories Using Conventional Reactors and Advanced Fuels: A Systems Study" (submitted to *Global '97 Int. Conf. Future Nucl. Systems*).

Ramsey, K.B., and H.T. Blair, "Fabrication of a Non-Fertile Fuel for the Disposition of Weapons Plutonium," *Fourth Int. Conf. Nucl. Eng. (ICONE-4)* **4**, 499 (1996).

Ramsey, K.B., and K. Chidester, "Fabrication of Non-Fertile and Evolutionary Mixed Oxide Fuels," *1996 ANS/ENS Fall Int. Meet.* **75**, 79 (1996).

Frequency-Resolved Optical Gating: A Diagnostic for the Characterization of Optoelectronic Materials and Devices

Antoinette Taylor

Frequency-resolved optical gating (FROG) is an ultrafast diagnostic technique that measures the full electric field, both instantaneous amplitude and phase, of an arbitrary ultrashort pulse. Because FROG is a general, broad-band optical diagnostic, it can be used in research areas for which characterizing the full electric field of an optical pulse would provide critical insight into physical processes. The objective of this research is to address issues relevant to high-speed optoelectronic materials and devices using FROG.

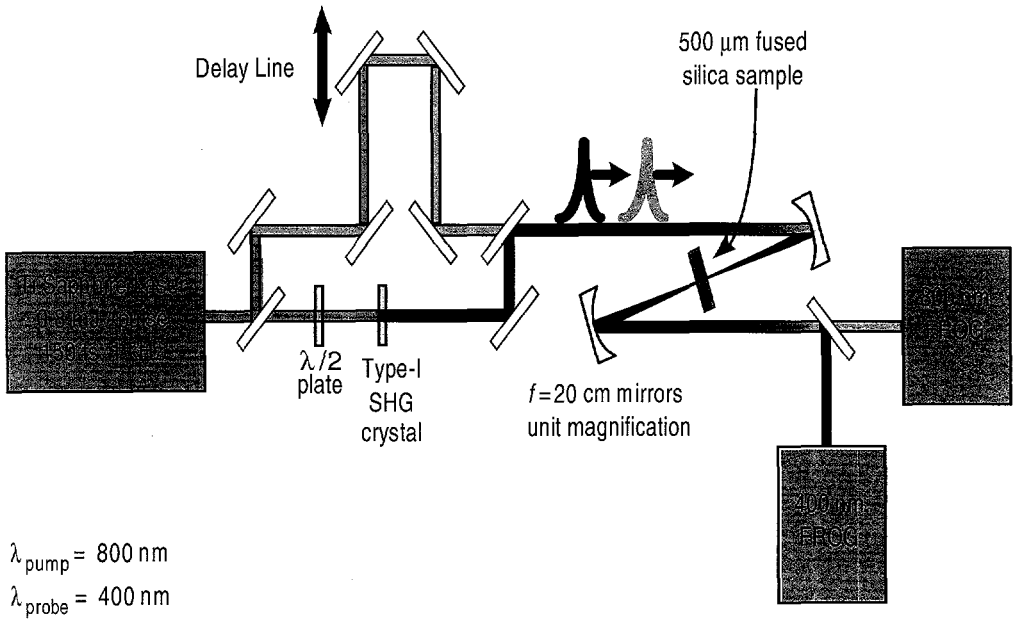
This year we continued to demonstrate FROG as a technique for determining basic properties of optical materials, such as dispersion, nonlinear refraction, two-photon absorption, and cross-phase modulation, by measuring the phase change of the optical pulse via FROG after the pulse traverses the material. The emphasis of this research is to determine the types of measurements possible with FROG, as well as the accuracy of those measurements.

We have completed a thorough set of measurements of cross-phase modulation in a variety of optical materials using the FROG setup shown in the first figure. We have demonstrated that this method results in the accurate measurement of cross-phase modulation coefficients in optical materials, with the error dominated by the uncertainty in the fluence measurement. The raw data, or FROG traces, for these measurements are shown in the second figure, while the analyses of the data used to determine the cross-phase modulation n_2 are shown in the third and fourth figures. However, the larger the frequency mismatch, the less accurate the measurement because walk-off of the two pulses limits the interaction region and hence the measured phase shift.

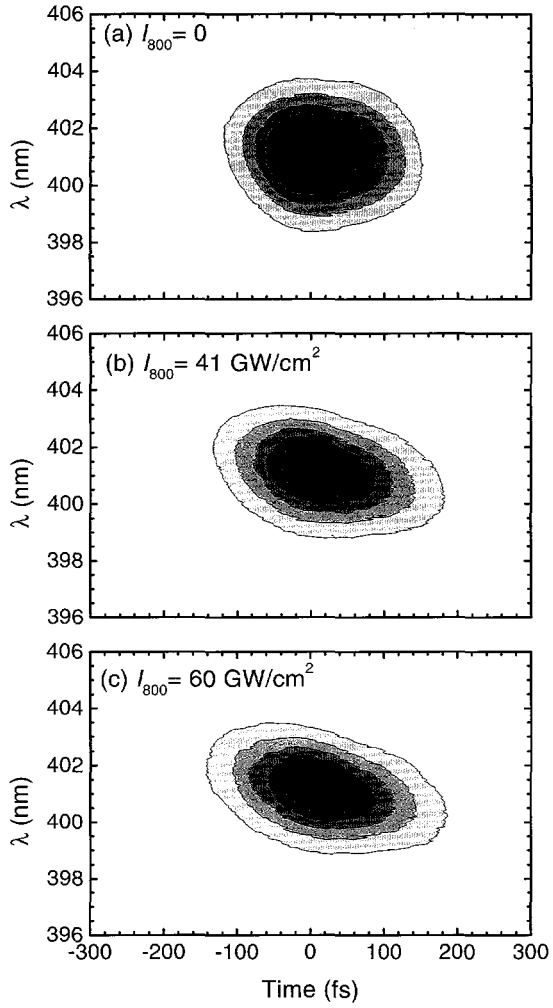
Publications

Kurnit, N.A., et al., "Measurement and Control of Optical Nonlinearities of Importance to Glass Laser Fusion Systems," *Proc. Soc. Photo-Opt. Instrum. Eng.* **3047**, 387 (1997).

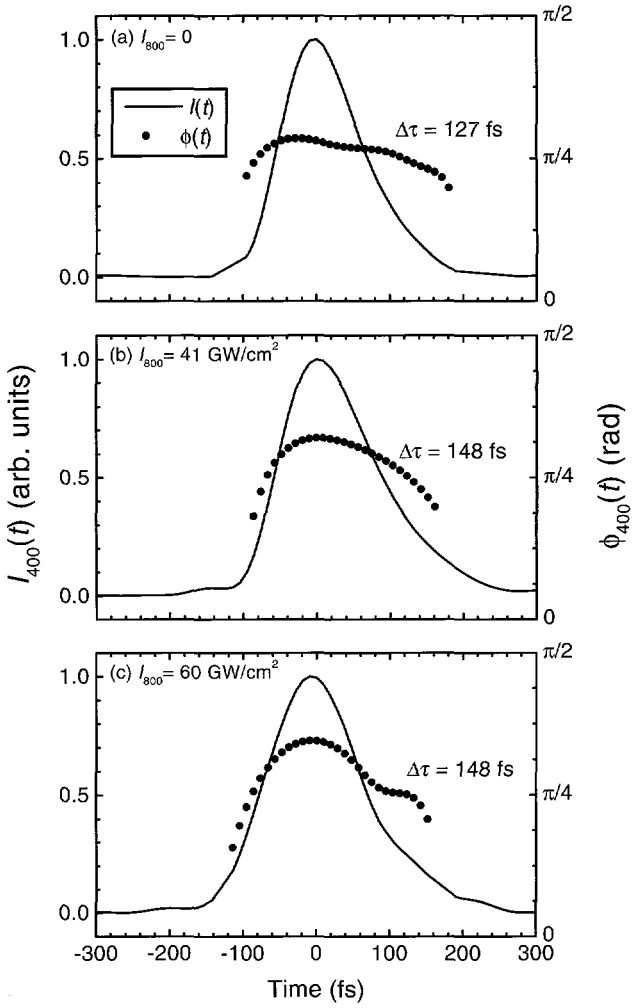
Rodriguez, G., and A.J. Taylor, "Measurement of Cross-Phase Modulation in Optical Materials through the Direct Measurement of the Optical Phase" (submitted to *Opt. Lett.*).



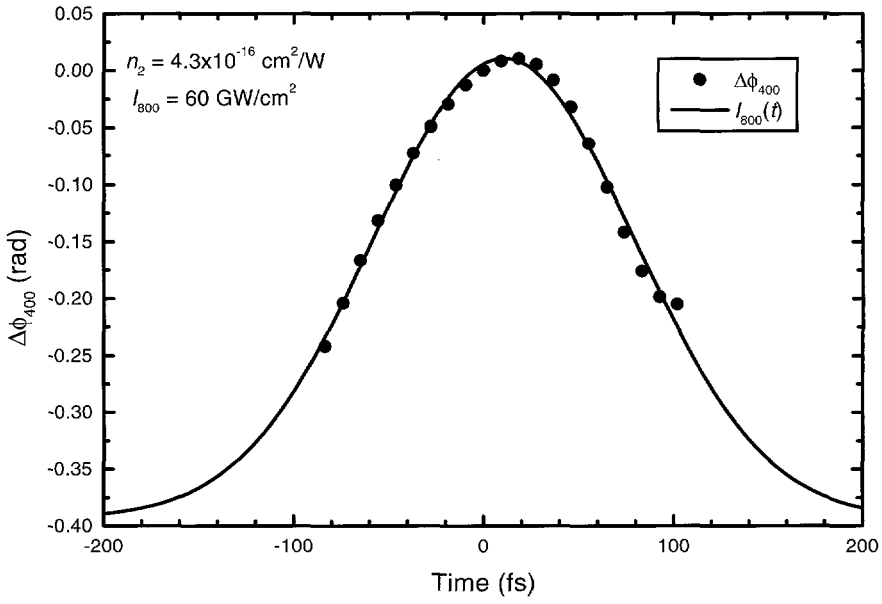
Experimental setup used to measure cross-phase modulation at wavelength λ of 400 nm by an 800-nm pulse. (SHG is second harmonic generation and *f* is focal length.)



Series of FROG traces with increasing pump pulse intensity (I_{800}). Increasing distortion of the FROG trace with increasing intensity is due to the effect of cross-phase modulation.



Retrieved amplitude $I(t)$ and phase $\phi(t)$ of the probe pulse's electric field with increasing intensity I_{800}. The pulse width $\Delta\tau$ remains 148 fs, while the phase changes significantly with increasing intensity.



Fit of phase change $\Delta\phi$ of 400-nm pulse to intensity profile I of 800-nm pulse determines value of cross-phase modulation n_2.

New Membrane Solutions for Hydrogen Isotope Issues

Robert Dye

Currently, there is no production of tritium in the United States. Until tritium production resumes, we need new, more efficient ways of handling and recovering tritium for use in current weapon systems containing this limited-lifetime gas. After tritium production resumes, we will also need novel approaches to safely capturing and collecting tritium. The use of new membrane strategies can lead to improved techniques for handling and processing tritium and to safe methods for cleaning up mixed waste. This project focuses on developing new membrane solutions that will provide alternatives to tritium supply and handling techniques.

This year we developed a metal membrane that has more than doubled the hydrogen flow rates. This dramatic improvement is the result of our improved understanding and processing of the metal membrane system. These membranes also show room-temperature permeation. Standard lead-membrane systems are operated at temperatures around 300°C, have 20 times lower flows, and break up at lower temperatures. So far in the current effort, we have measured deuterium/hydrogen selectivities of more than 1.6 (the ratio of hydrogen gas flow to deuterium gas flow).

While we did not expect the present metal membrane to display a large improvement in isotope separation, we are currently developing a membrane made from alloying palladium with 5% to 10% yttrium that we expect will improve this separation. In comparison with a membrane consisting of pure palladium, the palladium/yttrium membrane will decrease the hydrogen flow rate by a factor of 3 and will slightly improve the diffusivity of tritium. We are also constructing a composite metal membrane using lead/yttrium as the surface catalyst layer on both sides of a vanadium foil that may make possible the separation factors required to perform hydrogen isotope separation using membranes.

Publications

Dye, R.C., and T.S. Moss, "Advancing the Technology Base for High Temperature Hydrogen Membranes" (to be published in *Proc. 15th Annu. Membrane Technology/ Separations Planning Conf.*).

Moss, T.S., and R.C. Dye, "Composite Metal Membranes for Hydrogen Separation Applications," *Proc. 8th Annu. U.S. Hydrogen Meet.* **8**, 357 (1997).

Moss, T.S., and R.C. Dye, "Engineering Materials for Hydrogen Separation" (to be published in *Hydrogen and the Materials of a Sustainable Energy Future*).

Moss, T.S., and R.C. Dye, "Experimental Investigation of Hydrogen Transport through Metals," *NASA Conf. Publication 3354*, 351 (1997).

Point-of-Use VOC Control in Semiconductor Fabrication

Mark Cummings

Los Alamos National Laboratory and SEMATECH have evaluated a silent discharge plasma (SDP) device for point-of-use control of specific semiconductor volatile organic compound (VOC) emissions at the source. Destruction efficiencies were initially determined at the bench scale using controlled gas mixtures, and system performance was measured for simulated emissions containing a variety of VOCs and perfluoro-compounds (PFCs). Three potential applications of SDP equipment were explored: (1) treatment of emissions from a solvent clean operation, focusing on methanol, isopropanol, and acetone; (2) treatment of photoresist (lithography) streams containing mixtures of hexamethyldisilazane (HMDS), ethyl 3-ethoxy propionate, and propylene glycol monomethyl ether acetate (PGMEA); and (3) destruction of PFCs, focusing on the CF_4 and C_2F_6 compounds.

Based on this work, a field-pilot unit was designed and tested at a SEMATECH member site using two slip streams: (1) PGMEA and HMDS gas mixture from lithography tools and (2) acetone, PCE, and methanol from a wet bench cleaning tool. Based on the pilot test data, cost of ownership estimates for the SDP technology show annual operating expenses (including amortized capital and installation costs, maintenance, and utilities) are $8,300 for a single 250-scfm lithotrack tool. Point-of-use SDP system costs are $33,300 per 1000 scfm as compared with about $22,000 per 1000 scfm for a typical end-of-pipe (EOP) concentrator/thermal abatement system. Although replacing existing EOP systems with SDP is not recommended, SDP could easily be installed in niche circumstances for point-of-use control of VOCs from lithotrack tools.

Fundamental and Applied Studies of Helium Ingrowth in Plutonium

Michael Stevens

The most important issue regarding the integrity of plutonium in the stockpile is the inherent alpha decay, which causes constant helium accumulation and concomitant recoil damage to the lattice from the associated uranium atom. Of particular concern is the metallurgical condition of the lattice and the possible precipitation of either helium atoms or vacancies into voids or bubbles. This project's goals are to put in place the essential capabilities and investigations to better understand, model, and observe the possible consequences of aging in plutonium.

During this past year we completed a calculation to predict lattice relaxation around a gallium atom in plutonium. This calculation is important for predicting how phase stability may be affected by gallium microsegregation over time. This information is also important for future calculations that may need an interatomic potential for plutonium.

We wrote a code for calculating electronic and vibrational entropy in plutonium; developed a two-dimensional electronic structure code; provided an electronic potential for plutonium-gallium for use in a Los Alamos x-ray photoelectron code (important for interpreting x-ray photoelectron spectroscopy data on aged plutonium samples); and calculated the lattice constants and bulk modulus of plutonium, PuHe, and $PuHe_3$ using a full-potential local-density approximation. We also used an embedded atom model calculation of the elastic constants in plutonium. Phonon dispersion curves on plutonium were also calculated, indicating that these are smooth and expected to be like the dispersion curves for other face-centered cubic metals.

In addition, we synthesized alloys with entrapped helium to be used as surrogates for the development of the laser-driven miniflyer technique for assessing aging effects on shock response of plutonium, and we began preparing aged plutonium samples from the stockpile for transmission electron microscope examination. Development continued on the helium effusion cell for measuring helium outgassing from aged plutonium.

Publications

Becker, J.D., et al., "Calculated Structural Disorder in Delta-Stabilized Pu" (to be published in *J. Alloys Compd.*).

Eriksson, O., et al., "Theoretical Confirmation of Nearly Degenerate States in Delta-Pu" (to be published in *Phys. Rev. Lett.*).

Development of High Magnetic Fields for Energy Research

Joe D. Thompson

Our objective has been to establish the technological and scientific bases upon which the DOE Office of Basic Energy Sciences would form a partnership with the National Science Foundation (NSF) to build an entirely new type of 100-T nondestructive magnet and an associated high-field science program.

This year we have developed new, and improved existing, experimental-data acquisition techniques appropriate for measurements in 100-T fields. We discovered a field-induced magnetic-magnetic transition in $CeRh_2Si_2$ at 25 T and, from analysis of the resulting field-temperature phase diagram, resolved a long-standing controversy over the nature of magnetism in $CeCu_2Si_2$. From magnetization measurements to 60 T, we discovered evidence for the role of electronic correlations in producing a small energy gap in the "Kondo insulator" material, $Ce_3Bi_4Pt_3$. Lastly, we received new funding from NSF to develop a specific-heat measurement capability in pulsed magnetic fields.

In the course of this three-year project, we have achieved all of our major business, technical, and scientific objectives.

Publications

Boenig, H.J., et al., "Design Status of the US 100 Tesla Non-Destructive Magnet System" (to be published in *Proc. 7th International Conference on Megagauss Magnetic Fields*).

Harris, J.G.E., et al., "Fabrication and Characterization of 100-nm Thick GaAs Cantilevers" (to be published in *Rev. Sci. Instrum.*).

Lawrence, J.M., "Kondo-Hole Behavior in $Ce_{.97}La_{.03}Pd_3$," *Phys. Rev.* **53**, 12559 (1996).

Modler, R., et al., "H-T Phase Diagram of $CeRh_2Si_2$: Evidence for Local Magnetic Order in the Heavy Fermion Superconductor $CeCu_2Si_2$" (submitted to *Phys. Rev. B*).

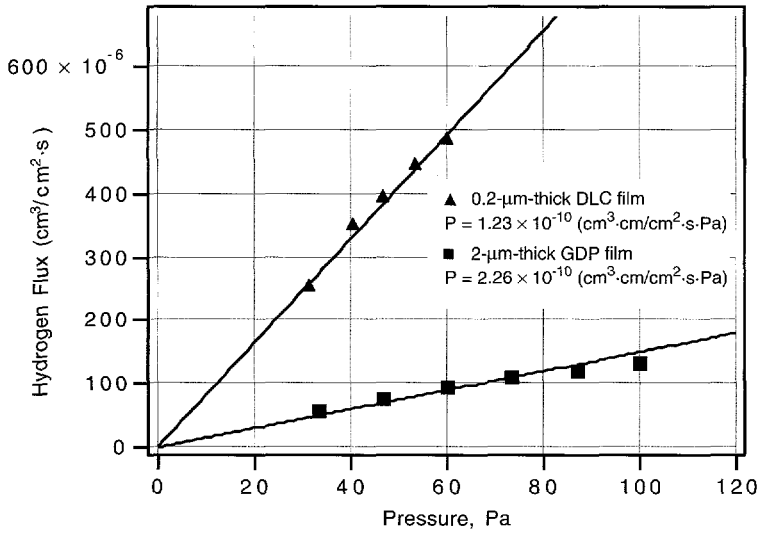
Modler, R., et al., "Intrinsic High Field Response of the Kondo Insulator $Ce_3Bi_4Pt_3$" (submitted to *Phys. Rev. B*).

Sarrao, J.L., et al., "Ferromagnetism and Crystal Fields in $YbInNi_4$" (submitted to *Phys. Rev. B*).

Diamond and Diamond-Like Materials as Hydrogen Isotope Barriers

Larry R. Foreman

The objective of this project is the examination of diamond and diamond-like carbon (DLC) as a hydrogen barrier to be used on implosion-target shells and tritium-processing equipment. To address the application of these materials to implosion targets, we used a radio-frequency, plasma-assisted chemical-vapor-deposition process to deposit DLC films and compared them with glow discharge polymer (GDP) films used by General Atomic for implosion shells. For measuring hydrogen through these membranes, we assembled an apparatus based on a calibrated mass spectrometer technique and calculated permeability from the slope of the hydrogen flux vs the pressure curve after correction for film thickness. For the results, see the accompanying figure.



Hydrogen flux measurements through DLC and GDP films. The permeabilities are 1.23×10^{-10} and 2.26×10^{-10} $cm^3 \cdot cm/cm^2 \cdot s \cdot Pa$, respectively.

Permeability measurements of hydrogen through polycrystalline diamond require free-standing thin films. We have deposited films on silicon wafers and attempted to remove the wafers by chemical etch. The films are brittle and stressed, and complete etching of the silicon substrate results in destruction of the diamond film. Our present approach is to etch a small hole in the wafer, leaving much of the substrate intact for support. We have designed and partly constructed a system for measuring isotope permeability under high pressures and temperatures. We will examine diamond coatings as well as a number of nitride and intermetallic systems. Reports indicate that the use of nitride and aluminide as barrier coatings can result in 1,000- to 100,000-fold decreases in the permeability of hydrogen.

Supporting Technologies for a Long-Pulse Spallation Source

Daniel Weinacht

We are studying the accelerator modifications, target-moderator-reflector concepts, neutron-scattering instrumentation, and facility modifications necessary for a long-pulse spallation source (LPSS) at Los Alamos. One objective is to evaluate from an engineering perspective the concepts for the linear accelerator (linac) modifications, target system, target station, and neutron-scattering instrumentation. We want to establish a basis for assessing the technical, cost, and schedule risks for the LPSS facility.

This year, in support of our project, we made the following progress: (1) We executed a special high-peak-current development run on the linac at the Los Alamos Neutron Science Center to further assess accelerator beam losses. (2) We performed shielding calculations to establish bulk shield size, chopper radiation environment, and beam-line shutter requirements; developed the bulk shield design; refined the cost estimate; and developed and evaluated base-line concepts for the target-moderator-reflector system, vacuum vessel and flight paths, and choppers, guides, and shutters. (3) In addition, we performed simulations of additional instruments for the initial LPSS instrument suite.

Thin-Film Sensors for Nerve and Mustard Agents

Basil Swanson and Karen Grace

The objectives of this project are to develop species-selective thin films that can detect chemical warfare (CW) agents, to test these films in surface-acoustic-wave (SAW) devices, and to develop advanced optical heterodyne transducer techniques. We obtain selectivity of the thin films to nerve and mustard agents by using cavity-shaped molecules (e.g., cyclodextrins) that form guest/host inclusion complexes with these agents. We chemically modify the cyclodextrins to optimize their binding affinities with CW agents and then form the modified cyclodextrins into self-assembled monolayers that we covalently bond to SAW or optical transducers. In addition to studying monolayer films, we are also studying ways to increase sensitivity by using multilayer films and polymeric films.

We have synthesized a number of cyclodextrin derivatives (CDs) and formed them into self-assembled monolayers or have attached them to polymer backbones before forming them into self-assembled monolayers. We have then tested these CDs in SAW devices against chemicals that simulate CW agents and chemicals that could potentially interfere with the detection process. We covalently attach the CD hosts to the backbones of polymers and then covalently bond the polymers in a functionalized self-assembled monolayer to the transducer surface. For chemicals that simulate nerve and mustard agents, the measured sensitivities of the polymer thin films are in the mid-to-low parts per billion range (10–100 ppb). The response of the films is highly specific to the molecules used to simulate CW agents and much less so to molecules with the potential to interfere with the detection process. Moreover, the response time of these films is quite fast (seconds to minutes), which makes them useful as continuous, near-real-time sensors.

We have also built a laboratory test bed to develop and test chemical sensors that are based on integrated optical techniques or chemically selective thin films. The test bed includes a gas-handling system, a user interface and data acquisition system, an imaging system to optimize the optical mode profiles, and environmental control and mechanical stabilization systems. We have used this test bed to successfully adapt optical transducer methods to thin-film sensor fabrication. Using waveguide Zeeman interferometry we have demonstrated reversible, real-time sensing of volatile organic compounds in the low parts per million range. Although these results have not yet been optimized, the sensitivities we have obtained thus far are comparable to those we have previously obtained with the same thin films using 250-MHz SAW transducers for detection. We believe that we can easily increase this sensitivity by 2–3 orders of magnitude.

We have submitted a patent application related to this work which is currently in the approval process.

Publications

Beregovski, Y., et al., "In Situ Chemical Detection Based on Photonic Devices" (SPIE Aerosense Symposium on Electro-Optic Technology for Chemical Detection and Identification, Orlando, FL, April 1997).

Grace, KM, et al., "Real Time Chemical Detection Using Species Selective Thin Films and Waveguide Zeeman Interferometry" (to be presented at the SPIE Aerosense Symposium on Electro-Optical Technology for Chemical Detection and Identification, Orlando, FL, April 13–17, 1998).

Grace, K.M., et al., "Thin-Film Chemical Sensors Using Zeeman Waveguide Interferometry" (12th International Conference on Optical Fiber Sensors, Williamsburg, VA, October 1997).

Grace, K.M., et al., "Waveguide Zeeman Interferometry for Thin-Film Chemical Sensors," *Electron. Lett.* **33**, 1651 (1997).

Yang, X., et al., "Growth of Ultrathin Covalently Attached Polymer Films: Uniform Thin Films for Chemical Microsensors" (to be published in *Langmuir*).

Yang, X., et al., "Molecular Host Sol-Gel Thin Films for Surface Acoustic Wave Chemical Sensors," *Sens. Actuators, B* **45**, 79 (1997).

Yang, X., et al., "Polyelectrolyte and Molecular Host Ion Self-Assembly to Multilayer Thin Films: An Approach to Thin Film Chemical Sensors," *Sens. Actuators, B* **45**, 87 (1997).

High-Magnetic-Field Research Collaborations

Jeffrey Goettee

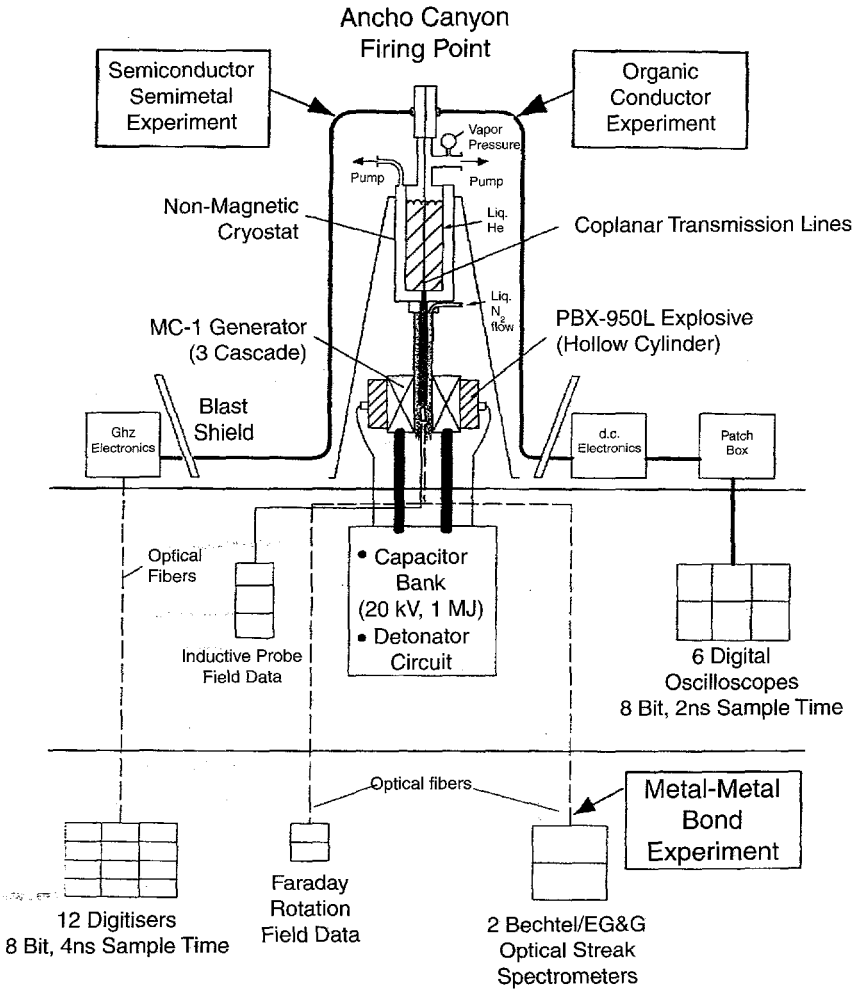
We initially set out to explore the applicability of current kilotesla technology to basic science. A series of experiments over the past two years built on initial work done in 1993 by a collaboration between Los Alamos and the All-Russian Institute of Experimental Physics (Arzamas-16) that brought the reproducible production of fields in excess of 1000 T to the United States. Our work has focused on developing other tools needed to use this high-magnetic-field technology for modern, fundamental research in chemistry and physics.

This year we completed four shots in the Russian-built MC1 generator and four shots in the Los Alamos–built, single-stage strip generator, and we performed countless experiments using more traditional capacitor-driven magnets in preparation for the final series of kilotesla shots. Each explosive shot contained 9 to 10 individual experiments (see the first figure), and each investigator repeated his experiment on a second shot. Collaborators from Russia, Australia, Japan, Belgium, Germany, England, Brazil, Yugoslavia, and the United States contributed to the technology advancement and were able to collect data on their experiments (see the second figure) in these ultrahigh-magnetic fields. Analysis of all these experiments is ongoing.

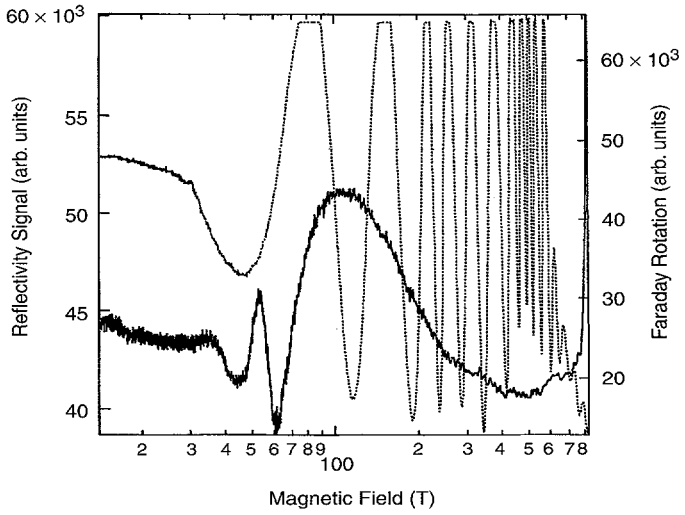
Publications

Mielke, C.H., et al., "Closing the Gap in Samarium Hexaboride (6) with Megagauss Fields," in *National High Magnetic Field Laboratory Annual Report* (Florida State University, Tallahassee, in press).

Mielke, C.H., et al., "Magnetotransport to 145 Tesla in Tetra Methyl Tetra Selena Fulvalene (2) Chloride Samarium Hexaboride (4)," in *National High Magnetic Field Laboratory Annual Report* (Florida State University, Tallahassee, in press).



A schematic of shots MC1-D and MC1-F, courtesy of the Australian team. Experiments in the upper half of the MC1 generator share space within the specially developed plastic cryostat that supplied a base temperature of 1.5 K. These experiments also employed the radio-frequency transmission lines developed by the Australian team to measure electron transport within several different condensed-matter systems. The lower half of the generator contained the quadruple metal/metal bond experiments from Louisiana State University, as well as the field diagnostics.



Data from shot MC1-D, courtesy of the Florida State University team, superimposed with a Faraday rotation signal. The solid trace shows the optical reflectivity of the two-dimensional electron gas confined by a quantum well. This signal can be related to the conductivity of the electron gas. The dotted trace is the Faraday rotation signal and is related directly to the intensity of the magnetic field.

Characterizing the Mechanical Effects of Aging Damage

Thomas D. Sewell

This project involves theoretical and experimental work aimed at identifying, characterizing, and understanding the mechanical effects of aging on organic composite materials. The focus of the work is PBX-9501, a plastic-bonded explosive formulation composed of 95 wt% high-melting explosive (HMX) and 5 wt% binder material. The binder is composed of 50 wt% Estane (a polyurethane-based block copolymer) and 50 wt% bis(2,2-dinitropropyl)-acetal/formal.

The goal is to incorporate molecular-level experimental data and computed atomistic-level information into a micromechanical model that can be used to understand the mechanical behavior of the composite and, in particular, to understand the effects of aging processes on the mechanical properties. We will use these results to construct a micro-mechanics-based constitutive model for particle-filled composites.

This year we assembled a Raman microscopy and imaging apparatus that uses 752-nm laser excitation. Near-infrared light is needed for our experiments because visible emission from organic complexes obscures the Raman spectra. We also began testing an IR microscopy and imaging setup. Initial comparisons were done for spectra of fresh and aged Estane, as well as PBX-9501. Based on these preliminary results, there are differences in the carbonyl region of the spectra that possibly reflect differences in hydrogen bonding caused by aging phenomena.

We used molecular dynamics and molecular mechanics simulations to compute the elastic constant matrix for the pure Estane polymer. We also used rigid-molecule Monte Carlo simulations to compute the bulk modulus of HMX. We extended our micromechanical model of composites to study various volume fractions of HMX with binders. The results showed that as the binder fraction increases, there is a decrease in the maximum stress that can be supported but an increase in the percent strain at final fracture. Finally, we obtained a more realistic microstructural model using a phase field model.

Publications

Fan, D., et al., "Computer Simulation of Grain Growth Kinetics with Solute Drag" (submitted to *J. Mater. Res.*).

Fan, D., et al., "Numerical Simulation of Zener Pinning with Growing Second Phase Particles" (submitted to *J. Am. Ceram. Soc.*).

Fan, D., et al., "Phase Field Formulation for Modeling the Oswald Ripening in Two-Phase Systems" (to be published in *Comput. Mater. Sci.*).

Sewell, T.D, "Monte Carlo Calculations of the Hydrostatic Compression of RDX and β-HMX" (to be published in *J. Appl. Phys.*).

Sewell, T.D., "Monte Carlo Calculations of the Physical Properties of RDX, β-HMX, and TATB," in *Shock Compression of Condensed Matter—1997*, S.C. Schmidt, D.P. Dandekar, and J.W. Forbes, Eds. (AIP Press, New York, in press).

Sewell, T.D., and E.M. Kober, "Understanding the Physical and Mechanical Properties of Plastic-Bonded High Explosive Formulations," in *Solutions* (Molecular Simulations Press, San Diego, in press).

Ceramic/Polymer, Functionally Graded Material (FGM), Lightweight Armor System

John Petrovic

The ability to produce FGMs (e.g., functionally graded ceramic/polymer and ceramic/metal materials) could form the basis for a quantum-leap improvement in lightweight body armor systems. The FGM is an enabling technology for such body armor improvements because it addresses the weight and threat issues. Our objective in the project was to demonstrate the ability to produce functionally graded ceramic/polymer and ceramic/metal lightweight armor materials. This objective involved two aspects. The first and key aspect was to develop graded-porosity, boron carbide ceramic microstructures. The second aspect was to develop techniques for liquid infiltration of lightweight metals and polymers into the graded-porosity ceramic.

During the past year, we were successful in synthesizing boron carbide ceramic microstructures with graded porosity. We achieved graded porosities in the range of 6% to 53% by using a combination of a furfuryl alcohol sintering aid for boron carbide, differently sized boron carbide powders, and ultrasonic vibration to obtain green bodies which were subsequently hot-pressed at 1900°C. These graded-porosity, boron carbide, hot-pressed pieces were then successfully liquid infiltrated in vacuum with molten aluminum at 1300°C and with liquid polymers at room temperature. Thus, we were able to demonstrate that producing boron carbide/aluminum and boron carbide/polymer FGMs is feasible.

Hydrogen Gas Getters

Eugene Mroz

Hydrogen getters are a class of materials that permanently remove hydrogen from gases. These materials are being considered for inclusion in transuranic waste containers destined for the Waste Isolation Pilot Plant (WIPP) to mitigate the risk of hazardous hydrogen buildup within the containers. The objective of this project was to study the chemical mechanism of the catalytic reaction of hydrogen with one commonly used getter, DEB—a di-alkyne compound wherein two alkyne linkages (carbon-to-carbon triple bonds) connect three phenyl rings (1,4-bis(phenylethynyl)benzene).

Two specific questions were addressed in work this past year: (1) Do the hydrogen atoms that are catalytically added across the unsaturated triple bonds stay localized on the carbon atoms or do they become delocalized across the phenyl ring structure? Nuclear magnetic resonance studies of DEB exposed to deuterium showed that the deuterium atoms remained localized on the sites of the alkyne linkages. (2) What is the nature of the intermediate species formed during the reaction? Gas chromography and mass spectroscopy analyses of DEB exposed to substoichiometric amounts of hydrogen in a slow, controlled fashion indicated that while small amounts of the alkene analogs are present, the majority of the reaction product is the fully saturated alkane analog. However, when DEB is instantaneously exposed to an excess of hydrogen, a diverse mix of unreacted alkyne is produced: cis- and trans- forms of the alkene are produced as well as completely saturated alkanes.

These results are helping us develop a mechanistic model of the reaction.

Decontamination of Chemical and Biological Warfare Agents in the Urban Arena

William L. Earl

The goal of this project is to develop and evaluate decontamination methods that can be used to combat chemical and biological terrorist attacks on civilian urban targets, an area of growing concern to DOE and the US government in general. Our specific objectives are to develop and use a low-temperature atmospheric plasma jet for decontaminating chemical and biological warfare agents and to develop high-surface-area catalysts for adsorption and catalytic destruction of chemical agents. The plasma and catalytic decontamination methods are being tested on simulants, not on actual chemical and biological warfare agents.

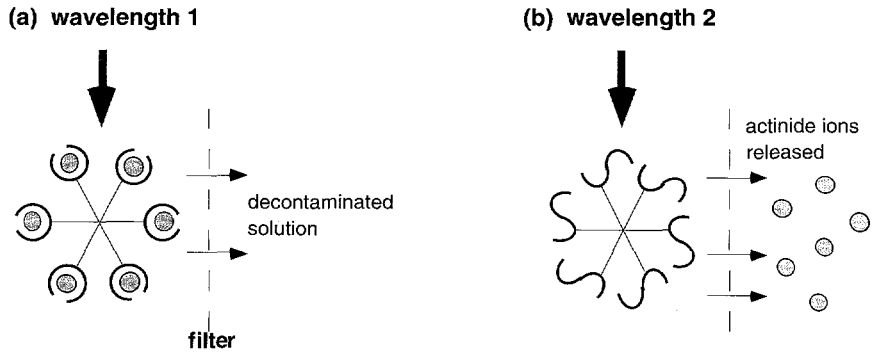
This past year, we advanced the technology of the plasma jet to make it more applicable to a decontamination scenario. We made a preliminary evaluation of an excited-state oxygen plasma for destroying malathion, a simulant for chemical nerve agents. The test was positive in that we were able to destroy some of the malathion, but it also pointed out the difficulties in sampling and analysis that prevent making a good quantitative measure of surface decontamination. We also tested several zeolite-based catalysts for their destruction of malathion; all were effective in adsorbing the chemical. We found that silver ions in the zeolites are effective at catalyzing the decomposition of the malathion; again, however, difficulties in sampling and analyzing results led to our being unable to quantify the ions' actual decontamination effectiveness.

Photo-Controlled Devices for Nuclear Materials Separation

Rebecca Chamberlin

This project was aimed at assessing the feasibility of using photo-controlled molecular devices to separate and recover actinide metal ions from aqueous solutions. A light-driven separation technology would be useful when the generation of secondary hazardous and/or mixed wastes is costly and problematic, such as in nuclear materials processing and environmental restoration activities.

We identified several challenges to implementing a photo-controlled membrane-transport separation process, including the thermal reversibility of the metal binding and release steps, and back diffusion of the metal ions through the membrane. As a result, we recommend an alternative engineered form that uses photo-reversible sequestration on water-soluble chelating polymers for the separation (see figure). In this process, ultrafiltration of the polymer-bound metal ion separates and concentrates the metal, while photo-induced release from the chelator allows recovery of the metal ion in a small waste volume and reuse of the chelator. Synthesis of potential actinide chelators, whose metal binding can be switched "on" and "off" using light of varying wavelengths, was shown to be feasible in two or three chemical steps from commercially available starting materials. We prepared photo-controlled chelators based on azobenzene and spiropyran molecular platforms. The spiropyran chelator was covalently attached to a water-soluble commercial polymer, PAMAM Generation 4.0, and used in the aqueous separation process shown in the figure.



Light-driven separation using water-soluble polymer ultrafiltration: (a) in its "uptake" form, the polymer retains the metal ions (grey circles), while the bulk of the water and other contaminants pass through the sievelike filter; (b) in its "release" form, the polymer allows the metal ions to pass through the filter. The specific wavelengths involved vary with the chelating polymer.

Advanced Nuclear Fuels Processing

Larry Avens

The objective of this project is to review the status of advanced nuclear processing technologies and approaches developed after 1977 that can minimize or eliminate key negative characteristics associated with aqueous (PUREX) processes—separated plutonium streams, complexity, waste generation, and economics. The impetus behind this review is to determine what nuclear fuel processing activities have occurred since President Carter's decision to split defense and commercial fuel cycles. This review is also an attempt to begin updating the Brookhaven National Laboratory review of alternative processing options, last published in 1977.

Since 1977, the only major programs that have attempted to develop alternate nuclear fuels processing methods are the Argonne National Laboratory Integral Fast Reactor Program, with its associated spent-fuel reprocessing flowsheet, and the Oak Ridge National Laboratory molten salt fluoride volatility process flowsheet. (The latter was actually begun earlier than 1977 and was nearly complete by this time.) The information derived from reviewing these programs will provide data to policymakers who may be evaluating the current US position on spent-fuel processing.

Separation Science and Technology

Barbara Smith

The focus of this project is to demonstrate and advance membrane-based separation and destruction technologies developed at Los Alamos. We are exploring the development of membrane systems for gas separations, for selective metal-ion recovery, and for separation and destruction of hazardous organics.

This past year we evaluated our existing water-soluble, chelating polymers and polymer formulations for recovery of toxic oxyanionic metals (such as chromate and arsenate) from selected waste streams, and we developed second-generation water-soluble polymeric systems for highly selective oxyanion removal and recovery. We also developed fission-product separations using cobalt dicarbollide for solvent extraction, ion exchange, and membrane filtration. We optimized the simultaneous removal of radioactive strontium and cesium from aqueous solutions using our new, nonhazardous separations agents, and we developed recyclable, redox-active extractants that permit us to recover the radioactive ions into a minimal waste volume.

In addition, we developed new, more-efficient membrane-based electrochemical reactors for destroying organics in the treatment of process waste. In another aspect of our research, we produced hollow fibers and fabricated hollow-fiber membrane modules for applications in gas separations and in the liquid-liquid extraction and recovery of actinides and nuclear materials from process streams. We also developed and fabricated cyclodextrin-based microporous materials that selectively absorb organic compounds in an aqueous environment; the resultant products gave pure water with organics at less than 0.05 parts per billion. Because we can easily regenerate the filter by air drying, we can use it repeatedly.

We addressed the need for advanced oxidation technologies based on molecular-level materials designs that selectively remove or destroy target species. We prepared and characterized surface-modified TiO_2 thin films using different linking approaches to attach ruthenium photosensitizers, and we started measuring the photodegradation products generated in the reaction of surface-modified TiO_2 films with chlorophenol.

Publications

Chamberlin, R.M., et al., "Butyllithium Deprotonation vs Alkali Metal Reduction of Cobalt Dicarbollide: A New Synthetic Route to C-Substituted Derivatives," *Inorg. Chem.* **36**, 809 (1997).

Espe, M., et al., "Packing in Amorphous T Regions of Hydrofluoric Acid Doped Polyaniline Powder by ^{15}N-^{19}F REDOR NMR" (to be published in *J. Am. Chem. Soc.*).

Fino, S.A., et al., "Condensation Polymerization of Cobalt Dicarbollide Dicarboxylic Acid," *Inorg. Chem.* **36**, 4604 (1997).

Ma, M., and D. Li, "Exceptionally Strong Non-Covalent Interactions and Transport of Organic Molecules in Nanoporous Polymers" (submitted to *J. Am. Chem. Soc.*).

Mattes, B.R., et al., "Electrically Conductive Polyaniline Fibers Prepared by Dry-Wet Spinning Techniques," in *Plastics—Saving Planet Earth (ANTEC '97)*, D. Bayley, Ed. (Society of Plastics Engineers, Brookfield, CT, 1997), p. 1463.

Mattes, B.R., et al., "Formation of Conductive Polyaniline Fibers Derived from Highly Concentrated Emeraldine Base Solutions," *Synth. Met.* **84**, 45 (1997).

Mattes, B.R., et al., "Temperature and Pressure Dependent Gas Transport through Polyaniline Membranes" (to be published in *Int. J. Thermophys.*).

Rais, J., et al., "Use of PVC Plasticized Membranes for Uptake of Radioactive Cesium and Strontium," *Sep. Sci. Technol.* **32**, 951 (1997).

Shennikova, I.N., et al., "Rigidity and Conformation of Poly-(1-trimethylsilyl-propyne) in Solutions" (to be published in *Macromolecules*).

Smith, B.F., et al., "Water-Soluble Metal-Binding Polymers with Ultrafiltration: A Technology for Removal, Concentration, and Recovery of Metal Ions from Aqueous Streams," in *Advances in Separations and Preconcentration* (American Chemical Society, Washington, DC, in press).

Winokur, M.J., and B.R. Mattes, "Determination of the Local Molecular Structure in Amorphous Polyaniline," *Phys. Rev. B* **54**, 18 (1996).

Winokur, M.J., and B.R. Mattes, "Differential Anomalous Scattering Studies of Amorphous HBr-Doped Polyaniline," *Synth. Met.* **84**, 725 (1997).

Winokur, M.J., and B.R. Mattes, "Polyaniline As Viewed from a Structural Perspective," *Plastics—Saving Planet Earth (ANTEC '97)*, D. Bayley, Ed. (Society of Plastics Engineers, Brookfield, CT, 1997), p. 1325.

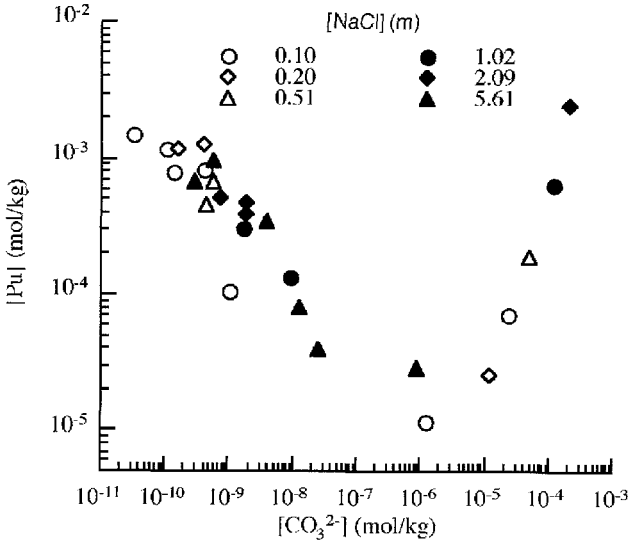
Integration of Advanced Nuclear-Materials Separations Processes

Gordon Jarvinen

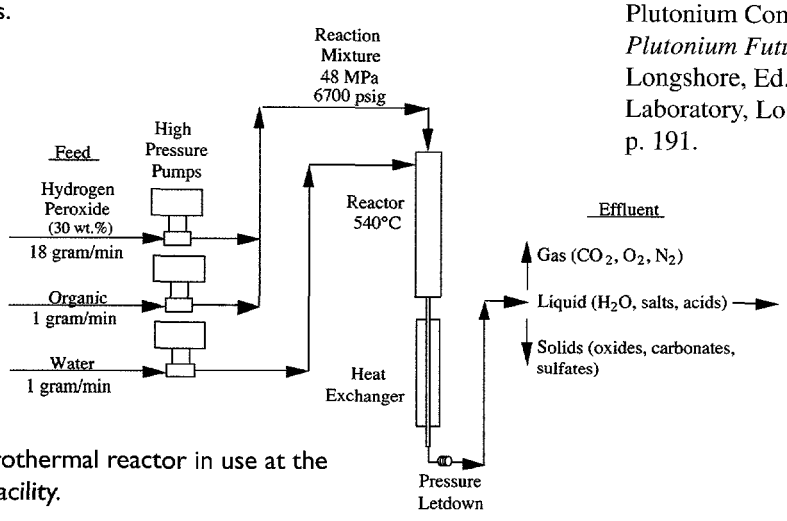
We have examined the fundamental chemistry of plutonium that affects the integration of hydrothermal technology into nuclear-materials processing operations. Chemical reactions in high-temperature water allow new avenues for waste treatment and radionuclide separation. Successful implementation of hydrothermal technology offers the potential to effectively treat many types of radioactive waste, reduce the storage hazards and disposal costs, and minimize the generation of secondary waste streams. Our focus has been on the plutonium(VI) carbonate chemistry that is expected to control the fate of plutonium following hydrothermal oxidation of plutonium-containing organic wastes.

We have collected spectrophotometric titration data on the plutonyl carbonate system and have used this data to calculate the formation constants for the tris- and biscarbonato complexes. These formation constants ($10^{17.7}$ and $10^{13.6}$, respectively) indicate that under conditions relevant to the hydrothermal process effluent, aqueous PuO_2CO_3 is the predominant species over a large pH range. We determined the solubility product, log K_{sp}, of PuO_2CO_3 to be –12.9 in 0.1-M NaCl (see data plot).

Installation and testing of flow and batch hydrothermal reactors in the Plutonium Facility required a formal DOE Readiness Assessment, which was concluded late in FY 1997. A flow reactor is depicted in the accompanying schematic. We processed two plutonium-contaminated organic solutions: pump oil and liquid scintillation cocktail. In both tests the effluent was a clear, colorless solution of pH 4 that was readily discardable. A new project starting in FY 1998 will build on the results obtained in this effort.



Solubility of plutonium(VI) in NaCl solution as a function of free-carbonate concentration. These measurements are used to determine the solubility product. The increasing solubility of plutonium(VI) at higher carbonate concentrations shows the influence of the formation of the bis- and triscarbonato complexes.



Schematic of a flow hydrothermal reactor in use at the Los Alamos Plutonium Facility.

Publications

Reilly, S.D., et al., "Plutonium Solubility and Speciation under Hydrothermal Waste Treatment Conditions," in *Plutonium Futures—The Science*, A. Longshore, Ed. (Los Alamos National Laboratory, Los Alamos, NM, 1997), p. 245.

Worl, L.A., et al., "Hydrothermal Oxidation for the Treatment of Plutonium Combustible Wastes," in *Plutonium Futures—The Science*, A. Longshore, Ed. (Los Alamos National Laboratory, Los Alamos, NM, 1997), p. 191.

Development of TRU-Waste Mobile Analysis Methods for RCRA-Regulated Metals

Cynthia Mahan

The objective of this project is to evaluate the effectiveness of several analytical techniques for potential mobile-laboratory-based transuranic (TRU) waste characterization. The techniques we are evaluating include laser-induced breakdown spectroscopy (LIBS), dc-arc atomic-emission spectroscopy (DC-ARC-AES) using a charge-injection device detector, glow-discharge mass spectrometry (GDMS), laser-ablation inductively coupled mass spectrometry (LAICPMS), and energy-dispersive x-ray fluorescence (EDXRF).

During the past year, we developed analytical methods for each of the above-listed techniques. To evaluate the techniques, we used surrogate cement samples spiked with metals regulated by the Resource Conservation and Recovery Act (RCRA). The surrogate samples were prepared by Argonne West for use in the DOE's Performance Demonstration Program (PDP). Laboratories must qualify their analytical methods by passing a PDP cycle; thus the same types of samples used in the performance demonstrations were used to evaluate the analytical techniques. The second phase of the study involved a round-robin analysis of the surrogate samples. We sent the samples to each technique developer for analysis. The results of the analysis will serve to measure the performance of each technique.

Thus far, we have finished the evaluation of EDXRF and DC-ARC-AES. The LAICPMS and GDMS development was temporarily halted because of the stand-down at the Chemistry and Materials Research (CMR) facility. This work will continue as soon as operations are restored. We anticipate that this work will be completed in 3 to 4 months after startup of the CMR. We expect the LIBS analytical data to be completed early next year.

After all the data has been collected, we will compare the results for each technique with traditional analysis methods. In addition, we will evaluate the figures of merit for each technique and will compare them with the regulatory data-quality objectives. After all the data has been pooled and studied, we will rank the techniques.

Publications

Mahan, C.A., "Evaluation of a Direct Current Arc Charge Injection Device Spectrograph for Direct Analysis of Soils," *J. Anal. At. Spectrom.* **12**, 247 (1997).

Heterogeneous Oxidation and Polymerization Processes in Supercritical Fluids: CO_2 as a Reagent, a Protecting Group, and a Solvent for Chemical Processing

William Tumas

Our technical objective is to study heterogeneous catalysis and chemistry using supercritical fluids in order to minimize energy use and waste generation and, in particular, to provide chlorine-free routes to chemicals. Our previous research in the application of supercritical carbon dioxide as a protecting group, a reactant, and a reaction medium will be applied to a variety of chemical transformations such as polymerization, production of agricultural chemical intermediates, and synthesis of bulk commodity chemicals.

This year we worked closely with individual chemical and energy companies to identify their current and future scientific and technological needs. We worked closely with potential customers of the DOE and the Environmental Protection Agency (EPA) to assess their needs in programs where catalysis either does or could play a role. On the national level, we became involved in the US chemical industry's Vision 2020 process to map the road forward for the industry. We ran a national workshop on catalysis as it relates to identifying and prioritizing the critical needs of catalysis in the chemical industry. We partnered with Sandia National Laboratories (Albuquerque) to organize and run this workshop. We called together fifty of the nation's top researchers in catalysis from academia, industry, and government.

The major finding of the road-mapping exercise was the prioritization of research needs in catalysis research. Eight major research areas were identified. Ranked by workshop participants, the top four critical needs are enabling catalyst design through mechanistic, experimental, and computational chemistry; in situ techniques for catalyst characterization; high-throughput catalyst synthesis and testing; and synthesis of catalysts with specific site architecture. This road map is a living document and will be updated on a regular basis through the Vision 2020 process.

These program development activities have resulted in four funds-in agreements with companies, the development of two substantial proposals to DOE with industrial collaborators, and the development of an EPA Green Chemistry Center at Los Alamos.

Mathematics and Computational Science

Simulation of Thin-Film Formation

Robert B. Walker

The purpose of this project is to develop a fully three-dimensional (3-D) integrated modeling, simulation, and design capability for the deposition of thin films of various materials. If successful, the results of this project will be of high value for both defense and industrial applications (see accompanying figure).

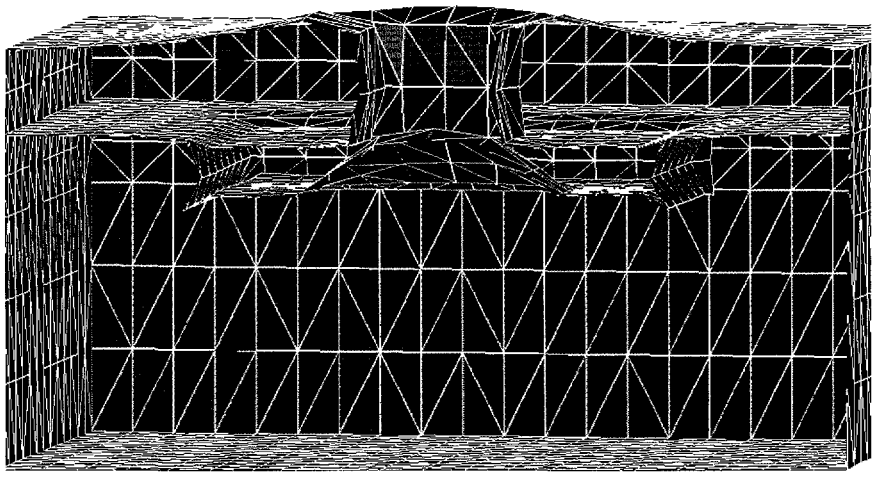
Version 1.0 of our feature-scale-profile simulation code, TopoSim-3D, is nearly ready to release to users. We have incorporated ChemKin and Surface ChemKin kinetics software (commercial) into TopoSim-3D, and preliminary tests indicate that the ChemKin/Surface ChemKin interface with TopoSim is working properly. We have also completed some preliminary code design experiments that will guide the future development of a parallel, modularized version of TopoSim-3D.

Based on electronic structure calculations for silicon- and chlorine-containing molecules, we have generated a new bond-counting-correction interatomic potential for silicon-chlorine. Molecular dynamics simulations of reactive ion etching using the new potential give a higher silicon etch yield relative to the previous potential of Garrison et al. (Pennsylvania State University) and, thus, are in better agreement with molecular beam experiments of Chang and Sawin (Massachusetts Institute of Technology). We're analyzing our molecular dynamics results in light of three recent molecular beam experiments: Balooch et al. (Lawrence Livermore National Laboratory), Levinson et al. (Stanford University), and Chang and Sawin.

Publications

Boehm, R.C., et al., "A Theoretical Study of Bond Energies in Model Si-H-Cl Molecules Using Density-Functional Approaches for Representing Si Surface Chemistry," *J. Compu. Chem.* **18**, 2075 (1997).

Hanson, D.E., et al., "Molecular Dynamics Simulation of Reactive Ion Etching of Si by Energetic Cl Ions," *J. Appl. Phys.* **82**, 3552 (1997).



Example of a calculation of the deposition of material onto a wafer containing an overhang structure that resembles a skylight in the roof of a house. Test structures of this type are manufactured by the semiconductor industry specifically to assist modeling efforts in distinguishing among mechanisms that transport material to surfaces.

Interconnect Performance Predictability

David Kilcrease

The semiconductor industry is facing a critical issue in the design of future generations of electronic components because traditional simulation techniques will not accurately predict either time delay or signal integrity. The issue is the lack of scalable, physically based design and simulation tools that can extract both capacitance (C) and inductance (L) from general three-dimensional (3-D) shapes of metal and predict the "cross talk" between real 3-D interconnect elements that leads to degeneration of signal integrity.

We are performing fundamental research needed to generate advanced algorithms for solving a wide class of applications in electrical performance predictability. The general technical issues of interest are the accurate prediction of the signal delay for any electrical path through the device and the time (frequency) character of the cross talk (signal integrity degeneration) for the entire chip.

Two aspects of electromagnetic field interactions with materials that are being evaluated in this project are (a) time-dependent solutions of Maxwell's equations using finite-difference algorithms and (b) L and C static parameter extraction from complex 3-D dielectric/metal configurations.

Technical accomplishments this year include evaluating the existing finite-difference codes for accuracy against new experimental test cases (from Motorola) to determine their range of validity for variations of the frequency, geometry, and dielectric constants, and developing the 3-D Poisson solver for extracting static C and L parameters for complex geometries and multiple dielectrics using the Los Alamos 3-D unstructured grid code.

Moment Equations for Two-Phase Flow in Random Porous Media

Dongxiao Zhang

Our goal is to develop partial differential equations that could be used as an alternative to the commonly used Monte Carlo simulation approach for the moments of two-phase flows in heterogeneous porous media. A successful derivation of workable moment equations would offer several benefits over the Monte Carlo approach. First, there would be a small number of equations to be solved: one for the mean and one each for a small number of variances and covariances. Second, the coefficients of the equations, as averaged quantities, would be smooth; thus these equations could be solved on relatively course grids. Third, the moment equations would be available in analytic form, even though they would be solved numerically in applications. This holds the potential for increased physical understanding of the mechanisms of uncertainty.

This year we studied the problems of single-phase flow and unsaturated flow and a special case of two-phase flow. In addition, we initiated and obtained good results for the case of two-phase (oil-water) displacement. With the results generated this year, we will be able to move firmly toward developing and solving the moment equations for two-phase displacement in random porous media.

Publications

Harter, T., and D. Zhang, "Water Flow and Solute Spreading in Heterogeneous Soils with Spatially Variable Water Content" (submitted to *Water Resour. Res.*).

Tartakovsky, D.M., et al., "Some Aspects of Head-Variance Evaluation" (submitted to *Adv. Water Resour.*).

Xin, J., and D. Zhang, "Stochastic Analysis of Biodegradation Fronts in Heterogeneous Media" (submitted to *Adv. Water Resour.*).

Zhang, D., "Numerical Solution to Statistical Moment Equations of Groundwater Flow in Nonstationary, Bounded Heterogeneous Media" (to be published in *Water Resour. Res.*).

Zhang, D., and H. Techelepi, "Stochastic Analysis of Immiscible Two-Phase Flow in Heterogeneous Media" (submitted to *Soc. Pet. Eng. J.*).

Zhang, D., and C.L. Winter, "Moment Equation Approach to Single Phase Fluid Flow in Heterogeneous Reservoirs" (submitted to *Soc. Pet. Eng. J.*).

Zhang, D., and C.L. Winter, "Nonstationary Stochastic Analysis of Steady-State Flow Through Variably Saturated, Heterogeneous Media" (to be published in *Water Resour. Res.*).

Zhang, D., et al., "Stochastic Analysis of Steady-State Unsaturated Flow in Heterogeneous Media: Comparison of the Brooks-Corey and Gardner-Russo Models" (to be published in *Water Resour. Res.*).

Physics-Based Damage Predictions for Simulating Testing and Evaluation (T&E) Experiments

Edward A. Rodriguez

This project addresses physics-based computational models to simulate damage to battlefield systems from ballistic threats. Computational modeling and simulation can assist in planning for critical benchmark tests and in designing and assessing system vulnerabilities.

Fighter aircraft are becoming more dependent on composite components to reduce weight, increase stiffness and thermal stability, and resist adverse conditions such as high temperature and corrosion. However, damage to composites can be severe and detrimental as evidenced by statistics from Desert Storm indicating that 75% of aircraft losses were attributed to fuel system vulnerability caused by the hydrodynamic ram (HRAM) phenomenon and/or threat explosion.

Therefore, this project primarily focuses on predicting the damage caused by HRAM created from a ballistic-threat penetration of the fluid-filled volume of a fighter aircraft wing tank constructed from fiber-reinforced composite materials. The highly complex physics include fluid-structure interaction, high-strain-rate dynamics, material failure mechanisms, and supersonic airflow over the wing.

This year we performed parallel-path modeling based on a small-scale, water-filled wing-tank HRAM and an explosion test conducted at Wright-Patterson Laboratory. We used two computational codes (CFDLIB/PIC and PRONTO-2D/CFDLIB) for comparative purposes.

Numerical results for the HRAM phenomenon and the threat explosion, coupled with the composite material failure, proved highly encouraging in showing the complex physical behavior. The CFDLIB/PIC method proved successful in performing both the HRAM and the explosive-threat penetration calculations. The PRONTO-2D/CFDLIB solution for a stationary explosive threat gave qualitative and quantitative predictions of test results, but the code failed to perform the fluid-penetration calculation.

Data Mining

Kenneth Lee

The objective of our project is to develop and implement data-mining technology suited to the analysis of large collections of unstructured data. Such analysis is becoming critical to decision-making in business settings. The focus of our work has been to develop highly parallel strategies for efficient data-accessing operations and for scalable pattern-extraction algorithms. This has taken the form of a software tool, PArallel Data Mining Agents (PADMA), which incorporates parallel data accessing; parallel, scalable, hierarchical clustering algorithms; and a Web-based user interface for submitting interactive data visualization and queries in structured query language. PADMA is written in C++ and Java. It uses the parallel portable file system for data access and the message-passing interface for interprocess communication.

Although PADMA is not specialized for any particular kind of data-mining domain, the current implementation is best suited to unstructured text document classification. We have demonstrated the viability and scalability of PADMA by applying it to a text database of 25,000 documents running on an IBM SP2 at Argonne National Laboratory. In collaboration with the University of New Mexico School of Medicine, we have also demonstrated the utility of PADMA for discovering patterns in data by applying it to laboratory test data for hepatitis C patients and autopsy reports.

Several corporations have expressed interest in licensing PADMA technology; the Laboratory's Civilian and Industrial Technology Program Office is currently handling the copyright process and technology transfer.

Publications

Kargupta, H., and B. Stafford, "From DNA to Protein: Transformations and Their Possible Role in Linkage Learning," in *Proc. Int. Conf. Genetic Algorithms* (Morgan Kaufmann, San Francisco, CA, 1997), p. 409.

Kargupta, H., et al., "PArallel Data Mining Agents for Scalable Text Classification," in *Proc. Symp. High Performance Computing* (Society for Computer Simulation International, San Diego, CA, 1997), p. 290.

Kargupta, H., et al., "Scalable, Distributed Data Mining Using an Agent Based Architecture," in *Proc. Am. Assoc. Arificial Intelligence* (AAAI Press, Menlo Park, CA, 1997), p. 211.

A Hierarchical Simulation R&D Test Bed for Test and Evaluation

Chris Barrett

The objective of this project is to develop an integrated collection of simulations and methodologies for the analysis of war-fighting systems, focusing first on the application to test and evaluation problems. More specifically, the objective is to provide an expanded analysis capability beyond what test and evaluation analysts currently possess by combining an existing state-of-the-art, composable architecture for war-fighting simulation; available statistical methodologies; existing physics-based codes; and current hardware. Our objectives for the past year were to develop mission and engagement simulations of a fighter aircraft in a survivability scenario and to identify the technical and analysis issues associated with integrating these simulations with physics-based modeling of the vulnerability of the aircraft.

This past year, we had three main accomplishments. First, we developed the necessary simulation actors (entities within a simulation representing a real-world counterpart) for a proof-of-concept mission-level simulation. Second, we developed the necessary simulation actors, and their interactions, for an engagement-level simulation; specifically, we simulated the interaction between a fighter aircraft and a surface-to-air missile. Finally, we began using the proof-of-concept engagement simulation as an example to develop techniques for addressing the variability and input uncertainty associated with the simulation. These techniques should lead to development of a methodology for approximating models of higher resolution for appropriate use in lower-resolution simulations. These same techniques should also lead to an integrated methodology for deciding the appropriate range of input variables given the context generated by the lower-resolution simulations.

National Transportation System Analysis Capability

Doug Anson

The National Transportation System (NTS)—planned and managed by the Department of Transportation (DOT)—includes the nation's highways, railways, airways, waterways, and pipelines and the carriers that use these diverse but interconnected systems. To better support the nation's economic, security, and safety goals, the Secretary of Transportation has proposed an NTS Initiative. Critical to meeting the objectives of this initiative is the development of a National Transportation Network Analysis Capability (NTNAC). An NTNAC will allow the DOT to view the NTS from the user's perspective and to make more-informed policy recommendations based on a holistic analysis of a single, integrated, intermodal system.

The objective of this project is to demonstrate to the DOT the feasibility and power of an NTNAC similar to the Transportation Analysis and Simulation System (TRANSIMS) that we are currently developing for metropolitan planning organizations across the nation.

During the past year we have examined the NTS Initiative, defined the NTS problem, and identified the essential attributes and technical characteristics of the problem. We then decomposed the large-scale transportation system into five modules that meet the DOT's needs and have the required resolution (geographic, network, carrier, and commodity), variable time scales, and the ease of modular development. The modules are supply, demand, planning, execution, and analysis. In addition, we have identified the data that is required to implement the model and the data that is available, and we have developed criteria for, and an outline of, a demonstration scenario for an initial prototype.

Atomic and Molecular Physics and Plasmas, Fluids, and Particle Beams

Application of New Techniques for Equation-of-State Data

Warren Hsing

One area of great uncertainty as well as high physics leverage is that of the material properties of plutonium and other materials (including zirconium, uranium-niobium, deuterium-tritium, tantalum, beryllium, and oralloy). These properties include plutonium Hugoniot and melt-curve data to 10 Mbars and the plutonium off-Hugoniot data.

Our goal is a proof-of-principle experiment on the Los Alamos Trident laser that will determine the technical and logistical issues involved with performing an accurate equation-of-state (EOS) and melt-curve experiment with plutonium, as well as performing initial measurements on the plutonium Hugoniot. This method can also be used to gather data at relatively low cost for other materials as well. Such a capability would complement existing gas-gun facilities and laser-driven miniflyers.

This year's activities included the following: (1) performing calculations on direct- and indirect-drive requirements to generate a planar and constant shock in a reference material and surrogate, (2) measuring shock planarity and strength generated by an indirect-driven hohlraum on Trident, and (3) performing preliminary experiments on Trident with surrogates to further develop the melt technique measurements using dynamic x-ray diffraction.

Plasma Source Ion Implantation for Advanced Manufacturing

Jay Scheuer

Plasma source ion implantation (PSII) and plasma immersion ion processing (PIIP) can be used to provide economical surface modification to satisfy industrial and DOE stockpile support needs. PSII is a non-line-of-sight ion implantation technique that reduces the complexity and cost of traditional ion implantation. PIIP is a logical extension of PSII that combines ion bombardment with plasma deposition, allowing deposition of conformal, adherent coatings with thicknesses of tens of microns. Our project is dedicated to developing and demonstrating PSII/PIIP processes for US industry and DOE stockpile support applications.

This year we performed research and development in five areas: (1) We conducted PSII and PIIP recipe development and materials characterization (including chemical, microstructural, and surface mechanical properties) for industrial applications. (2) We continued demonstration processing on full-scale industrial components provided by our industrial partners. (3) We continued equipment development of PSII/PIIP pulsed-power technology to provide 50-kV, 10-μs pulsed-bias capability for the PIIP system and further development of a high-power, solid-state pulse modulator. (4) We applied advanced inductive plasma sources to the PIIP system to provide the ion densities and species composition required, and we constructed plasma diagnostics capable of analyzing the ion mass during the PIIP process. (5) There is a pressing need for simple, quick, and inexpensive diagnostics indicating incident ion dose and retained ion dose in an industrial facility. Calorimetry of the cooling water supplied to the component may allow correlation between incident ion dose and heat extracted from the target. We correlated the color change of anodized tantalum coupons with the retained nitrogen dose.

Comparison of Cyclotrons and Linacs for High-Intensity-Beam Applications

Tarlochan Bhatia

The goal of this work was to gain a theoretical understanding of the capabilities and limits of cyclotrons in their ability to accelerate very high ion currents without significant beam loss. In doing this, we collaborated with scientists working at the large cyclotron facilities at the Yerevan Physics Institute and the Paul Scherer Institute.

The key concern identified is the role played by space-charge forces, because they determine the limiting current and the energy spread. The energy spread, in turn, determines losses at extraction. Losses throughout the overall cycle of operation must be kept to an absolute minimum to ensure maintainability of the facility without the need for remote handling. It is essential to evaluate the effects of space-charge forces on the beam-loss problem in isochronous cyclotrons, specifically one designed to achieve 10-MW operation.

We have chosen to investigate the applicability of the code PARMILA to our cyclotron studies. PARMILA is an established, well-documented code for calculating phase and radial motion in linear accelerators. With some modifications, this code can be used effectively to track particles in a ring cyclotron, such as the 590-MeV machine at the Paul Scherer Institute. We have documented the modifications needed for this application.

We have successfully initiated a collaboration with the Yerevan Physics Institute, which will be seeking its own internal funds for ongoing studies and experiments. Under this collaboration, we will study a design of a low-energy, radio-frequency, quadrupole accelerator suitable for producing ions for injection into the first stage of a major cyclotron ring, such as the one located at Yerevan.

Engineering Science

Heatpipe Power System (HPS) Development

Michael Houts

The heatpipe power system (HPS) is a power system for space applications that is potentially capable of providing up to 1,000 kWt to power converters, resulting in the capability to produce maximum electric power in excess of 100 kWe. The objective of this project was to determine if the design approach we used for the HPS (see accompanying figure) could lead to a near-term, low-cost power supply with good safety and performance features.

During this past year, we further developed the methods to ensure that the power system would remain subcritical during all credible launch accidents, thus helping to ensure launch safety. Our detailed neutronic models showed that enriched boron, rhenium, or a combination of the two materials could increase the shutdown margin during launch accidents without an unacceptable increase in power system mass. We calculated the peak axial and radial heat fluxes and showed them to be well within the existing database for heatpipe operation. We also evaluated shield designs and found that certain innovative designs appear feasible.

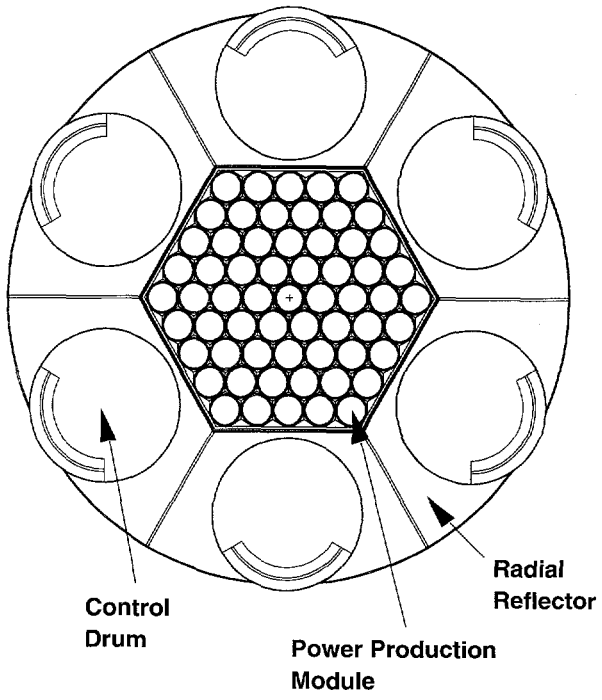
Our results show that significant axial and radial power flattening is possible through proper location of the boron or rhenium used to ensure launch accident subcriticality. We found that a beryllium oxide reflector would have a smaller volume than a beryllium reflector with an equivalent reactivity value, but reflector mass would be comparable because of beryllium oxide's higher density. We also confirmed the feasibility of performing full-system, electrically heated tests. Control of the power system can be accomplished by control drums or sliding reflectors (the latter would be less massive).

Our accomplishments indicate that our design approach could lead to a near-term, low-cost, space fission-power supply with good safety and performance features.

Publications

Houts, M.G., and D.I. Poston, "Terrestrial Applications of the Heatpipe Power System," *Am. Institute Phys. Conf. Proc. 387* **1**, 299 (1997).

Houts, M.G., et al., "Heatpipe Power System and Heatpipe Bimodal System Design and Development Options," *Am. Institute Phys. Conf. Proc. 387* **3**, 1317 (1997).



Cross section of a 12-module HPS heat source.

Detection of Underground Structures, Tunnels, and Land Mines

Joseph Mack

There is a continuing need in US defense, intelligence, and drug interdiction programs for effective overt and covert means of detecting underground tunnels, structures, voids, and land mines. Examples are the tunnels discovered in the Korean demilitarized zone, tunnels across the US-Mexican border east of San Diego, underground facilities for clandestine weapons operations, and operations in Bosnia to find and disable land mines. A significant improvement in the ability to detect underground facilities and land mines would benefit several federal agencies, the International Atomic Energy Agency, as well as the mining, energy, construction, and waste cleanup industries.

Our effort this year focused on assessing, optimizing, and adapting the 5- to 100-kHz radio-imaging method (RIM), developed by Raton Technology Research in Raton, NM. It has been demonstrated experimentally (from prior results) and understood theoretically that the RIM system of electromagnetic scattering can detect conducting materials found in tunnels. Related technologies for land-mine detection and underground communications are also being developed in this project.

We have achieved a basic understanding and demonstration of the RIM as it applies to the detection of underground structures. Sensing buried conductors from near-surface to ~30-m depths is possible. (Depth and resolution limits of this system are evolving.) We have developed, applied, and are now documenting our wave-propagation assessment computer code. It is being expanded to model a variety of transmitter configurations, receivers, and long or compact targets. We have continued our analysis of our previously reported small-scale experiments aimed at quantifying the many issues and baseline technologies noted above.

As we prepare for our next set of experiments, improved modeling will allow a better understanding of the signal-to-noise ratio, dynamic range, and clutter suppression associated with the hardware development.

Technology for CO_2 Emissions Monitoring and Control

Edward Joyce

Our objective is to examine three specific issues relative to CO_2 emissions and controls: the effect on emissions of deregulating the utility industry, the role of advanced power systems in reducing emissions, and the development of sequestration technologies. Deregulating, or more generally restructuring, the electric utility industry has been widely debated on the national scene over the past several years. The largest impact of deregulation will be on how electric power is generated. Traditional, rate-of-return-regulated public utilities are already losing their monopoly franchises in their service territories, with industry entry now open to any and all suppliers (generators). After restructuring, suppliers of electric power will be motivated by an entirely new set of incentives. Consumers will get to choose their supplier and will make that choice based upon relative price and other marketing incentives. Price per kilowatt-hour will be the most important selection criterion, and this will force generators to choose the most cost-effective technologies and fuels. Standard private-industry cost analyses will prevail, which may put coal and coal conversion technologies at a disadvantage.

We established a rationale and the desirable parameters for the modeling, simulation, and analysis (MS&A) of end-to-end coal-based electric power production and delivery systems, from fuel source through generation stations with CO_2 capture to transmission of electricity and transport of CO_2 for sequestration. The MS&A is intended to provide information in such categories as material flows (including emissions), energy flows, performance (including efficiencies), cost, and risks that decision-makers need to effectively allocate resources and to direct the research, development, demonstration, evaluation, testing, and commercialization of new technologies.

We also evaluated the merits of sequestration technologies for CO_2 disposal that are currently being developed at Los Alamos and other institutions. At Los Alamos, these technologies principally involve mineral sequestration of CO_2, the conversion of CO_2 to methanol via high-temperature photolysis, and the enhancement of natural sinks via biotechnology methods. Another interesting and promising technique that we evaluated is the formation of clathrate hydrate compounds through reaction with water at near-freezing conditions. It is too soon, however, to make any firm decisions about the relative merits of these technologies.

Removal of Transuranic Materials from Contaminated Equipment Using Plasma Decontamination

Carter Munson

Our goals for this project include developing and demonstrating plasma-based decontamination techniques for metallic surfaces. This work is relevant to DOE decontamination, decommissioning, and legacy waste issues.

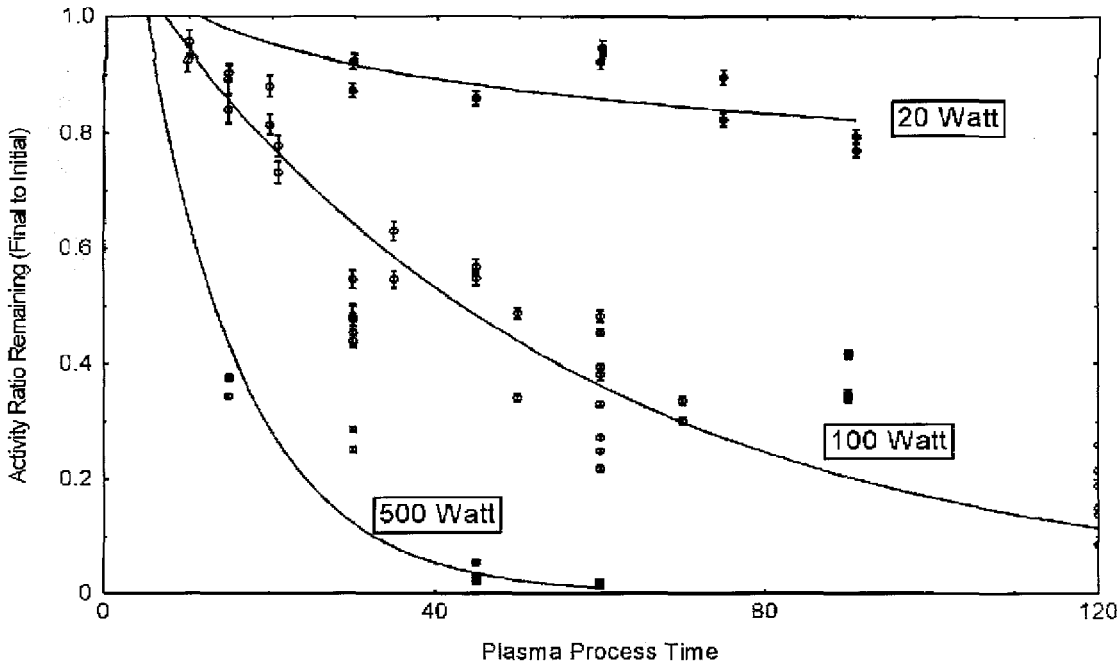
During the past year, we conducted experiments to quantify the efficiency of the plasma-based removal of radioactive contaminants from surfaces (see accompanying figures). We examined both the contaminant materials expected to be volatilized by the action of the fluorine-containing plasma (isotopes of uranium used as surrogates for plutonium) and the materials not known to form volatile fluorides (radioactive daughter products of the uranium decay). We achieved contaminant removal of better than 98.5% for volatile-forming uranium isotopes in one hour of plasma processing with a moderate RF power input of 500 W, which generates a plasma sheath potential of approximately –400 V. (Sheath potentials of >1 kV are necessary to obtain significant physical sputtering.) To perform decontamination experiments, we fabricated a dedicated modulator that is capable of generating sheath potentials in excess of 1 kV. The modulator is available for continued experimental work.

We have generated interest in using resources at the Idaho National Engineering and Environmental Laboratory for the engineering design and construction of a pilot facility. This facility could ultimately be incorporated in the upgrade of the Center for Materials Research building at Los Alamos. In addition, we submitted a successful Waste Management Education and Research Consortium proposal, obtaining support for additional collaborative research to be performed at the University of New Mexico.

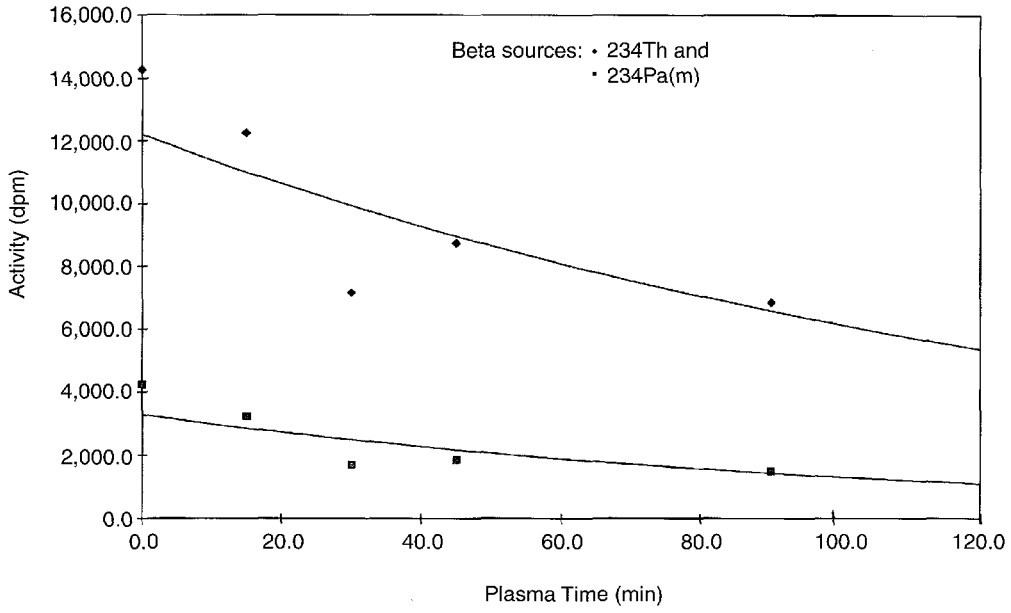
Publications

Veilleux, J.M., et al. "Plasma Decontamination of Uranium from the Interior of Aluminum Objects" (Joint Conference on the Environment, Waste Management Education & Research Consortium, Albuquerque, NM, April 22–24, 1997).

Veilleux, J.M., et al., "Plasma Decontamination of Uranium Oxide from Stainless Steel Surfaces" (American Nuclear Society Meeting, Albuquerque, NM, November 16–20, 1997).



Decontamination of uranium oxide from stainless steel by NF_3 plasma. The ratio of final-to-initial alpha contamination is a function of time and the RF power level to target.



Beta activity versus plasma immersion time. This graph demonstrates removal of materials that do not form volatile fluorides, even under conditions with minimal physical sputtering (plasma sheath is approximately −130 V). The beta sources are ^{234}Th and ^{234}Pa (primarily ^{234}Th because of half-life considerations). The RF power is 100 W.

Environmental Technology Analysis Using Complex System Simulation Techniques

R. Wayne Hardie

The purpose of this work is to research and apply a fundamentally new approach to environmental management policy analysis and assessment. Our goal is to use advanced simulation techniques for analyzing complex systems to develop an environmental technology evaluation toolbox for use by environmental management policymakers. The toolbox is being designed to evaluate such issues as how to spend the budget to achieve the best set of prevention/remediation options over the next decade, given various constraints and new treatment and mitigation technologies.

This year we (1) studied the DOE's needs and goals for environmental management as expressed in the 1995 and 1996 Baseline Environmental Report, the Office of Environmental Management's critical performance measures, and other publications; (2) prepared a software requirements document to guide the development of simulation software that would directly address the published analysis needs of the environmental management policy; (3) developed an initial C++ based simulation toolbox; (4) developed pit production and waste transportation and disposal models; and (5) performed sample analyses using the models. Our constructionist approach supports analysis of global properties of complex systems that emerge as a result of the unpredictable interactions of the various system components.

Waste characteristics of DOE production and environmental management systems analyzed in this first year of our project include location, medium, concentration, and volume. We also prepared an initial design to include cost analysis in the simulation toolbox.

Reactor-Based Tritium Production

John R. Ireland

The viability of using fast-spectrum reactors to produce tritium is strongly dependent on target design and associated production rates. To fully understand the production potential of these reactors requires understanding the issues connected with each type of target (lithium aluminate, lithium oxide, and helium-3) in a fast-spectrum reactor.

We have updated our findings on the use of fast-spectrum systems, such as the Fast Flux Test Facility at Hanford, WA, to produce tritium to meet future nuclear weapons stockpile requirements. The work focuses on information and analysis results that have become available since earlier studies, principally the 1996 JASON Committee study, and is a continuation of our previous studies on other reactor systems and tritium targets. We looked at target performance, effects of fuel type on performance, tritium production rates, impacts on the reactor safety envelope, and other systems issues.

Development of Predictive Simulation Capability for Reactive Multiphase Flow

Brian VanderHeyden

The objective of this project is to develop a self-sustained research program for advanced computer simulation of industrial, reactive multiphase flows. Reactive multiphase flows are found in many of the processes across the spectrum of manufacturing industries. Examples of interpenetrating multiphase flows include riser reactors used in petroleum refining, bubble column reactors used in chemical manufacturing, and sparged aqueous precipitators used in aluminum production and in plutonium recovery. The prototype research problem we are focusing on is a three-phase alumina precipitator used in the Bayer process, a key step in aluminum refining.

Accomplishments this year include the development of an improved reaction mechanism of the alumina precipitation growth process, the development of an efficient method for handling particle-size distribution in multiphase flow simulation codes, the incorporation of precipitation growth and agglomeration kinetics in the Laboratory's multiphase flow code library (CFDLIB), and the evaluation of multiphase turbulence closure models for bubbly flow simulations. While only in the first year, the project has already stimulated interest from Alcoa, the largest aluminum producer in the United States.

Molten-Metal Target Development for High-Power Neutron Spallation Sources

Eric Pitcher

The goal of this project is to develop the technology for high-power, spallation-neutron-production targets. These targets can be used for intense neutron sources for material science, nuclear science, accelerator production of tritium, and accelerator-driven transmutation technologies. We chose lead-bismuth eutectic (LBE) as the production fluid because it has a low neutron-absorption cross section, a high boiling point, and a low melting point. Considerable experience exists at the Institute for Physics and Power Engineering in Obninsk, Russia, where LBE was developed as a coolant for submarine reactors.

As a first step in developing this technology, we constructed a medium-size molten LBE test loop. The loop consists of a steel cask that contains approximately 3 tons of LBE that can be resistively heated. Above the cask is a pump bowl with a 25-hp impeller motor. The loop is approximately 1 m on a side and is connected to the pump bowl. The entire loop is heated with heat tape and is insulated. Attached to the loop are level sensors, thermocouples, and automatic safety systems.

We performed an initial test of the loop, including its associated control and auxiliary systems, heating the LBE in the cask and transferring it to the pump bowl by pressurizing the cask. The LBE was circulated in the test loop for approximately 3 hours. The test was a complete success. Future work will involve a series of experiments to address corrosion and material compatibility issues.

Instrumentation and Diagnostics

Advanced Ignition and Propulsion Technology

James Early

In this project, we have previously demonstrated several technologies that use lasers as a viable alternative to capacitive discharge for igniting jet-turbine engines. These innovative ignition technologies reliably ignited jet fuel aerosols over a broad range of fuel/air mixtures and at fuel temperatures as low as –40°F. The fuel ignition performance is highly superior to that obtained with state-of-the-art laser-spark ignition at comparable laser energy. We have also developed a laser-based method for effectively removing optically opaque deposits of hydrocarbon combustion residues from laser window surfaces. To date, this R&D project has resulted in Laboratory action on four pending patents and one invention disclosure.

This past year, we developed a multiple-laser ignitor concept that will reduce laser hardware cost and size while enhancing its durability. This dual-pulse laser design uses multiple, passively Q-switched, monolithic Nd:YAG ignitor lasers that require no cooling water or electrical input. Each laser is pumped by a single, remotely located multiplexed laser. Tests using a prototype fuel-injection nozzle, developed for the Air Force's advanced turbine engine program, demonstrated the superior fuel ignition achieved with our dual laser-pulse approach compared with that obtained with the conventional single-pulse laser ignitor. These tests were run at reduced fuel temperatures as low as –40°F. A patent is pending on this technology.

We were also invited by Allied Signal, Inc., to demonstrate our multiplexed, dual-pulse laser ignitor under actual turbine engine operating conditions using the combustor test rig facility at its Technology Center in Morristown, New Jersey. The test showed a significant increase in the envelope of fuel and air flow over which fuel ignition could be achieved compared with Allied Signal's best results using a single-pulse laser approach.

Development of a System for Endoscopic Imaging and Spectroscopy of Pit Interiors

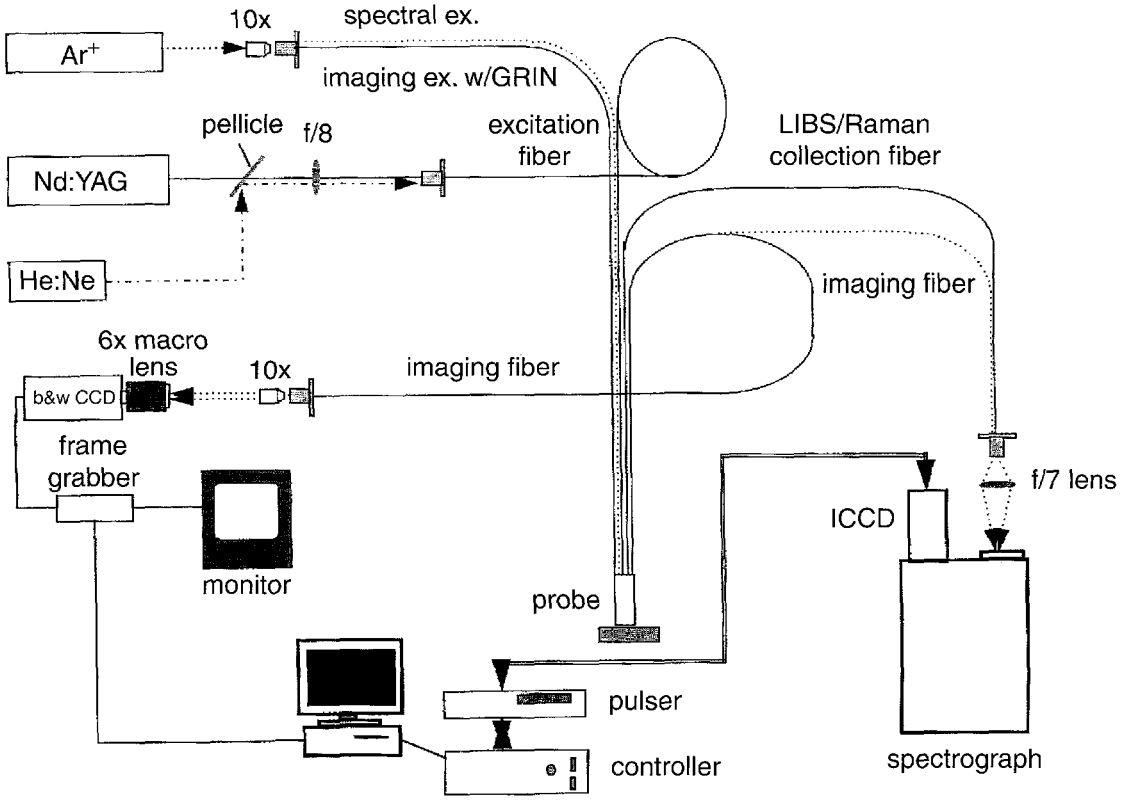
D. Kirk Veirs

Our objective is to develop a method for visually imaging the inner surface of intact plutonium pits and spectroscopically analyzing their features. During this past year, we tested the designs for the major subsystems. We used a fiber-optic imaging bundle to image the interior surface of a surrogate pit through a fill tube. Linear, rotational, and bending motions were used to control the probe tip and define the area being imaged. We designed and began to fabricate a mechanical device to hold a pit and precisely control the motions of the fiber-optic imaging bundle. Computerized motion control will allow us to raster the fiber tip across the pit's inner surface in order to collect overlapping images for a nearly complete image of the surface.

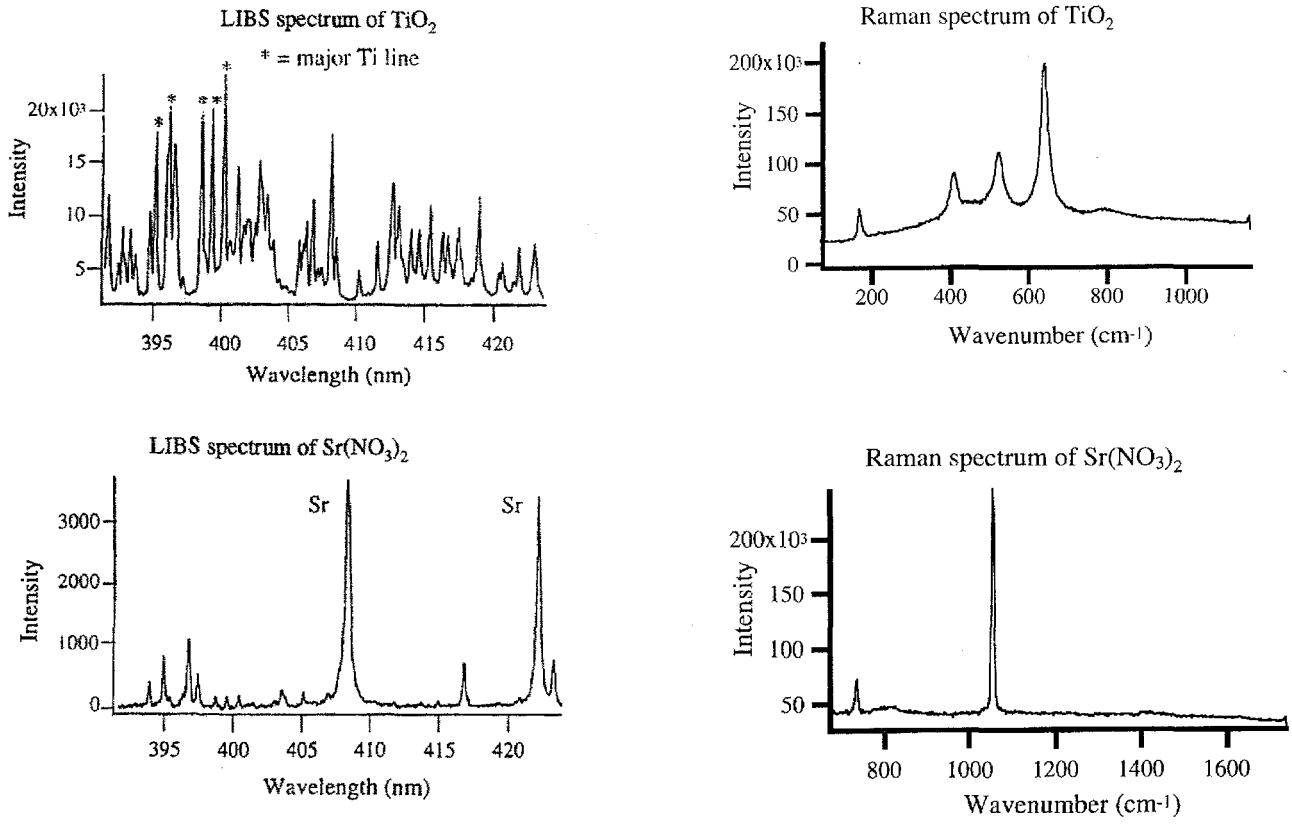
We fabricated and tested a fiber-optic microprobe that incorporates imaging optics, laser-induced breakdown spectroscopy (LIBS) for elemental analysis, and Raman spectroscopy for chemical analysis. The microprobe consists of three silica multimode fibers, one for the Nd:YAG laser for LIBS excitation, one for the continuous-wave, argon ion laser for Raman excitation, and one to collect and deliver the emitted or scattered light to a spectrometer (see first figure). The probe's imaging bundle simultaneously identifies features and aligns the excitation lasers onto the area to be analyzed. We collected LIBS and Raman spectra of small, discrete particles, demonstrating the power of our analytical approach (see second figure).

Publications

Marquardt, B.J., et al., "A Novel Probe for LIBS and Raman Measurements Utilizing Fiber-Optic Imaging" (submitted to *Appl. Spectrosc.*)



Schematic diagram of the microprobe apparatus. A Q-switched Nd:YAG laser operating at 1064 nm was used for LIBS excitation. A continuous-wave argon ion laser operating at 514.5 nm was used as the excitation source in the Raman experiments. The helium-neon laser was used to target the region of interest on the sample. Optical fibers are used to link the excitation sources, the sample, the imaging video, and the spectroscopic instruments.



The fiber optic microprobe was used to analyze fine powders of TiO_2 and $Sr(NO_3)_2$ embedded in a clay soil sample. It identified particles for analysis and targeted the excitation lasers. On the left are the LIBS spectra for elemental analysis; on the right are the Raman spectra for chemical analysis. The combination of the two spectra allows unambiguous identification of the particles as TiO_2 and $Sr(NO_3)_2$.

Field Detection of Chemical Agents by Membrane-Introduction Mass Spectrometry

Philip Hemberger

This year we developed technology for the on-line detection of chemical-agent surrogates based on membrane-introduction mass spectrometry (MIMS). We met all our project goals: we interfaced a membrane-sampling system to an ion-trap mass spectrometer and demonstrated the rapid detection of semivolatile chemical-agent surrogates in air.

In MIMS, a polymer membrane is the interface between the sample and the vacuum of the mass spectrometer. Sample analyte adsorbs on the outer surface of the membrane, diffuses through it, then evaporates from the inner surface into the mass spectrometer. For chemical-warfare (CW) surrogates, the diffusion process is slow and prevents rapid detection and quick recovery. We used a commercially available composite membrane made by plasma deposition of a 0.5-μm layer of polydimethylsilicone on a microporous polypropylene support fiber. This ultrathin membrane leads to high permeation rates and limits the amount of analyte that dissolves in the membrane.

We used the CW surrogate dimethyl methylphosphonate (DMMP) to evaluate this membrane. Diffusion of DMMP through the silicone membrane that we used in our earlier studies was extremely slow, and the recovery time lasted for several days. With the ultrathin membrane, however, we have achieved very brief sampling periods and, thus, reduced recovery periods. In the case of DMMP, recovery time has been reduced from days to minutes.

Publications

Cisper, M.E., and P.H. Hemberger, "The Direct Analysis of Semivolatile Organic Compounds by Membrane Introduction Mass Spectrometry," *Rapid Commun. Mass Spectrom.* **11**, 1449 (1997).

Cisper, M.E., and P.H. Hemberger, "Organometallic Analysis by Membrane Introduction Mass Spectrometry," *Rapid Commun. Mass Spectrom.* **11**, 1454 (1997).

Assessment of Cold-Neutron Radiography Capability

Thomas McDonald

The purpose of this project is to demonstrate cold-neutron radiography techniques at the Los Alamos Neutron Science Center (LANSCE) and to investigate their potential applications. Neutron radiography is an attractive means of nondestructive testing and evaluation. Measurement of the neutron transmission as a function of energy and position can be used to give elemental and spatial information over a broad band of neutron energies. In addition, by using time-of-flight and gated imaging, radiography at specific neutron energies is possible.

We developed a radiography technique that uses only a narrow range of neutron energies and with it demonstrated the potential of the Bragg cutoff phenomena for discriminating between materials of different densities (see accompanying figure). The use of this technique appears to have possible applications in the DOE stockpile maintenance and science-based stockpile stewardship (SBSS) programs and in industry. We used it to resolve a crack in a fireset sample for the SBSS program and produced an image of a jet engine fuel nozzle.

In stockpile maintenance and SBSS, cold-neutron radiography could be used to determine imperfections in bulk assemblies, the degree of saturation of desiccant, cracks and inhomogeneities in fireset cases, and cracks in PZT (lead zirconate titanate) electrical generators. The technique also has at least one industrial application—determining the extent of coking in jet engine nozzles. We presented our radiography technique at three technical meetings that had industrial participation and explained it to potential industrial users.

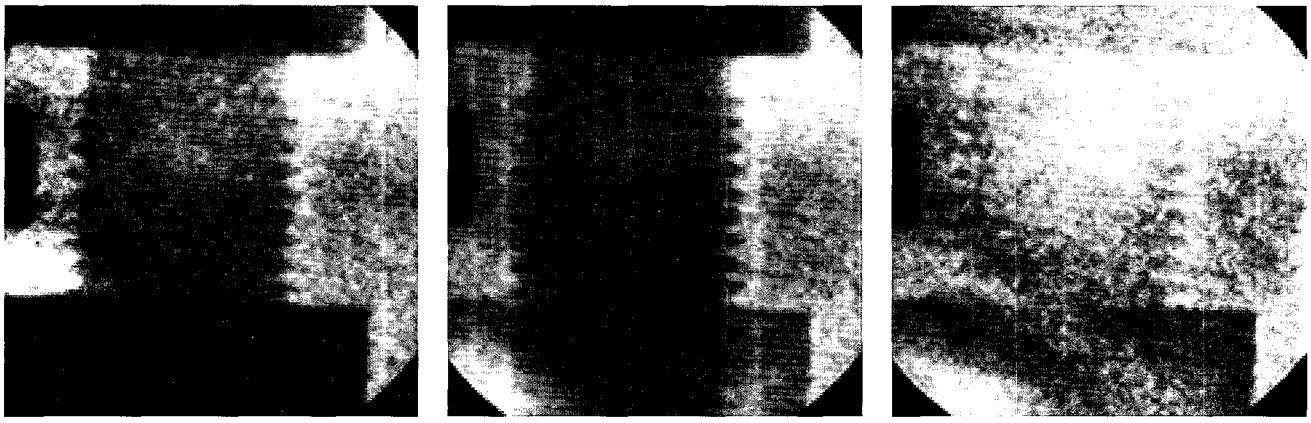


Image A Image B Image C

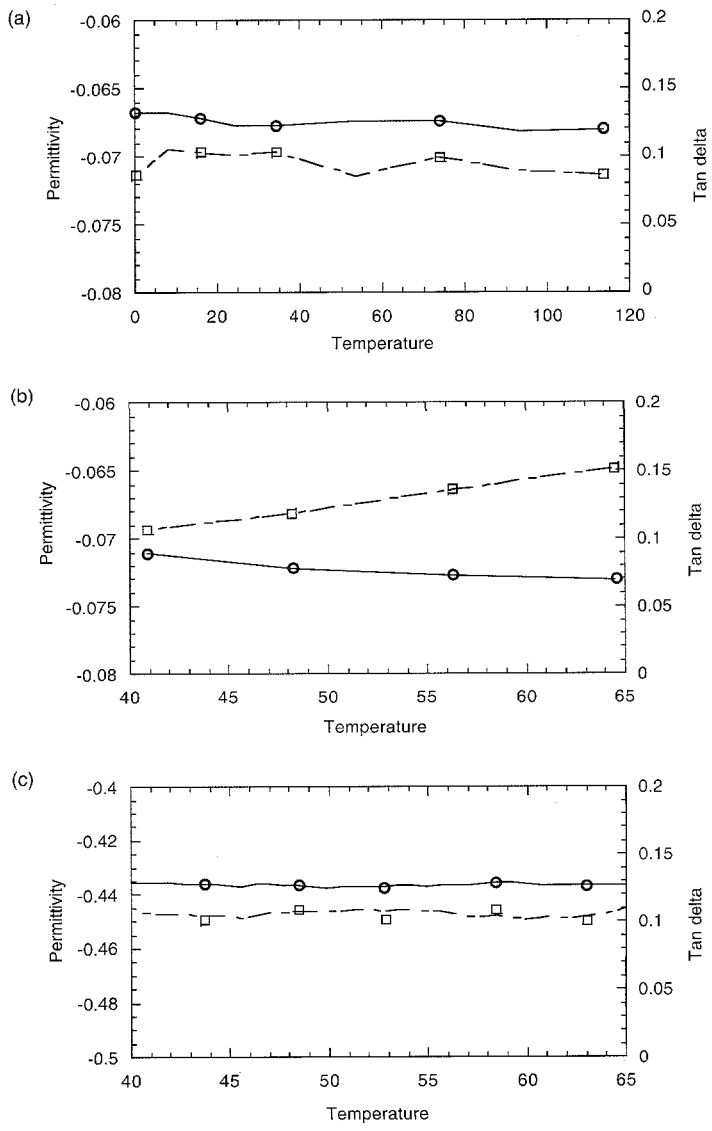
Demonstration of cold neutron radiography using time-of-flight and gated imaging to obtain radiographs at specific neutron energies. These images—a series of radiographs of a carbon bolt threaded into a beryllium block—demonstrate materials discrimination using the Bragg cutoff phenomenon. Image A is a radiograph taken at a neutron energy of approximately 7.5 meV, which is above the Bragg cutoffs of both beryllium (6 meV) and carbon (2 meV). Both the beryllium block and carbon bolt are relatively dark. Image B is a radiograph taken at approximately 2.9 meV. The beryllium is lighter because of the reduction in scattering cross-section across the Bragg cutoff, and the carbon bolt inside the beryllium is now more visible. Image C is a radiograph taken at approximately 1.5 meV, which is below the Bragg cutoffs of both beryllium and carbon. Both materials are much lighter: if any denser material had been obscured by them, it could now be seen.

Nonintrusive Characterization of High Explosives in Nuclear Weapons Systems: Dielectric Relaxation Analysis of PBX

Mark Smith

Our objective is to develop dielectric relaxation techniques for determining the physical state of the high explosive PBX 9501 that has a compositional formulation of 95/2.5/2.5/0.1 wt% HMX/Estane/nitroplasticizer/inhibitor. This measurement process will ultimately require little or no disassembly of the stockpile warhead. The measurements will describe the physical properties and mechanical changes in PBX 9501. We are performing proof-of-principle work on the polymer components of the binder system in PBX 9501 that span the range of the war reserve specifications and the expected changes associated with aging. We are correlating dielectric measurements to changes in nitroplasticizer content, Estane molecular-weight reduction, and inhibitor (diphenylamine or Irganox) depletion.

The accompanying figure shows results of the dielectric analysis of Estane, Estane with nitroplasticizer, and a physical mock of PBX 9501 comprising Estane, nitroplasticizer, and sugar particles. The greatest challenge is to increase the sensitivity of the measurement in order to determine the feasibility of the dielectric technique. Although the samples under examination are visually and chemically very different, their dielectric responses do not vary significantly. The results over a broad temperature range show a greater variation; however, this variation would not serve the goal of developing a nonintrusive diagnostic for stockpile applications. Without greater sensitivity, the dielectric technique will not be suitable as a diagnostic for PBX 9501 in the stockpile.



In these dielectric relaxation analyses of high-explosive components, the circles represent the permittivity and the squares correspond to the loss tangent (tan delta). When pure Estane (a) is heated to 120°C, a slight decrease in permittivity is indicated; the loss tangent remains nearly constant over the temperature range. For Estane with nitroplasticizer (b), a clear decrease in the permittivity is apparent; the increase in the loss tangent is due to the presence of the plasticizer acting as an internal lubricant with respect to the polymer chains. The sugar-mock (c) shows a constant permittivity, greatly decreased compared with those of the other samples, and a constant loss tangent. The sugar filler masks the signatures of the polymer/plasticizer component.

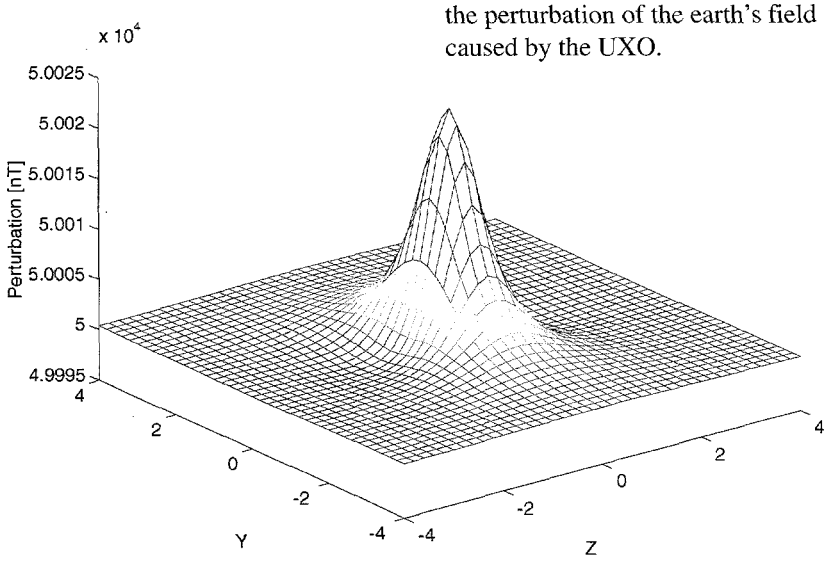
Miniature Flux-Gate Magnetometers for Use on Small Mobile Platforms

R. Clayton Smith

Recently, the need for remediation and reclamation of military proving grounds containing unexploded ordnance (UXO) has been recognized. The goal of this project is to develop a miniature flux-gate magnetometer capable of being carried on a robot to measure the static magnetic field of the UXO and use the field information to determine its location. A flux-gate magnetometer is a linear sensor for measuring an arbitrary vector component of a magnetic field. The instrument we are developing is a three-axis device for measuring three orthogonal components of a magnetic field both in total field and field gradient modes. The earth has a terrestrial magnetic field magnitude of approximately 50,000 nT. A major thrust of this project is to determine the perturbation of the earth's field caused by the UXO.

We generated a magnetic model of a ferrous sphere placed in the terrestrial magnetic field to determine the field perturbation in a plane located an arbitrary distance from a ferrous sphere. The model was evaluated for a cold-roll steel sphere 155 mm in diameter located 1.5 m from the measurement plane. The data from the model indicates that the instrument must be capable of detecting perturbations from 100 to 500 ppm in order to detect such a ferrous body.

The instrument developed to detect such magnetic field perturbations is a low-power device (less than 50 mW) with an absolute field measurement range of 60,000 nT and a voltage output sensitivity of 65 μV/nT. Our work characterizing and testing the instrument is ongoing.



The magnetic field perturbation caused by a 155-mm, cold-roll steel sphere a normal distance of 1.5 m from the measurement plane. The sphere is centered at the origin. The terrestrial magnetic field is 50,000 nT.

Sensors for Point Detection of Biological and Chemical Warfare Agents

Thomas Zawodzinski

The goal of this project is to develop enabling technology for electrochemical devices capable of point detection of chemical and biological warfare agents with high sensitivity. Our approach is to develop thin-film structures that enable the use of various sensing methods, though our emphasis is on electrochemical transduction methods.

During the past year, we prepared prototype electrochemical sensors for the detection of chemical warfare agents, and we tested the sensors against compounds that simulate the activity of these agents. Various types of applications were explored. In addition to sensors for HCN (a blood agent) and mustard agents, we demonstrated electrochemical detection of wintergreen oil, a common test agent. We also showed that an electrochemical sensor could be deployed to achieve extremely sensitive detection of dipicolinic acid, a signature component of anthrax spore coats. We envision using our sensor in conjunction with pyrolysis to provide a relatively rapid indicator for anthrax.

Finally, we have carried out steps toward practical realization of these sensors by (1) collaborating in the development of a breadboard minipotentiostat to be integrated into a small hand-held sensor system (we are currently developing the remainder of the required hardware), and (2) developing a self-contained electrochemical sensor. The latter has been demonstrated in the context of the tests with wintergreen oil present in aqueous solution.

Geoscience, Space Science, and Astrophysics

Improved Atmospheric Transport for Risk Assessment

Daniel Cooper

The goal of this project was to develop and test a model with high spatial and temporal resolutions that simulates plume development and transport of mass and energy between the earth's surface and the atmosphere. We are currently using this high-gradient (HIGRAD) model to characterize the turbulent-convective behavior in the first few meters of the atmosphere. Applications of the model are currently related to ocean-atmosphere interactions and the mechanisms for the development of forest fires.

During this past year, we began testing the HIGRAD model with lidar data collected in FY 1996 over the ocean. The model has a spatial resolution of 2 m in all three dimensions and includes a small-eddy-turbulence simulation capability (a Smagorinsky code) with a fully integrated radiation section. It is the highest-resolution model of its kind in the world.

The model was able to reproduce the horizontal and vertical microscale convective elements that the lidar observed over the ocean. In addition, we can expand the model's spatial grid to cover any scale, including global scales. Further, because the model is modular, we can incorporate new codes into the simulation scheme. For instance, the new LDRD project on "Coupled Environmental Modeling of Wild Fires" uses the HIGRAD code to simulate fires. We will continue to refine the HIGRAD model and will produce a paper on the interactions of the ocean-atmosphere system with lidar data and model results.

A Simulator for Copper Ore Leaching

Bryan Travis

Demand for copper, a strategic metal, is growing worldwide and may outstrip the ability of the mining industry to keep up in the coming decades. Extraction of copper from the earth is an energy- and pollution-intensive operation. To improve recovery, several improvements in the current acid-leaching technology are required, the most important of which is the ability to quantify how fluids move through heterogeneous materials in a complex chemical environment.

There is a strong conviction in the mining industry that copper extraction technology can be improved through model simulation. Models provide a mechanism for organizing in a systematic way what we know about a site and the processes involved. We have developed several simulation tools that can be applied directly to the copper-leaching process. We already have an advanced, three-dimensional grid-generation capability (GEOMESH) that allows us to simulate very accurately the potentially complex geology found in mining environments. We also have state-of-the-art reactive flow and mass and energy transport simulators (FEHM, TRACR3D) for modeling porous/fractured rock. The primary need is to couple a comprehensive copper chemistry package with our reactive flow and transport models. This is the goal of our project.

This year we worked with chemists and hydrologists at Broken Hill Proprietary, formerly Magma Copper, of Tucson, Arizona, to define a set of simulations to test their designs for in situ copper leaching. We also worked with engineers from Kennecott to define simulations that will address problems with heap pad leaching, in particular, channeling of flow. In addition, we worked with Kennecott to address contamination remediation for one of their sites. Further, we expanded the capability of our numerical simulators to include the relevant chemical reactions for copper leaching.

In Situ Bioremediation of Trichloroethylene-Contaminated Groundwater

Nina Rosenberg

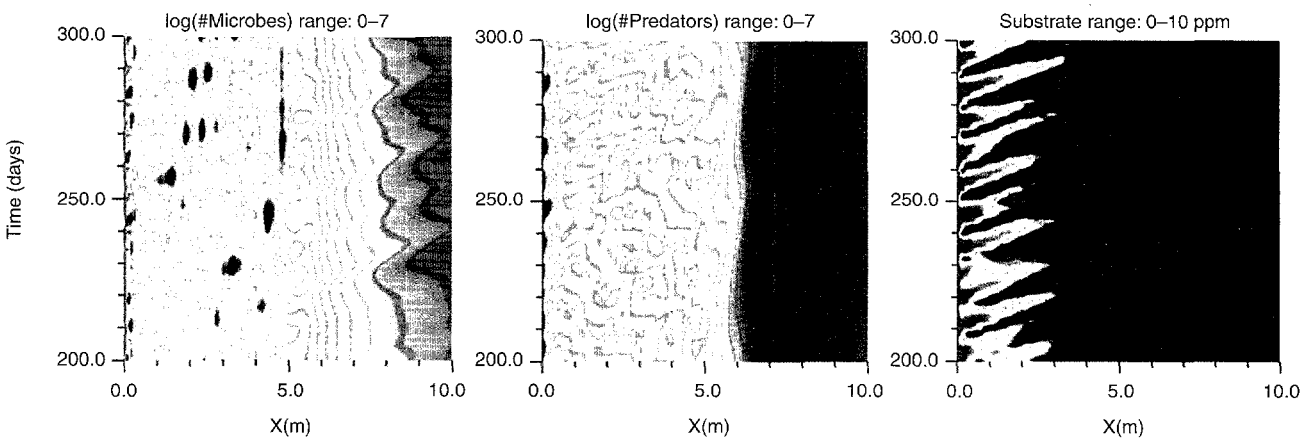
The primary objective of this project is to improve computational tools used to assess the field performance of in situ bioremediation technology. During the past year, we added microbial species interactions to TRAMPP, our bioremediation model. Recent observations indicate that predator species can have a significant impact on in situ bioremediation (see figure) by feeding on the microbial species that degrade contaminants. Simulation studies carried out with our model indicate that predator grazing of microbes can decrease efficiency by about 25%. Laboratory experiments were begun to verify this.

Predator grazing also has implications for barrier methods. Here, a soil region into which a contaminant plume will migrate is enriched with nutrients and growth substrate. The goal is to stimulate microbes so that when the plume arrives, the microbes will be at sufficiently high numbers to rapidly degrade the contaminant. Simulation studies indicate that the behavior of barrier systems can become complex because of predator grazing. For example, barriers may operate properly for several weeks and then fail almost completely for about a week, and then recover. This alternating pattern of a long working phase and a shorter failure phase can repeat indefinitely.

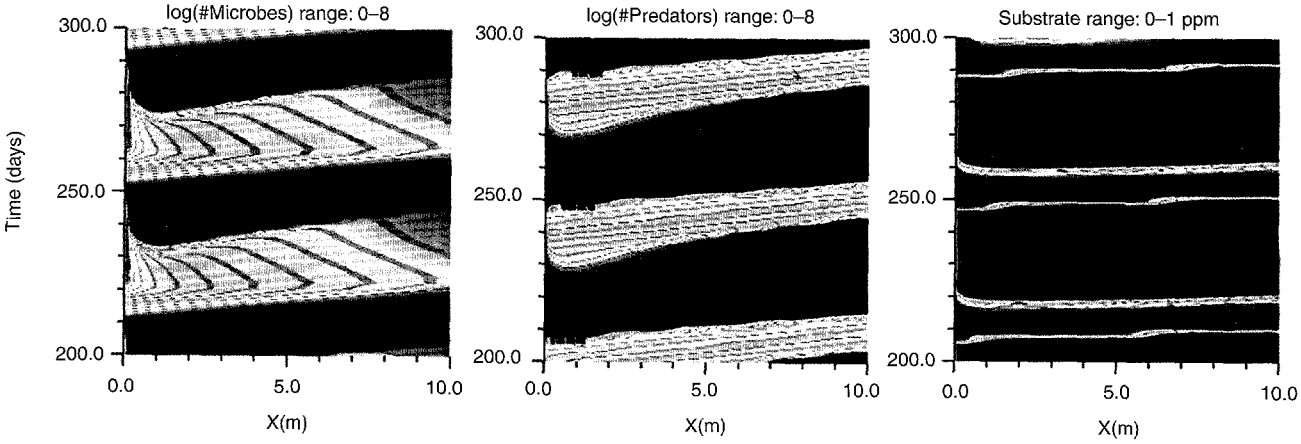
We held a workshop on the "Mathematical Issues of In Situ Bioremediation" at Los Alamos in June 1997. The effects of soil heterogeneity and biofilm growth processes were identified as two major challenges. Finally, we are implementing two promising ideas for capturing small-scale heterogeneity: homogenization and fractal integration. This will yield better calculation of transport pathways and more-accurate bioremediation simulations.

Publications

Travis, B.J., and N.D. Rosenberg, "Modeling In Situ Bioremediation of TCE at Savannah River: Effects of Product Toxicity and Microbial Interactions on TCE Degradation," *Environ. Sci. Tech.* **31**, 3093 (1997).



One-dimensional simulation of a barrier system that works. The contaminant (substrate) enters from the left at a constant rate in a steady groundwater flow. The vertical axis indicates time after startup of the barrier system. Although the substrate penetration is variable due to predator growth, the microbes are able to consume the substrate within 2 to 4 m downstream (horizontal axis).



In this case, the predator-microbe pair is different and the groundwater flow rate higher. The barrier system is characterized by two alternating phases, a long one (about 30 days), in which substrate (contaminant) is completely removed as fast as it enters, and a shorter one (about 10 days), in which predators grow vigorously and decimate the microbes, allowing most of the contaminant to break through.

Underground Communication

David Reagor

In underground areas, the need to communicate with mobile personnel or equipment arises frequently. Conventional radio signals do not propagate underground, but it has been demonstrated that radio signals with ultralow frequencies (ULFs) propagate significant distances. However, conventional semiconductor-based receivers for these ULF signals are either too large or too noisy. A possible alternative would be to use high-temperature superconducting components to form compact, low-noise receivers for the ULF signals.

In this project, we are studying the underground communications potential of high-temperature superconducting quantum interference devices (SQUIDs). Our goal is to develop the scientific and technological foundation necessary to provide a compelling case for fully developing this technology. We are focusing on (1) continued development of SQUIDs, (2) development of control and interface electronics for SQUIDs, (3) development of a transmission antenna, (4) preliminary design and testing of orientation-independent receiver technology, (5) engineering of cryogenic packaging, (6) development of novel refrigeration technology, and (7) field testing and demonstration of a simple receiver.

In FY 1997 we made substantial progress. We demonstrated a SQUID-based pager-type receiver that can detect information on a 100-pT ULF signal. We also designed, fabricated, and successfully tested control electronics for stable, sensitive SQUID receivers. In addition, we completed the preliminary design of a magnetic-dipole transmission antenna capable of hand-held operation. We conducted extensive thermal analyses on dewar design and clearly identified potential design improvements that would lead to a longer cryogenic lifetime. Finally, we performed field tests of a receiver in an actual mine environment and received signals in a limestone mine over a distance of 500 m.

Publications

Jia, Q.X., et al., "Characterization of Ramp Edge-Geometry Ag:$YBa_2Cu_3O_{7-x}$/$PrBa_2Cu_3O_{7-x}$/Ag:$YBa_2Cu_3O_{7-x}$ Junctions and dc SQUIDs," *IEEE Trans. Appl. Supercond.* **7**, 3005 (1997).

Jia, Q.X., et al., "High Temperature Superconductor Edge-Geometry SNS Junctions with Engineered Normal-Metal Layers," *Supercond. Sci. Technol.* **9**, 985 (1996).

Reagor, D.W., et al., "A High Temperature Superconducting Receiver for Low Frequency Radio Waves" (to be published in *IEEE Trans. Appl. Supercond.*).

Reagor, D.W., et al., "Noise and Operational Characteristics of Magnetometers Made from Superconducting-Normal-Superconducting Josephson Junctions" (to be published in *J. Mater. Res.*).

Modeling and Assessment of Concrete and the Energy Infrastructure

George D. Guthrie Jr.

The purpose of this project is to apply innovative geoscience methods to the development of high-performance concrete having enhanced strength and durability. To establish the research program, we used proof-of-principle studies to demonstrate the applicability of geoscience methods to solving long-term problems in concrete. Our initial study addressed one of the most significant issues in concrete durability for the nation and, in particular, New Mexico: the alkali-silica reaction (ASR). This deleterious reaction occurs because of a geochemical incompatibility between the rock aggregate and the cement pore fluids.

This year we demonstrated that our staining method, which allows ASR to be identified in concrete structures (see the figure), also permits evaluation of the extent and point of attack of ASR. We also demonstrated that our staining method detects two different chemical compositions for the gel resulting from ASR in the concrete. We discovered evidence for a model of ASR attack in which potassium-rich ASR gels (yellow-staining) form when alkaline pore fluids attack siliceous aggregate, followed by the migration of the gels into the matrix along fractures (possibly formed by the gels) where the ASR gel reacts with calcium ions in the pore fluids to create a calcium-rich gel (pink-staining). This work resulted in a patent application (90% of which has been granted to date) and an R&D 100 Award. The test is a significant advance over existing technologies in that it is simple and has already yielded a new understanding of the mechanism of ASR.

We presented this work in a variety of forums, including meetings of the Transportation Research Board, the International Center for Aggregate, and the Strategic Highway Research Program (topical meetings on High-Performance Concrete and ASR) and presentations to state departments of transportation (including Iowa and New Mexico) and the Federal Highway Administration. The response has been very positive.

Potential sponsors also expressed significant interest in other research programs we presented. One program we continued to explore is a new evaluation technique that uses nonlinear acoustic spectroscopy to reveal ASR. Our goal is a field-deployable, nondestructive evaluation tool for deterioration in concrete. The other program is a collaboration with the Organic Analysis Group to create a new kind of high-performance concrete using supercritical CO_2 treatment. The resulting concrete is more chemically stable with a lower pH, a possible advantage in a variety of chemical environments in which long-term stability is needed.



The field application of ASR Detect™ to the identification of the alkali-silica reaction. The researcher applies ASR Detect™ to the surface of a concrete core taken from a bridge in Albuquerque, NM. The procedure revealed the presence of ASR in this structure.

Publications

Guthrie, G.D. Jr., and J.W. Carey, "A Simple, Environmentally Friendly, and Chemically Specific Method for the Identification and Evaluation of the Alkali-Silica Reaction," *Cem. Concr. Res.* **27**, 1407 (1997).

Neutron Sensors for Locating Sites of Planetary Water Deposits

William Feldman

The purpose of this project is to explore the feasibility of improving the sensitivity of neutron spectrometers specifically designed for deep-space missions. These improved spectrometers will be able to detect and identify present-day deposits of near-surface water ice and evidence for ancient deposits of standing water on solar-system bodies. The strongest and most unique signature of water is a low flux of leakage epithermal neutrons. Because water deposits are expected to be small relative to the altitudes of orbiting spacecraft, some form of collimation is needed to enhance the signal-to-background ratio. Our goal is to build a collimator that enhances this ratio and requires minimal weight and volume.

During the past year, we concentrated our effort on optimizing a slat collimator for the neutron sensor used on the Mars Global Surveyor-2001 (MGS-01) gamma-ray/neutron spectrometer. Such a collimator, if it is effective for epithermal neutrons between about 1 eV and 500 keV, will enhance the capability to detect hydrated silicates in the Valles Marineris, which is a very long and deep canyon thought to have been filled with running water early in Martian history.

Our collimator design uses borated polyethylene slats that fit the MGS-01 neutron sensor. We have written a computer code to generate input decks containing parameterized lengths and thicknesses of these slats. To date we have run seven geometries for plane-wave neutron inputs that cover 10 angles between 0 and 90 degrees and 10 energies that span 0.01 eV to 7 MeV. Our next step is to survey the results of these runs to iterate the geometries.

Technologies for the Oil and Gas Industry

Gregory Swift

Electronic instruments are used in the hot, high-pressure environment of drilled wells at depths up to several kilometers, where temperatures are over 200°C and pressures are thousands of pounds per square inch. Using such instruments on drill collars instead of merely in logging tools is one of the most interesting technical challenges for the next generation of drilling equipment. The difficulty lies in maintaining cool enough temperatures for these electronic instruments to operate in the hostile drilling environment.

In this short project, we performed a preliminary design study to explore the plausibility of using pulse-tube refrigeration to cool instruments in a hot downhole environment for the oil and gas industry or the geothermal industry. The orifice pulse-tube refrigerator is the simplest, cheapest, most rugged, and most reliable low-power cryocooler. We performed design calculations for three target criteria: (1) cooling at 30°C in a 300°C environment, (2) cooling at 75 K in a 50°C environment, and (3) cooling at both 75 K and 30°C in a 250°C environment. We chose these specific temperatures as representatives of the conditions possible in a drilling environment. The primary objectives were low cost, reliability, and small package diameter. At the end of the study, we concluded that all three designs are promising.

Nuclear and Particle Physics

Development of a Gamma-Ray Spectroscopy Capability at LANSCE

Ronald Nelson

The objective of this project is to explore the feasibility of an upgrade to the Germanium Array for Neutron-Induced Excitations (GEANIE) in the Weapons Neutron Research Facility at the Los Alamos Neutron Science Center (LANSCE). Such an upgrade will make the facility more attractive to university researchers and will aid in building a user program that the DOE Office of High-Energy and Nuclear Physics (OHENP) will support. From discussions with university researchers, we concluded that the potential resultant high data-acquisition rates would make the facility more attractive to outside users. The addition of neutron detectors was also seen as important for certain experiments.

In FY 1997, we successfully upgraded electronics with resulting improvement in the detector timing resolution, and we upgraded the data-analysis computer system. We also increased the number of detectors on the system for greater data-acquisition efficiency. Finally, we designed and fabricated a new sample holder that will allow us to use large neutron detectors close to the experiment samples. Representatives from OHENP will be visiting in early FY 1998 to discuss plans for GEANIE with us.

Publications

Becker, J.A., and R.O. Nelson, "New Opportunities in Nuclear Science with GEANIE at LANSCE/WNR," *Nucl. Phys. News Int.* **7** (2), 11 (1997).

Nelson, R.O., et al., "GEANIE at WNR/LANSCE: A New Instrument for Neutron Science" (International Conference on Nuclear Data for Science and Technology, Trieste, Italy, May 19–24, 1997).

Target Irradiation for Radioisotope Production

Eugene Peterson

Our objective in this project is to assess the technical feasibility of a new 100-MeV isotope production facility at the Los Alamos Neutron Science Center (LANSCE). We achieved this objective by evaluating potential show-stopping design issues associated with the diversion of 100-MeV protons to a new beam line and its subsequent use for target irradiation in a new facility.

This past year, we completed the design calculations for a kicker magnet needed to extract a 100-MeV proton beam from LANSCE. This work established the feasibility of building a kicker magnet that is compatible with the existing accelerator and resolved a critical technical concern for the facility concept. We also completed magnetic optics calculations for the beam line. In this work, we evaluated several concepts and ultimately revealed an acceptable beam-line design, which was used as a basis for creating the physical layout of the proposed facility.

Using a Unigraphics CAD/CAM system, we developed a three-dimensional design model for the target station, target shield, and target handling systems. This model resolved a number of design questions and will provide the basis for a technical design review of the target systems.

We used Monte Carlo computer codes for particle transport to carry out a radioisotope inventory analysis. We used the results of those calculations to establish the radiological classification of the proposed facility. We carried out similar calculations to determine the effectiveness of various radiation shielding configurations. These results identified shield wall and entryway geometries that will satisfy all known regulatory requirements.

Nuclear Measurements, Analysis, and Safeguards Science

Mark M. Pickrell

This project focuses on the creation and development of a new generation of measurement and analysis methods for characterizing nuclear materials. The changing mission of DOE has shifted the safeguards challenge from relatively pure, homogeneous materials to materials that are highly impure, contaminated, inhomogeneous, and embedded in highly shielding matrices. Current measurement and safeguards technologies are rapidly becoming obsolete and will need to be replaced by new technologies that can characterize complex materials, including residues and waste.

The physics issues we are addressing are (1) highly shielding matrices that attenuate gamma and neutron signals, sometimes completely; (2) heterogeneous materials that introduce bias errors because of the spatial dependence; and (3) impure materials that contain contaminants that emit copious gamma rays or neutrons that can mask the signal. The research topics we have selected focus on these broad physics issues rather than on any particular application or programmatic driver. The essential scientific challenge of this project is to solve these existing measurement limitations.

We have identified six specific scientific developments that will be necessary for us to reach our goal (see first figure): a sensitivity study, data fusion techniques to combine assay results from different detectors, advanced CdZnTe gamma-ray detector analysis, neutron detectors that have short die-away times and that use new types of scintillators and fibers, reduced-bias active interrogation using delayed-neutron reinterrogation, and neutron-induced gamma-ray signatures (spectral and spatial analysis of gamma rays induced by neutrons). The second figure shows some early results from our CdZnTe development work: a plot of the calculated and measured gamma-ray spectra for a CdZnTe detector.

Publications

Luke, P.N., et al., "Electrode Design for Coplanar-Grid Detectors," *IEEE Trans. Nucl. Sci.*, **44**, 713 (1996).

Prettyman, T.H., et al., "Advances in Nuclear Instrumentation for Safeguards" (Joint ESARDA-INMM Workshop on Science and Modern Technology for Safeguards, Arona, Italy, October 28–31, 1996).

Prettyman, T.H., et al., "Applications of PGNAA to Plutonium Surveillance" (to be published in *Trans. Am. Nucl. Soc.*).

Prettyman, T.H., et al., "Optimization of CdZnTe Detectors for Safeguards" (38th Annual INMM Meeting, Phoenix, AZ, July 20–24, 1997).

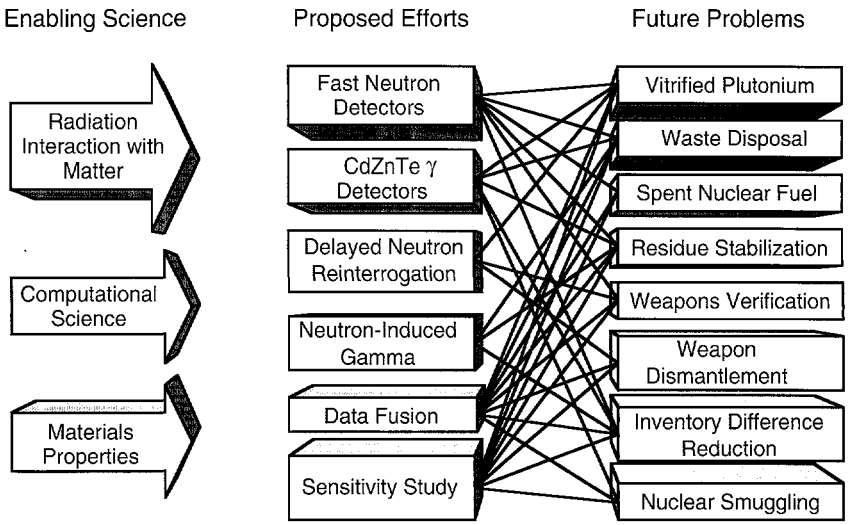
Prettyman, T.H., et al., "Physics-Based Generation of Gamma-Ray Response Functions for CdZnTe Detectors" (to be published in *J. Radioanal. Nucl. Chem.*).

Prettyman, T.H., et al., "Physics-Based Simulation of the Response of CdZnTe Radiation Detectors" (5th Scientific Symposium on Room-Temperature Semiconductor X-Ray, Gamma-Ray, and Neutron Detectors, Livermore, CA, March 11–12, 1997).

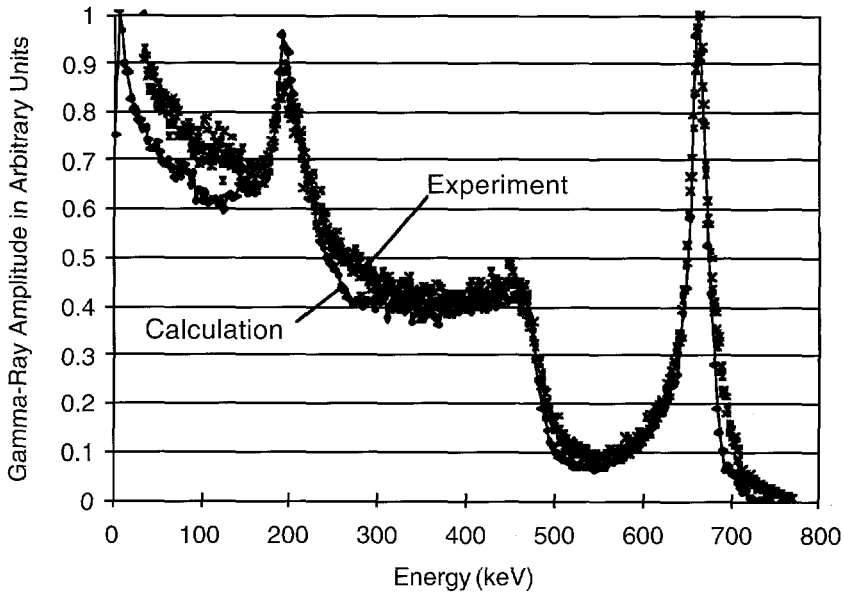
Prettyman, T.H., et al., "Radiation Imaging Technology for Nuclear Materials Safeguards" (38th Annual INMM Meeting, Phoenix, AZ, July 20–24, 1997).

Russo, P.A., et al., "A New, Room-Temperature Gamma-Ray Detector for Improved Assay of Plutonium" (38th Annual INMM Meeting, Phoenix, AZ, July 20–24, 1997).

Staples, P., et al., "Application of PGNAA to Safeguards and Surveillance" (38th Annual INMM Meeting, Phoenix, AZ, July 20–24, 1997).



Schematic of the relationship between the future measurement problems and the specific scientific developments we are proposing. Each solution depends on several, perhaps all, of the physics issues we have identified.



Comparison of measured and calculated spectra for the CdZnTe detector. We developed the calculated spectrum from a full physics model of the detector.

An Ultracold Neutron Facility at the Manuel Lujan Jr. Neutron Scattering Center

Thomas Bowles

Ultracold neutrons (UCNs) are neutrons whose wavelengths are long enough (typically greater than 500 Å) to undergo total internal reflection at all angles from the surfaces of a variety of materials. This characteristic suggests that UCNs can be confined in a bottle for more than 100 s, making a compact source of stored neutrons for use in fundamental physics measurements. One of our objectives is to simulate a UCN rotor source based on a Doppler-shifted, Bragg-scattered source. The simulations will demonstrate that we can produce UCNs with the liquid-hydrogen cold-neutron source at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC). Our second objective is to implement the first stage of such a source at MLNSC to demonstrate that we can reproduce our calculated UCN density.

During the first two years of this project, we carried out analytical and Monte Carlo design studies of a UCN source based on Doppler-shifted Bragg scattering from a moving crystal. We optimized the design using the Monte Carlo calculations and began the first stage of an experimental UCN source.

This year we completed the design, construction, and testing of a UCN source at MLNSC. The source performed reliably, and measurements of the UCN production rate agreed reasonably well with our Monte Carlo calculations. The source now provides the basis for additional study of polarized neutron beta decay in fundamental physics research and for possible applications in materials science.

Development of an Isotope Separator for Studies of Radioactive Species

Jerry Wilhelmy

The objective of this project is to develop and construct an isotope separator that can be dedicated to the separation of radioactive isotopes. Separated isotopes are an important product for many chemical, physical, and biological applications. At Los Alamos, we possess capabilities and facilities to handle highly radioactive species. With this project we are attempting to augment these capabilities to include the electromagnetic separation of the chemically isolated elements into their isotopic constituents. A contained, high-efficiency separator that can deliver milligram quantities of separated isotopes is required. The initial customers for these products include the medical isotope program, the science-based stockpile stewardship program, and energy research applications for astrophysical processes. Once the separator has been established and demonstrated, it is expected that there will be more users of this unique Laboratory capability.

After careful consideration of various deployment options, we decided to install the isotope separator in Wing 9 of the Chemistry and Materials Research (CMR) building at the Laboratory. Two adjacent, newly refurbished hot cells were identified, and detailed engineering design was begun to allow deployment in the confined space. The design objectives were to produce a robust separator that could be operated and serviced in a remote manner using hot cell manipulators.

Major milestones that have been accomplished this year include (1) separator design, (2) component procurement, (3) component assembly, (4) initial testing, and (5) demonstration of separated isotope collection. Remaining tasks include (1) debugging and optimization of beam transport; (2) construction, programming, testing, and debugging of the control monitor and alarm system; (3) ion source and chemical procedure development for the variety of elements to be separated; and (4) disassembly of the separator and transport to, and reassembly in, the CMR building.

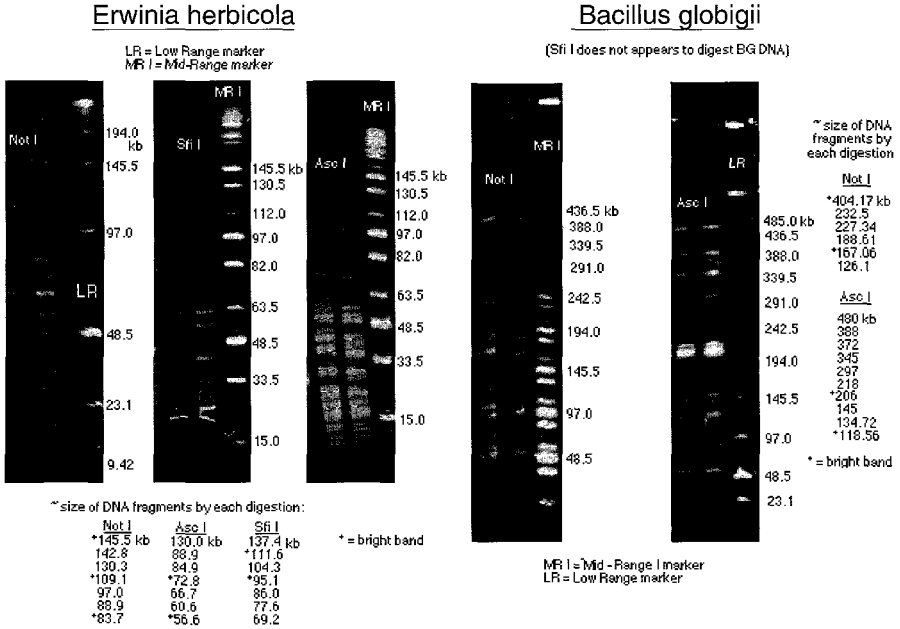
Universal Biological-Agent Point-Sensor Development

Gary Salzman

This project involves research on detection systems for biological-warfare microorganisms and on toxins and methodologies that will enable us to identify biological-warfare microorganisms. This year we tested the hypothesis that bacteria can be identified by performing a DNA restriction digest followed by a fragment-size analysis with gel electrophoresis or a DNA-fragment-sizing flow cytometer. We performed all of the chemistry within an agar plug to prevent further breakage of the fragments created by the restriction digests of *Bacillus globigii* and *Erwinia herbicola* (a simulant for *Y. pestis*). As part of this research, we developed methods for (1) inserting the bacteria into the agar plugs, (2) extracting the DNA in situ, (3) performing the restriction digests using 6- to 10-base cutters, and (4) electroeluting the DNA fragments from the agar plug into a pulsed-field gel-electrophoresis apparatus.

Examples of fragment-size analyses for *Bacillus globigii* and *Erwinia herbicola* are shown in the accompanying figure. The fragment-sizing flow cytometer gave good results in one instance for bacterial DNA fragments. However, the fragmentation of the DNA before and after the restriction digest causes a background of fragments that reduces the resolution on the instrument.

We redirected our work at midyear to focus on instrumentation for rapidly detecting biological aerosols on the battlefield. The first concept we developed was for a flow cytometer that measures scattered light and ultraviolet-excited fluorescence directly from single aerosol particles in a flow stream. The second concept uses fluorescence nephelometry to produce a biological smoke alarm.



Pulsed-field electrophoresis of the restriction fragments for *Erwinia herbicola* and *Bacillus globigii* generated by the restriction enzymes Asc I, Not I, and Sfi I. *Erwinia* and *Bacillus* are bacteria that are similar to those that may be detected during biological warfare. For each bacteria, the 8- and 10-base cutter restriction enzymes give distinct bands of DNA fragments that are recovered consistently. The bands, which indicate the sizes in kilobases (kb) of DNA fragments that will be differentiated on the flow cytometer, provide a unique pattern with quantitative data for each type of bacteria.

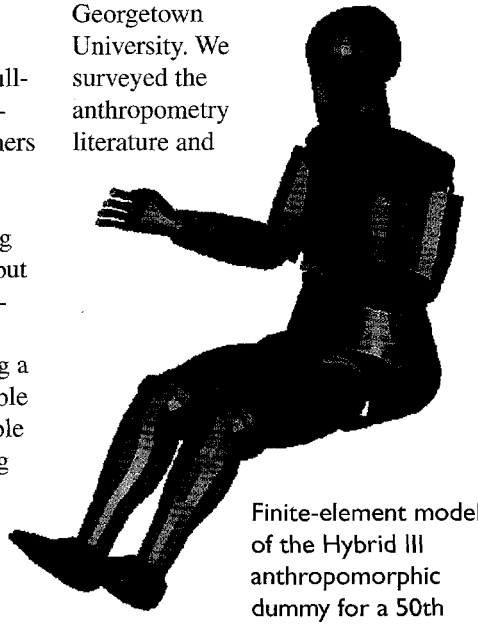
Deformable Human Body Model Development

William O. Wray

Crash victim simulator models developed to simulate the response of occupants to vehicular collisions initially were based entirely on rigid body mechanics. More recently, vehicle occupants have been represented by finite-element models of deformable anthropomorphic crash test dummies. This development was motivated by the fact that impact criteria that must be met by new automobile designs are based on full-scale crash tests with anthropomorphic dummies. In parallel, researchers have developed relatively realistic biomechanical models of critical human body components, including the head, neck, thorax and pelvis, but these models are generally inaccessible to automobile designers.

In this project, we are developing a realistic, fully accessible, deformable human body model that is applicable to automobile design by integrating existing anthropomorphic dummy models with existing, detailed biomechanical component models. This year we obtained a high-resolution, finite-element model of the Hybrid III, 50th percentile male anthropomorphic dummy (see the first figure) from the National Crash Analysis Center at Georgetown University. We surveyed the anthropometry literature and obtained data that represent the world population as 20 distinct regional/ethnic groups and provide size information for male and female members of the 5th, 50th, and 95th percentile groups within each region projected to the year 2000. Furthermore, we developed scaling procedures that allow us to scale the Hybrid III dummy model on a segment-by-segment basis to represent any size/sex group within any of the 20 regions worldwide.

We also obtained access to the Visible Human Dataset, provided by the National Library of Medicine, and downloaded the head and neck regions of the dataset. By scanning each cross-sectional image of the head and identifying boundaries between the various physiological structures, we produced a set of component geometries that may be assembled into a high-resolution finite-element model of the human head. The skull portion of the head model is presented in the second figure, and the brain component is shown in the third figure. Note that the brain model is sufficiently detailed to represent the convolutions of the cerebral cortex and the longitudinal cerebral fissure between the right and left hemispheres. The assembled head model will ultimately reside on top of the neck of an appropriately scaled Hybrid III dummy model when used in computational studies of blunt body trauma to the head.



Finite-element model of the Hybrid III anthropomorphic dummy for a 50th percentile male.



Skull component of the human head model based on the Visible Human Dataset.



Brain component of the human head model based on the Visible Human Dataset.

Advanced Spectroscopic and Imaging Diagnostics for Breast Cancer

Irving Bigio

The goals of this project were to accomplish proof of principle or to advance the state of the art for two advanced diagnostic techniques for breast cancer. The first technique uses elastic-scattering spectroscopy to provide real-time in vivo "biopsy" diagnosis with a newly designed optical-fiber needle (to replace surgical lumpectomies that are performed for diagnosis of breast lesions). The fiber-optic needle is designed to take interstitial cancer measurements with larger volume sensitivity than is possible with conventional fine-needle aspiration.

The second advanced diagnostic tool is time-resolved techniques for mammographic imaging with near-infrared (NIR) light (to replace x-ray mammography). NIR imaging uses nonionizing light and has the potential to provide images of the functional condition and structure of tissue. This technique is based on a high-speed, gated image intensifier coupled to a cooled charge-coupled-device (CCD) camera. This combination provides spatial sampling of NIR light used to transilluminate the tissue and captures information regarding photon path lengths.

This year we had many accomplishments in both areas of research. In our work on the technique using elastic-scattering spectroscopy, we designed and constructed optical needle probes. We took measurements in tissue "phantoms" to determine the optical geometry of the zone examined by the probe. In work with medical collaborators, optical measurements were made on freshly excised breast tissues (subsequent to mastectomies). We then compared these measurements with histopathology results from the same tissue sites, thus providing information about the spectral signatures that correlate best with the state of the tissue.

In our work on the NIR imaging technique, we designed and constructed an ultrafast, gated intensifier. When combined with a cooled CCD camera, this intensifier will enable much better resolution in NIR tomographic imaging. The gating times measured were under 100 ps and can be further reduced. Separately, we demonstrated a variation of this technique by taking external measurements of blood oxygenation changes in different regions of the brain, related to changes in visual and motor stimulation. We made these measurements with continuous broadband light from 600 to 900 nm.

Publications

Hielscher, A.H., et al., "Influence of Particle Size and Concentration on the Diffuse Backscattering of Polarized Light from Tissue Phantoms and Biological Cell Suspensions," *Appl. Opt.* **36**, 125 (1997).

Mourant, J.R., et al., "Measuring Absorption Coefficients in Small Volumes of Highly Scattering Media," *Appl. Opt.* **36**, 5655 (1997).

Human Brain Mapping: Experimental and Computational Approaches

Charles Wood

The Human Brain Project (HBP), a joint program developed by a consortium of federal agencies, supports the integration of noninvasive brain-imaging techniques with advanced computer modeling, visualization, and database techniques. The purpose of our project is to combine the experimental and computational strengths of Los Alamos and its collaborators to contribute significantly to the goals of HBP. The experimental component of our work emphasizes the optimization of spatial and temporal resolution of functional brain imaging by combining (a) structural magnetic resonance imaging (MRI) measurements of brain anatomy; (b) functional MRI measurements of blood flow, oxygenation, and volume; and (c) magnetoencephalography measurements of neuronal population currents. The computational component emphasizes the development of a high-resolution, three-dimensional volumetric model of the brain based on anatomical MRI, in which structural and functional information from multiple forms of brain imaging can be integrated into a single computational framework for modeling, visualization, and representation in distributed-database form.

This year we developed and completed initial validation studies of a new approach to the electromagnetic inverse problem based on Bayesian probabilistic inference. Rather than calculating a single "best" solution according to an arbitrary criterion (the strategy employed in existing approaches), the Bayesian approach samples the entire distribution of solutions using Markov chain Monte Carlo (MCMC) techniques. Features that are highly probable across a large number of MCMC solutions represent likely regions of brain activity. The

Bayesian approach also provides a natural means for incorporating information from other functional imaging modalities such as positron emission tomography or functional MRI.

Publications

Aine, C.J., et al., "Retinotopy of Cingulate Cortex? An MEG Study," *Neuroimage* **5**, 152 (1997).

George, J.S., et al., "Cingulate Sources Observed During Photosensitive Epileptic Discharges, Observed with Magneto-encephalography," *Neuroimage* **5**, 319 (1997).

Ranken, D., and J.S. George, "MRIVIEW: A Software Tool for Integrating Brain Structural and Functional Information," *Neuroimage* **5**, 388 (1997).

Schmidt, D.M., et al., "Bayesian Inference Applied to the Electromagnetic Inverse Problem" (submitted to *Hum. Brain Mapp.*).

Schmidt, D.M., et al., "Estimating Active Regions of Variable Extent from MEG Data," *Neuroimage* **5**, 433 (1997).

Carbon-Based Prosthetic Devices

David Devlin

The technical goal of this project is to demonstrate the feasibility of using carbon-carbon composites as prosthetic devices such as artificial hips, knees, and finger joints. Specifically, we are investigating the application of these materials to the metacarpophalangeal joint of the finger. We are carefully evaluating the mechanical properties of these composites and the relation of these properties to processing.

Carbon materials are known to be biocompatible and have been used for the past 25 years in the fabrication of artificial heart valves. Experience with heart valves demonstrates good wear properties for these materials. Because carbon composites can be fabricated with a stiffness similar to that of bone, they can prevent bone resorption around a stem. The porous structure of carbon composites has the added advantage of providing pores for bone ingrowth, thereby eliminating the need for a cement fixation. These improvements are expected to increase the performance and lifetime of implants.

In the past year we investigated the use of three-dimensional, fiber-reinforced materials as stem and head materials. Our results suggest that these materials have adequate strength to meet the requirements for the articulating head. We have developed a new material consisting of a unidirectional carbon-fiber-composite core with an outer carbon-composite braid. This material has a core strength higher than that of commercially pure titanium, and the outer braid provides a convenient way to introduce porosity. We have also demonstrated a method of joining the new stem to the head. Samples of these materials are being tested at Tulane University for biocompatibility. We anticipate that by the end of 1997, we will have the initial biocompatibility results, wear results on diamondlike-carbon-coated and noncoated composites, and data on the mechanical properties of our preferred material.

Publications

Devlin, D., et al., "Carbon Based Prosthetic Devices," in *Advances in Bioengineering (BED 36): Proceedings of the 1997 ASME International Mechanical Engineering Congress*, B. Simon, Ed. (American Society of Mechanical Engineers, New York, 1997), p. 265.

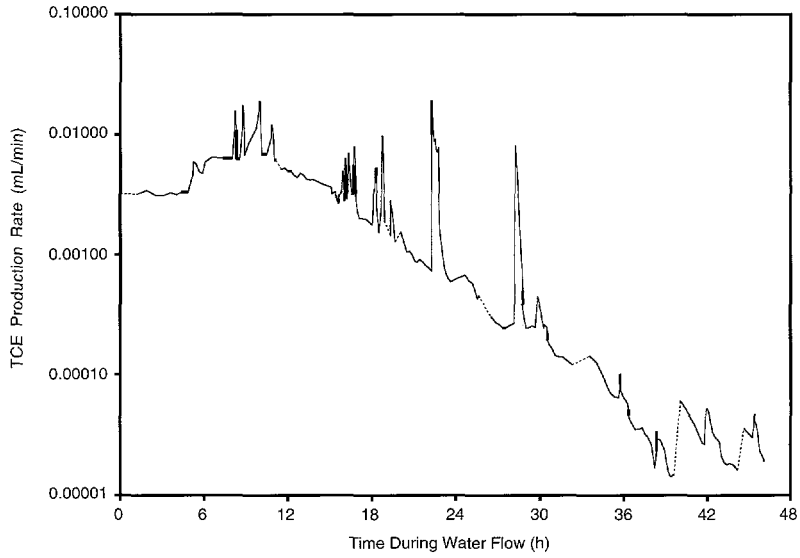
Directed-Energy Methods for Enhanced Transport of Subsurface Organics

Peter M. Roberts

The objective of this project was to evaluate a novel concept focused on using low-frequency seismic waves to enhance the transport and extraction of dense, nonaqueous-phase liquids (DNAPLs) trapped in groundwater aquifers. Extraction of DNAPLs is a major problem in environmental remediation, and existing field techniques have limited effectiveness.

We have performed laboratory experiments to investigate the use of low-frequency (10–100 Hz) seismic stimulation for enhanced transport of trichloroethylene (TCE) during water flow in a sand core with a 1-in. diameter and a 12-in. length. In these experiments, we packed clean 20- to 40-mesh sand into a rubber sleeve and confined the sand at 120-psi radial and 60-psi axial pressures. We then saturated the core with water and found the pore volume to be 65 mL. Next, we pumped water through the sample at constant, pulse-free flow rates after injecting a 7-mL aliquot of TCE into the core. The TCE concentration of the effluent tailed off from an initial value of 900 ppm to less than 10 ppm over 46 h. We applied axial pressure cycling at 25–100 Hz by direct mechanical coupling of a magnetostrictive actuator to the core. During the stimulation, pure-phase TCE droplets were produced, and the net concentration jumped to values well above the saturation limit of 1100 ppm (see figure). TCE production remained high during the stimulation and dropped back to low values after the stimulation. These treatments produced no sand fines or changes in the sample permeability.

Our results indicate that acoustic stimulation could increase both the pure-phase mobility and the dissolution rates of TCE trapped in groundwater aquifers. We have identified potential field sites and existing surface and downhole acoustic devices that we could use to test this new remediation method in a field demonstration.

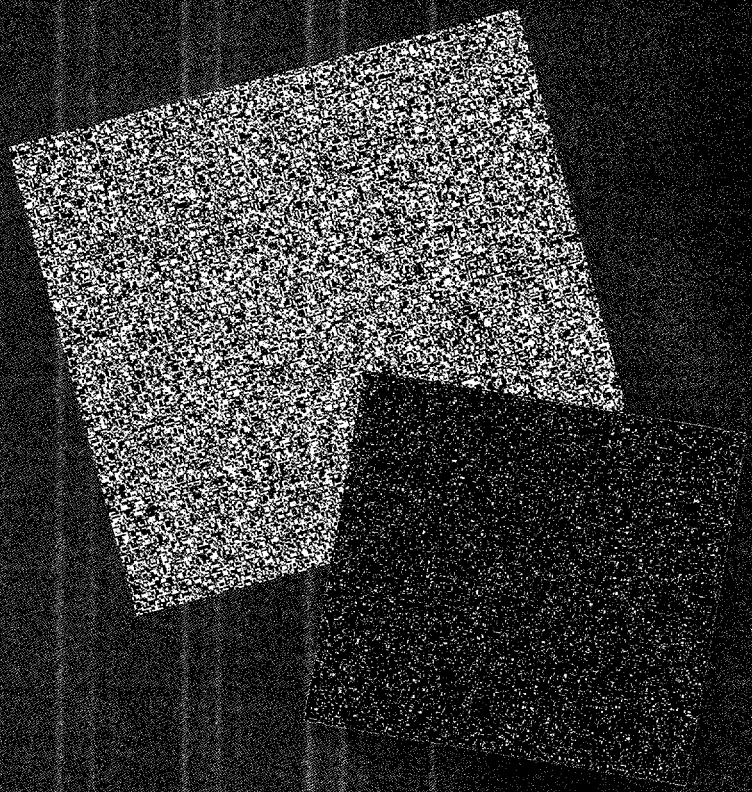


TCE production rate plotted vs time during water flow. The dashed lines indicate stagnant periods of one to several days between separate flow runs. All of the production peaks during the flow were caused by stimulation. The large peaks during the first 30 h correspond to production of pure-phase TCE, and the smaller peaks during the last 16 h correspond to enhanced dissolution that occurred because the amount of TCE remaining was too low (8% of the injected volume) to produce the pure phase.


Cl
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under N2
C6H6
THF/NaClO4
PPh2
Rh
P
H
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Individual Projects



Optimum Design of Ultrahigh-Strength Nanolayered Composites

Huijou Harriet Kung

The objective of this research is to elucidate the relationship between macroscopic mechanical behavior and microscopic strengthening mechanisms in nanolayer composites. Specifically, the atomistic mechanisms involving the movement, storage, and recovery of dislocations in the crystalline phases and the interfaces between them need to be clarified and quantified. Through this understanding we seek to obtain general scaling laws that describe the transitions between mechanisms in terms of the modulation wavelength and interfacial structure and bonding. The major goal of this study is to develop a comprehensive theory that will relate changes in structure to changes in mechanical behavior.

This year we have successfully synthesized epitaxial copper-nickel multilayered films on NaCl and copper single-crystal substrates by using the electron-beam evaporation technique. By employing transmission electron microscopy (TEM) techniques, we have characterized the multilayers to be single crystalline, as shown in the figure. The atomic-scale interfacial structure and the misfit dislocation structure have been characterized by high-resolution TEM. Preliminary results on deformation structure suggest that the interfaces play a major role in dislocation movement and slip propagation in multilayers.

In our modeling work, we have written a workstation-based molecular dynamics computer program capable of the million-atom simulations necessary for this project. We have also constructed a copper-nickel interatomic potential based on the embedded atom method. We have begun simulations of the interface misfit dislocations, showing, for example, that while the extra planes lie on the nickel side, the core spreads as much as possible into the copper, which has a lower modulus.

Publications

Fayeulle, S., et al., "Ion Beam Induced Modifications in DC Sputtered TiN/B-C-N Multilayers," *Nucl. Instrum. Methods B* **127**, 198 (1997).

Fayeulle, S., et al., "Thermal and Ion Irradiation Stability of DC Sputtered TiN/B-C-N," *Appl. Phys. Lett.* **70**, 1098 (1997).

Kern, K.T., et al., "Boron and Nitrogen Implantation of Steels," *Nucl. Instrum. Methods B* **127**, 972 (1997).

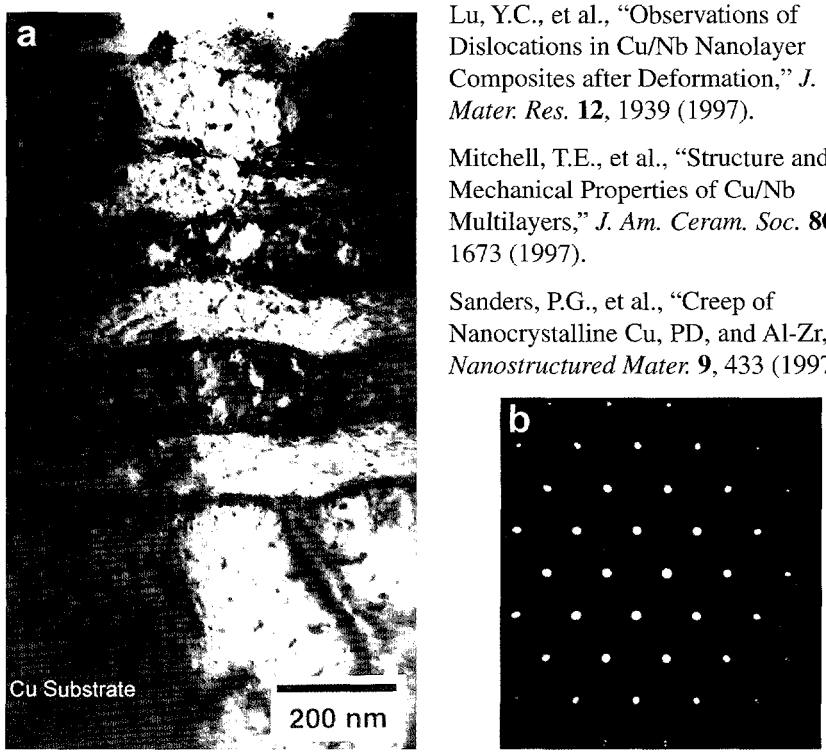
Kung, H., et al., "Observation of b.c.c. Cu in Cu/Nb Nanolayered Composites" (to be published in *Appl. Phys. Lett.*).

Lu, Y.C., et al., "Effect of 690 keV Xe Ion Irradiation on the Microstructure of Amorphous $MoSi_2$/SiC Nanolayer Composites," *Nucl. Instrum. Methods B* **127**, 648 (1997).

Lu, Y.C., et al., "Observations of Dislocations in Cu/Nb Nanolayer Composites after Deformation," *J. Mater. Res.* **12**, 1939 (1997).

Mitchell, T.E., et al., "Structure and Mechanical Properties of Cu/Nb Multilayers," *J. Am. Ceram. Soc.* **80**, 1673 (1997).

Sanders, P.G., et al., "Creep of Nanocrystalline Cu, PD, and Al-Zr," *Nanostructured Mater.* **9**, 433 (1997).



(a) Cross-sectional TEM bright field image of a copper-nickel multilayer grown on a copper single-crystal substrate. The corresponding electron diffraction pattern in (b) shows the good epitaxial relationship between the multilayer and the substrate.

Development of a Prototype Optical Refrigerator

Richard Epstein

We have carried out a range of tasks directed toward the construction and testing of a proof-of-principle optical refrigerator. We procured and tested new cooling elements that are at the heart of an optical refrigerator. The cooling element absorbs pump radiation and then fluoresces with nearly unity quantum efficiency. We constructed and tested a cooling chamber with low-thermal-emissivity walls that reduce the parasitic heating.

We tested cooling elements composed of a fluoride glass doped with ytterbium ions. Laser light enters the sample through a small hole in one of the mirrors and remains trapped until the ytterbium ions absorb it. We tested and characterized one mirrored cooling element and found measurable cooling. Using these results, we designed an improved cooling element.

Publications

Fajardo, J.C., et al., "Electrochemical Purification of Heavy Metal Fluoride Glasses for Laser-Induced Fluorescent Cooling Applications," *J. Non-Cryst. Solids* **213**, 95 (1997).

Mungan, C.E., et al.., "Internal Laser Cooling of Yb-Doped Glass Measured between 100 and 300 K," *Appl. Phys. Lett.* **71B**, 1458 (1997).

Mungan, C.E., et al., "Laser Cooling of a Solid by 16 K Starting from Room Temperature," *Phys. Rev. Lett.* **78**, 1030 (1997).

Mungan, C.E., et al., "Spectroscopic Determination of the Expected Optical Cooling of Ytterbium-Doped Glass," *Mat. Sci. Forum* **239**, 501 (1997).

New Deposition Processes for the Growth of Oxide and Nitride Thin Films

Kevin M. Hubbard

Organometallic chemical vapor deposition (OMCVD) has been slow in gaining acceptance for coatings applications despite the possibilities for significant advantages over metal-halide-based CVD processes. The major underlying limitation is the lack of understanding of the chemistry important to the deposition process. Control of the fundamental chemistry of a deposition process is the key to advancement of the OMCVD technique. Recently, the efficient transamination of homoleptic metal amides, $M(NR_2)_x$, with ammonia to yield highly reactive metal amido/imido intermediates has been used to develop low-temperature, high-growth-rate processes for the deposition of high-purity metal nitrides and oxides.

The general goal of our project is to understand the details of the chemistry involved in the deposition process of nitrides and oxides from homoleptic metal amido precursors. This understanding will lead to new routes for the synthesis of high-quality metal nitrides and oxides.

This year we studied the deposition of GaN thin films from the precursor hexakis(dimethylamido)digallium, or $Ga_2(NMe_2)_6$. As part of the effort, we generated a matrix of film properties as a function of deposition conditions. We also developed and performed synthesis of the AlN precursor compound hexakis(dimethylamido)-dialuminum, or $Al_2(NMe_2)_6$ and studied the deposition of AlN thin films from that precursor.

Intrinsic Fine-Scale Structure in Complex Electronic Materials: Beyond Global Crystallographic Analysis

Albert Migliori

The principal aim of this project is to provide a robust analysis tool for (1) neutron- and x-ray-derived pair-distribution-function data and (2) calibration against macroscopic properties (elastic constants, thermodynamics). In order to accomplish our goal, we studied single crystals of the colossal magnetoresistance material $La_{.83}Sr_{.17}MnO_3$ by using resonant ultrasound spectroscopy. Fine structure was found to be present and strongly connected to the magneto-transport in this compound. We also made neutron measurements on this system to look at the fundamental distortions at a unit-cell level and used resonant ultrasound to estimate length scales.

We conducted theoretical studies of fine-scale pattern formation, including work on stripes and related unit-cell-level patterns. In addition, we also studied the effect of certain distortions that must occur in a system containing very fine structures. This work will be coupled to upcoming neutron and ultrasound studies of nickel-rich NiAl, a martensite system that we are in the process of measuring.

Publications

Bussmann-Holder, A., and A.R. Bishop, "Competing Length Scales in Anharmonic Lattices: Domains, Stripes and Discommensurations" (to be published in *Phys. Rev. B*).

Darling, T.W., et al., "Measurement of the Elastic Tensor of a Single Crystal of $La_{.83}Sr_{.17}MnO_3$ and Its Response to Magnetic Fields" (submitted to *Phys. Rev. B*).

Dimitrov, D., et al., "The Crystallography of a Two-Dimensional Binary Crystal with Local Distortion," *Phys. Rev. B* **56**, 2969 (1997).

Louca, D., et al., "Local Jahn-Teller Distortion in $La_{1-x}Sr_xMnO_3$ Observed by Pulsed Neutron Diffraction" (submitted to *Phys. Rev. B*).

Millis, A.J., et al., "Quantifying Strain Dependence in 'Colossal' Magnetoresistance Manganites," *J. Appl. Phys.* **83**, 1588 (1998).

Saxena, A., et al., "Hierarchical Pattern Formation in Elastic Materials," *Physica A* **239**, 18 (1997).

Understanding Interfacial Charge Transport in Organic Electronic Materials: The Key to a Revolutionary New Electronics Technology

Darryl Smith

Our primary aim is to obtain a basic scientific understanding of electrical transport processes at interfaces that contain an organic electronic material as a constituent. These interfaces are the essential active elements of every organic electronic and electrooptic device. Because of their processing advantages, the tunability of their electronic properties, and their flexibility in both materials and device design, organic electronic materials and devices are poised to revolutionize major technological areas, such as information display and optical communication. However, the lack of fundamental understanding of the properties of these materials and devices limits the progress of the numerous design options available at both the molecular and mesoscopic levels. Because interfaces are the essential active elements in every organic electronic device, understanding interfacial charge transport is the key to realizing the potential of these materials and devices.

In the second year of the project, we developed a theory to describe the current-voltage (*I-V*) characteristics of organic diodes. This theory predicts how these *I-V* characteristics depend on the Schottky energy barriers of the metal contacts used to fabricate the devices. We made a series of electrical measurements on devices fabricated from the conjugated polymer MEH-PPV (a derivative of poly[phenylene vinylene]) and a series of metals. The results of these measurements were successfully interpreted in terms of our theoretical model.

Publications

Campbell, I.H., et al., "Measuring Internal Fields in Organic Electronic Devices Using Electroabsorption Spectroscopy," *Polym. Adv. Technol.* **8**, 417 (1997).

Cornil, J., et al., "Photoluminescence Spectra of Oligo (Paraphenylene-vinylenes)s: A Joint Theoretical and Experimental Characterization" (to be published in *Chem. Phys. Lett.*).

Davids, P.S., et al., "Device Model for Single Carrier Organic Diodes" (to be published in *J. Appl. Phys*).

Lee, E.Y., et al., "Monte-Carlo Simulations of BEEM Imaging and Spectroscopy of Buried Mesoscopic Structures," *Phys. Rev.* **55**, 16033 (1997).

Experimental and Theoretical Investigation of Fracture and Deformation of a Revolutionary High-Temperature Gamma-TiAl Alloy

George Gray III and Zhe Jin

The objective of this study is to seek a clear understanding of the deformation mechanisms leading to the low ductility in gamma-TiAl and to provide a scientifically driven, physically based rationale for how to pursue increased tensile ductility in gamma-TiAl.

In our experimental work, we successfully grew polysynthetically twinned TiAl. We found that the deformation of gamma-TiAl alloys was dominated by 1/2[1 1 0] ordinary dislocation slip and 1/6<1 1-2>{1 1 1} true twinning at high strain rates. We determined experimentally all six domain orientations in lamellar structures. We observed that individual domains in the lamellar structures responded differently to the external loading. Accordingly, we observed both interlamellar and translamellar microcracks in gamma-TiAl. However, with increasing compression strain, both microcracks propagated little else than new nucleate microcracks (see accompanying tables and figures).

We found that the formation of microcracks under compression was caused by the strain mismatches between domains in lamellar structures. We determined that the final fracture of the samples was caused by propagation of one or two macrocracks that were formed after certain large amounts of strain. We also found that the ductile-brittle transition temperature increases with increasing strain rate.

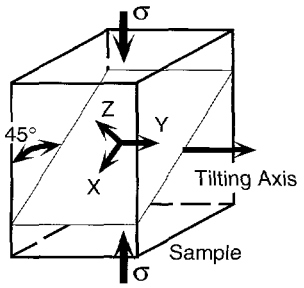
A series of three-dimensional molecular dynamics simulations of microcrack propagation showed that crack-tip blunting by the emission of Shockley partials was insufficient to effectively block crack propagation. Our simulation of dislocations and

Deformation modes in individual domains observed in 45°<–3 2 1> oriented polysynthetically twinned TiAl crystals deformed at high strain rates.

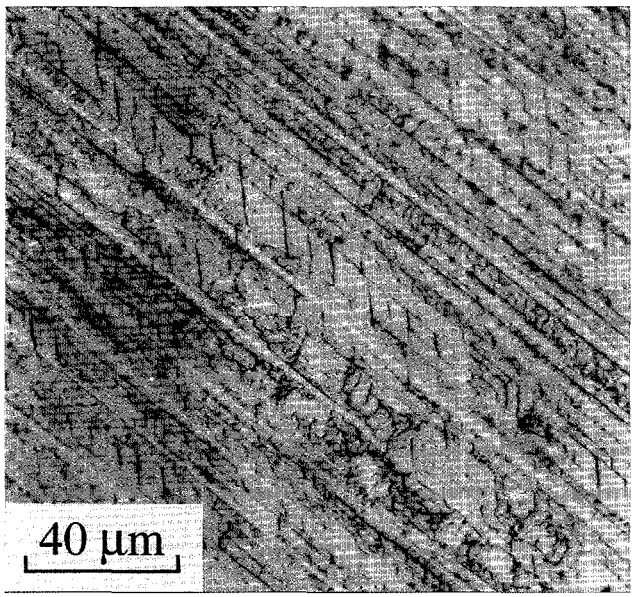
Domain	Dominant Deformation Mode	Complementary Deformation Mode
[I]	No Deformation	
[II]	1/2[1 -1 0]	1/2[1 1 0], 1/6[1 -1 -2](1 -1 1), 1/6[-1 1 -2](-1 1 1)
[III]	1/6[1 1 -2](1 1 1)	1/2[1 1 0], [1 0 -1]
[IV]	1/6[1 1 -2](1 1 1)	1/2[1 -1 0]
[V]	1/2[-1 1 0]	1/2[-1 -1 0], [0 1 1], 1/6[-1 1 -2](-1 1 1) 1/6[1-1-2](1 -1 1)
[VI]	1/2[-1 -1 0]	1/2[-1 -1 0], [0 -1 1]

Strain mismatches along Y-direction between domains for a unit shear strain in X-direction by dominant deformation modes.

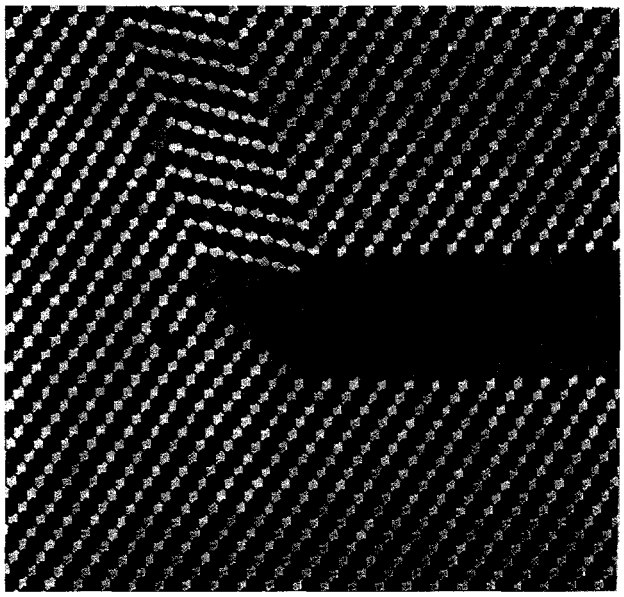
Domain	[I]	[II]	[III]	[IV]	[V]	[VI]
[I]	0					
[II]	0.192	0				
[III]	0.866	0.674	0			
[IV]	0.346	0.539	1.213	0		
[V]	0.192	0	1.059	0.539	0	
[VI]	1.155	1.347	2.021	0.808	1.347	0



lamellar interface interactions showed that lamellar interfaces were an effective barrier for translamellar deformation.



Interlamellar and translamellar microcracks observed in a soft-mode-oriented polysynthetically twinned TiAl crystal deformed at 2,500/s and 25°C to 10% strain. The compression-loading direction is vertical in the figure.



The emission of 1/6[-1 1 2] Shockley partials at a crack tip with [1 1 0] crack front in (-1 1 -1) crack plane. The crack was observed to continuously propagate, although many Shockley partials were emitted at the crack tip.

Publications

Jin, Z., and G.T. Gray III, "On Deformation Twin and Twin-Related Lamellae in TiAl" (to be published in *J. Mater. Sci.*).

Jin, Z., and G.T. Gray III, "Experimental Determination of Domain Orientation and Domain Orientation Relationships across Lamellar Interfaces in Polysynthetically Twinned (PST) TiAl Crystals," *Mater. Sci. Eng., A* **231**, 62 (1997).

Jin, Z., et al., "Deformation of Polysynthetically Twinned (PST) TiAl Crystals at High Strain Rates and High Temperatures," in *High-Temperature Ordered Intermetallic Alloys VII*, C.C. Koch, et al., Eds. (MRS, Pittsburgh, PA, 1997), p. 189.

Jin, Z., et al., "Deformation of Polysynthetically Twinned (PST) TiAl Crystals at High Strain Rates and High Temperatures" (submitted to *Philos. Mag. A*).

Jin, Z., et al., "Mechanical Behavior of a Fine-Grained Duplex Gamma-TiAl Alloy" (submitted to *Metall. Mater. Trans. A*).

Jin, Z., et al., "Mechanical Behavior and Microcrack Formation in Gamma-TiAl Alloys as a Function of Strain Rate and Temperature," in *Structural Intermetallics 1997*, M.V. Nathal, R. Darolia, et al., Eds. (TMS, Warrendale, PA, 1997), p. 225.

Jin, Z., et al., "Mechanical Response and Microcrack Formation in a Fine-Grained Duplex TiAl at Different Strain Rates and Temperatures," *Deformation and Fracture of Ordered Intermetallic Materials III*, W.O. Soboyejo, T.S. Srivatsan, and H.L. Fraser, Eds. (TMS, Warrendale, PA, 1996), p. 101.

Jin, Z., et al., "Mechanical Twinning in a 45°<-3 2 1> Oriented Polysynthetically Twinned (PST) TiAl Crystal at High Strain Rate and High Temperature" (to be published in *Philos. Mag. A*).

Jin, Z., et al., "Texture Evolution during Alpha-Forging of Gamma-TiAl Alloys" (to be published in *Mater. Sci. Eng.*).

Characterization and Manipulation of Broken-Symmetry Materials at Phase Boundaries

Robert Donohoe

The low-dimensional, halide-bridged metal chains (MX) provide an idealized testing ground for examining the interactions between charge, lattice, and spin energies, and their combined effects upon the ground and defect states. The two primary objectives of this project are a detailed analysis of the effects of applied magnetic fields to weak charge-density-wave (CDW) MX materials and a careful examination of the spatial and kinetic properties of magnetic defect centers in MX chains.

Last year we carefully analyzed the electron paramagnetic resonance (EPR) data from photo-induced defects in PtCl, a strong CDW material, with respect to temperature, power, modulation frequency, and orientation, and as a function of isotopic enrichment of platinum and chlorine centers. The dependence of the data on the chlorine isotope reveals the spectrum is fundamentally composed of a superposition of superhyperfine features in individual chlorines. Modeling of the data demonstrates that neutral excitons are the primary source of photo-induced EPR signals.

In addition to the exciton signal, we observed two sample-dependent signals, one of which originates from a triplet (biexciton) and the other of which appears at elevated temperatures and is unusually stable. Although we observed activation and recombination of the excitons, we could not detect any motional broadening of the exciton signal, which is consistent with the strong lattice pinning expected for defects in PtCl.

We also observed a new dynamical process at low temperatures that may result from charge-dissociated tunneling of spin between metal sites. We observed an extraordinary dependence of this dynamic process on the platinum isotope.

Publications

Wei, X., et al., "ESR Study of Photoinduced Defects in Isotopically Enriched Quasi-One-Dimensional Chlorine-Bridged Platinum Complexes," *Phys. Rev. B* **56**, 8257 (1997).

Wei, X., et al., "Quantum Spin Fluctuations in Quasi-One-Dimensional Chlorine-Bridged Platinum Complexes" (to be published in *SPIE Proceedings*).

Experimental Determination of Statistical Parameters for Improving a Micromechanical Model of Ductile Fracture

Anna Zurek and Rich Thissell

The objectives of this project are the development and application of experimental techniques in order to determine quantitative microstructural descriptors of the ductile fracture process. The quantitative descriptors will further the development of a micromechanical model of ductile fracture.

A wide quantitative chasm exists in the methodology currently used in studying dynamic ductile fracture. A fine level of microstructural detail is built into numerical micromechanical models of the kinetics of ductile fracture, but the overall experimentally quantifiable parameters (such as impact velocity, shock pressure, and spalled surface velocity) are macroscopic. Sample recovery techniques permit posttest microscopy. Current microscopic techniques enable qualitative validation of the types of micromechanical processes that the model should emulate. Our work will bridge this gap and permit detailed quantification of the micromechanical features and processes of ductile fracture. As a result, our work will propel micromechanical model development to a far greater level of physical accuracy.

This year, we concentrated on defining and implementing quantitative descriptors and appropriate theoretical constructs for the micromechanical model. We implemented image analysis procedures using commercial and custom software. We developed procedures to combine data obtained from optical micrographs and optical profilometry. The descriptors developed in the past year include aspect ratio, void size, porosity, and void-clustering information. Finally, we implemented damage visualization and information portability by writing ACIS solid-model files of voids.

Publications

Thissell, W.R., et al., "Quantification of Damage Evolution for a Micromechanical Model of Ductile Fracture in Spallation of Copper," in *21st International Symposium on Shock Waves* (available online, *http://aerodec.anu.edu.au/~sue/start.htm*, 1997).

A Molecular Architectural Approach to Novel Electrooptical Materials

DeQuan Li

Electrooptical thin-film materials are important for optical technologies, including signal processing, telecommunication, and data storage. The goal of this project is to use innovative molecular self-assembly techniques to fabricate multilayer thin films that will directly benefit electrooptical devices. Improved thin films in such devices would lead to faster and cheaper routing of information along fiber-optic networks and would enable users to be interconnected economically to high-quality telecommunication systems.

We have been exploring many practical pathways to multilayer thin-film materials. In particular, we have been successful at building multilayer structures with alternating donors and acceptors. These structures include conjugated polymers as electron donors and buckyballs (C_{60}) as acceptors. We have fabricated thin films of 0.1-μm thickness with control at the molecular level (1 nm). Another approach is the formation of spontaneously self-assembled multilayers using a complementary hydrogen-bonding motif. From this approach we have obtained polar and highly ordered structures, and we have observed nonlinear optical effects (d_{33} = 3 pm/V).

We also modeled linear molecules and calixarenes using quantum chemical techniques (e.g., the semiempirical codes PM3 and MNDO) implemented in the molecular-orbital code MOPAC93. We then studied two finite chains, $C_{24}H_{26}$ and $C_{40}H_{42}$, and created a breather—a time-periodic, localized, persistent nonlinear excitation of the system with the symmetry of the ground state—by initially localizing energy in the molecular dynamics simulations. We found that the nonlinear optical susceptibilities of the excited chains were significantly enhanced relative to the optical susceptibilities of their ground states. This study provided the first evidence that breather formation would enhance optical nonlinearity.

Publications

Brazovskii, S., et al., "Stability of Bipolarons in Conjugated Polymers" (to be published in *Opt. Mater.*).

Li, D., "Self-Assembled, Molecular and Macromolecular p-Conjugated Optical Materials and Their Nonlinear Optical Properties," *Synth. Met.* **86**, 1849 (1996).

Li, D., "Supramolecular Architectures for Nonlinear Optical Materials" (Second International Workshop on Molecular and Electronic Nanostructures, Hatoyama, Japan, November 13–14, 1997).

Li, D., et al., "A Molecular Architectural Approach to Nonlinear Optical Materials," *Opt. Soc. Am. Tech. Digest Ser.* **11**, 409 (1996).

Li, D., et al., "Molecular and Macromolecular Multilayer Self-Assemblies as Electronic and Optical Materials" (American Chemical Society National Meeting, Las Vegas, NV, September 7–11, 1997).

Li, D., et al., "Multilayer Self-Assemblies as Electronic and Optical Materials" (Materials Research Society 1997 Fall Meeting, Boston, MA, December 1–5, 1997).

Luett, M., et al., "X-Ray Reflectivity Study of Self-Assembled Multilayers of Macrocycles and Macromolecules" (to be published in *J. Phys. Chem.*).

Shi, X., and D. Li , "Self-Assembled Multilayers and Photoluminescence Properties of a New Water-Soluble Poly(*para*-Phenylene)" (Materials Research Society 1997 Fall Meeting, Boston, MA, December 1–5, 1997).

Smilowitz, L., et al., "Imaging Nanometer-Thick Patterned Self-Assembled Monolayers via Second-Harmonic Generation Microscopy," *J. Appl. Phys.* **81**, 2051 (1997).

Yang, X., et al., "A Molecular Architectural Approach to Self-Assembled Monolayers as NLO Materials," *Mat. Res. Soc. Symp. Proc.* **413**, 241 (1996).

Yu, Z.G., et al., "Excitons in Quasi–One-Dimensional Organics: Strong Correlation Approximation," *Phys. Rev. B* **56**, 3697 (1997).

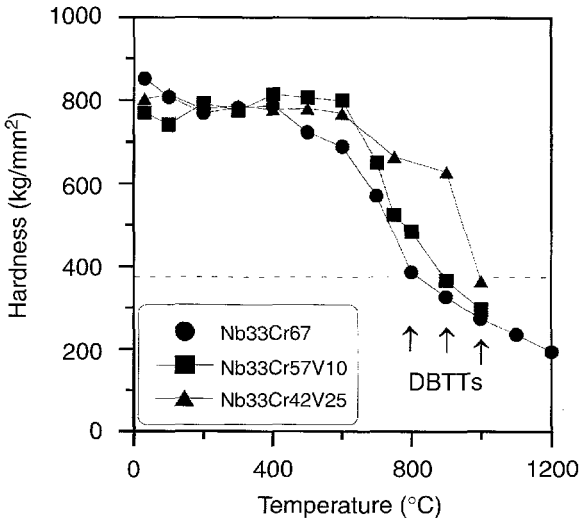
Theoretical and Experimental Investigation on the Low-Temperature Properties of the $NbCr_2$ Laves Phase

Dan J. Thoma

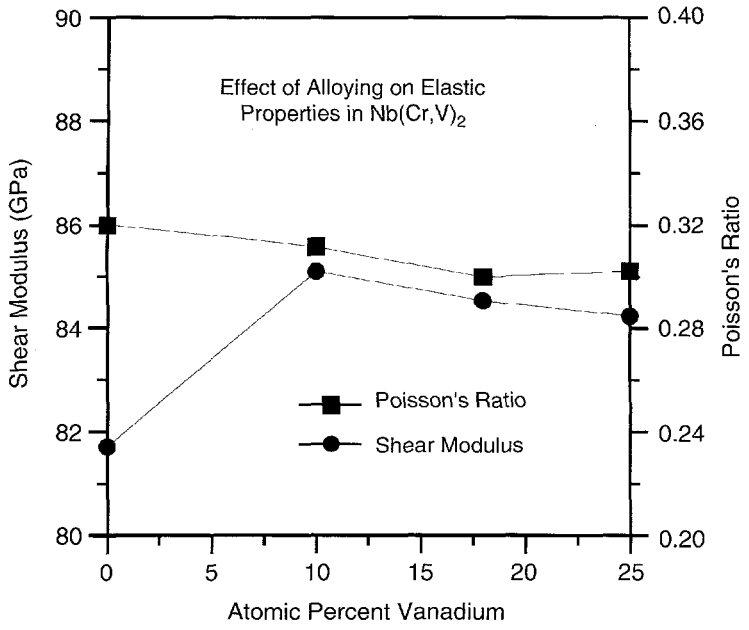
Laves phases are the most abundant yet least utilized intermetallic phase. Unfortunately, this abundance has not been exploited, largely because of their tendency for low-temperature brittleness. However, recent advances have been made in improving Laves-phase toughness and ductility through understanding (1) the defect structure and deformation mechanisms in Laves phases; (2) the electronic and geometric contributions to phase stability and alloying behavior; and (3) dual-phase (Laves/body-centered cubic) structures. Our research pursues a scientific methodology—from atomic to bulk scale—that improves the low-temperature ductility and toughness of Laves phases.

We had a number of major accomplishments this year. Work is currently under way on synthesizing and growing single crystals of $NbCr_2$ and $Nb(Cr,V)_2$ as well as directional solidification of $NbCr_2$/Nb and $NbCr_2$/Cr eutectic composites. We used electron microscopy to characterize defects in HfV_2 + Nb, $NbCr_2$, and $HfCo_2$ Laves phases and determined the phase diagrams and alloying behavior of C15 $Nb(Cr,V)_2$ Laves phases (C15 refers to the crystal structure). In addition, we identified point-defect structures in $NbCr_2$ and $Nb(Cr,V)_2$ Laves phases by high-resolution synchrotron x-ray powder diffraction and Rietveld structure refinement, and we conducted mechanical tests of $NbCr_2$ and $Nb(Cr,V)_2$ Laves phases (see plot of hardness vs temperature).

Our work also included first-principles total-energy and electronic-structure calculations of $HfCo_2$ and $TiCr_2$ Laves phases; elastic-property measurement for $Nb(Cr,V)_2$ and $HfCo_2$ Laves phases by resonant ultrasound spectroscopy (see plot of shear modulus vs atomic percent of vanadium); supercell calculations for vacancy and antisite point-defect energies in C15 $NbCr_2$; and elastic-constants calculations for C15 HfV_2. These calculations for C15 HfV_2 showed a tetragonal instability that agrees with the results from total-energy calculations, which found the C14 structure stable over the C15 structure.



Vickers hardness plotted vs temperature for alloys with vanadium substituting for chromium in monolithic C15 Laves phases. The ductile-to-brittle transition temperature (DBTT) increases with vanadium content, consistent with theories for synchro-shear (a cooperative movement of multiple atom planes during deformation) that we developed in this program.



Elastic properties, determined from resonant ultrasound spectroscopy, plotted vs vanadium content for alloys with vanadium substituted for chromium in monolithic C15 Laves phases. The values illustrate stiffening (increased shear moduli) and increased directional bonding (lower Poisson ratios) with vanadium additions.

Publications

Chen, K.C., et al., "Assessment of the Compositional Influences on the Toughness of $TiCr_2$-Base Laves Phase Alloys," *Mater. Res. Soc. Symp. Proc.* **460**, 695 (1997).

Chen, K.C., et al., "Factors Affecting the Room-Temperature Mechanical Properties of $TiCr_2$-Base Laves Phase Alloys" (to be published in *Mater. Sci. Eng. A*).

Chu, F., et al., "Phase Stability and Defect Structure of the C15 Laves Phase $Nb(Cr,V)_2$" (to be published in *Acta Mater.*).

Chu, F., et al., "Phase Stability and Defect Structure of the Laves Phases in the Hf-V-Nb System" (to be published in *Phil. Mag. A*).

Chu, F., et al., "Phase Stability of C15 MV_2 (M = Zr, Ta, or Hf): An Electronic Structure Investigation" (to be published in *Phil. Mag. B*).

Chu, F., et al., "Resonant Ultrasound Spectroscopy: Elastic Properties of Some Intermetallic Compounds," in *Nondestructive Evaluation (NDE) and Materials Properties (III)*, P.K. Liaw et al., Eds. (TMS, Warrendale, PA, in press).

Chu, F., et al., "Structural and Defect Analysis of V-Alloyed C15 $NbCr_2$ from High Resolution Synchrotron X-Ray Powder Diffraction" (to be published in *Phil. Mag. A*).

Kotula, P.G., et al., "ALCHEMI of $NbCr_2$/V C15 Laves Phase," in *Proceedings of Microscopy and Microanalysis 1996*, G. W. Bailey et al., Eds. (San Francisco Press, San Francisco, CA, 1996), p. 554.

Kotula, P.G., et al., "Orientation Relationships in the Nb-$NbCr_2$," in *Proceedings of Microscopy and Microanalysis 1997*, G. W. Bailey et al., Eds. (Springer-Verlag, New York, 1997), p. 707.

Kotula, P.G., et al., "Site Occupancies of Alloying Additions in C15-Structured Laves Phase Materials," *Mater. Res. Soc. Symp. Proc.* **460**, 617 (1997).

Ormeci, A.H., et al., "Elastic Constants of a Laves Phase Compound: C15 $NbCr_2$," *Mater. Res. Soc. Symp. Proc.* **460**, 623 (1997).

Ormeci, A.H., et al., "Total-Energy Study of Electronic Structure and Mechanical Behavior of C15 Laves Phase Compounds: $NbCr_2$ and HfV_2," *Phys. Rev. B* **54**, 12753 (1996).

Perepezko, J.H., et al., "Phase Stability in Processing of High-Temperature Intermetallic Alloys," *Mater. Res. Soc. Symp. Proc.* **460**, 3 (1997).

Thoma, D.J., "Comparison of $NbCr_2$ and HfV_2 Laves Phases," *Mater. Res. Soc. Symp. Proc.* **460**, 689 (1997).

Thoma, D.J., et al., "Elastic and Mechanical Properties of C15 Laves Phase $Nb(Cr,V)_2$" (to be published in *Mater. Sci. Eng. A*).

The Electrochemical Properties of Bundles of Single-Walled Nanotubes

Thomas Zawodzinski

Recently, the Smalley group at Rice University announced that they have produced long bundles of single-walled fullerene nanotubes, which they call "ropes." The bundles form spontaneously when certain metals are added to the mix commonly used to synthesize buckyballs. The ropes are quite stable and have a further interesting property: they are electrically conducting.

The objective of this project is to study the electrochemical properties of the ropes by attempting to electrochemically intercalate alkali metal cations such as lithium into the ropes. Our research objectives are (1) to try to confirm the electrical conductivity values reported by the Smalley group; (2) to explore the use of these materials as electrodes in various solvents, focusing on the redox window of these materials; and (3) to assess the possibility of reversibly inserting various alkali metal cations into the ropes.

This year we observed that the conductivity of the ropes is highly anisotropic. In addition, electrochemical cycling in solutions of alkyl ammonium salts showed that the ropes have a fairly high accessible surface area. We observed double-layer charging currents corresponding to roughly 30 μF/g, somewhat lower than the values obtained with the state-of-the-art materials used in ultracapacitors. Finally, we attempted to electrochemically insert lithium into the ropes. Although we observed several features in slow voltammetric scans, the features were generally not reversible, indicating that any insertion that occurred is largely irreversible.

Innovative Composites through Reinforcement Morphology Design—A Bone-Shaped Short-Fiber Composite

Yuntian Zhu

The objective of this project is to improve the strength and toughness of conventional short-fiber composites by using innovative, "bone-shaped" short fibers as reinforcement. We have made significant progress in composite fabrication, characterization, and modeling in the past year.

On the experimental front, we have optimized to some extent the fiber morphology (fiber length, diameter, and ball size at fiber ends) to obtain the best combination of composite strength and toughness. Both nickel fibers and polyethylene fibers have been used to make composites (see first figure). Mechanical testing of bone-shaped polyethylene short-fiber-reinforced polyester composites demonstrated, for the first time, that the difficulty of pulling out the bone-shaped short fibers is a toughening mechanism. Our results showed that the bone-shaped nickel fibers are 120% more effective than the straight fibers in reinforcing the composites (see second figure).

On the modeling front, we have studied the stress profiles both in intact and broken bone-shaped fibers using the finite-element method (FEM). We have also simulated the failure process of the bone-shaped short-fiber composites with our CLASFAM code. The code has been modified to deal with both straight and bone-shaped short fibers randomly distributed in an elastic-plastic matrix. The role of the "sphere" at the end of the short fiber is described by frictional forces provided by FEM. By comparing the stress-strain curves of a bone-shaped fiber composite with those of a straight-fiber composite, we found that the composite reinforced with bone-shaped fibers has both better toughness and strength, which is in fairly good agreement with the experimental results.

Publications

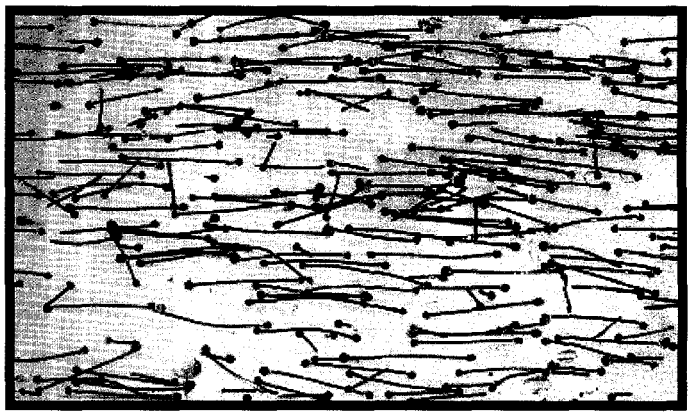
Taylor, S.T., et al., "New Perspectives on the Fracture of Nicalon Fibers" (Twenty-Ninth National Fracture and Fatigue Symposium, Stanford University, Palo Alto, CA, June 24–26, 1997).

Zhu, Y.T., et al., "Evaluation of a Modified Weibull Distribution in Characterizing the Strength of Ceramic Fibers and Whiskers with Varying Diameters" (to be published in *J. Test. Eval.*).

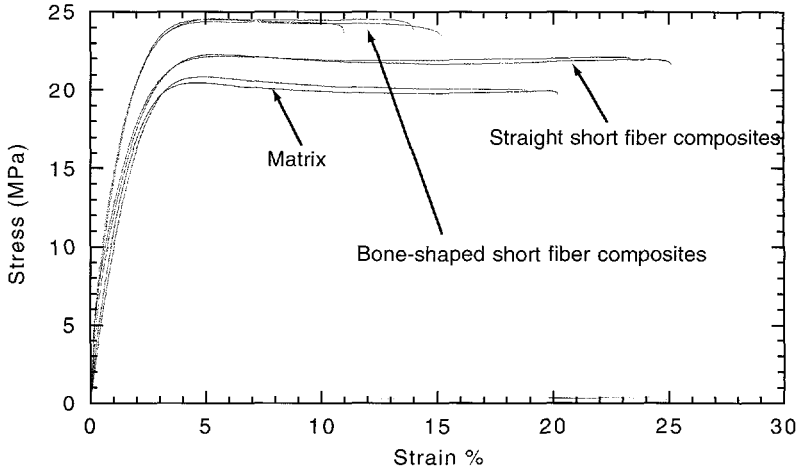
Zhu, Y.T., et al., "Influence of Reinforcement Morphology on the Mechanical Properties of Short-Fiber Composites" (The Minerals, Metals, Materials Society (TMS) Spring Meeting, San Antonio, TX, February 16–19, 1998).

Zhu, Y.T., et al., "Kinetics for Thermal Oxidation of Cylindrical Fibers" (to be published in *J. Am. Ceram. Soc.*).

Zhu, Y.T., et al., "On the Statistical Strength of Nicalon Fibers and Its Characterization," *Ceram. Eng. Sci.* **18**, 119 (1997).



Nickel bone-shaped short fibers are well aligned in a polyethylene matrix. The average fiber length is 2.5 mm.



Stress-strain curves of bone-shaped and straight short nickel fiber composites and polyester matrix. We used 0.6% methyl ethyl ketone peroxide hardener to harden polyester matrix for all samples. Nickel fiber length = 2.5 mm, diameter = 76.2 mm. Fiber volume fraction = 1.7%. The bone-shaped short fibers are 120% more effective than the straight fibers in reinforcing the composites.

Texture Characterization for High-Strain-Rate Deformations

Sheila K. Schiferl

The objective of this project is to develop the capability to investigate the effects of crystallographic texture—the preferred orientation of single-crystal grains in a polycrystalline material—on the behavior of systems undergoing high-strain-rate plastic deformations. Preferred orientations are consequences of metal-working and other thermomechanical processes, and result in both strength and elastic anisotropies.

We developed a model for anisotropic material strength and for changes in the anisotropy due to plastic strain. The model has been developed for use in high-rate, explicit, Lagrangian multidimensional continuum-mechanics codes. The model handles anisotropies in single-phase materials, in particular the anisotropies due to crystallographic texture. The material model includes a texture code, or micromechanical calculation, coupled to a continuum code. The texture code updates grain orientations as a function of tensor plastic strain and calculates the yield strength in different directions. For each computational cell in the continuum simulation, the texture code tracks a particular set of grain orientations. The orientations will change due to the tensor strain history, and the yield function will change accordingly. Hence, the continuum code supplies a tensor strain to the texture code, and the texture code supplies an updated yield function to the continuum code. Since significant texture changes require relatively large strains—typically, a few percent or more—the texture code is not called very often, and the increase in computer time is not excessive. We implemented the model using a finite-element continuum code and a texture code specialized for hexagonal close-packed crystal structures.

Publications

Schiferl, S.K., and P.J. Maudlin, "Evolution of Plastic Anisotropy for High-Strain-Rate Computations," *Comput. Methods Appl. Mech. Eng.* **143**, 249 (1997).

Thermodynamic and Electrodynamic Studies of Unusual Narrow-Gap Semiconductors

Albert Migliori

Our objectives for the past year were to produce materials, based on the principles developed previously in this project, that include single crystals of $Bi_{1-y}Sb_ySn_{.0000x}$ alloys and to measure their thermopower, resistivity, and thermal conductivity tensors by using systems constructed in previous years. We also continue to advance our theoretical understanding of the Ettingshausen effect and our understanding of procedures to tune materials for optimum refrigeration below 100 K.

This past year we completed the growth of many samples of these alloys with y near 0.03. Measurements were made of all the transport quantities described above for several orientations of magnetic field. Results strongly indicate that for x near 1 (1 part per million of tin), dramatic changes in the transport properties occur and that our predictions that such alloys can exceed the performance of conventional ones, if a nonconventional orientation for magnetic field is used, are borne out.

Publications

Freibert, F., et al., "New Results for the Adiabatic Thermoelectric and Conductivity Tensors in Bi-Sb Alloy Single Crystals" (American Physical Society Spring Meeting, Kansas City, MO, April 12–21, 1997).

Migliori, A., et al., "New Approaches to Thermoelectric Cooling Effects in Magnetic Fields" (to be published in *J. Heat Transfer*).

Migliori, A., et al., "New Directions in Materials for Thermomagnetic Cooling" (to be published in *Proceedings of the 1998 Space Technology and Applications International Forum*).

Migliori, A., et al., "Optimization of Materials for Thermomagnetic Cooling" (to be published in *J. Mater. Res.*).

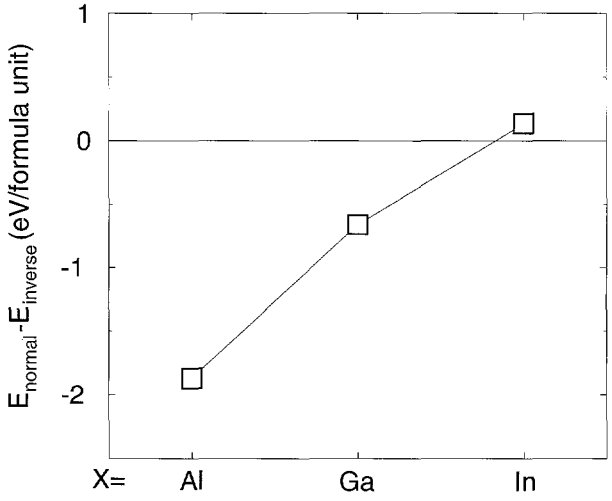
Development of a Fundamental Understanding of Chemical Bonding and Electronic Structure in Spinel Compounds

Kurt Sickafus

Hundreds of ceramic compounds possess the spinel crystal structure and exhibit a remarkable variety of properties, ranging from compounds that are electrical insulators to compounds that are superconducting or from compounds with ferri- and antiferromagnetic behavior to materials with colossal magnetoresistive characteristics. The unique crystal structure of spinel compounds is in many ways responsible for the widely varying physical properties of spinels. The objective of this project is to investigate the nature of chemical bonding, point defects, and electronic structure in compounds with the spinel crystal structure.

This year, we performed x-ray and neutron diffraction studies on nonstoichiometric, alumina-rich spinels to determine where charge-compensating cation vacancies reside in the lattice. For a single crystal with the composition $MgO{\bullet}3.2Al_2O_3$, crystal-structure refinements indicate that all cation vacancies reside on octahedral sites. Similar x-ray and neutron diffraction studies on chromia-doped magnesio-aluminate spinels revealed that cation disorder (prevalent in pure $MgO{\bullet}Al_2O_3$) disappears upon addition of chromium.

We used shell-model total-energy calculations to assess point-defect migration energies in magnesio-aluminate spinel, and we determined that O^{2-} and Al^{3+} are the mobile species in this material with migration energies of 1.19 and 1.70 eV, respectively (compared with 8.58 eV for Mg^{2+}). Using fully relaxed, *ab initio* electronic-structure calculations, we also examined the cation-disordering energetics and electronic structure of magnesiospinels with the isoelectronic octahedral constituents aluminum, gallium, indium, and thallium. The difference in total energy between normal and inverse spinel structures is shown in the accompanying graph. The normal structure $MgO{\bullet}Al_2O_3$ is stable by almost 2 eV per formula unit. This stability is reduced to about 0.7 eV per formula unit in $MgO{\bullet}Ga_2O_3$, and the inverse structure is stable by about 0.1 eV per formula unit in $MgO{\bullet}In_2O_3$. These theoretical results are consistent with our experimental observations.



The difference in energy between the normal spinel structure and the fully inverse structure for $MgO{\bullet}X_2O_3$ (where X = Al, Ga, and In) calculated by an *ab initio*, full-potential, electronic-structure method. All internal degrees of freedom were relaxed in these calculations.

Tandem Metal-Mediated Synthesis

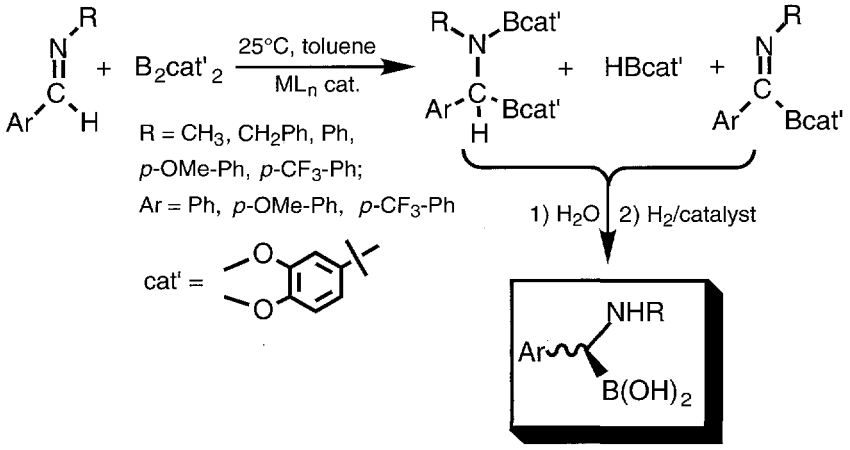
Tom Baker

In this project we use a series of metal-mediated reactions to prepare complex molecules from simple precursors in one pot without isolation or purification of intermediates. This strategy will reduce energy usage and especially waste generation in the manufacture of fine chemicals, pharmaceuticals, and agrochemicals. Boronic acids [$RB(OH)_2$] are currently of interest for sensor applications, and alpha-aminoboronic acids [$NHRCR'R''B(OH)_2$] are particularly selective inhibitors of serine protease enzymes. As known synthetic routes do not allow a wide variation in substituents R, R', and R", we sought to prepare alpha-aminoboronic acids by direct routes that rely on metal-catalyzed addition of diboron compounds to imines and nitriles.

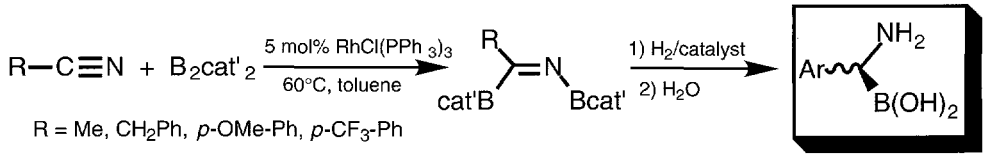
Metal-catalyzed diboration of imines gives a mixture of diboration and borylation depending on the regiochemistry of imine insertion into the M-B bond (first figure). We are developing a hydrolysis-hydrogenation work-up procedure to obtain the desired alpha-aminoboronic acids.

Metal-catalyzed diboration of nitriles gives addition of the B-B bond across the C≡N triple bond to give diborylimines (second figure). Subsequent hydrogenation of these diborylimines should provide a direct route to alpha-aminoboronic acids, which also has potential for controlling the stereochemistry by employing an asymmetric metal-catalyzed hydrogenation.

We have also used diboron additions to effect the reductive coupling of imines to new diamines, as a mild sulfoxide reduction method, and for tandem deoxygenation/C-C bond forming chemistry with carbonyl compounds. This work has clearly demonstrated the utility of diboron additions for novel tandem metal-mediated synthesis, and we will be expanding this work in partnership with several pharmaceutical companies.



Tandem imine diboration/borylimine hydrogenation route to alpha-aminoboronic acids.



Tandem nitrile diboration/diborylimine hydrogenation route to alpha-aminoboronic acids.

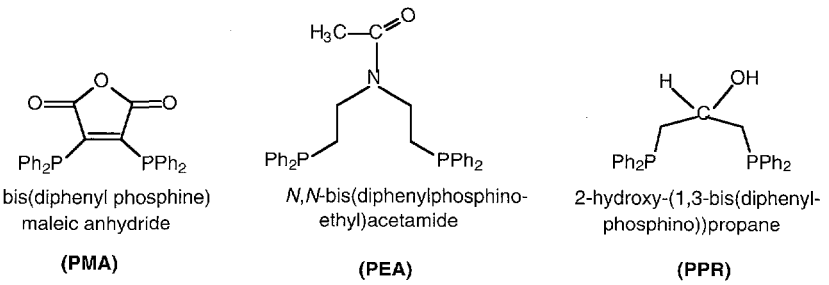
Reactivity at Metal Centers Bound to Water-Soluble Catalysts

Nancy N. Sauer

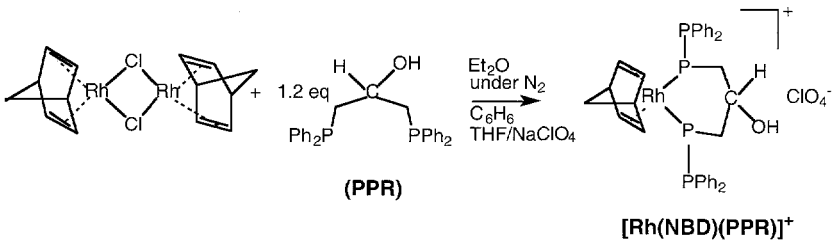
The development of new catalysts that function in aqueous solution is an area of growing interest. Typically, such systems provide much easier product separation and generate less hazardous waste. Our goals are to evaluate the reactivity of metal centers bound to water-soluble polymers and understand the effects the macromolecular environment has on the active site. The practical objective is to provide economically feasible and environmentally benign catalysts in which the selectivity and activity can be controlled through modification of the polymer.

This year we focused on synthesizing and characterizing rhodium hydrogenation catalysts with attachment points for coupling with water-soluble polyamines. Our work continued with a ligand prepared during the previous year, bis(diphenyl phosphine) maleic anhydride (PMA). We also prepared and evaluated two new ligand systems: *N,N* -bis(diphenylphosphinoethyl)acetamide (PEA) and 2-hydroxy-(1,3-bis(diphenylphosphino))propane (PPR) (see the first figure). After reaction with rhodium sources, these systems can be tethered to the amine polymers via the aldehyde and hydroxyl carbons (see the second and third figures).

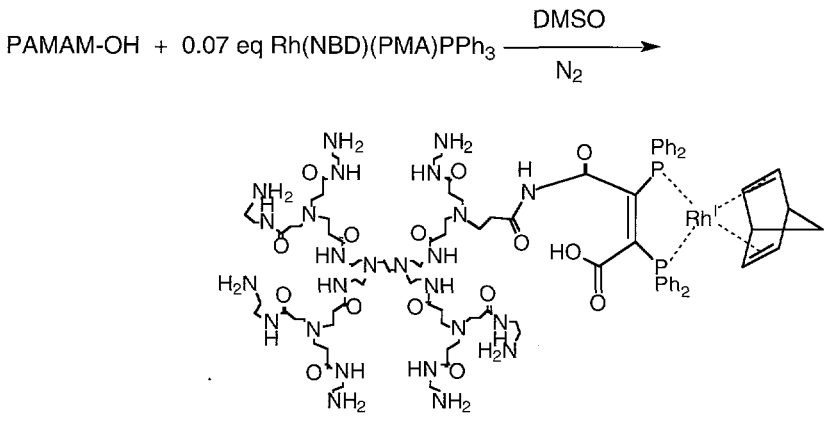
We fully characterized the resultant catalysts using standard techniques and measured the hydrogenation capabilities of these systems. Comparison of the reactivity of free and polymer-bound catalysts that we have prepared will allow us to evaluate the efficacy of using soluble polymer-supported catalysts for a wide variety of reactions, including hydrogenation, hydroformylation, and carbon-carbon bond-forming reactions.



Structures of the three phosphine ligands for attachment to soluble polymers.



Reaction of PPR with the rhodium dimer to give an active hydrogenation catalyst [Rh(NBD)(PPR)]$^+$, where NBD is norbornadiene.



Ring-opening reaction of PMA rhodium complex with a dendrimer to give a tethered hydrogenation catalyst.

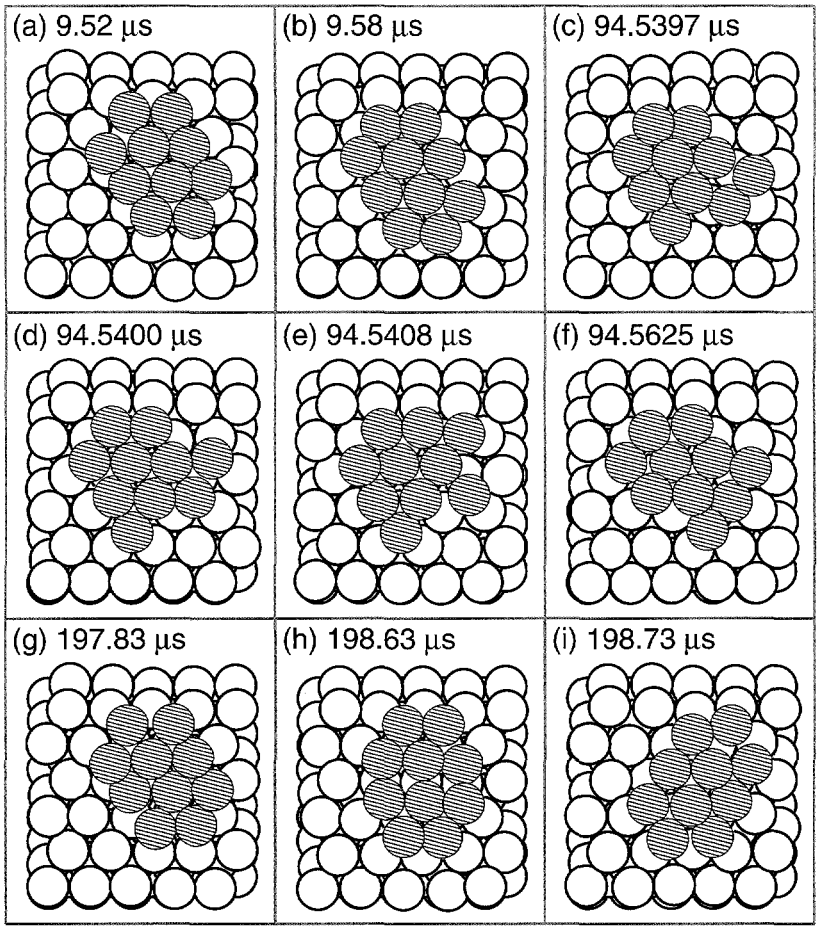
Decay of Surface Nanostructures via Long-Time-Scale Dynamics

Arthur Voter

Atomistic simulations are playing an increasingly important role in chemistry and materials science. However, despite recent advances in computer speed, dynamical methods, and empirical potentials, an important class of unsolvable problems remains. These problems are characterized by intrinsically long time scales, which exceed the time available to direct molecular dynamics (MD), and a complexity that precludes the use of catalogued, kinetic Monte Carlo methods. Examples include post-ion-implant damage annealing and diffusion on a rough surface. The goal of our research is to develop a general method for solving problems of this type.

Our main objective is to develop a dynamical method for accelerating the MD simulation of infrequent events. We achieved a significant breakthrough in the development of the "hyperdynamics" method, in which a bias potential augments the interatomic potential for the system. The bias potential is designed to raise the energy of the potential basins but not affect the energy in the regions of the transition states. We can accomplish the selective energy increase, even if the locations of the transition states are not known, by using the position-dependent matrix of second derivatives. The time scale of the accelerated dynamics is also recovered.

This year, we have successfully shown that with an iterative procedure, we can compute the bias potential (and its derivatives, which give the necessary interatomic forces) by using only first derivatives of the original (unbiased) potential function. Therefore, we can apply the hyperdynamics method relatively easily, using iterative calls to the standard MD force subroutine. The figure shows a sample calculation for the diffusion of a cluster on a silver surface. This new approach is generating considerable interest in the condensed-phase simulation community.



Snapshots from the hyperdynamics simulation of a 10-atom silver cluster on the Ag(111) surface at T = 300 K. The snapshots show boosted time values. Three distinct mechanisms contributing to the diffusion of the cluster are visible: gliding (a)–(b), edge-running (c)–(f), and dislocation-mediated motion (g)–(i). A direct MD simulation of this system for the same length of time (200 μs) would require several years of computer time on the fastest available workstation.

Publications

Voter, A.F., "Hyperdynamics: Accelerated Molecular Dynamics of Infrequent Events," *Phys. Rev. Lett.* **78**, 3908 (1997).

Voter, A.F., "A Method for Accelerating the Molecular Dynamics Simulation of Infrequent Events," *J. Chem. Phys.* **106**, 4665 (1997).

Classical Kinetic Mechanisms Describing Heterogeneous Ozone Depletion

Bryan Henson

Since the first report of the polar "ozone hole," field and laboratory measurements have shown that several heterogeneous (gas/surface) reactions on polar stratospheric clouds lead to ozone depletion and that the surface concentration of reagents and products in the stratosphere couples these reactions. This project uses classical kinetic descriptions of the heterogeneous interface to provide complete models of these coupled stratospheric reactions. In one of the first examples of an experimentally consistent mechanism relevant to these processes, we have shown that an important reaction, the hydrolysis of chlorine nitrate, proceeds by the classic Eley-Rideal kinetic mechanism (see first figure).

In a series of volumetric adsorption experiments, we have shown that under stratospheric conditions HCl is not adsorbed on ice in appreciable quantities, a result that contradicts the current belief that reactions with concentrated HCl on ice are extremely important (second figure). We presented our work at a symposium on stratospheric chemistry at the fall meeting of the American Chemical Society and argued that the role of HCl in heterogeneous chemistry must be completely reconsidered.

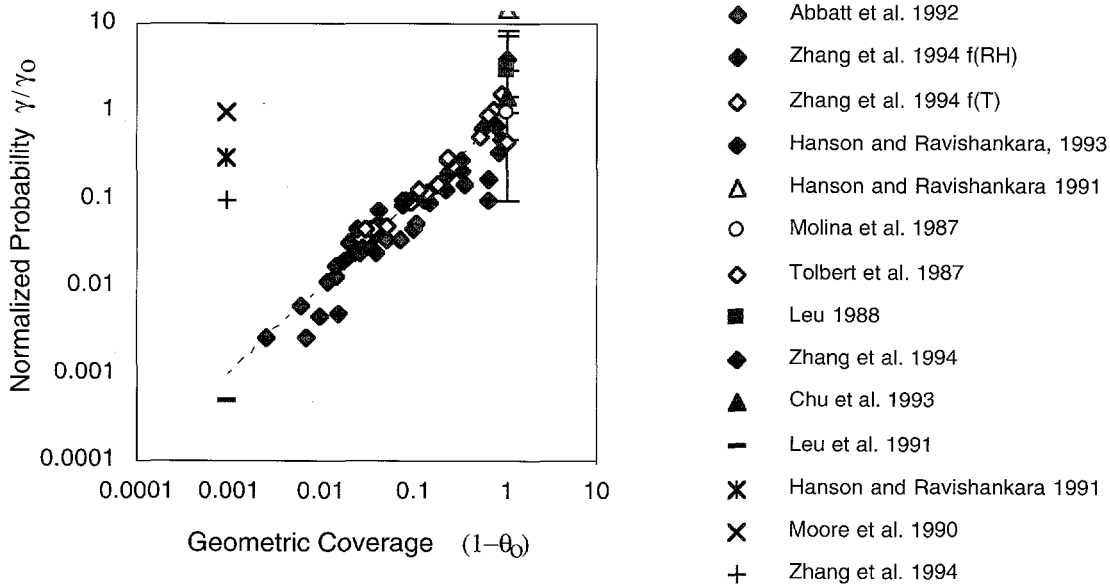
We have also performed studies on the morphology of laboratory-grown ice surfaces, a subject of continuing debate in the *Journal of Physical Chemistry*. Our detailed measurements, particularly of surface area, for samples grown in our laboratory have contributed significantly to understanding the adsorption measurements described above.

Publications

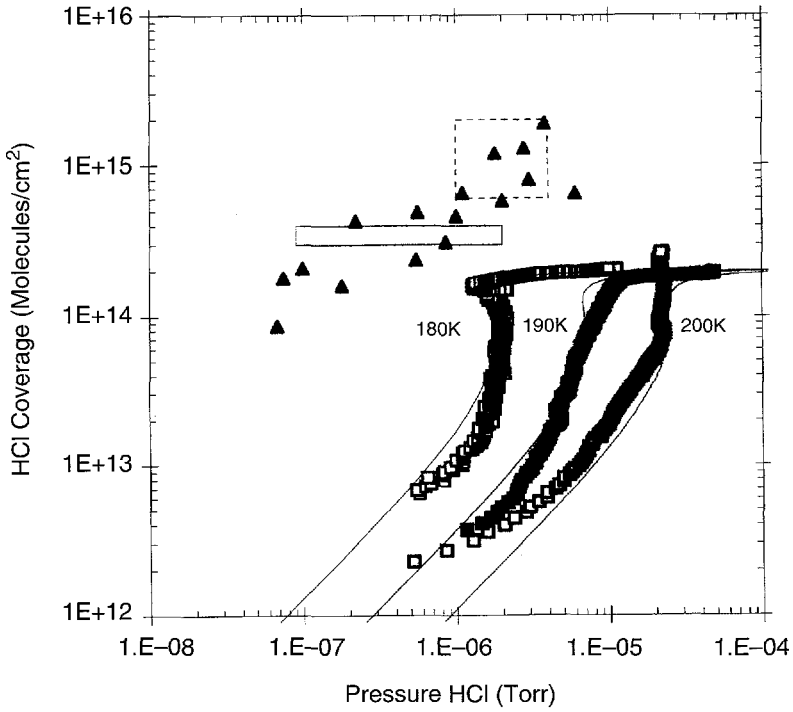
Henson, B.F., et al., "Measurements of HCl Adsorption Isotherms on H_2O Ice from 180 K to 200 K at Near Stratospheric Pressures," *Abstr. Pap. Am. Chem. Soc.* **214**, 400 (1997).

Henson, B.F., et al., "Models of Hydrogen Chloride Adsorption and Chlorine Nitrate Reactivity on Simulated PSC Surfaces," *Eos, Trans., Am. Geophys. Union* **77**, 95 (1996).

Henson, B.F., et al., "Quantitative Measurements of Multilayer Physical Adsorption on Heterogeneous Surfaces from Nonlinear Light Scattering," *Phys. Rev. Lett.* **79**, 1531 (1997).



Summary of experimental data from several laboratories on the hydrolysis reaction of chlorine nitrate. The experiments measure reaction probability on a number of surfaces with varying water coverage. The data are plotted as the normalized reaction probability—the measured probability of reaction divided by the probability of reaction on pure ice—against the geometric surface coverage of H_2O on the surface. The dashed line is the fit predicted by the Eley-Rideal reaction mechanism.



Measured adsorption isotherms for HCl on ice at 180, 190, and 200 K. The data are plotted as the surface concentration of HCl as a function of HCl pressure. The solid lines are calculations of coverage from a Langmuir-Bragg-Williams statistical treatment of classic physical adsorption. The data indicate monolayer HCl uptake of 2×10^{14} molecules/cm^2. It is clear from these data that under stratospheric conditions—200 K and 1×10^{-7} torr pressure—HCl coverage is of the order of 0.01% of a monolayer.

Heterogenization of Homogeneous Catalysts: The Effect of the Support

William L. Earl

Our goals are to covalently bind known solution (homogeneous) catalysts to the surfaces of high-surface-area, solid materials and to investigate the effects due to binding. We vary the structure of the solid surface and the chemical link between the catalyst and surface and measure the catalytic activity. This process allows us to determine if there is a relation between the final structure and catalytic activity.

This project includes two phases of investigating heterogeneous catalysts. The first is to duplicate unusual data that already exists, as reported in the literature, and then understand the cause of the unusual behavior. The second is to use this knowledge to develop new catalysts and reactivity. In the primary phase of this work, we use the asymmetric hydrogenation of an enamide as a probe of the interaction of the supported catalyst with the support.

As reported last year, we have been unable to duplicate the unusual results that appear in the literature. This year we continued to synthesize the compounds to be supported on surfaces as described in the literature by a number of routes, carefully characterized them by nuclear magnetic resonance and infrared spectroscopy, and verified that we indeed had the desired compounds in hand. These compounds were catalytically active by themselves in homogeneous phase but did not yield any enantiomeric excess above what is detectable by gas chromatography. After several attempts and altering of the reaction conditions, we must conclude that the literature reports are in error. We have subsequently found that other researchers at a pharmaceutical company have also been unable to reproduce these results.

Consequently, this year we decided to take a different approach to heterogenized catalysts and examine the differences among solution and immobilized vanadium oxidation catalysts. A comparative study was made to determine the best choice of polymer support for the immobilization of the vanadyl ion catalyst species for use in epoxidation of allylic alcohols in CO_2. We studied in particular the epoxidation of *trans*-2-hexen-1-ol to 2,3-epoxyhexanol using *tert*-butyl hydroperoxide as the oxidant and using both hexane and CO_2 as solvents. We find 2- to 5-fold differences in initial rate among the reactions using the two solvents, with reactions in hexane being somewhat faster, and find that vanadium does leach from the catalysts over time. The amount leached depends upon the support. Some supports exhibit only a small degree of leaching, so the catalyst may be recycled multiple times with little loss in activity. Preliminary work indicates that the reaction follows first-order reaction kinetics and that there is little, if any, inhibition due to water that is a byproduct of the reaction.

Establishment of a Room-Temperature Molten-Salt Capability to Measure Fundamental Thermodynamic Properties of Actinide Elements

Wayne Smith

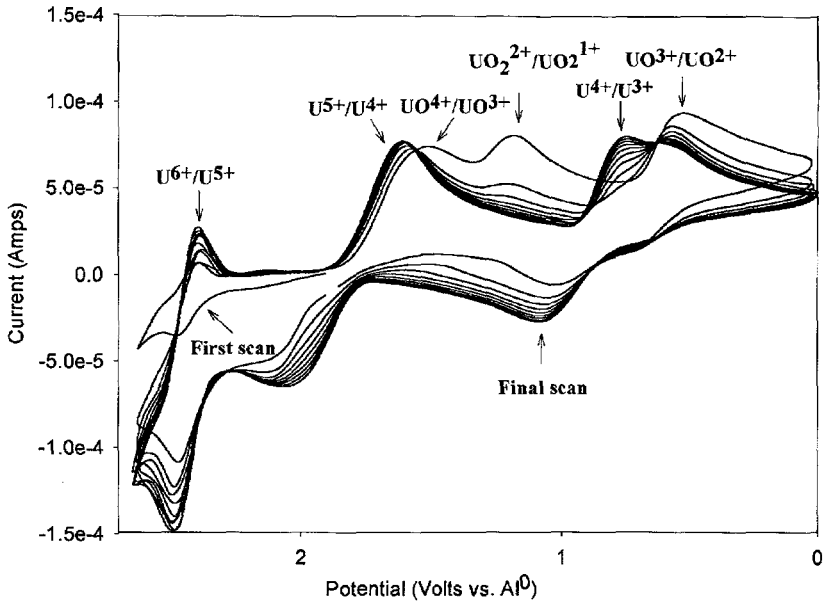
The primary goal of this project was to develop the capability to work in room-temperature molten-salt (RTMS) media and to obtain an in-depth understanding of the fundamental chemical behavior of actinide elements in molten-salt systems. The initial emphasis of this study was to develop a working methodology for the purification of melt components, $AlCl_3$ and 1-ethyl-3-methylimidazolium chloride, and to understand the essential electrochemical behavior of actinide elements in acidic, basic, and buffered melts. Uranium was chosen as the first element to be evaluated in the RTMS systems.

In acidic melts, UCl_4 produces one reversible reduction and two reversible oxidation processes to yield U^{3+}, U^{5+}, and U^{6+}, respectively. This result demonstrates the enhanced stability of high-oxidation-state species in this media. The uranyl ion UO_2^{2+} demonstrates a markedly more complex electrochemical behavior in an acidic melt (see first figure). By comparing this voltammogram with that obtained with UCl_4, we were able to deduce the electrochemical reaction scheme for UO_2^{2+}, which is presented in the second figure.

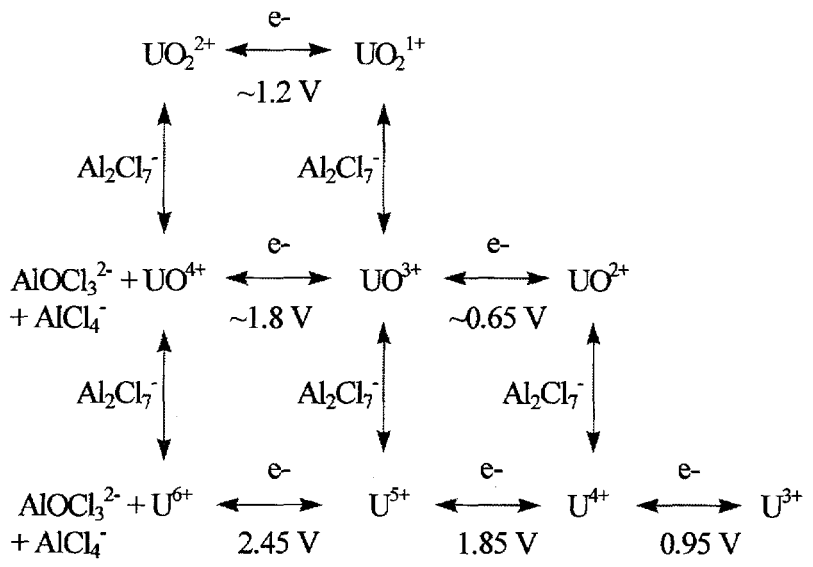
We have also observed a slow homogenous reaction between UO_2^{2+} and the acidic melt leading to the formation of unoxygenated U^{5+}. A mechanism for this reaction is presented in the third figure. The first step is due to distribution of U^{6+} between the di-oxo, mono-oxo and non-oxo species, a typical base dissociation reaction. The second, rate-determining step is an electron transfer reaction between the de-oxygenated U^{6+} species and the solvent.

Publications

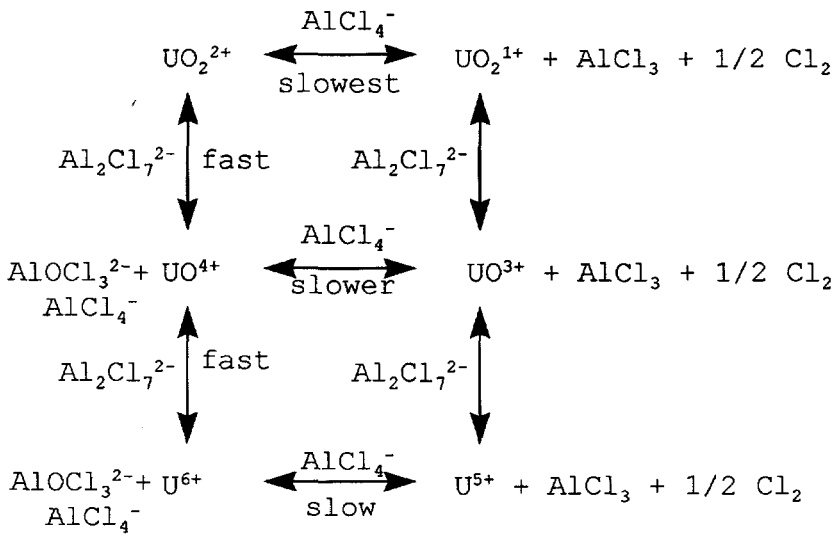
Smith, W.H., et al., "Determination of Thermodynamic Properties of Actinide Elements in Room Temperature Molten Salts" (Plutonium Futures—The Science: Topical Conference on Plutonium and Actinides, Santa Fe, NM, August 25–27, 1997).



Repetitive scan cyclic voltammogram of UO_2^{2+} in an acidic RTMS melt.



Electrochemical reaction mechanism for UO_2^{2+} in an acidic RTMS melt.



Homogeneous reaction mechanism between UO_2^{2+} and an acidic RTMS melt.

Solvation and Ionic Transport in Polymer Electrolyte Membranes

Thomas Zawodzinski

Ion exchange membranes such as Nafion, a teflon-like polymer with acid functionality attached to it, are key components of emerging devices such as fuel cells, batteries, and membrane-based reactors. Nonetheless, transport mechanisms in these materials are incompletely understood at the molecular level. This project seeks to combine theoretical and experimental studies to develop such understanding.

We are using a recently developed solvation theory to probe processes fundamental to water and ion transport in the membranes. We are also carrying out high-frequency dielectric studies of the membranes in various states of hydration to probe water, ion, and polymer dynamics.

This year we made substantial progress in obtaining electronic structures of important portions of Nafion, the perfluorosulfonic acid polymer electrolyte of primary interest. We are also comparing the perfluorosulfonic acid polymer electrolytes to aromatic sulfonates to understand significant differences in the transport properties of these two electrolytes. Building on the electronic structure calculations, we are in the process of conducting molecular dynamics studies to understand proton transfer in hydrated polymer electrolytes.

In addition, we have completed apparatus and analysis method development for high-frequency dielectric measurements on the polymer electrolytes. Water-uptake-dependent dielectric response has been observed. The combination of the above studies is yielding new insights into dominant properties determining transport in polymer electrolytes.

Publications

Paddison, S.J., et al., "Molecular Modeling of the Pendant Chain in Nafion" (submitted to *Solid State Ionics*).

Paddison, S.J., et al., "Molecular Modeling of Trifluoromethanesulfonic Acid for Solvation Theory" (to be published in *Fluid Phase Equilib.*).

Zawodzinski, T.A. Jr., et al., "Mechanism of Transport in Proton-Conducting Membranes" (submitted to *Solid State Ionics*).

Asymmetric Catalysis in Organic Synthesis

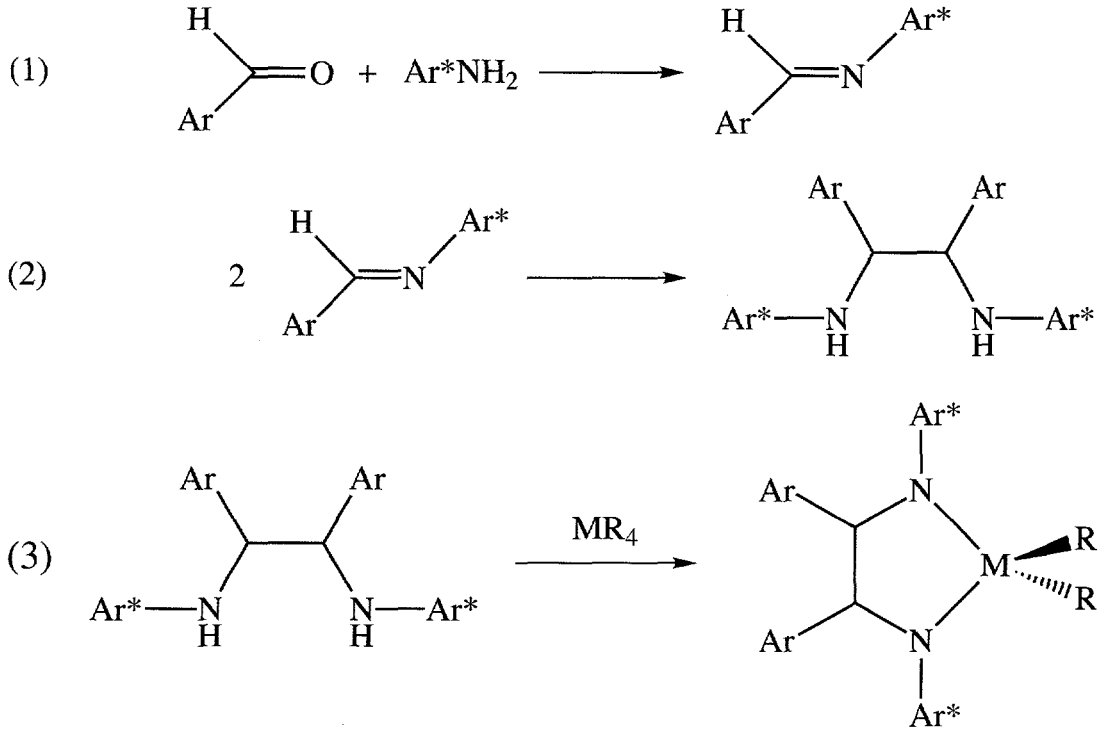
John Watkin

This project is aimed at developing new catalysts for the enantioselective formation of organic molecules. As well as being of broad interest to our defense mission, organic catalysis is relevant to the pharmaceutical and fine-chemical industries.

In the past year, we have continued to examine the use of lanthanide complexes containing chelating diolate and Schiff-base ligands as enantioselective catalysts for reducing aldehydes to optically active alcohols. We have also extended our investigations from enantioselective catalysis of organic reactions to catalysis of polymerization reactions, and we have explored catalyst systems that have the potential to polymerize prochiral monomers such as propylene in a stereospecific manner.

We have developed a new class of noncyclopentadienyl ligands that may be attached to a Group IV metal center to produce a catalytically active species. The synthesis of this class of ligands (see the accompanying reaction scheme) involves the initial reaction of an aldehyde with an aromatic amine to form an aldimine. We then reductively couple two equivalents of aldimine to produce a 1,2-diamine species in both meso and racemic forms. Separation of the two diastereoisomers by crystallization allows us to isolate the racemic form of the diamine. When we then attach the diamine to a Group IV metal center, we can produce metal complexes that catalyze the polymerization of olefins. Currently, we are investigating the effectiveness of this class of ligands in controlling the stereoregularity of polymers such as polypropylene.

This work has generated significant interest from the chemical industry. We have recently filed a patent application to cover the use of these bis-amine ligands in forming catalytically active complexes of the Group IV and lanthanide metals.



Three-step route to bis-amine complexes of the Group IV metals: (1) aldimine formation, (2) reductive coupling, and (3) attachment to a metal center. In these reactions, Ar and Ar* represent any aromatic group, M is a Group IV metal, and R is an alkyl group such as methyl.

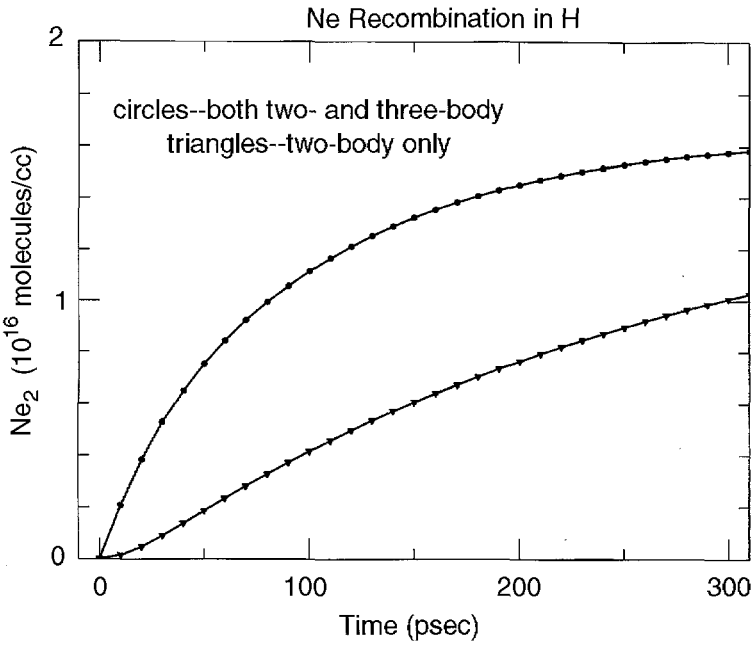
Recombination Kinetics: Correcting the Textbooks

Russell Pack

The overall objective of this project is to change the way chemists think about and treat recombination reactions. Virtually all the textbooks and literature assume that these reactions occur only through sequences of two-body collisions. We are showing by actual calculations on real reactions that true three-body collisions also contribute significantly.

Progress during this past year was faster than expected. To get results rapidly, we used the Vibrational Rotational Infinite Order Sudden Approximation, which is valid for a number of reactions and treats all recombination mechanisms on equal footing. With it we calculated cross sections for all possible mechanisms for the simple but relevant reaction $Ne + Ne + H \rightarrow Ne_2 + H$. The results, which have just been published, show clearly that true three-body collisions are as important as sequential two-body mechanisms and cannot be neglected.

Next, we thermally averaged the cross sections to get rate constants and did kinetics studies which show that, for the above reaction (and probably also for reactions such as $H + H + He \rightarrow H_2 + He$), three-body collisions actually dominate. As the accompanying figure shows, inclusion of only the traditionally popular energy-transfer mechanism gives a recombination rate (bottom curve) that is more than a factor-of-4 too small! We also made good progress toward exact calculations on the reaction: We have found new ways to choose the basis wave functions more efficiently and to make calculations for large total angular momenta feasible.



Concentration of neon dimers as a function of time in the recombination of neon atoms in hydrogen atoms at a temperature of 30 K. The circles are the results of complete calculations including all recombination mechanisms. The triangles result when only the traditional sequences of two-body collisions are included.

Publications

Pack, R.T., et al., "Mechanisms of Atomic and Molecular Recombination and Collision-Induced Dissociation," *Chem. Phys. Lett.* **276**, 255 (1997).

Characterization of Propane Monooxygenase: Initial Mechanistic Studies

Pat Unkefer

Industrial use of chlorinated hydrocarbons has led to their widespread contamination of soils and groundwater. Despite the persistence of chlorinated compounds in the environment, a number of bacterial strains have been shown to degrade chlorinated compounds including trichloroethylene (TCE). For example, methanotrophic bacteria degrade TCE via the enzyme methane monooxygenase (MMO), which converts TCE to its epoxide. However, the effectiveness of methanotrophic bacteria in bioremediation schemes is limited because of the toxicity of TCE epoxide.

We are investigating the propane monooxygenase (PMO) from *Mycobacterium vaccae*, which is similar to the MMO from methanotrophs in that it oxidizes a wide variety of organic compounds including TCE. Because it does not degrade TCE via formation of the epoxide, PMO could provide an important alternative to MMO for bioremediation of TCE-contaminated sites. However, to date, PMO has not been isolated and is largely uncharacterized. Our objective is to further PMO's bioremediation potential by developing a clear understanding of its properties, mechanism, and substrate specificity.

We have demonstrated strong progress in scaling up the production of the organism when grown on propane. This scale-up was required to produce the quanitites of organism required to supply the mechanistic studies. Previous work had been largely conducted in small-scale (<5-L) fermentations, and a dependable, larger-scale (>20-L) fermentation production system had to be developed.

Furthermore, the organism's observed growth rate has increased significantly in the process that we have developed. This increase in growth rate is significant because it indicates that the induction system for the PMO had not been operating at maximal velocity and that the induction system had not been saturated. The substrate, propane, is the inducer, and thus our results indicate that failure to saturate the aqueous fermentation medium with propane has limited the growth. This finding has important implications for practical application of this organism because its activity is much greater than previously observed.

Ultrafast Solid-State Electron Transfer in Donor-Acceptor Conducting Polymers

Duncan McBranch

Electron transfer in conducting polymers is a promising new field of photochemistry. In these polymers, charge transfer has been demonstrated to occur in less than 200 fs with near-unity quantum efficiency and to be metastable with an asymmetry in the forward and back transfer rates of up to 9 orders of magnitude. We are applying femtosecond spectroscopy to study the dynamics of the various steps involved in the charge transfer: the excitation events, the charge transfer itself, and the stabilization mechanism. Using this knowledge, we plan to explore the practical applications of these materials by studying methods to utilize the charge-transfer state in order to drive nonlinear optical effects.

During this past year, we have unambiguously assigned, over a wide range of excitation densities, the visible-near-infrared transient absorption features in polymers and oligomers from the poly(p-phenylene vinylene) family. We have shown that emissive excitons are the dominant species created after photoexcitation in these materials. In addition, we have identified new, ultrafast decay mechanisms that are operative at high excitation densities, including exciton-exciton annihilation and creation of intrachain-bound polaron pairs via doubly excited excitons (biexcitons). Our report of biexcitons is the first experimental verification of these species in conducting polymers. Based on new spectral results of linear and nonlinear absorption, we have developed a new theory of the optical properties of conducting polymers. Finally, we have pursued initial studies of the spatial range of excitons in charge-transfer blends.

Publications

Kirova, N., et al., "Understanding the Excitations and Optical Properties of Phenylene-Based Conjugated Polymers: A Hybrid Molecular and Band Model" (submitted to *Phys. Rev. Lett.*).

Klimov, V.I., and D. McBranch, "Auger-Induced Charge Separation in Semiconductor Nanocrystals," *Phys. Rev. B* **55**, 13173 (1997).

Klimov, V.I., and D. McBranch, "Electron and Hole Trapping Dynamics in Semiconductor Nanocrystals: Femtosecond Nonlinear Transmission and Photoluminescence Study," *Mat. Res. Soc. Symp. Proc.* **452**, 317 (1997).

Klimov, V.I., and D. McBranch, "Femtosecond High-Sensitivity, Chirp-Free Transient Absorption Spectroscopy" (to be published in *Opt. Lett.*).

Klimov, V. I., et al., "Exciton and Biexciton Signatures in Femtosecond Transient Absorption of π-Conjugated Oligomers," *SPIE Proc., Opt. Probes Conjugated Polym.* **3142**, 58 (1997).

Klimov, V. I., et al., "Femtosecond Dynamics of Excitons in π-Conjugated Oligomers: The Role of Intrachain Two-Exciton States in the Formation of Interchain Species," *Chem. Phys. Lett.* **277**, 109 (1997).

Maniloff, E.S., et al., "Intensity-Dependent Recombination and the Nature of Excited States in Solid-State Conducting Polymers," *Phys. Rev. B* **56**, 1876 (1997).

McBranch, D., and M.B. Sinclair, "Ultrafast Photoinduced Absorption in Nondegenerate Ground-State Conjugated Polymers: Signatures of Excited States," in *Nature of the Photoexcitations in Conjugated Polymers: Semiconductor Band vs Exciton Model,* N.S. Sariciftci, Ed. (World Scientific, New York, in press).

New Fullerene-Based Mixed Materials: Synthesis and Characterization

Duncan McBranch

Since the first isolation of buckminsterfullerene (C_{60}), there has been extensive research into this new form of carbon. The unique nonlinear optical properties of the fullerenes are due to the highly polarizable, conjugated p-electrons that are delocalized over the surface of the C_{60} sphere. Our goal has been to gain a fundamental understanding of these materials and their properties in selected hosts.

We have incorporated fullerenes into inorganic and organic hosts to form new composite materials, and we have studied the optical-limiting properties of the composites to see whether these materials can offer eye protection from intense, pulsed laser light. Using sol-gel techniques, we have formed glass composites containing either C_{60} or soluble derivatives of C_{60}. Derivatized-C_{60} sol gels show enhanced optical-limiting performance in the near-infrared portion of the spectrum. We have demonstrated that the concentration of these optically clear glasses can be controlled over a wide range and that the resulting materials can be polished to high-quality surfaces.

In addition, we have studied in detail the excited-state absorption dynamics and optical-limiting behavior of neat (underivatized) and derivatized fullerenes in solution, in thin films, and in solid-state matrices such as sol-gel glasses and optical plastics. We have investigated the underlying mechanisms for optical limiting in these composite materials and have identified the advantages and limitations for practical solid-state optical limiters that contain fullerenes. We have also patented a new mechanism for optical limiting in composite materials that relies on photo-induced charge transfer. Based on this concept, we have developed a new, proprietary optical-limiting fullerene derivative with a much stronger optical-limiting performance than the other fullerenes that we have measured.

Publications

Kohlman, R., et al., "Optical Limiting and Excited-State Absorption in Fullerene Solutions and Doped Glasses," in *Optical and Electronic Properties of Fullerenes and Fullerene-Based Materials* (Marcel Dekker, New York, in press).

Kohlman, R., et al., "Ultrafast and Nonlinear Optical Characterization of Optical Limiting Processes in Fullerenes" (to be published in *SPIE Proc. Fullerenes and Photonics IV*).

McBranch, D., et al., "Femtosecond Excited-State Absorption Dynamics and Optical Limiting in Fullerene Solutions, Sol-Gel Glasses, and Thin Films," *SPIE Proc. Fullerenes and Photonics III* **2854**, 140 (1997).

McBranch, D., et al., "Femtosecond to Nanosecond Dynamics of C_{60}: Implications for Excited-State Nonlinearities," *Res. Chem. Intermed.* **23**, 587 (1997).

Wang, H.L., et al., "Variations in the Optical Properties of Poly(3-hexylthiophene)/C_{60} Blends and Poly(3-hexylthiophene)/Sol-Gel Composites," *Synth. Met.* **84**, 781 (1997).

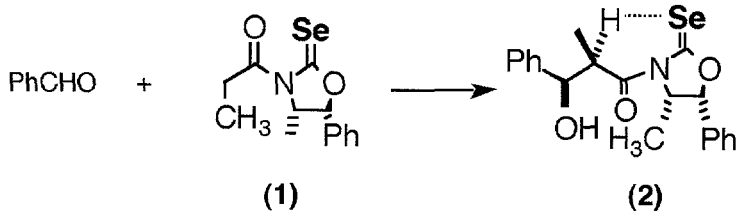
Uses of Novel Selenium-Containing Chiral Derivatizing Agents

Louis A. Silks

The major goal of this research is to investigate new chemistries of chiral selone reagents. We have focused our efforts on novel aldol chemistries and chiral oxidations of simple alkenes. Our major accomplishment during the past year has been the discovery of a new type of aldol reaction using chiral selone reagents. Reaction of an aldehyde with the titanium-based enolate of a propionyl selone gave rise to a product in which the two new chiral centers are in a *syn* relationship (see reaction scheme); that is, they are both protruding from the same side of the carbon backbone. Two different *syn* products are possible, and this reaction has given rise to the uncommon *syn* product that is called a non-Evans aldol product. Although they are rare, there are a few other examples of non-Evans aldols. The reaction proceeded in good yield (85%), and the stereoselectivity was greater than 98%.

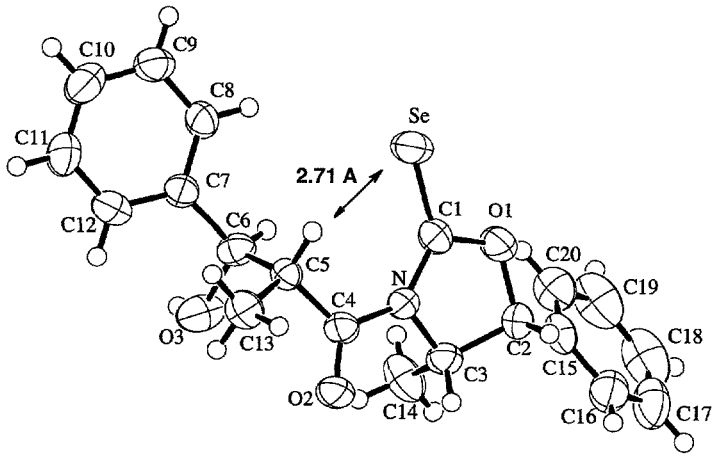
The most exciting feature of this work has been the additional apparent discovery of a C-H···Se through-space interaction. Although C-H···O and C-H···S interactions are well known (C-H···S less so than C-H···O), there is only one report of a C-H···Se interaction in the literature. We have examined all of our aldol products for this type of interaction, and every one has displayed this unique characteristic. Single-crystal x-ray analysis has indicated a H···Se distance of 2.71 Å (see diagram of the crystal structure), and two-dimensional $^{1}H/^{77}Se$ nuclear magnetic resonance (NMR) spectroscopy has clearly indicated a unique correlation between this proton and selenium.

In addition, we have performed aldol reactions with alpha-alkoxy groups on both reacting partners of this selone-mediated aldol reaction. Preliminary results indicate that aldol products are generated. However, the relationship of the two new centers that are generated in this carbon-carbon-bond-forming reaction is apparently *anti*. A proton-proton coupling constant of 9.6 Hz in our NMR experiments indicated the *anti* relationship. Synthetic methods for generation of *anti*-1,2-diol units are rare, and this method may prove valuable for constructing this type of unit. Moreover, many natural products, such as carbohydrates and some pharmaceuticals, possess such units, and our method may provide synthetic access to these important compounds.



(1) **(2)**

Chiral-selone-promoted aldol reaction with $TiCl_4$ (Ph is a phenyl group). The product, molecule 2, is a non-Evans aldol with a *syn* relationship between the newly created chiral centers. The dashed line indicates the C-H···Se interaction we found.



X-ray crystal structure of molecule 2. This structure provides evidence for a C-H···Se interaction.

Mathematics and Computational Science

Diffusion in Porous Media and Stochastic Advection

Shi-Yi Chen

This project involves combined numerical simulation and theoretical research of diffusion by intermittent stochastic velocity fields. The principal intended application is diffusion of substances in porous media. There will also be an impact on treatment of pollutant diffusion in the atmosphere or oceans and chemical reactions in fluids.

The work exploits the unmatched parallel-computation capability at Los Alamos together with recent advances in the analytical treatment of anomalous scaling phenomena in passive scalar turbulence advection. It is intended that this research will both increase fundamental knowledge of the advective process and help provide improved modeling. The technical aspect that distinguishes this work is the development of sound analytical and numerical treatment of advection by extremely intermittent stochastic advection velocities.

This year we have completed research in the following areas:

- An intermittency model for passive scalar turbulence
- Anomalous scaling and structure instability in three-dimensional (3-D) passive scalar turbulence (see figure)
- Kinematic effects on local energy dissipation rate
- Inertial range scalings of dissipation and entrophy in isotropic turbulence
- A refined similarity hypothesis for transverse structure functions

Publications

Cao, N., and S. Chen, "An Intermittency Model for Passive Scalar Turbulence," *Phys. Fluids* **9**, 1203 (1997).

Chen, S., and N. Cao, "Anomalous Scaling and Structure Instability in Three-Dimensional Passive Scalar Turbulence," *Phys. Rev. Lett.* **78**, 3459 (1997).

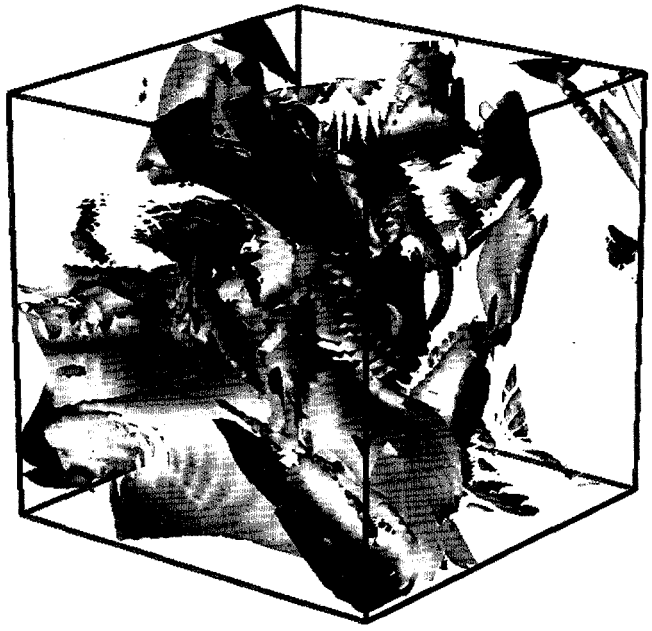
Chen, S., and R.H. Kraichnan, "Inhibition of Turbulence Cascade by Sweep," *J. Plasma Phys.* **57**, 187 (1997).

Chen, S., et al., "Inertial Range Scalings of Dissipation and Entrophy in Isotropic Turbulence," *Phys. Rev. Lett.* **79**, 1253 (1997).

Chen, S., et al., "A Refined Similarity Hypothesis for Transverse Structure Functions," *Phys. Rev. Lett.* **79**, 2253 (1997).

Doolen, G.D., et al., "Energy Spectrum in the Dissipation Range of Fluid Turbulence," *J. Plasma Phys.* **57**, 95 (1997).

Kraichnan, R., "Passive Scalar: Scaling Exponents and Realizability," *Phys. Rev. Lett.* **78**, 22 (1997).



Isosurface of the scalar dissipation at the value of 10 times the average dissipation from a simulation of 3-D turbulent diffusion.

Elliptic Solvers for Adaptive Mesh Refinement Grids

Daniel Quinlan

The goal of this project is to develop multigrid methods that will efficiently solve elliptic problems with anisotropic and discontinuous coefficients on adaptive grids. The final product will be a library that provides for the simplified solution of such problems.

The focus of this work is research on serial and parallel elliptic algorithms and the inclusion of our black-box multigrid techniques into this new setting. The approach applies the Los Alamos object-oriented class libraries that greatly simplify the development of serial and parallel adaptive mesh refinement applications.

This year our efforts have been focused on both numerical issues and the development of the adaptive mesh refinement (AMR) technology onto parallel architectures. The numerical work has focused on how to improve the performance of the parallel AFACx algorithm and reduce the amount of previously inherent serial behavior; this work is still in progress.

Other numerical work has been done to resolve issues of the use of the elliptic solvers specific to time-dependent problems in which optimal time steps are used, namely, the efficient solution of the intermediate levels at time steps different from those of the global grid. Our development work within the parallel environment has focused on simple demonstrations of the working parallel infrastructure.

We published several papers this year as part of the work over the previous years on this LDRD research. One paper was on parallel load balancing, which was an important part of the support for AMR within the parallel environment. Another paper was specific to the use of the parallel infrastructure we are using within AMR++ for our parallel AMR work. A third and fourth paper were specific to the use of AMR++ and its design for use within complex geometry applications. A fifth paper, currently in preparation, outlines the use of AMR++ within the broader context of overlapping grid computations. As a note, the presentation of the combined work at the 1997 SIAM Parallel Conference resulted in an award for best poster session.

Publications

Balsara, D., and D. Quinlan, "Parallel Object-Oriented Adaptive Mesh Refinement" (SIAM Conference on Parallel Processing and Scientific Computing, Minneapolis, MN, March 1997).

Brown, D., et al., "OVERTURE: An Object-Oriented Software System for Solving Partial Differential Equations in Serial and Parallel Environments" (SIAM Conference on Parallel Processing and Scientific Computing, Minneapolis, MN, March 1997).

Quinlan, D., "AMR++: A Design for Parallel Object-Oriented Adaptive Mesh Refinement" (IMA Workshop on Structured Adaptive Mesh Refinement, Minneapolis, MN, March 1997).

Quinlan, D., et al., "MLB: Multilevel Load Balancing for Structure Grid Applications" (SIAM Conference on Parallel Processing and Scientific Computing, Minneapolis, MN, March 1997).

Advanced Algorithms for Information Science

Daniel Strottman

In a modern, information-controlled society, the importance of fast computational algorithms facilitating data compression and image analysis cannot be overemphasized. For these algorithms to be useful, they need to meet a variety of criteria: they need to yield high data compression rates but still be of low computational complexity to allow for fast implementation. They should be robust with respect to data loss and error (possibly arising from noisy or faulty transmission). And with the increasing availability of multiprocessor and parallel systems, they should allow for easy parallelization. It was the purpose of this project to assess the Laboratory's capabilities in this area and to begin the effort to enhance and exploit them.

Feature extraction and pattern recognition are key to many Los Alamos projects, and the same types of dimensionality reduction and compression used in source coding are also applicable to image understanding. We have begun developing wavelet coding that decomposes data into different length-scale and frequency bands. New transform-based source coding techniques that use joint optimization techniques appear in practice to offer potential for achieving better combined source-channel coding performance.

We initiated work on a system that not only compresses the video stream in real time, but that also takes the additional step of analyzing the video stream concurrently. By using object-based compression schemes (where an object is an identifiable feature of the video signal, repeatable in time or space), we found that the analysis is directly related to the efficiency of the compression.

A Self-Consistent Multiscale Theory of Internal-Wave, Mean-Flow Interactions in the Ocean

Darryl Holm

Our project is directed toward applications of nonlinear dynamics and kinetic theory in developing new model equations and better analysis tools for nonlinear problems, such as ocean dynamics, involving multiple time and length scales. At present it is not feasible to resolve all these scales and their interactions in numerical simulations. We have chosen a fundamental approach to describing the effects of these interactions; that is, we are developing a self-consistent multiscale theory of internal-wave, resolved-flow interactions.

This year we studied the general theory of reduction of variational principles with respect to their invariance groups. These group-reduced variational principles are mathematically interesting because they involve constraints on the allowed variations, analogous to what we find in the theory of nonholonomic systems with the Lagrange-d'Alembert principle. These equations generalize earlier work by Poincaré for dynamics on a Lie algebra, in that they depend on a parameter. The parameter in fluid dynamics examples is interpreted as advected, or Lie-dragged, as with the density in compressible fluid flow. In addition, we derived the basic abstract theorem about fluid circulation for these variational equations, which we call the Kelvin-Noether theorem.

We also derived a series of approximate models of ocean circulation dynamics by using asymptotic expansions of Hamilton's principle for the most accurate theory in the small dimensionless parameters that typically appear in the large-scale, rapidly rotating situations characteristic of oceanographic applications. Our approach to deriving these approximate models is particularly effective because it preserves the invariance properties of the action principle that are responsible for the Kelvin-Noether theorem. Thus, each of the approximate model equations we derive for internal-wave, mean-flow interactions in the ocean possesses its own circulation theorem and its own attendant conservation law for potential vorticity, which is an important and useful central concept in geophysical fluid dynamics at every level of approximation.

Publications

Allen, J.S., et al., "A Note on Kelvin Waves in Balance Models," *J. Phys. Ocean* **27**, 2060 (1997).

Cendra, H., et al., "The Maxwell-Vlasov Equations in Euler-Poincaré Form" (submitted to *J. Math. Phys.*).

Holm, D.D., and V. Zeitlin, "Hamilton's Principle for Quasigeostrophic Motion" (to be published in *Phys. Fluids A*).

Holm, D.D., et al., "The Euler-Poincaré Equations and Semidirect Products with Application to Continuum Theories" (submitted to *Advances in Mathematics*).

Designing a Micromechanical Transistor

Ronnie Mainieri

Micromechanical electronic systems are chips with moving parts. They are fabricated with the same techniques that are used to manufacture electronic chips, sharing their low cost. Micromechanical chips can also contain electronic components. The combination of mechanical parts makes it possible for chips to process signals mechanically. To achieve designs comparable to those obtained with electronic components, it is necessary for these chips to have a mechanical device, i.e., a mechanical transistor, that can change its behavior in response to a small input.

In this project, we are using the geometrical ray to develop the design tools for these complex-shaped resonant structures. To overcome the limitations of geometrical ray chaos, we are studying the dynamics of the rays by using the methods developed for the study of nonlinear dynamical systems. This work will lead to our development of numerical methods that execute well in parallel computer architectures, using a limited amount of memory and no interprocessor communication.

We have shown that the amount of computational work required to compute the nth acoustic resonance of a system to a fixed precision grows exponentially with n. This is an important result as it shows that the techniques being developed in the project may be the only method for the reliable computation of resonances. The other result is that we have extended the ray technique from scalar waves to vector waves, as needed for typical mechanical vibration problems. We introduced a very unusual coordinate system and a moving-frame coordinate system into the ray technique and found that the conditions for resonance in the vector case are the same as in the scalar case.

Dispersive, Internal, Long-Wave Models

Roberto Camassa

This project is a joint analytical and numerical study of internal, dispersive, water-wave propagation in a stratified two-layer fluid, a problem that has important geophysical fluid dynamics applications. During the past year, we studied our model equations for a stratified two-layer fluid in the fully nonlinear regimes for a finite and infinite depth of the lower layer.

In particular, we derived closed-form traveling-wave solutions of the fully nonlinear model for finite depth configuration and showed that they are graphically indistinguishable from the numerically computed solutions of the Euler equations. This result confirms that the dynamics supported by our model has the potential of accurately reproducing that of the original Euler system. Since solutions to Euler equations can only be obtained numerically and are notoriously expensive to compute, it is extremely interesting to observe that our model's accuracy can be achieved over a wide range of wave amplitudes.

For sufficiently long time scales, dictated by the fluid viscosity, no inviscid model can compare favorably with experimental observations. Therefore, we have undertaken the task of introducing viscous corrections in some of our models, and we have begun to study them analytically and numerically. An unexpected result from this preliminary work was the discovery of small, shelflike waves that propagate at speeds much higher than those of the primary waves and whose existence solely results from viscosity. This suggests that the usual paradigm of inviscid models providing instantaneously a good approximation to the solution of the viscous system might need some reexamining.

We have reported these results in a comprehensive article that we recently submitted to the *Journal of Fluid Mechanics*, and we are currently preparing another article concerning the viscous effects.

Publications

Camassa, R., and W. Choi, "Fully Nonlinear Internal Waves" (submitted to *J. Fluid Mech.*).

Camassa, R., and W. Choi, "Long Internal Waves of Finite Amplitude," *Phys. Rev. Lett.* **77**, 1759 (1996).

Camassa, R., and W. Choi, "Nonlinear Internal Waves in a Two-Layer Fluid," *J. Fluid Mech.* **313**, 83 (1996).

Camassa, R., and C.D. Levermore, "Layer-Mean Quantities, Local Conservation Laws, and Vorticity," *Phys. Rev. Lett.* **78**, 650 (1997).

Camassa, R., et al., "Long-Time Effects of Bottom Topography in Shallow Water," *J. Fluid Mech.* **349**, 173 (1997).

Camassa, R., et al., "Long Time Evolution Equations for Shallow Water with a Varying Bottom," *Physica D* **98**, 258 (1996).

Choi, W., "On the Fission of Algebraic Solitons," *Proc. R. Soc. Lond. A* **453**, 1753 (1997).

Particles and Patterns in Cellular Automata

Erica Jen

Cellular automata are discrete dynamical systems consisting of lattices of sites whose values evolve according to a local interaction rule. Although simple in construction, cellular automata generate a diverse range of highly complicated spatiotemporal behavior. Cellular automata are now widely recognized to be powerful simulation tools for the modeling of certain types of complex behavior, such as percolation through porous media, dendritic growth of crystals, or the spread of disease through a population. The objective of our work is to develop methods for analyzing the relationship between an automaton's microscopic-level specification through its rule table and certain macroscopic features—including pattern formation and computational capabilities—that characterize its evolution.

This year our research focused on the decomposability of an automaton into multiple automata of smaller interaction radius. The primary results obtained relate to the relationship between decomposability and the ability to perform computational tasks such as synchronization and density classification. We established the fundamental decomposability of a specific rule of high computational ability. We are presently exploring the implications of decomposability for the nature of particles and domains generated by the automaton rules.

Another aspect of our research focused on the spatiotemporal behavior of two-dimensional cellular automata. Extending our earlier work on one-dimensional cellular automata, we are investigating techniques for discovering and characterizing the domains, domain boundaries, and their interactions in the spatiotemporal dynamics of a two-dimensional cellular automaton. The long-term objective of this work is to predict the computational capabilities of a two-dimensional cellular automaton from the macroscopic features of its behavior.

Fiber-Optic Communications Using Solitons (FOCUS)

Darryl Holm and Benjamin Luce

Industry is currently developing soliton-based optical-fiber networks in the hope of sending many gigabits per second of digital data over fiber-optic links between computers and into homes and businesses. Such networks would take advantage of the intrinsic stability of soliton solutions of the nonlinear Schrodinger equation, which to leading order describes the evolution of optical pulses in a lossless fiber. However, even in the highest-purity fibers, unavoidable energy losses cause soliton pulses to decay and disperse, thereby limiting data transmission rates. To overcome this dispersion, devices are needed that can reshape pulses. We have discovered that such devices can be designed to actually exploit losses and dispersion in order to stabilize optical pulse transmission. These results indicate that a much wider class of nonlinear pulse behavior, far beyond that of the leading-order soliton solutions, is of great practical interest. We are studying this wider class of behavior and are applying our results directly to the theory of fiber-optic networks. Our study ranges from the analysis of individual devices to the modeling and optimization of complete networks.

In the past year, we made significant progress in developing a perturbation method for studying the nonlinear partial differential equations that govern the propagation of optical-fiber pulses. We will soon publish a series of three papers (the first three listed publications) on these developments. Additionally, we have been studying various types of pulse transmission techniques. This year we discovered a very promising power enhancement phenomenon using a technique called sliding-frequency guiding filters. Specifically, we found that the use of super-Gaussian filters in this technique produces a profound enhancement of the pulse energy for a fixed pulse width.

Publications

Cruz-Pacheco, G., et al., "Complex Ginzburg-Landau Equations as Perturbations of Nonlinear Schrodinger Equations: A Case Study" (submitted to *Physica D*).

Cruz-Pacheco, G., et al., "Complex Ginzburg-Landau Equations as Perturbations of Nonlinear Schrodinger Equations: A Melnikov Approach" (submitted to *Physica D*).

Cruz-Pacheco, G., et al., "Complex Ginzburg-Landau Equations as Perturbations of Nonlinear Schrodinger Equations: Traveling Wave Persistence" (submitted to *Physica D*).

Cruz-Pacheco, G., and B.P. Luce, "On the Relationship of Periodic Wavetrains and Solitary Waves of Complex Ginzburg-Landau Type Equations" (to be published in *Phys. Lett. A*).

Gabitov, I., et al., "Low Noise Picosecond Soliton Transmission Using Concatenated Nonlinear Amplifying Loop Mirrors," *JOSA B* **14**, 1850 (1997).

Luce, B.P., "Power Enhancement with super-Gaussian Sliding-Frequency Guiding Filters" (submitted to *Opt. Lett.*).

3-D, Unstructured, Hexahedral-Mesh S_n Transport Methods

Jim E. Morel

The purpose of this work is to develop a discrete-ordinates, or S_n, method for solving the radiation transport equation on three-dimensional (3-D), hybrid, finite-element meshes consisting of arbitrary combinations of hexahedra, wedges, pyramids, and tetrahedra. Many fundamental difficulties arise in solving the discrete-ordinates equations on this type of mesh because the cell faces are generally curved rather than flat. We believe that we have effective numerical strategies to deal with these difficulties. The use of hybrid finite-element meshes will result in a unique and powerful capability for modeling complex, 3-D geometries.

This year we developed a trilinear-discontinuous spatial-discretization scheme for the S_n equations on hybrid finite-element meshes, together with an algorithm for solving these equations via source iteration. One important component of this solution technique is an algorithm for ordering the flux unknowns so that the associated source-iteration matrix is lower-triangular. Another important component is a diffusion-synthetic algorithm for accelerating the convergence of the source iterations. We implemented this algorithm in a computer code, called PERICLES, which runs on workstation platforms.

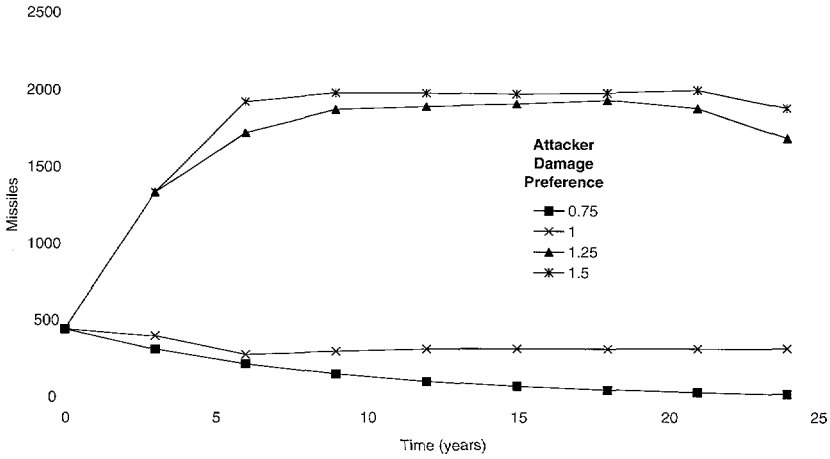
Advanced Techniques for the Analysis of Nuclear-Crisis Stability, Deterrence, and Latency

Gregory Canavan

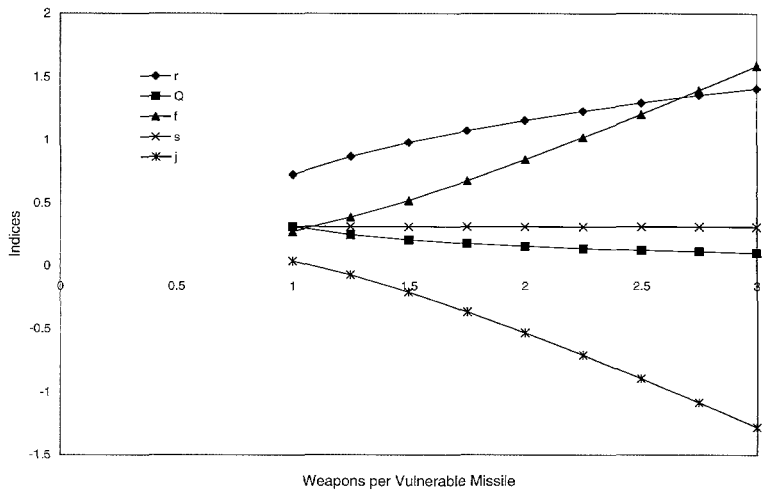
The major goal of this project was improving strategic stability and deterrence theory by building time dependence and nuclear latency into bipolar stability analysis. We fully met this goal. Our study established the first connection with Russian analyses and extended them to large force levels to study the transition from multiple independently targetable reentry vehicles to singlet missiles. It showed that defenses decrease the stability at intermediate levels but improve it for large levels, that stability falls for large attack preferences (a major concern of Russian analyses), and that the growth and decay of missile forces is controlled by attack preferences (see first plot).

We found that using the conventional objective of "unacceptable damage" removes some freedom in the attack optimization and produces attacks on military targets at low missile levels, and we found that unilateral US weapons reductions would increase the overall stability at the expense of US first and second strikes. In addition, hidden Russian weapons would formally increase the stability, reductions in vulnerable weapons and missiles would decrease the US stability and the overall stability during the transition, and maximizing the overall stability rather than minimizing the first-strike cost would increase the stability but would require that the first striker degrade the effectiveness of the strike.

We achieved automatic attack optimization by analytic optimization within linear subspaces of the nonlinear overall exchange space. This process of linearization and optimization, which is asymptotically exact for deep reductions, is sufficiently accurate to bridge the gap between the reductions specified in the Strategic Arms Reduction Treaty and the deep reductions ultimately required. Stability indices are proportional to the difference between the second and first strikes by vulnerable forces. Thus, increasing the number of weapons per vulnerable missile decreases the stability, and the number of latent weapons required is not large (see second plot).



The optimal growth or decay of missiles with time depending on the attack damage preference (that is, the attacker's relative preference for inflicting damage on others vs preventing self-damage).



A plot showing how the stability varies with *m*, the number of weapons per vulnerable missile. The stability index *j* increases to zero as *m* approaches unity. The stability index is affected by changes in the number of weapons attacking each missile (*r*), the probability that these weapons survive (*Q*), the number of weapons used for first strikes on military targets (*f*), and the number of surviving weapons per missile (*s*).

Quantum Information Capacity in the Presence of Noise

Raymond Laflamme

Within the past two years, researchers have found that specific mechanical techniques of error correction can protect quantum computation against errors, at least to some extent. In fact, we have recently established that by using hierarchical ("concatenated") quantum codes, one can successfully perform arbitrarily long computations, as long as the rate at which errors occur per elementary operation is below a certain threshold. Rigorous upper bounds on this tolerable error rate range from 10^{-6} to 10^{-12} errors per operation, but numerical simulations suggest that thresholds as forgiving as 10^{-3} may be obtained, if errors are predominantly of a certain single type (the "depolarizing channel").

We studied small quantum-error-correcting codes in view of their applications to a nuclear magnetic resonance (NMR) quantum computer. Our aim was to understand quantum information capacity in the presence of noise, for small numbers of quantum bits ("qubits"), both analytically and by using computer simulations. We performed a systematic study of the effects of decoherence and noise on a small number of qubits. We investigated in detail the performance of the perfect 5-bit code and the 7-bit code in the presence of specific types of noise. We also examined how well these codes perform when subjected to spontaneous emission, phase decoherence, and systematic errors induced during unitary transformations. We also investigated how quantum-error correction can be implemented in a specific quantum computer implementation, the one using NMR.

Publications

Knill, E., et al., "Resilient Quantum Computation" (to be published in *Science*).

Laflamme, R., et al., "NMR GHZ" (submitted to *Phys. Rev. Lett.*).

The Fundamental Role of Solitons in Nonlinear Differential Equations

James Hyman

Numerical simulations and mathematical analysis have proved crucial to understanding the fundamental role of solitons in the evolution of general initial data for quasi-linear, dispersive partial differential equations, such as the Korteweg-de Vries (KdV), nonlinear Schrödinger, and the Kadomtsev-Petviashvili equations. These equations are all integrable and have linear dispersion, and the solitons have infinite support. Integrability and the infinite number of conservation laws help explain the extraordinary stability of solitons.

We have discovered a class of solitary waves with compact support (which we call compactons) that are solutions of a multiparameter family of fully nonlinear, dispersive partial differential equations. Compactons are solitary waves with the remarkable soliton property that after colliding with other compactons, they reemerge with the same coherent shape. These particle-like waves exhibit elastic collisions that are similar to the soliton interactions associated with completely integrable equations supporting an infinite number of conservation laws.

Compacton equations, however, are not integrable; they satisfy only a handful of conservation laws. Thus, it is not expected that the compactons will remain coherent when two of them collide. The robustness of these compactons and the inapplicability of the inverse scattering tools, which worked so well for the KdV equation, make it clear that a nonlinear mechanism causes these structures to be robust.

This past year, we derived a new KdV-like Hamiltonian equation with nonlinear dispersion from a four-parameter Lagrangian. This equation supports a new class of solitons with compact support (compactons) as well as the usual (noncompact) solitons. The new equation is not integrable and only conserves mass, momentum, and energy; yet, the solitons display the same modal decompositions and structural stability observed in integrable partial differential equations.

We found explicit formulas for multiple classes of compact, traveling-wave solutions for this equation. When more than one solitary-wave solution exists for the equation, we have observed that the wider compacton is a minimum of the Hamiltonian and is the only one that is stable. In addition, there is a two-parameter subclass of these equations in which the dispersion balances the advection such that the width of the compactons is independent of the amplitude.

Our research opens the way to a far-reaching and new understanding of the central role of solitons in nonlinear dispersion.

Publications

Hyman, J., and P. Rosenau, "Pulsating Multiplet Solutions of Quintic Wave Equations" (to be published in *Physica D*).

Hyman, J., et al., "Compacton Solutions in a Class of Generalized Fifth Order Korteweg-de Vries Equations" (submitted to *Physica D*).

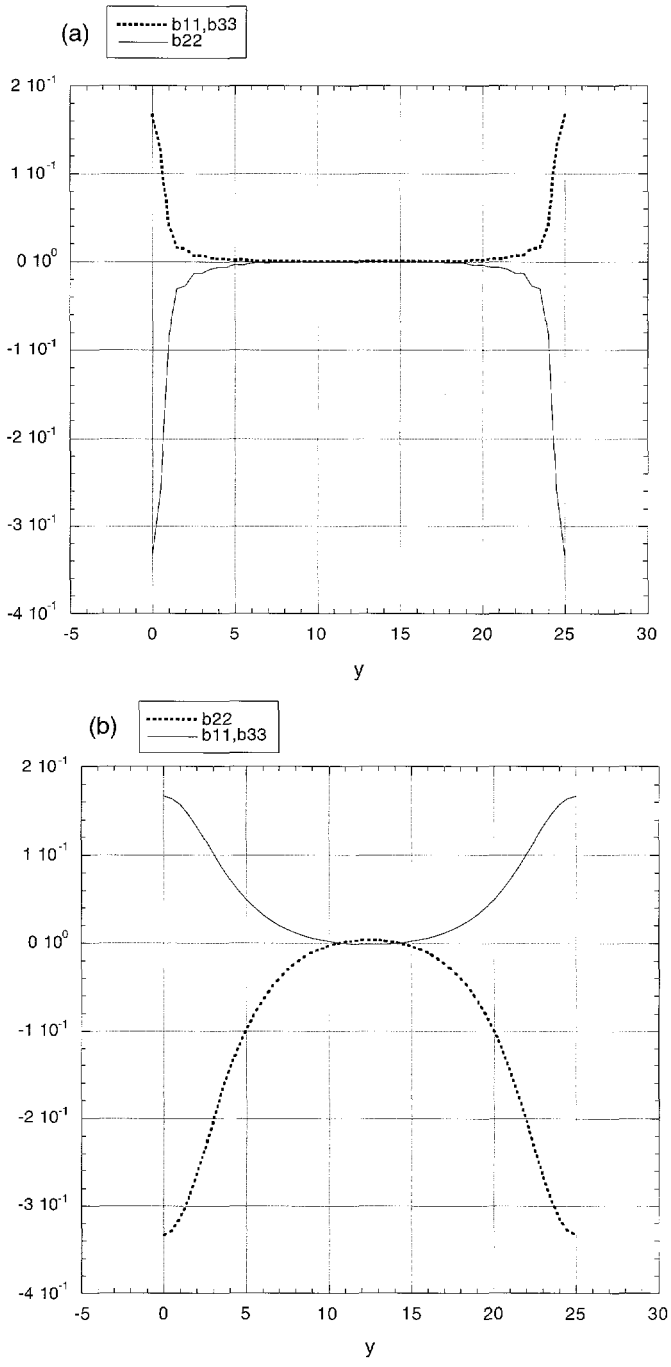
A Theoretical Description of Inhomogeneous Turbulence

Leaf Turner

We are developing a new formalism for describing inhomogeneous turbulence by using a helicity representation that we used previously for describing anisotropic, but homogeneous, turbulence. We are also interested in applying our formalism to the magnetohydrodynamic dynamo problem, with applications to astrophysics and magnetic fusion. This work will likely have engineering applications including drag forces on transportation vehicles, energy efficiency in automotive combustion, meteorological phenomena, forest fires, eruptions of volcanoes, and weapons physics.

Using a helicity-based Galerkin representation of flows, we studied and compared the dynamics of evolving fluid turbulence in three geometries (three-dimensional, homogeneous; two-dimensional, homogeneous; and three-dimensional inhomogeneous turbulence of free-slip channel flow) using three standard models of statistical evolution of a viscous, incompressible fluid. We have coded one of these models (a so-called eddy-damped quasi-normal Markovian model) to analyze the evolution of turbulence in a free-slip channel.

Highlights of what we learned are the following: (1) We can use past data of evolving energy spectra of homogeneous, isotropic, mirror-symmetric turbulence to find the corresponding evolution of turbulence in the physical coordinate space of free-slip channel flow, which will be neither homogeneous nor isotropic. (2) We obtained formulas for the Reynolds stress and the mean pressure gradient in terms of the energy spectrum for the free-slip channel flow (see accompanying figures). (3) We demonstrated that the random-phase approximation yields excellent agreement with the results



The nonvanishing elements of the normalized anisotropic Reynolds stress tensor, b{ij}, are shown as a function of the position y between the boundaries that are located at y = 0 and y = 25. When the turbulence is isotropic, these elements vanish. In (a), at time $t = 0.507$, the elements are nearly null. In (b), at $t = 12.3$, the presence of the boundaries has caused the anisotropy of the Reynolds stress to become significant. Even at the midpoint, there is a slight anisotropy.

of an average of over 16 direct numerical simulations of a free-slip channel flow.

Publications

Turner, L., "Helicity Representation of Evolution of Incompressible Turbulence. I. Arbitrary Homogeneous, Anisotropic, Helical Case" (submitted to *J. Fluid Mech.*).

Turner, L., "Helicity Representation of Evolution of Incompressible Turbulence. II. Inhomogeneous Case—Free-Slip Channel" (submitted to *J. Fluid Mech.*).

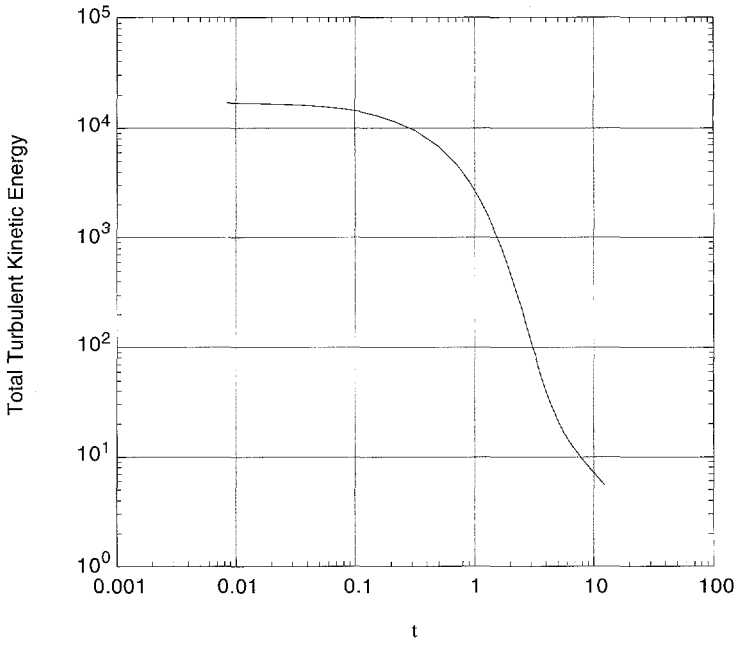
Turner, L., "Helicity Representation of Evolution of Incompressible Turbulence. III. Direct-Interaction Approximation and Test-Field Models of Two- and Three-Dimensional Homogeneous and Free-Slip Channel Cases" (submitted to *J. Fluid Mech.*).

Turner, L., "Incompressible, Inhomogeneous Turbulence: Lassoed in a Slab" (ICASE Colloquium, NASA/Langley Research Center, Hampton, VA, October 9, 1996).

Turner, L., "Inhomogeneous Turbulence: A Laboratory for the Principles of Homogeneous Turbulence" (Seminar to the Center for Turbulence Research, Stanford University, Palo Alto, CA, July 16, 1997).

Turner, L., "The Principles of Homogeneous Turbulence: A Beacon for a Theoretical Description of Inhomogeneous Fluid Turbulence" (Minisymposium on MHD and Related Issues, SIAM 45th Anniversary Meeting, Stanford, CA, July 14–17, 1997).

Turner, L., "A Yellow-Brick Road from Homogeneous to Inhomogeneous Turbulence" (Fiftieth Annual Meeting of the Division of Fluid Dynamics, San Francisco, CA, November 23–25, 1997).



The associated turbulent kinetic energy as a function of time.

New Ways of Representing Functions

George Zweig

In this project we are creating new sets of expansion functions that efficiently represent functions with structure at many scales. Unlike the wavelets of continuous wavelet transforms, the new sets of expansion functions have a longest scale and do not have similar shapes at all scales. We have developed fast transform algorithms based on the new expansion functions by solving a partial differential equation for the transform coefficients. We expect the new functions to find applications in pattern recognition and noise removal.

The primary question addressed during the first year of the project was the following: Under what conditions can expansion functions have separate dependences on both their characteristic frequencies and times? Wavelet transform expansion functions, for example, depend only on the product of these two variables. Perhaps our most interesting finding is that we can construct a broad class of transforms that are in some sense the square root of a weighted average of the square of a wavelet transform and the square of a moving Fourier transform. The weighting is such that functions containing primarily low frequencies are transformed with a moving Fourier transform and those containing primarily high frequencies are transformed with a wavelet transform. The underlying expansion functions have complex dependences on the characteristic frequencies and times but simple forms at both high and low characteristic frequencies.

The chimeric transform described above has all the desirable mathematical properties that we seek, but efficient ways to compute this transform are not known. Next year we will find efficient ways to compute this transform and search for additional classes of expansion functions.

Geometry in the Large and Hyperbolic Chaos

Brosl Hasslacher

In the study of strongly chaotic systems, global geometrical information from the original dynamical system is entangled in an unknown way throughout the one-dimensional shift map projection. The calculation of observables by using orbit expansions is blocked by the lack of a workable orbit classification for strong chaotic systems. Our research uses insights from geometrical methods in modern mathematics and recent connections between global geometry and modern quantum field theory to study the natural geometrical objects belonging to hard chaos, i.e., hyperbolic manifolds.

We have attempted to embed strong chaotic systems in higher dimensional spaces by exploiting their connection to knot theory. The relationship between a knot and its space complement—a hyperbolic manifold—has led to the study of the monodromy groups of representations for knot polynomials. The action of each group element on the punctured Poincaré disc can be interpreted as a class of dynamical systems. Utilizing the analogy between group elements and dynamical systems allows one to consider the case with an infinite number of punctures.

Our main target was to now come at the problem from the infinite degree-of-freedom case, using flavors of topological field theories whose classical limits are strongly chaotic systems. Our initial approach was a metric-free one, so we dealt with chaotic geometries described by such quantum field theories. These geometries appeared to be exactly integrable in the sense of Duestermat-Heckman functional integrals over equivariant cohomologies.

High-Quality Finite-Difference Schemes for Partial Differential Equations and Discrete Vector and Tensor Analysis

Mikhail Shashkov

Our primary goal in this project is to develop new, general techniques for performing large-scale numerical simulations based on approximating the solution of partial differential equations. The basis of these new techniques is the design of discrete operators that preserve certain essential properties of, and relationships between, their corresponding analytic operators. The techniques will significantly extend the well-known and useful finite-volume methods and are designed to more faithfully represent important properties of physical processes and the continuum mathematical models for such processes. Algorithms based on these techniques can be used for modeling high-speed flows, porous-media flows, diffusion processes, and geophysical flows.

This year we constructed a full system of discrete analogs of the invariant first-order operators—div, grad, and curl—on a logically rectangular grid and proved a discrete analog of an orthogonal decomposition theorem. That is, we proved that any discrete vector can be presented as a sum of a discrete gradient of some scalar function and a discrete curl of a vector function. Using discrete analogs of divergence and gradient, we developed new, local finite-difference schemes for two- and three-dimensional diffusion equations with a discontinuous, nondiagonal matrix of coefficients on a logically rectangular grid. We also tested our new schemes on practical problems. Based on our theoretical investigations, we have constructed new, conservative finite-difference methods for Lagrangian gas dynamics that do not contain parasitic modes. We also suggested a new algorithm for local reconstruction of a vector field from its components on the faces of grid cells. Such reconstruction gives us a tool to compare different spaces of discrete vector functions.

Publications

Caramana, E.J., and M. Shashkov, "Elimination of Artificial Grid Distortion and Hourglass-Type Motions by Means of Lagrangian Subzonal Masses and Pressures" (submitted to *J. Comput. Phys.*).

Caramana, E.J., et al., "The Construction of Compatible, Mimetic Hydrodynamics Algorithms Utilizing Conservation of Total Energy" (submitted to *J. Comput. Phys.*).

Caramana, E.J., et al., "Formulation of Artificial Viscosity for Multi-Dimensional Shock Wave Problems" (submitted to *J. Comput. Phys.*).

Hyman, J., and M. Shashkov, "The Adjoint Operators for the Natural Discretizations of the Divergence, Gradient, and Curl on Logically Rectangular Grids," *IMACS J. Appl. Numer. Math.* **25**, 1 (1997).

Hyman, J., and M. Shashkov, "Discrete Analogs of Invariant First-Order Operators on Logically Rectangular Grids," *Int. J. Comput. Math. Appl.* **33**, 81 (1997).

Hyman, J., and M. Shashkov, "The Orthogonal Decomposition Theorems for Mimetic Finite Difference

Methods" (submitted to *SIAM J. Numer. Anal.*).

Hyman, J., et al., "The Numerical Solution of Diffusion Problems in Strongly Heterogeneous Non-Isotropic Materials," *J. Comput. Phys.* **132**, 130 (1997).

Morel, J.E., et al., "A Local Support-Operator Diffusion Discretization Scheme for Quadrilateral r-z Meshes" (submitted to *J. Comput. Phys.*).

Shashkov, M., et al., "Local Reconstruction of a Vector Field from its Normal Components on the Faces of Grid Cells" (to be published in *J. Comput. Phys.*).

Completely Parallel ILU Preconditioning for Solution of Linear Equations

Wayne Joubert

We propose to develop completely parallel versions of incomplete Cholesky and incomplete lower and upper (ILU) triangular decomposition preconditioners for solving sparse systems of linear equations. The solution of linear equations is the most computationally expensive part of many modeling and simulation processes. However, the parallelization of popular serial preconditioners such as ILU has been problematic. The purpose of this project is to (1) develop parallel versions of these algorithms for structured-grid problems; (2) perform basic research on the convergence properties and parallelization possibilities of ILU preconditioners; and (3) explore the possibility of applying these parallelization techniques for structured problems to the preconditioning of unstructured problems.

During the past year, we completed the development and investigation of parallel ILU preconditioning for three-dimensional (3-D), seven-point stencil matrices, a case that is in heavy use in structured-grid modeling and simulation codes. We also developed a new, effective parallelization strategy and working code for general, 3-D structured problems. The strategy makes it possible both to perform parallel ILU preconditioning for highly accurate finite-difference discretizations and to use level fill-in preconditioning, which is very important for solving difficult, structured simulation problems.

We performed an exhaustive orderings study for a class of small structured-grid problems. The insights we obtained from this study have led to the development of several new theoretical results that explain the limitations of certain parallel grid orderings. These results suggest ways to improve convergence of reordered, parallel ILU preconditionings.

Publications

Joubert, W., et al., "Fully Parallel Global M/ILU Preconditioning for 3-D Structured Problems" (submitted to *SIAM J. Matrix Anal. Applications*).

Solution Adaptive Method for Low-Speed Flows and All-Speed Flows

Jeffrey Saltzman

Our objective is to develop methods to efficiently solve fluid problems that involve low-speed flows and those that involve a mixture of low-speed and high-speed flows. An example of a flow problem that can be solved with such methods is that found in the combustion chamber of a diesel engine. The approach we take extends recent advances in techniques for incompressible flows to low Mach numbers.

We have developed two approaches for solving all-speed flows. The first approach is a novel extension of the Godunov-projection algorithm for incompressible flows. We decompose the velocity and pressure fields into two new fields, one representing the incompressible part of the flow and the other holding the compressible effects. To avoid having a small time step based on the speed of sound, we advance the compressible part implicitly. Extensive numerical convergence tests have verified the accuracy and convergence properties of the method, and we are in the final stages of preparing a paper on this scheme.

The second approach we have developed couples a partially implicit method for low Mach numbers with a high-order Godunov method for intermediate and high Mach numbers. The partially implicit method requires the solution of only one scalar elliptic problem. In the limit of small Mach numbers, this method reduces to the standard approach for the incompressible equations. This second approach has been incorporated into a general-purpose solver for overlapping grids with moving geometries in two and three space dimensions. Significant progress has been made toward treating chemically reacting flows in a quite general fashion by utilizing the Chemkin package.

Combinatorics, Geometry, and Mathematical Physics

William Chen

Combinatorics and geometry have been among the most active areas of mathematics over the past few years because of newly discovered interrelations between them and their potential for applications. In this project, we set out to identify problems in mathematical physics where these methods could impact significantly. In particular, our experience suggested that the areas of unitary symmetry and discrete dynamical systems could be brought more strongly under the purview of combinatorial methods. Unitary symmetry deals with the detailed description of the quantum mechanics of many-particle systems, and discrete dynamical systems deal with chaotic systems.

The depth and complexity of the mathematics in these physical areas of research suggested not only that significant advances could be made in these areas but also that there would be a fertile feedback of concept and structure to enrich combinatorics itself. During the three years of this project, we have confirmed these projections by a number of discoveries that have been published in some two dozen papers and reported at a number of international conferences. In this third-year progress report, we list these publications from 1996 forward, except for those publications listed in last year's progress report. We have made substantive contributions in the cross-fertilization of combinatorics and its applications.

Publications

Beyer, W.A., and J.D. Louck, "Transfinite Function Iteration and Surreal Numbers," *Adv. Appl. Math.* **18**, 333 (1997).

Chen, W.Y.C., "The Skew, Relative and Classical Derangements," *Discrete Math.* **160**, 235 (1996).

Chen, W.Y.C., and Z.G. Liu, "Parameter Augmentation for Basic Hypergeometric Series, II," *J. Combin. Theory, Series A* **80**, 175 (1997).

Chen, W.Y.C., and J.D. Louck, "Enumeration of Cubic Graphs by Inclusion-Exclusion" (submitted to *J. Combin. Theory, Series A*).

Chen, W.Y.C., and J.D. Louck, "Necklaces, MSS Sequences and DNA Sequences," *Adv. Appl. Math.* **18**, 18 (1997).

Chen, W.Y.C., et al., "The Double Schur Function" (submitted to *J. Alg. Combin.*).

Louck, J.D., "Combinatorial Aspects of the Representations of the Unitary Group," in *Proc. Fourth SSCTP,* T. Lulek, et al., Eds. (World Scientific, Singapore, 1997), p. 231.

Louck, J.D., "Conway Numbers and Iteration Theory," *Adv. Appl. Math.* **18**, 181 (1997).

Louck, J.D., "Doubly Stochastic Matrices in Quantum Mechanics" (to be published in *Found. Phys.*).

Louck, J.D., "Future of the Theory of Angular Momentum: Discrete Mathematics and Combinatorics" in *Metropolis Memorial Volume* (Birkhauser, Boston, MA, in press).

Louck, J.D., "Power of a Determinant with Two Physical Applications" (to be published in *Int. J. Math. Math. Sci.*).

Louck, J.D., "Problems in the Theory of Words on Two Letters Originating from Discrete Dynamical Systems," *Ann. Combin.* **1**, 88 (1997).

Louck, J.D., "Unitary Symmetry, Combinatorics and Generating Functions" (to be published in *Discrete Math.*).

Louck, J.D., "Unitary Symmetry, Combinatorics, and Special Functions," *Group 21: Physical Applications and Mathematical Aspects of Geometry, Groups, and Algebras* **1**, 50 (1997).

Atomic and Molecular Physics and Plasmas, Fluids, and Particle Beams

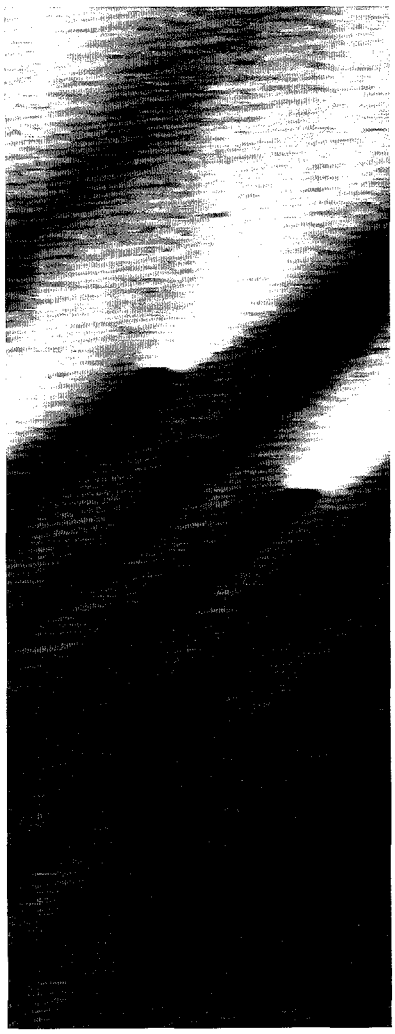
Geometric Phase, Spatial Resonance, and Control in Spatially Extended Nonlinear Systems

Robert Ecke

Thermal convection with rotation provides a unique system for studying novel phenomena in nonlinear dissipative systems. In particular, a one-dimensional, nonlinear traveling wave that we discovered is well described by the Complex Ginzburg-Landau (CGL) equation. This thermal wave is confined near the walls of a cylindrical convection cell and is effectively one-dimensional in the azimuthal coordinate. It has azimuthal symmetry in the base state and is characterized by a wavelength and a wave speed. Studying this simple, one-dimensional system will offer insight into real-world problems such as the geophysical coupling at the earth's core-mantle boundary, the interaction of storms with mountain ranges, and industrial materials processing in which controlling spatial homogeneities is important.

This year we used our thermal wave to study several experiments. The first was an experimental search for the geometrical phase in dissipative systems, which was previously shown to apply to nondissipative (frictionless) systems, both quantum mechanical (Berry Phase) and classical (Hannay Angle). The second experiment was a model for topographic forcing of the wave motion that can arise in geophysical systems, such as the waves that form on the surface of the earth's core. Finally, the thermal wave will respond to a more general application of temporally and spatially distributed perturbations, allowing for the possibility of controlling a spatially extended nonlinear system. Such control is extremely important in manufacturing, in process control in chemical engineering, and in the broader area of engineering control theory and application.

We experimentally determined the parameters of the CGL equation, demonstrated the agreement between theory and experiment for the Eckhaus-Benjamin-Feir instability, and measured for the first time the properties of the phase diffusion of wave-number distortions (see accompanying figure). We have numerically calculated the linear stability for the traveling-wave mode and derived a phase equation that includes higher-order terms necessary because of the symmetry breaking of rotation. We have shown experimentally that these higher-order terms account for the small discrepancies with the pure CGL description.



Grey-scale-coded, wave-number distortions (the dark grey, angled stripes) of nonlinear traveling waves in rotating convection. The vertical scale is time; the horizontal is distance. The distortions grow until they induce dislocation nucleation in the waves to relieve the local wave-number stress (i.e., until two waves coalesce, seen here as the abrupt termination of the first and third stripes).

Publications

Liu, Y., and R.E. Ecke, "Eckhaus-Benjamin-Feir Instability in Rotating Convection," *Phys. Rev. Lett.* **78**, 4391 (1997).

A Target-Plasma Experiment for Magnetized Target Fusion

Frederick Wysocki

Magnetized target fusion (MTF) is an approach to controlled fusion in which a premagnetized, preheated target plasma is near-adiabatically compressed to fusion conditions. The objective of this project is to develop the ability to generate suitable target plasmas for MTF. Our approach involves driving a 1-MA fast-rising current through a cryogenically frozen deuterium fiber of about 200 μm in diameter. The fiber rapidly turns to plasma, heats, and expands to fill a plasma-containment chamber, thus becoming confined by the chamber walls. This project relies heavily on existing Los Alamos facilities and equipment that are adapted to our needs. Our MTF target-plasma generator is shown in the accompanying cross-sectional diagram.

We have continued the plasma-formation experiments that we started in FY 1996. These experiments use a static-fill of hydrogen gas. This year we made a HeCd-laser interferometer operational and measured the plasma density (n_e) as a function of time (see first plot). These measurements show the density rising to at least 1×10^{18} cm^{-3} in a period of 1 μs. After this time, the interferometer signal is lost, most likely because of beam desteering that results from spatial gradients in the plasma density.

In addition, we used filtered and unfiltered silicon photodiodes to determine soft x-ray emission properties (see plots of diode current vs time). These data indicate that the hot plasma cools off in 8–10 μs and that the maximum plasma temperature may be roughly 50–100 eV. We have measured plasma parameters vs capacitor-bank charge voltage, and these measurements show increased plasma lifetime and higher light emission (perhaps implying higher temperature) with increased charge voltage.

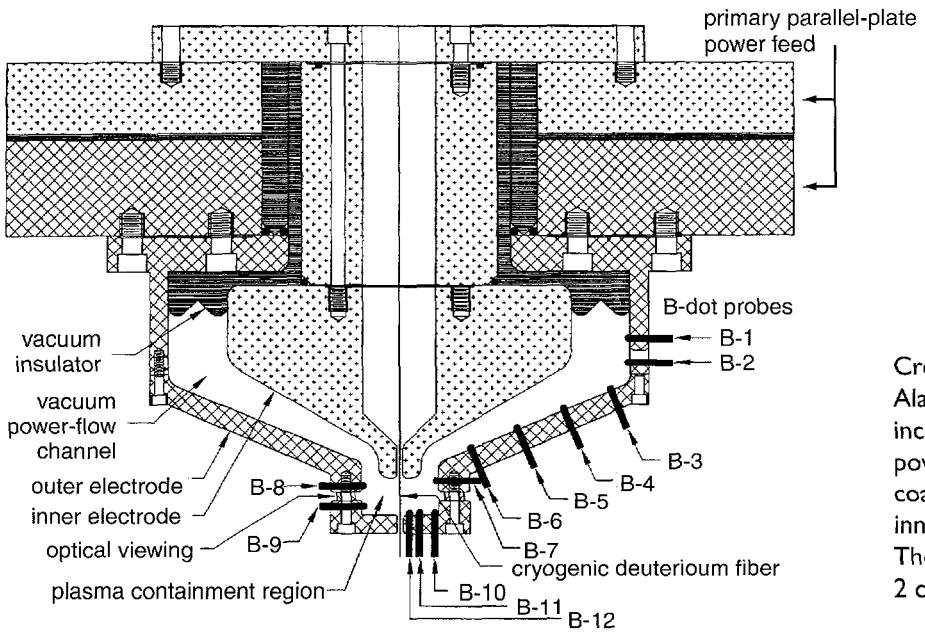
During the past year we have also performed experiments using electrodes coated with 10-mil-thick tantalum. These electrodes gave minor improvements over our past experiments, as inferred from the soft x-ray emission. The cryostat used for making the cryogenically frozen deuterium fiber has been successfully operated on a separate test stand, and we will install it on the Los Alamos Colt capacitor-bank facility as soon as the adapter pieces arrive.

Publications

Lindemuth, I.R., et al., "Magnetic Compression/Magnetized Target Fusion (MAGO/MTF): A Marriage of Inertial and Magnetic Confinement" (16th IAEA/Plasma Physics and Controlled Nuclear Fusion Research Conference, Montreal, Canada, October 7–11, 1996).

Lindemuth, I.R., et al., "MAGO/MTF: Controlled Thermonuclear Fusion with Existing Pulse Power Systems?" (11th IEEE Pulsed Power Conference, Baltimore, MD, June 29–July 2, 1997).

Lindemuth, I.R., et al., "Role of Z-Pinches and Other Related Configurations in MAGO/MTF, a Low-Cost Approach to Controlled Thermonuclear Fusion" (4th International Conference on Dense Z-Pinches, Vancouver, Canada, May 28–30, 1997).



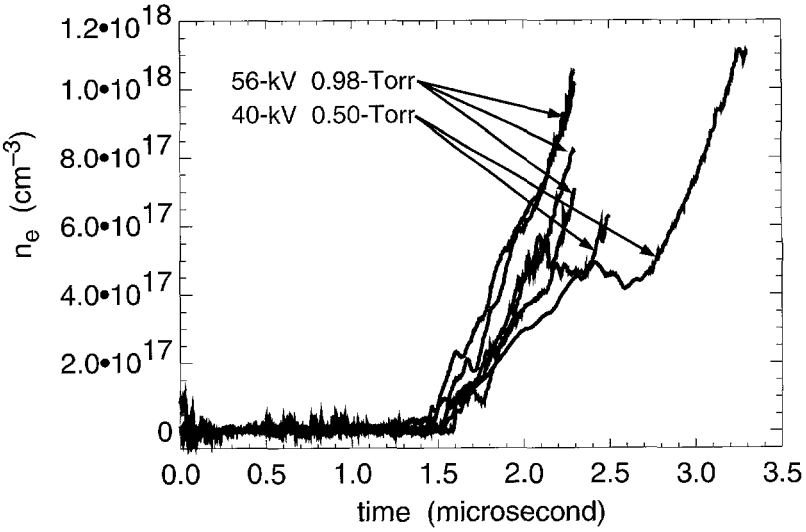
Cross-sectional view of the Los Alamos MTF target-plasma generator, including the primary parallel-plate power feed, region of conversion to coaxial feed, electrical insulators, and inner and outer coaxial electrodes. The plasma-containment region is 2 cm in radius × 2 cm high.

Sheehey, P.T., et al., "Computational Modeling of Wall-Supported Dense Z-Pinches" (DOE Office of Fusion Energy Innovative Confinement Concepts Workshop, Marina del Ray, CA, March 3–6, 1997).

Sheehey, P.T., et al., "Computational Modeling of Wall-Supported Dense Z-Pinches" (4th International Conference on Dense Z-Pinches, Vancouver, Canada, May 28–30, 1997).

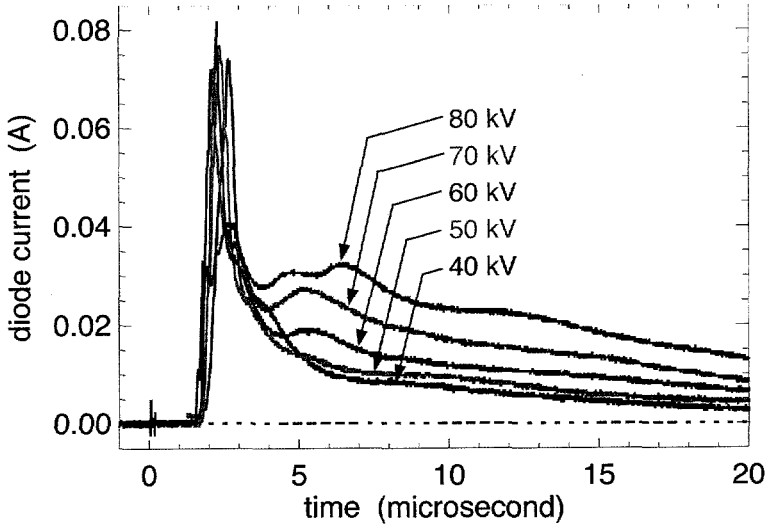
Wysocki, F.J., et al., "Progress with Developing a Target for Magnetized Target Fusion" (1997 IEEE International Conference on Plasma Science, San Diego, CA, May 19–22, 1997).

Wysocki, F.J., et al., "Progress with Developing a Target for Magnetized Target Fusion" (11th IEEE Pulsed Power Conference, Baltimore, MD, June 29–July 2, 1997).

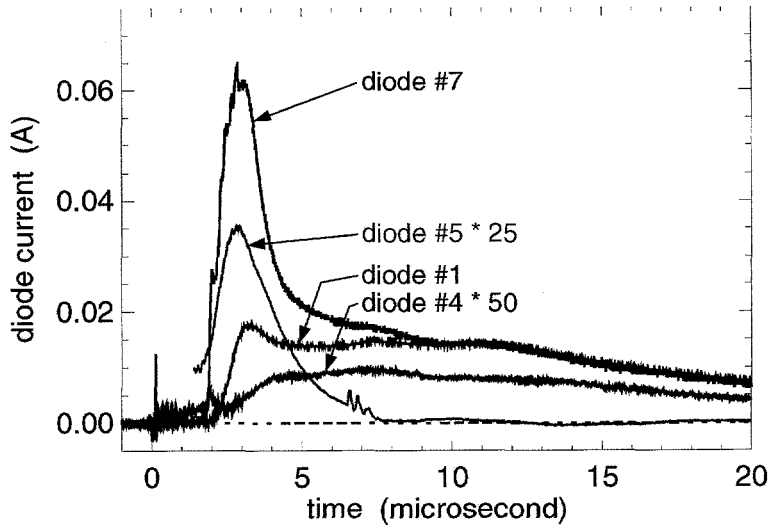


Plasma density (n_e) data from the HeCd-laser interferometer. The four upper curves are from discharges with an erected bank voltage of 56 kV, whereas the two lower curves are from discharges at 40 kV. These data show the density rising to at least 1×10^{18} cm^{-3} within a period of 1 μs.

Unfiltered silicon photodiode data for discharges ranging in erected bank voltage from 40 kV (lowest curve at 10 μs) to 80 kV (highest curve at 10 μs). These data show increased plasma lifetime and higher light emission (implying higher temperature) with increased bank voltage.



Filtered photodiode data for discharges with an erected bank voltage of 66 kV. In order of decreasing magnitude of the peak signal (as plotted with the stated multipliers) is diode #7 (unfiltered), diode #5 with a 452-mg/cm^2 nickel filter, diode #1 with a 66,000-mg/cm^2 polystyrene filter (which allows only visible light through), and diode #4 with a filter of 323-mg/cm^2 kimfol ($C_{16}H_{14}O_3$) plus 10-mg/cm^2 aluminum. These data indicate that the hot plasma cools off in 8–10 μs.



High-Intensity Laser-Matter Interaction Physics

James Cobble

The objective of this project is to examine the penetration of a high-intensity laser beam (greater than 10^{18} W/cm^2) into a preformed plasma. The motivation for this is to investigate basic assumptions of the fast-ignitor-fusion concept. In the first year, we measured the transmission and backscatter of a 1.054-μm infrared laser into plasmas of various densities. We found that when the laser irradiance exceeds a critical value, the beam is refracted such that its effective f-number is increased; that is, the beam tends to be less divergent upon exiting the plasma than it would be if the plasma were not there. This effect is due to electron cavitation—the expelling of electrons out of the region of higher intensity by the ponderomotive force of the beam.

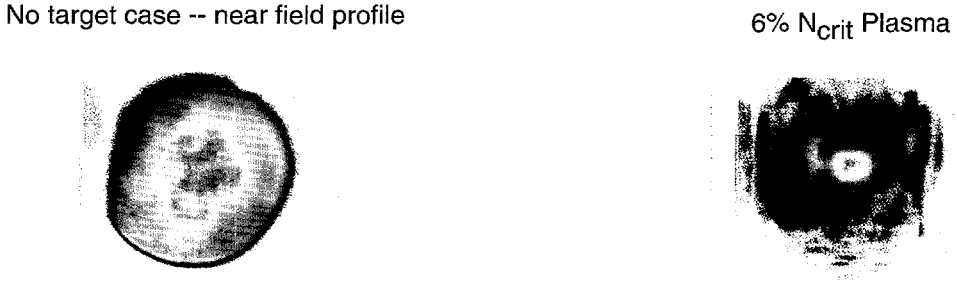
In this second year of the project, we repeated the experiment with green light at 0.527 μm. We discovered that the shorter-wavelength light behaves in a way similar to the 1-μm laser: the f-number is increased when the critical power is exceeded (see accompanying figure). We developed a model for scaling the transmission, which is actually higher for the green light than for the infrared light even though in the former case the absolute plasma density is four times higher at a constant fraction of the critical density. Our experimental results suggest that the modeling is valid.

Publications

Cobble, J.A., et al., "Exploded Foil Transmission Experiments with an Ultra-Intense Laser" (26th Anomalous Absorption Conference, Fairbanks, AK, August 28, 1996).

Cobble, J.A., et al., "f number of a High Intensity Green Laser Beam in a Plasma," *Bull. Am. Phys. Soc.* **42**, 1839 (1997).

Cobble, J.A., et al., "High-Intensity Illumination of an Exploding Foil," *Phys. Plasmas* **4**, 3006 (1997).



Near-field channeling of a green beam indicating a four-fold increase in the f-number. The left-hand profile shows the beam with no target plasma, and the right-hand profile shows the beam passing through a plasma at 6% of the critical density (n_{crit}).

Dynamics of Quantum Wave Packets

Timothy R. Gosnell

The goal of this work is to utilize existing ultrafast laser technology and atomic and molecular physics expertise to demonstrate the coherent control of wave-packet dynamics in the reactions of small molecules.

The time-domain investigation of localized vibrational wave functions in diatomic molecules is possible with the use of femtosecond laser sources. We have developed an experiment in which a femtosecond pump pulse creates a wave packet on an excited potential surface of gas-phase K_2 molecules. The packet then oscillates within the well, much as one expects classically, but disperses in time as a result of rotational effects and the mild anharmonicity of the potential well. The objective of the experiment is not just to create and observe such packets but to control their shape by manipulating the amplitude and phase of the exciting pump pulse.

This year we further improved the signal-to-noise ratio in the wave-packet detection apparatus and remeasured the chirp-induced effects on the shape of K_2 packets. Specifically, we observed wave-packet focusing during the second period of return of the wave packet. To analyze the data, we used a radical new approach called pixon-based reconstruction, which represented its first application outside of the astronomical community. This approach significantly improved data quality. Our previous theoretical investigation had helped clarify the specifics of the wave-packet dynamics, but this year we increased the sophistication of our model by adopting a Liouville-equation approach, which has the advantage of providing a natural way to include finite temperature and rotational effects.

Determination of Optical-Field Ionization Dynamics in Plasmas through Direct Measurement of the Optical Phase Change

Antoinette Taylor

The interaction of ultrashort, high-intensity laser pulses with solid and gas targets is a field of growing interest. New ultrafast diagnostics are needed to measure the ionization and subsequent plasma dynamics that give rise to these conditions. The goal of this project is to develop a technique for characterizing laser-plasma interactions with femtosecond resolution based on the direct measurement of the phase change of an optical pulse. We will use this technique to investigate femtosecond field ionization dynamics in atomic and molecular gases and to study solid-target laser-plasma interactions.

This year we performed the first femtosecond coherent spectroscopy experiments of ultrafast high-field ionization in atmospheric noble gases and simple molecules both near and above the barrier ionization threshold. When the barrier suppression threshold is below the focal intensity, a blueshifted diagonal is observed in the time-resolved frequency shift which is strongest for xenon (see the accompanying figure). When the barrier suppression threshold is above the focal intensity, strong cross-phase modulation is observed with red-shifting/blueshifting on the leading/trailing edge of the pump. Two aspects of the results are unexpected and represent potentially new physics: (1) The previously hypothesized contribution of collisional ionization to ionization-front blueshifting is inconsistent with our results, whereas the role of the neutral gas appears to be enhanced. (2) We observe highly structured redshifts on the leading edge of the pump with a 25-fs period suggestive of ultrafast coherent oscillations in electronic structure near ionization.

Publications

Downer, M.C., and C.W. Siders, "Ultrafast Terawatt Laser Sources for High-Field Particle Acceleration and Short Wavelength Generation," *Adv. Accel. Concepts* **7**, 214 (1997).

Kane, D.J., et al., "Simultaneous Measurement of Two Ultrashort Laser Pulses from a Single Spectrograph," *J. Opt. Soc. Am. B* **14**, 935 (1997).

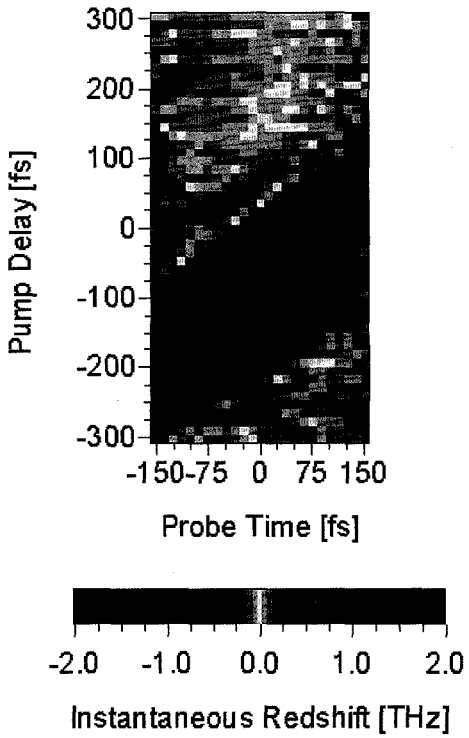
Siders, C.W., and A.J. Taylor, "Femtosecond Coherent Spectroscopy at 800-nm" (submitted to *Phys. Rev. Lett.*).

Siders, C.W., and A.J. Taylor, "Multi-Pulse Interferometric Frequency-Resolved Gating: Coherent Spectroscopy with the Ultrafast Boxcar Advantage," in *Proceedings of Ultrafast Optics '97* (Optical Society of America, Washington, DC, 1997), p. 29.

Siders, C.W., et al., "First Measurements of Laser Wakefield Oscillations by Longitudinal Interferometry," *Adv. Accel. Concepts* **7**, 372 (1997).

Siders, C.W., et al., "Measurement of Ultrashort Pulses Using Fast Principal Component Generalized Projections" (submitted to *Opt. Lett.*).

Siders, C.W., et al., "Multi-pulse Interferometric Frequency Resolved Gating: Real-Time Imaging of Ultrafast Dynamics," *Opt. Lett.* **22**, 624 (1997).



Time-resolved instantaneous frequency shift of a 175-fs, 800-nm probe pulse in 600-torr xenon due to a 400-nm pump pulse. Horizontal axis is time within the probe pulse, whereas the vertical axis is pump-probe delay time. The frequency shifts are predominately to higher frequency. The dark diagonal from lower left (pump on trailing edge of probe) to upper right (pump on leading edge of probe) corresponds to the ionization front.

Laser Cooling of Solids

Timothy R. Gosnell

The purpose of this project is to develop materials and techniques for the laser cooling of solids. By the mechanism of anti-Stokes fluorescence in Yb^{3+}-doped fluorozirconate glass, we have demonstrated the cooling of a condensed phase for the first time. It is the primary objective of this project to demonstrate laser cooling at as low a temperature as possible. Some of our additional objectives are to develop thermodynamic models of the cooling process, understand the fundamental limits of the cooling process, and develop new materials for optimizing the cooling effect.

In the past year, we established a new world record for a laser-cooled solid. The temperature reached was 21 K below room temperature. We also discovered that performance of the cooling mechanism was within 0.1% of the ideal performance. Moreover, we noticed the presence of optical saturation, a phenomenon exhibited by the cooling ions, whose effect was to reduce the cooling power and hence the temperature drop. We constructed a new apparatus for performing cooling measurements. This apparatus consists of a vacuum chamber pumped by an "oil-less" turbomolecular vacuum system, an ionization pressure gauge, a residual gas analyzer, a more accurate thermometer, and a charge-coupled-device detector array cooled by liquid nitrogen.

Publications

Fajardo, J.C., et al., "Electrochemical Purification of Heavy-Metal Fluoride Glasses for Laser-Induced Fluorescence Cooling Applications," *J. Non-Cryst. Solids* **213**, 95 (1997).

Mungan, C.E., et al., "Internal Laser Cooling of Yb(3+)-Doped Glass Measured Between 100 and 300 K," *Appl. Phys. Lett.* **71**, 1458 (1997).

Mungan, C.E., et al., "Laser Cooling of a Solid by 16 K Starting from Room Temperature," *Phys. Rev. Lett.* **78**, 1030 (1997).

Mungan, C.E., et al., "Spectroscopic Determination of the Expected Optical Cooling of Ytterbium-Doped Glass," *Mater. Science Forum* **239**, 501 (1997).

Plasma-Wakefield Accelerator

Bruce Carlsten

The purpose of this project is to demonstrate key aspects of an electron-driven plasma-wakefield accelerator that are required to move this technology from a laboratory experiment to a practical device. The technology holds promise for reducing the overall length of the accelerators required in high-energy physics applications. Our specific goals are to (1) demonstrate accelerating gradients on the order of 1 GV/m, (2) demonstrate large transformer ratios between the decelerating gradient seen by the drive pulse and the accelerating gradient seen by the witness pulse, and (3) demonstrate acceleration linearity in the blowout regime (where the witness pulse is accelerated for applications downstream).

Our efforts this year were focused on modifying the drive laser, developing a theoretical understanding of the anomalous ionization of the gas from the train of drive electron bunches, and fabricating a differential plasma-density interferometer. For our plasma-wakefield experiment, drive-laser stability needs to be improved and a smaller witness pulse has to follow the larger drive pulse. Toward these improvements, we brought the drive laser closer to the accelerator and have designed the witness-pulse optics.

In terms of our second focus, the experimentally observed ionization occurs much faster than what is predicted by simple ionization theory. We now understand that the major physical result is that the plasma electrons are radially accelerated by the radial electric field of the drive bunch, increasing ionization by about a factor of 20. Finally, the differential plasma-density interferometer we are fabricating is based on using the differential phase of laser light through the plasma. It will measure the buildup of plasma density from shot to shot. Understanding and measuring the ionization buildup is critical to the performance of the plasma-wakefield accelerator in the blowout regime.

Delta-f and Hydrodynamic Methods for Semiconductor Transport

Lester E. Thode

One of the major problems encountered in applied research is the need to develop new codes or use existing codes in order to test new approaches. The development or modification of such codes often dominates the research effort, thereby diminishing the amount of time that can be devoted to the new ideas. That is why we have developed a more efficient approach toward the development and testing of new algorithms.

The two major thrusts of our research were (1) to develop a proof-of-principle approach for a plug-and-play architecture with application to research in gas-core nuclear rockets, neutron diffusion, magnetic-coil design, and high-power accelerators and (2) to investigate the possibility of applying the delta-f method to semiconductor transport in order to cut the high computational costs associated with the traditional Monte Carlo method. With respect to the delta-f method, our approach is the numerical analog of the Chapman-Enskog expansion of statistical mechanics, in which the unknown distribution described by a Boltzmann equation is divided into a local equilibrium part and a deviation. It thus becomes possible to treat the nonequilibrium character of the distribution function more accurately and with less computation and noise.

This year, we had three objectives: (1) to complete the integration of a Monte Carlo code into the prototype plug-and-play environment, (2) to develop the diagnostics necessary to evaluate the transport coefficients needed for the delta-f method, and (3) to introduce a delta-f method into the common environment. We met the first milestone by integrating the drift diffusion, energy balance, hydrodynamic, and Monte Carlo models into the plug-and-play environment. To complete the second milestone, we developed new diagnostics for the higher-order transport moments. In one dimension, a sufficient number of electrons could be utilized, and thus we could obtain a reasonable approximation for the higher-order transport coefficients. However, in two dimensions, the noise level remained large, meaning that the fluid equations would be driven by a noisy source. Estimates indicated that without smoothing, which would have led to a smearing of the depletion region, the noise would have propagated into the fluid solution.

Based on these evaluations, we concluded that, in two and three dimensions, the delta-f approach for microelectronics was questionable. For this reason, we abandoned our third objective. Nonetheless, a major success was the demonstration of a plug-and-play environment in which a large number of existing FORTRAN, C, and C++ programs could be integrated, leading to common methods for input, output, restart, graphics, and algorithm development. This environment is now being used extensively in gas-core nuclear rocket research for NASA and high-power accelerator research in the weapons program.

Publications

Thode, L.E., et al., "Vortex Formation and Stability in a Scaled Gas-Core Nuclear Rocket Configuration" (submitted to *J. Propulsion and Power*).

Strongly Coupled Dusty Plasmas

Dan Winske

This project uses several unique experimental, computational, and theoretical capabilities developed at Los Alamos to investigate fundamental aspects of strongly coupled dusty plasmas.

Dusty plasmas are ionized gases that have been contaminated by small (micron-size) particulates. In the presence of the ionized gas (plasma), the particulates become electrically charged, and the interaction between these charged particles can exhibit strongly correlated behavior. For example, under some conditions, the particulates can form a regular lattice array. As the conditions of the plasma are changed, the system can make a phase transition from the solid to the liquid or gaseous state.

During the first year of this project, we carried out three-dimensional molecular dynamics simulations of plasma crystal formation using a simplified physics model. We also began to investigate how plasma flowing through the crystal affects the crystal's structure. We designed and built an experiment to study plasma crystal formation and started preliminary measurements of the plasma properties.

Publications

Murillo, M.S., et al., "MD Simulations of Dusty Plasma Crystal Formation: Preliminary Results" in *Strongly Coupled Coulomb Systems* (Plenum Press, New York, in press).

Winske, D., and M. Rosenberg, "Nonlinear Development of the Dust Acoustic Instability in a Collisional Dusty Plasma" (to be published in *IEEE Trans. Plasma Sci.*).

Transient Quantum Mechanical Processes

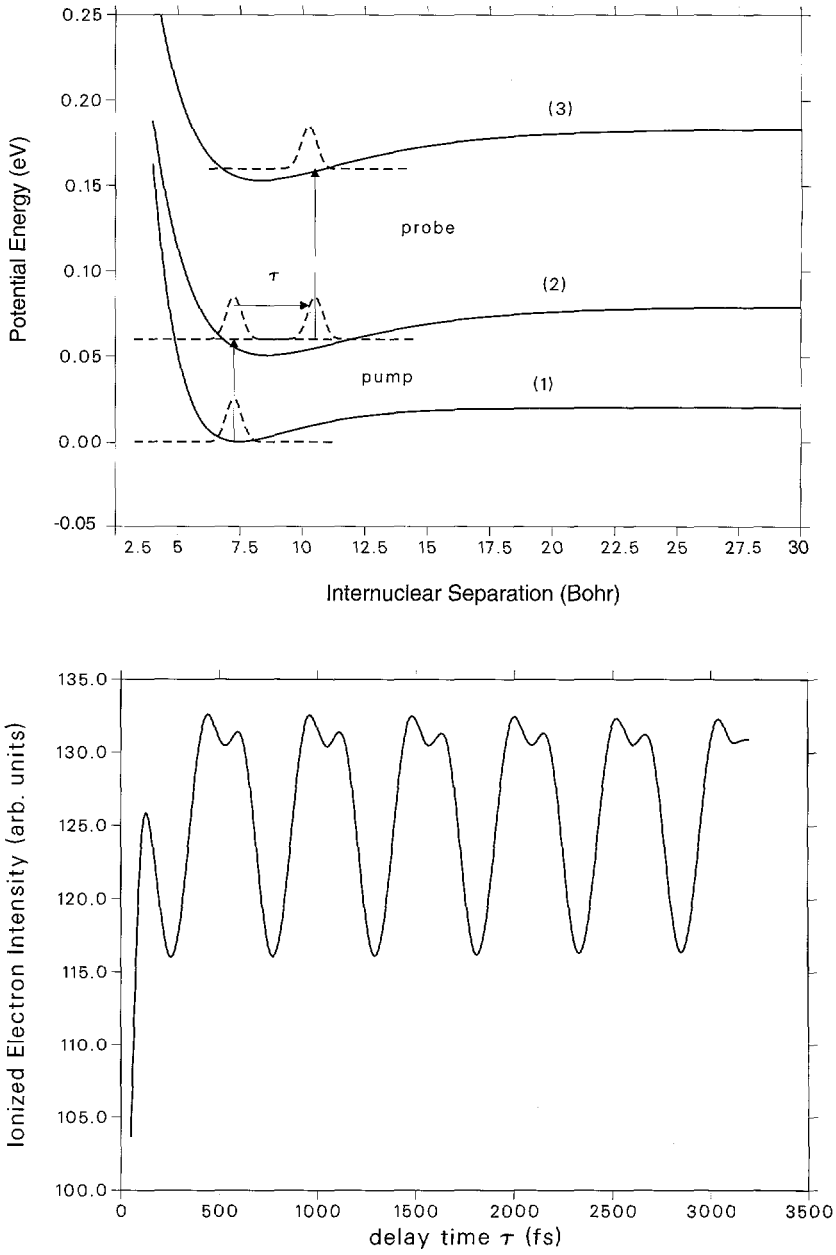
Lee Collins

Publications

Collins, L.A., et al., "Control of Molecular Processes," *Phys. Electron. At. Collisions* **20**, 54 (1997).

We continued our investigation of transient quantum mechanical processes using sophisticted methods to solve the time-dependent Schrödinger equation, which governs the evolution of such processes. Two mechanisms have commanded particular attention: (1) the control of molecular processes by laser interactions and (2) the tunneling of electrons through quantum dots. The first mechanism has served as a test bed for our basic approaches. In particular, a change in the shape and arrival time of the laser pulse may induce a related change in the outcome of the molecular process. Such changes not only provide an intricate probe of the fundamentals of quantum mechanics but also may lead to ways of modifying chemical reactions.

We have developed two independent, general methods to solve the Schrödinger equation and performed calculations with them on the pump-probe interaction of two laser pulses with a diatomic potassium molecule. The first pulse excites the molecule into a state that begins to vibrate; the second pulse catches the molecule at some stage of its vibrational excursion and ionizes it. Varying the delay time between the two pulses allows us to map the quantum mechanical path of the vibrating system through the detected electrons, as seen in the accompanying figure. Most quantum mechanical measurements produce only averaged quantities; in our simulation, we can actually follow the system's temporal evolution. We have found a strong frequency dependence in the shape of the electron signal, which has also been observed in experiments at Los Alamos, and have shown that a multistate treatment of the contiuum is important to obtaining quantitatively accurate results.



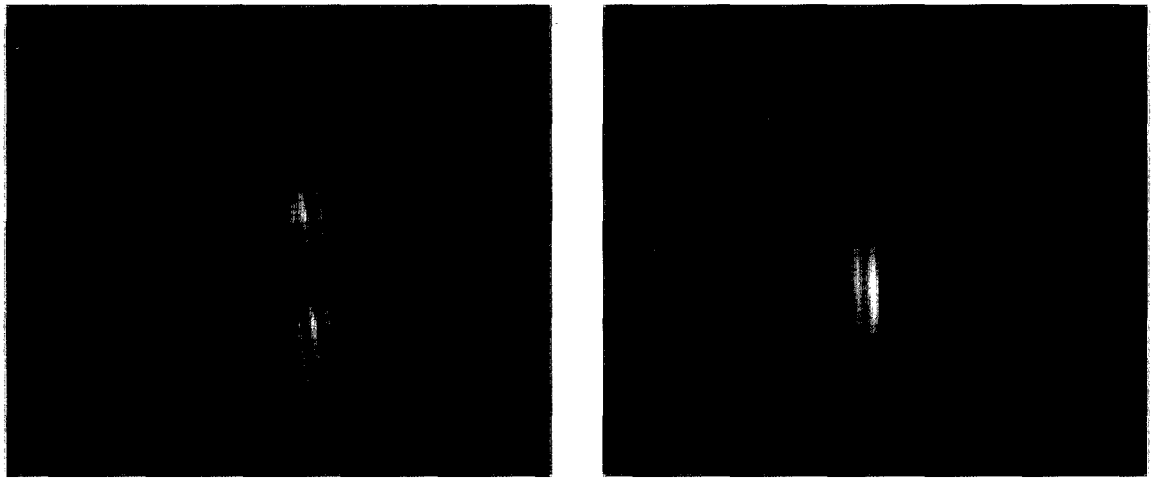
(Top) A pump laser pulse excites the wave packet of the ground-state molecule to an upper state [(1)→(2)]. The wave packet in the upper state begins to move. At a delay time (τ), a probe laser pulse catches the wave packet at its new position and excites it to the ground state of the molecular ion (3), thereby releasing an electron. The intensity of the electron signal, which can be experimentally measured, is proportional to the probability of the wave packet being in state (3). (Bottom) The electron signal as a function of delay time gives direct information on the position and shape of the wave packet in the upper state [i.e., state (2)]. Changing the form of the laser pulses can affect the nature of the upper-state wave packet, allowing control of its basic properties.

Optical Wave Packets (Optical Bullets): A New Diffraction-Free Form of Light Travel

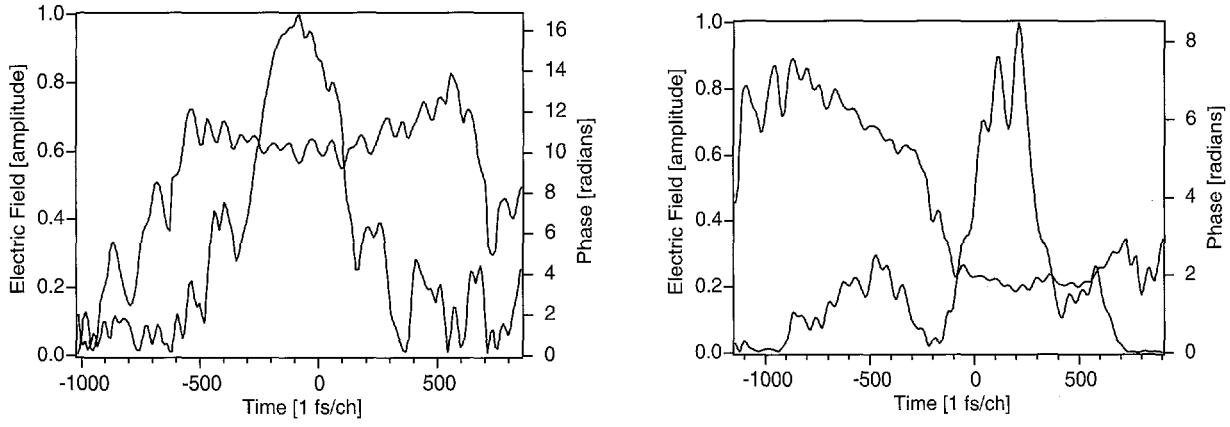
David Funk

We are studying the propagation of self-confined packets of light through the atmosphere. These packets are self-forming and require no active optics or guiding media. At sufficiently high intensities, light will not only self-focus but will propagate as fine filaments until the intensity of the pulse falls below a measurable critical value. We are analyzing the phase changes and structure of the pulse above, near, and below the critical value to resolve the origin of this phenomenon. Our studies have concentrated on the demonstration and understanding of the fundamental physics associated with this phenomenon and the proof of the existence of these wave packets in air.

In the past year, we analyzed frequency-resolved optical-gating (FROG) data and are presently comparing the data with numerical simulations of the nonlinear Schrödinger equation performed by our collaborators at the University of Arizona (see figures). We upgraded the laser so that it will now be capable of generating 60-mJ, 150-fs pulses, allowing us to study the effects of scaling on the filamentation process.



Experimental FROG trace (left) and reconstruction (right). The interference fringes arise from the pulse splitting.



Electric field envelope and phase at two distances (3.4 m on the left and 11.9 m on the right). The splitting is absent at 3.4 m but is clearly evident in the pulse at 11.9 m.

A Compact Compton-Backscattering X-Ray Source for Mammography and Coronary Angiography

Dinh Nguyen

Our goal is to develop a compact, tunable, monochromatic x-ray source for use in mammography and coronary angiography. Using monochromatic x-rays in mammography will reduce the patient's x-ray dose and at the same time enhance the film image by reducing the unwanted scatter. Last year's objective was to test a new Compton-backscattering design that can generate a large flux of monochromatic x-rays. Our new approach is through Compton backscattering with a regenerative amplifier free-electron laser (FEL). This new FEL approach is capable of generating a large flux of infrared photons to be used in the Compton-backscattering process.

We built a specially designed wiggler that maintained the electron beam in a small, focused spot throughout the scattering region. We completed a design and fabricated the retroreflection optics to overlap the infrared photons with the electrons in both space and time. These advanced techniques result in a significant increase in the Compton-backscattering x-ray flux.

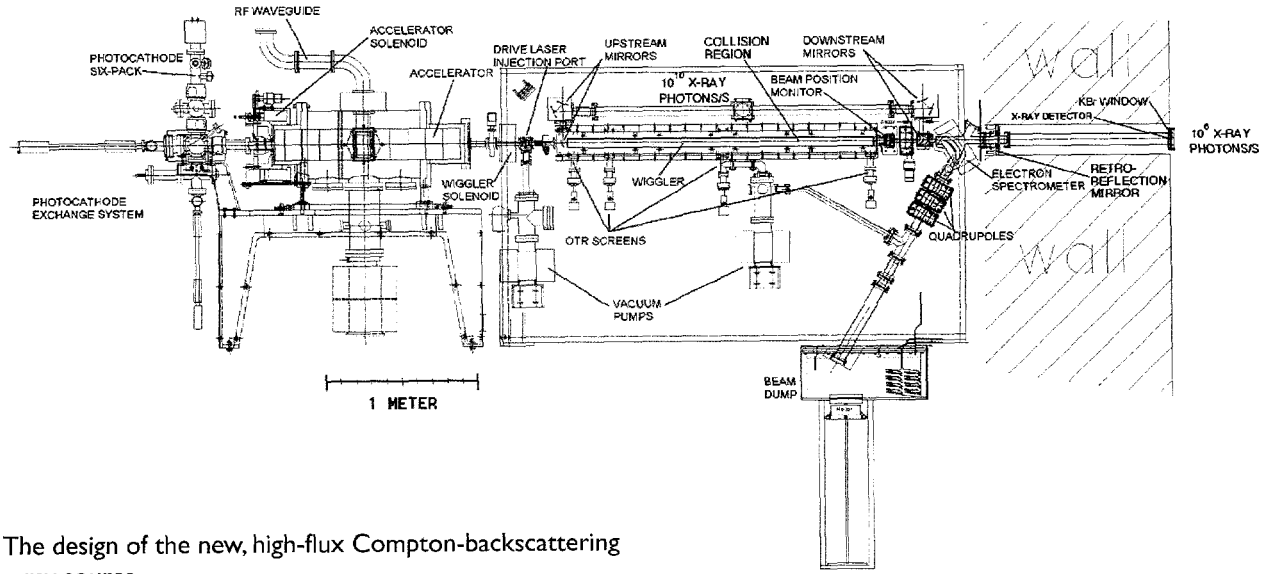
We have successfully achieved the following goals of the project: the production of a large photon flux for Compton scattering; the focusing of a high-current electron beam to a small, focused spot in the focusing region; and the final design of a high-flux Compton-backscattering x-ray source. With the regenerative amplifier FEL, we generated 10^{15} infrared photons per 10-ps micropulse. We have also produced 10^{10} electrons per bunch focused to a 200-μm-radius spot. We completed the integration of the regenerative amplifier FEL into the high-flux Compton-backscattering x-ray source. The accompanying figure shows the new Compton-backscattering design with the calculated numbers of x-rays generated and detected.

Publications

Nguyen, D.C., "Coherent Smith Purcell Radiation as a Diagnostic for Sub-picosecond Electron Bunch Length," *Nucl. Instr. Meth. A* **393**, 514 (1997).

Nguyen, D.C., et al., "A High-Power Compact Regenerative Amplifier FEL" (1997 Particle Accelerator Conference, Vancouver, British Columbia, Canada, May 12–16, 1997).

Nguyen, D.C., et al., "Synchronously Injected Amplifiers, A Novel Approach to High-Average-Power FEL," *Nucl. Instr. Meth. A* **393**, 252 (1997).



The design of the new, high-flux Compton-backscattering x-ray source.

Nonlinear Atom Optics

Peter Milonni

This project is exploring the possibilities in nonlinear atom optics of demonstrating atomic matter wave analogs of nonlinear optical effects such as wave mixing, solitary wave formation, phase conjugation, and complex spatial patterns. Our original research proposal alluded to the possibility of observing Bose-Einstein condensation in cold-atom systems, and since then at least five groups have reported observations of such condensation. There have been many theoretical papers written on the subject, with most of them relying on some form of the so-called Gross-Pitaevskii equation, or, in effect, on the nonlinear Schrödinger equation (NLSE), to explain the condensation phenomena.

It is easy to predict nonlinear atom-wave effects based on the NLSE approximation. Our work has evolved toward justifying this approximation, which, in essence, amounts to factoring the expectation value of a product of field operators into a product of expectation values. The NLSE is so widely used in studies of nonlinear atom optics that a general procedure for justifying its use in any given context, such as for the condensation phenomena, is necessary.

During the past year, we have developed what we believe to be a systematic approach to this difficult justification problem. Our approach is based on Green-function techniques developed in other many-body problems by Schwinger, Keldysh, and others. Our work has focused on these techniques during the past six months partly because of our collaboration with Professor R. Y. Chiao at the University of California, Berkeley, who has undertaken an experimental program to study the limits on applying the NLSE to nonlinear atom optics as well as to effects such as the "fermionization" of photons.

Enhancements of Diagnostics for Plasma Processing of Materials

Gary S. Selwyn

The ultimate capability of plasma processes in treatment of surfaces could potentially be limited by the ability to control and/or measure the presence of small amounts of trace element contamination or additive in the plasma. These small amounts of contaminants can result in surface properties not reaching the theoretical limits that ought to be attainable. The goal of this project is measuring those contaminants with much higher precision than that previously achievable.

Our efforts this year focused on enhancing the diagnostic capabilities of the plasma processing activities in the Physics Division at Los Alamos and resulted in successfully developing and implementing in situ laser-based Raman spectroscopy and near-infrared emission spectroscopy. We used these methods to detect the presence and nature of ground-state and electronically excited molecular oxygen formed in an atmospheric-pressure, nonthermal plasma source used for environmental, industrial, and decontamination applications.

Publications

Babayan, S.E., et al., "Deposition of Glass with an Atmospheric-Pressure Plasma Jet" (submitted to *Appl. Phys. Lett.*).

Babayan, S.E., et al., "Plasma-Assisted Chemical Vapor Deposition of Silicon Dioxide at Atmospheric Pressure" (submitted to *J. Vac. Sci. Technol.*).

Jeong, J.Y., et al., "Etching of Materials with an Atmospheric-Pressure Plasma Jet" (submitted to *Appl. Phys. Lett.*).

Jeong, J.Y., et al., "The Evolution of the Chemical Species in a Parallel-Plate Plasma with Pressure" (submitted to *J. Vac. Sci. Technol.*).

The Calculation of Satellite Line Structures in Highly Stripped Plasmas

Joseph Abdallah Jr.

Our goal is to understand the role of hot electrons created in laser-produced plasmas. Hot electrons influence the level populations that determine spectral line structure, and they are usually ignored in spectral simulations.

In pursuit of our goal, we irradiated magnesium targets, using three different types of laser pulses at the Trident laser facility. We compared the observations with collisional-radiative calculations that used an electron energy distribution function comprising a thermal component and a hot-electron component. We estimated plasma parameters, including electron temperature, density, and hot-electron fraction, by choosing best fits to the experimental measurements.

The calculations show that hot electrons can significantly alter the calculated spectrum and must be considered when the satellite spectrum is used as a diagnostic. The figures show comparisons between theory and experiment. We have obtained some unique experimental results. Trident shots using a high-contrast laser pulse show an unusually broad satellite spectrum, whereas others show emission, possibly from "hollow atoms." These results are as yet unexplained and should be the subject of future study. Additional research involving the use of blue satellite lines for diagnostics and the analysis of argon plasma focus experiments are under investigation.

Publications

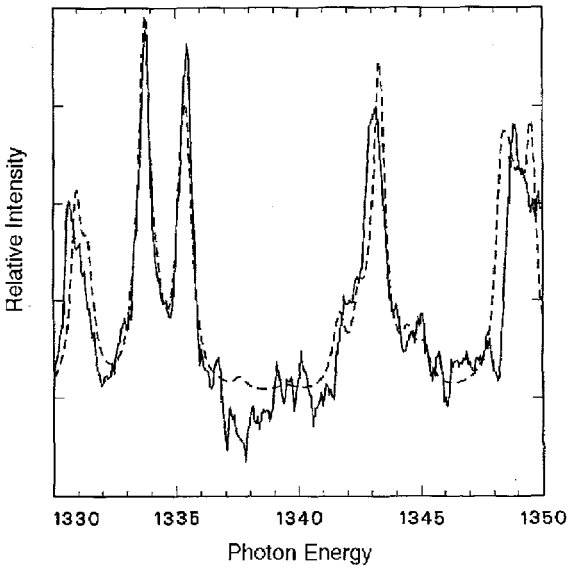
Abdallah, J. Jr., et al., "Hot Electron Effects on the Satellite Spectrum of Laser Produced Plasmas" (to be published in *J. Quant. Spectros. Radiat. Transfer*).

Abdallah, J. Jr., et al., "Various Applications of Atomic Physics and Kinetics Codes to Plasma Modeling," in Proceedings of the 10th APS Topical Conference on Atomic Processes in Plasmas, *AIP Proceedings* **381**, 131 (1996).

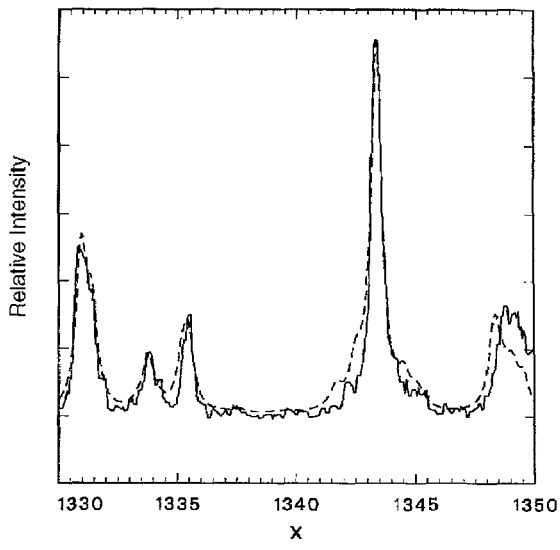
Faenov, A. Ya., et al., "High-Resolution X-Ray Spectroscopy of Hollow Atoms Created in Plasma Heated by Subpicosecond Laser Radiation," in *SPIE Proceedings* (1997).

Kyrala, G.A., et al., "Intense Sub-nm X-Ray Generation for Diagnostic Applications" (International Workshop on Measurements of the Ultrafast Dynamics of Complex Systems with Short Wavelength Radiation, Esterel, Montreal, Canada, June 1997).

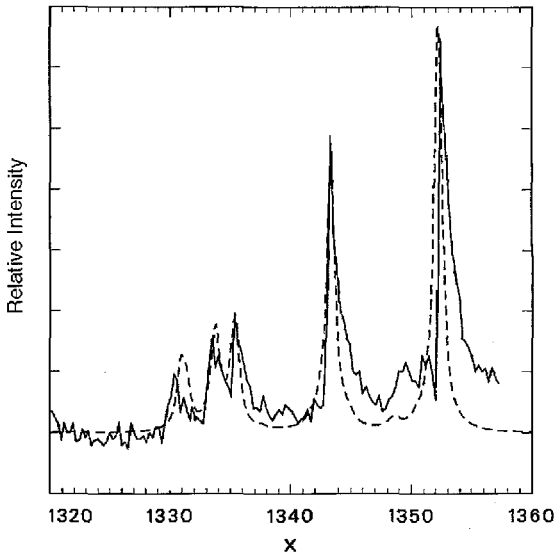
Rosmej, F.B., and J. Abdallah Jr., "Blue Satellite Structure and Redistribution near Helium-Alpha and Helium-Beta" (submitted to *Phys. Lett. A*).



A comparison of the measured 600-fs Trident spectra (solid line) and theory (dashed line) as a function of photon energy. Units for spectra are arbitrary.



A comparison of the measured 1-ns Trident spectra (solid line) and theory (dashed line) as a function of photon energy. Units for spectra are arbitrary.



A comparison of the measured 2-ns carbon dioxide spectra (solid line) and theory (dashed line) as a function of photon energy. Units for spectra are arbitrary.

High-Power, High-Frequency, Annular-Beam Free-Electron Maser

Michael Fazio

Free-electron masers (FEMs) operating in the microwave region have demonstrated both high beam-to-RF power extraction efficiencies (~30%) and high output power (gigawatts). In addition, FEMs are potential candidates for driving advanced accelerators at relatively high frequencies (greater than 10 GHz). However, current FEM technology does not lead to adequate phase stability for this application. Furthermore, high output power has been demonstrated only with FEMs driven by induction accelerators at voltages higher than those practical for driving long accelerators.

The objectives of this project are to demonstrate high output power at 17 GHz by using a high-current but relatively low-voltage electron beam and to develop a phase-stability scheme in which the RF phase of the output power is not affected by small fluctuations in the electron beam voltage.

In the first year of this project, we theoretically derived phase-stability conditions and designed a high-power FEM using the axial FEM interaction that satisfied these conditions. In the second year, we designed and fabricated experimental hardware, including the interaction circuit, input coupler, directional couplers, and high-power calorimeters. In this last year of the project, we demonstrated gain using the axial FEM interaction mechanism. The experiments appeared to demonstrate some characteristics of the phase-stability mechanism, but high power was not achieved because of RF breakdown where we were driving the device with an input signal. In addition, we investigated a new interaction mechanism, the rippled-beam free-electron laser, which theoretically demonstrated superior phase-stability and RF properties.

Publications

Carlsten, B., "Rippled Beam Free-Electron Laser Amplifier Using the Axial Free-Electron Laser Interaction," *J. Appl. Phys.* **81**, 6570 (1997).

Equation of State of Dense Plasmas

John Benage

This project involves the measurement of the equation of state (EOS) of a dense, strongly coupled plasma. These plasmas are at conditions such that the Coulomb interaction energy between particles is greater than their thermal energy. It is under just such conditions that the EOS of materials is least known.

The technique we will use is a standard shock-driven EOS measurement, with the exception that the shock will be produced in a plasma jet by a laser. We will use state-of-the-art x-ray radiography to measure the shock velocity and the density of the shocked plasma. If our technique is successful, these will be the first results to address the EOS of materials at dense plasma conditions. Our primary goal is to demonstrate the feasability of this new technique for measuring the EOS of a material in the dense plasma regime.

This year we have completed the development of our pulsed power supply and the development of the laser backlighters necessary for the shock velocity and density measurements required for this experiment. This involved the development of new imaging techniques using curved crystals for x-ray imaging at very fine resolution (~2 μm). We are beginning the measurements of the plasma plume that will be used as a target plasma for launching a strong shock and creating the dense plasma for the EOS measurements. We have also nearly completed the construction of the laser that will be used for generating the shock in the plasma. We expect to obtain our first EOS measurements for aluminum in early spring 1998.

Engineering Science

Tritium Recovery and Isotope Separation Using Electrochemical Methods

Scott Willms

Our objective is to demonstrate the feasibility of electrochemical techniques for recovering tritium (a radioactive hydrogen isotope) from mixed gas, tritiated water, and hydrocarbons, by using proton-conducting oxides. We are evaluating several doped cerate, zirconate, and beta alumina materials that exhibit protonic conductivity at elevated temperatures. An important aspect of this new technology is the electrochemical destruction of tritiated hydrocarbons and the electrochemical control of the water-shift and steam-reforming reactions currently used in palladium membrane reactors. We will collect the engineering data necessary to design and construct systems for tritium recovery.

This year we synthesized doped cerate and zirconate perovskite ceramic membranes and evaluated the crystal structure and morphology of these materials. X-ray diffraction reveals an orthorhombic distorted perovskite structure. Electrochemical transport measurements of the ionic mobility of hydrogen in these materials have confirmed the high mobility for protonic species within the materials at temperatures from 500°C to 800°C. Using alternating-current impedance spectroscopy, we measured proton conductivities of alkaline-earth cerates and zirconates and found that the activation energy for proton migration was approximately 0.56 eV.

Experiments demonstrated that hydrogen gas permeated across the membrane only when a current was allowed to flow between two platinum electrodes on opposing sides of the solid-electrolyte membranes. Rates for electrochemical permeation obeyed Faraday's law, indicating an ionic transport mechanism. We observed hydrogen-gas permeation rates of 0.5 $cm^3/min \cdot cm^2$ at current densities of 70 mA/cm^2.

In addition, we demonstrated water vapor electrolysis, using ceramic membranes made of alkaline-earth cerates and zirconates. Water electrolysis rates on platinum electrodes were approximately 0.2 $cm^3/min \cdot cm^2$ at current densities of 40 mA/cm^2. The water vapor electrolyzed to dry hydrogen gas at the cathode and to wet oxygen gas at the anode—an especially significant result because conventional types of electrolysis cells produce wet hydrogen gas. The production of dry tritium gas from tritium oxide electrolysis is very desirable because the biological hazards of tritiated water are much greater than those of tritium gas.

Virtual Bandwidth via Stochastic Polyspectra

Murray A. Wolinsky

We are investigating a novel, relatively high-risk approach to better utilizing existing communications channels—not through the use of data compression but through an effective expansion of bandwidth. The research involves using polyspectra to provide noise-resistant communications.

We made significant progress toward theoretically resolving a long-standing controversy concerning the ability to detect aliasing using bispectra, a key objective for the first year's work. Our new arguments support the use of polyspectra to detect aliasing. These arguments were discussed at the IEEE Higher-Order Statistics Workshop (HOS'97, Banff, Alberta, Canada).

We have designed two systems to convey information through band-limited noisy channels using polyspectra. One system embeds information directly into the bispectrum of the transmitted signal and should allow us to examine the aliasing question experimentally. The second system is a novel multicarrier modulation scheme that relies on trispectral signal processing for noise immunity. Soon we should have both systems fully implemented in MATLAB (a commercial software) and begin assessing their performance.

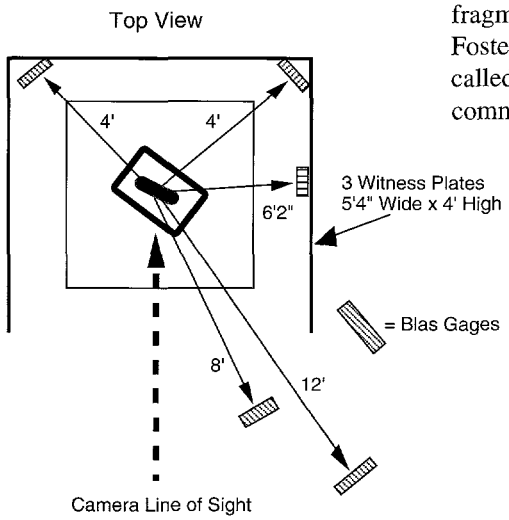
Our effort has not yet resolved the existence of "constraints" on polyspectra. Discussions at HOS'97 indicate that this question is still little understood. Results from the general time-series model we constructed suggest that no such constraints exist. Our results, combined with the work of others, are promising for the general program of conveying information using higher-order spectra in a noise-resistant fashion.

Exploration of Technologies of Use to Civil Security Forces

Eugene H. Farnum

Police forces routinely encounter dangerous situations in which their ingenuity and the state of their equipment determine the life or death of bystanders and officers involved. Often, the officers recognize areas where improvements in equipment could greatly enhance their performance but do not have access to advanced technologies or to the laboratory facilities needed to develop and test prototypes. The objective of this project is to determine whether armor tile technology could be used to solve problems of civil law enforcement as identified by the New Mexico State Police.

Most of our effort focused on the design and construction of a lightweight, portable box that could contain the shrapnel and redirect the blast from a steel-pipe/black-powder bomb. We collaborated on the bomb box with two companies, Foster Miller, Inc., and Ordnance Body Armor Company, who constructed most of the boxes tested. The accompanying figure shows the test setup.



Schematic diagram of test setup for evaluating pipe-bomb boxes. The bomb is shown inside the rectangular test box. The plywood witness plates preserve the distribution pattern of the flying fragments. Blas gages record the blast pressure.

The results of the tests indicated that soft, flexible fabrics are superior to hard tiles in containing the bomb fragments. After our experiments, Foster Miller developed a bomb box, called the "Frag Bag," that they have commercialized and are selling to law enforcement agencies. The New Mexico State Police are currently field testing and evaluating this box.

The Plasma Fluidized Bed

Robert P. Currier

The project goal is to develop novel fluidized-bed technologies using cold plasmas. The basic concept is to couple radio-frequency or microwave energy into the fluidizing gas to form a nonequilibrium plasma. Depending on conditions, the plasma may contain high concentrations of ions and reactive free-radical species, all at modest bulk temperatures. We intend to use these species to initiate or promote the desired chemistry.

Our effort is divided into three major topical areas: fluidized-bed hydrodynamics, diagnostics, and electromagnetic design considerations. To determine the optimal hydrodynamic conditions, we conducted experiments in a dense bubbling bed (with input of microwave energy). The high solids concentration typically makes it difficult to sustain a glow discharge.

We then constructed fluid-bed systems consisting of a "riser" section in which we can operate in the so-called fast fluidized regime. This vertical tubular device has solids distributed along the axis such that the bed is dense in solids at the bottom and lean in solids at the top. Experiments using this device are under way to determine the optimal and maximum solids concentration that still permits the plasma to be easily sustained. We also constructed a "downer" reactor in which the solids fall through the fluid, are collected, and may be pneumatically transported back to the top of the downer section.

Our work on diagnostics has led to construction of a fluidized-bed unit, microwave applicator, and port for mass spectral sampling. This configuration permits analysis of gas composition as a function of system parameters.

Electromagnetic design has focused primarily on issues related to the ultimate scale-up of the laboratory reactors. This effort has led to the preliminary design of a slitted enclosure that would surround a reactor and introduce electromagnetic energy uniformly over the entire length. We also developed a conceptual design for other configurations suitable for establishing and maintaining a glow discharge. In addition, we began implementing an automated control system and installing flow visualization (optical) diagnostics.

A Comprehensive Monitoring System for Damage Identification and Location in Large Structural and Mechanical Systems

Charles Farrar

Identifying the onset of damage to many large structures and mechanical systems at the earliest possible stage would be very helpful. Methods that can perform such assessments on a global basis offer many advantages over traditional, local nondestructive testing methods. This project focuses on developing a suite of robust, state-of-the-art algorithms that can process measured vibration data from structural systems, statistically analyze this data, and estimate damage according to changes in identified modal properties. The primary methods for quantifying and locating damage from global vibration response measurements have been to look at changes in strain energy stored in the structure and to identify the nonlinear characteristics using time-frequency analysis. We have developed algorithms for nonlinear time-frequency analysis, such as wavelet transforms. To verify the accuracy of these tools, this year we conducted numerous vibration tests on in situ bridge structures in damaged and undamaged configurations with varying environmental conditions.

There are several indications of this project's success. First, each week we receive requests from around the world for the software package that we developed. Second, we have been invited as keynote speakers to present this work at three international conferences on vibration analysis and testing. Third, we have developed a short course on vibration-based damage identification, which has been or will be taught in the United States, Australia, and Japan. Sponsors for the course include the Society for Experimental Mechanics and the Japan Society for the Promotion of Sciences. We are currently applying this technology to vibration tests conducted as part of the nuclear weapons program.

Publications

Doebling, S.W., and L.D. Peterson, "Computing Statically Complete Flexibility from Dynamically Measured Flexibility," *J. Sound Vib.* **205** (5), 631 (1997).

Farrar, C.R., and T.A. Duffey, "Bridge Modal Properties Using Simplified Finite Element Analysis" (to be published in *J. Bridge Eng.*).

Farrar, C.R., and G.H. James, "System Identification from Ambient Vibration Measurements on a Bridge," *J. Sound Vib.* **205** (1), 1 (1997).

Farrar, C.R., and D.V. Jauregui, "Comparative Study of Damage Identification Algorithms Applied to a Bridge: Part I, Experimental" (to be published in *J. Smart Mater. Struct.*).

Farrar, C.R., and D.V. Jauregui, "Comparative Study of Damage Identification Algorithms Applied to a Bridge: Part II, Numerical" (to be published in *J. Smart Mater. Struct.*).

The Compliance Method for Measuring Residual Stress

Michael Prime

The purpose of this project is to develop the compliance method for measuring residual stress. Subsequently, we wish to turn this fledgling method into a proven and versatile technique that can fill the gaps in current residual-stress measurement technology. In addition to extending measurement capabilities, the results of our project will greatly aid the development and verification of predictive models.

In the past year, we had three major accomplishments. First, we completed and published a literature review in which we identified the state of the art in methods similar to the compliance method and provided the necessary direction for innovative research in this field. Second, we prepared a unique specimen with a known residual-stress distribution and measured it using all the techniques available at Los Alamos. This achievement has led to objectively quantifying our measurement capabilities and deficiencies, and it has provided a well-characterized specimen for use in developing the theoretical tools for implementing the compliance method. Finally, we extended the compliance method to a new capability—measuring residual stress in a layer with elastic constants different from those of the parent material. We applied this technique to a hard copper alloy that was clad to an aluminum substrate. Such engineered layers and coatings are becoming increasingly critical to developing optimized thermal barrier coatings on such structures as diesel engines or jet aircraft turbines.

Publications

Prime, M.B., and Ch. Hellwig, "Residual Stresses in a Bi-Material Laser Clad Measured Using Compliance" (Fifth International Conference on Residual Stress, Linköping, Sweden, June 16–18, 1997).

Prime, M.B., "Residual Stress Measurement by Successive Extension of a Slot: A Literature Review," Los Alamos National Laboratory report LA-13283-MS (May 1997).

Quantum Cryptography for Secure Communications to Low-Earth-Orbit Satellites

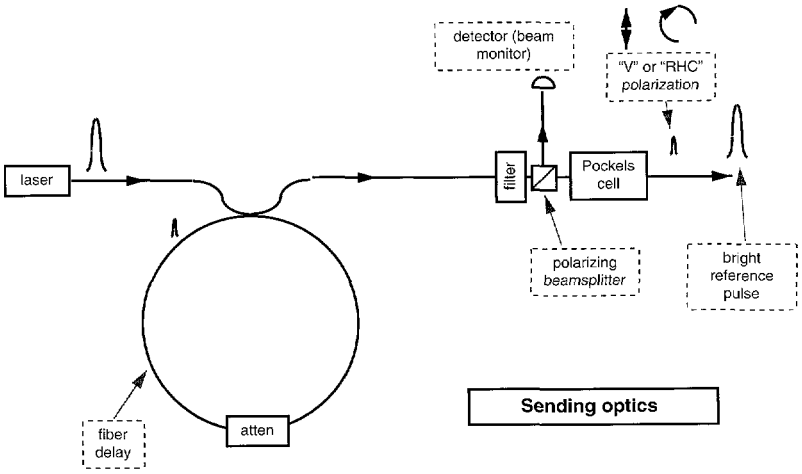
Richard Hughes

Cryptanalysis techniques and algorithms are advancing rapidly and by the start of the twenty-first century will make information security for orbital assets difficult to ensure without the use of new technologies. The aim of this project is to develop quantum cryptography to provide absolute encryption security in communications to low-earth-orbiting satellites. Quantum cryptography uses single photons to create and transfer information in a way that uses the laws of quantum mechanics to afford absolute security. Calculations show that completely secure random "seeds" (cryptographic key material) can be generated between a ground station and satellite in real time. This means that there is no prior record of a key and no possibility of hostile interception, making the entire encryption process both simpler and more assured. This project will develop and demonstrate technology to a stage where it can be incorporated into new satellites.

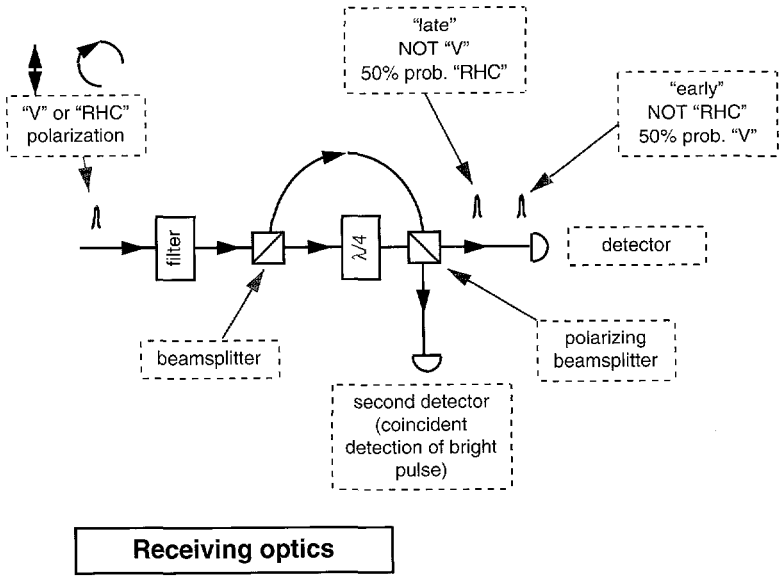
This year we developed our quantum cryptography system (see the first two figures) to perform key generation over long optical paths, starting with indoor experiments under fluorescent lighting conditions that demonstrated key generation over distances up to 205 m. We incorporated an encryption/decryption stage into the computer control system that allowed us to demonstrate the key-generation process with the transmission of encrypted images (see the third figure). Also, we investigated and incorporated error correction schemes into the control system. After demonstrating satisfactory performance of the system indoors, we transferred the whole quantum cryptography system into an instrumented vehicle. We then demonstrated the feasibility of key generation outdoors over a 1-km path (see the fourth figure).

Publications

Buttler, W.T., et al., "Free-Space Quantum Key Distribution" (submitted to *Phys. Rev. A*).



Schematic of the quantum cryptography transmitter.

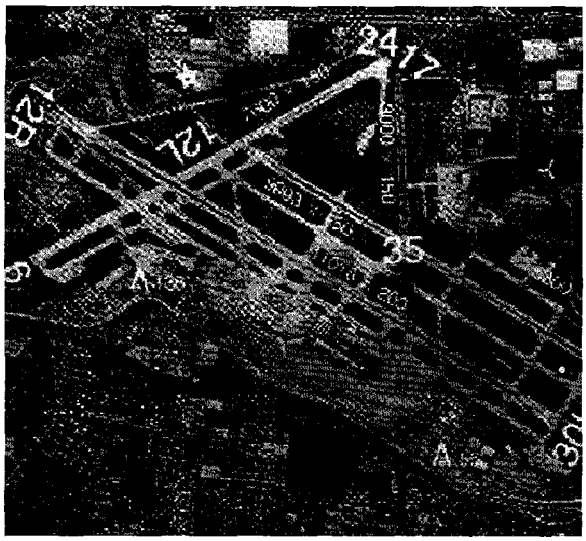


Schematic of the quantum cryptography receiver.

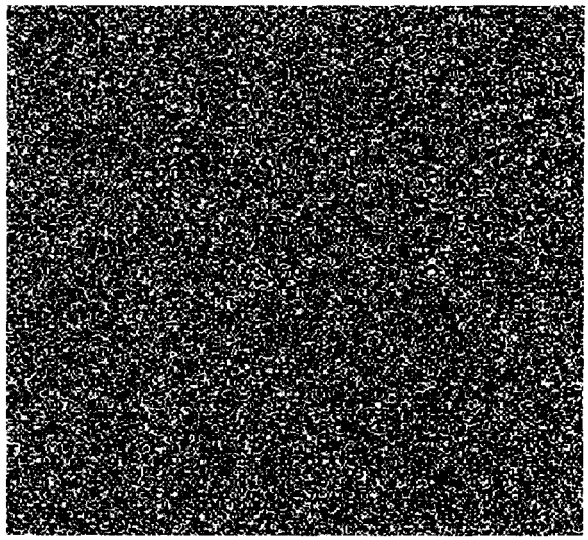
A	11100011	11111010	11001111	10000000
B	11100011	11111110	11001111	10000000
A	00001001	01111001	11001100	10111011
B	00001001	01111001	11001100	10111011
A	01110110	10110111	11101101	10111011
B	01110110	10110111	11101101	10111011
A	10110111	11010111	10111011	11111111
B	10110111	11010111	10111011	11111111

A sample of key material obtained in a 40-m outdoor transmission, with a 1% bit error rate.

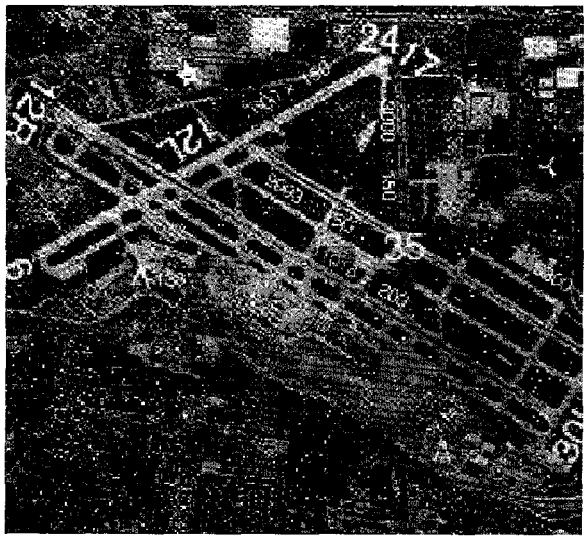
(a)



(b)



(c)



An example of encryption/decryption of an image using quantum key material: (a) is the image to be transmitted, (b) is the encrypted image, and (c) is the decrypted image.

Material Processing for Self-Assembling Machine Systems

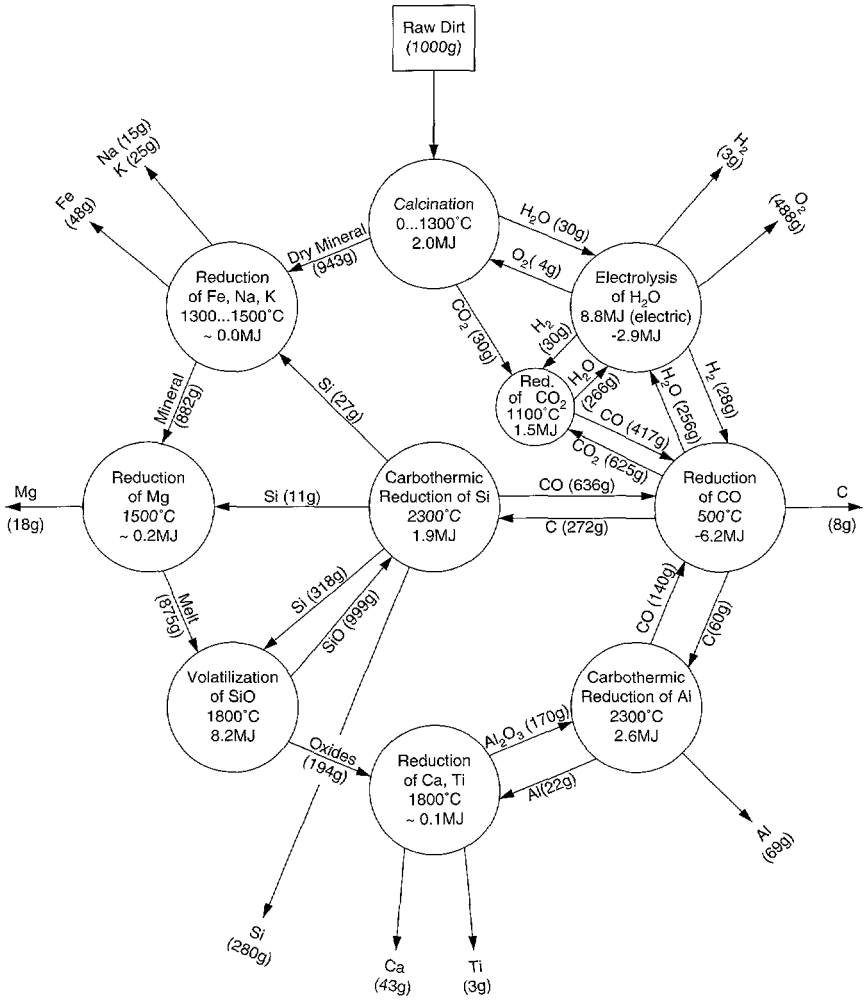
Klaus Lackner

Major problems for the coming decades include limitations in energy and nonrenewable resources and the need to control deleterious side effects of human activities on the environment. Self-reproducing machine systems, systems that can build themselves, could be the solution to these problems. Our project focuses on developing the foundation for a new technology based on self-reproducing machine systems. There are numerous issues that need to be addressed before our vision of this new technology can be implemented; we are researching the issue of material extraction.

Machine systems that build themselves promise an increase in industrial productivity as dramatic as that of the industrial revolution. A system that can build itself must procure all raw materials in its surroundings. Thus, next to automation, one of the most critical issues in developing this new technology is the design of the process that will extract all the important chemical elements from raw dirt. This process differs from conventional extraction metallurgy in that it uses only abundant elements, because the entire mass of the self-replicating system is channeled through the process (see figure). However, such extraction processes are of interest even in conventional metallurgy because they do not involve use of expensive and sometimes hazardous materials.

This year's work was dominated by the development of experimental techniques to operate at very high temperatures and with very small samples. We greatly expanded our development of small furnaces and have gone through several generations of small furnace designs. We have operated these furnaces in air, argon, and vacuum. We were able to separate silica and iron from complicated mixtures, and we also have succeeded with the carbothermic reduction of aluminum. We are currently weighing the advantages (much lower temperature) and disadvantages (increased complexity) of vacuum processes. In addition, we have started a theoretical analysis of the interactions of the various ions in the melt. These interactions have a noticeable effect on the overall thermodynamics and thus can affect the outcome of the separation process.

While our preliminary experiments have succeeded in performing most steps required in the material separation, a more controlled set of experiments is required. What is still missing is a demonstration of the entire separation cycle. We expect to achieve this in the coming year.



Outline of a high-temperature extraction scheme that can decompose average rock into its elemental constituents while using only materials that are commonly available in average rock.

Development of a Fullerene-Based Hydrogen Storage System

Shimshon Gottesfeld

The objectives of this project are to evaluate hydrogen uptake by fullerene substrates and to probe the potential of the hydrogen/fullerene system for hydrogen fuel storage. We have completed and tested a fully automated, computer-controlled system for measuring hydrogen uptake that is capable of handling both a vacuum of 1×10^{-6} torr and pressures greater than 200 bars. We have previously established conditions for significant uptake of hydrogen by fullerenes.

During this year, we further studied hydrogenation and dehydrogenation of pure and catalyst-doped C_{60} to probe its suitability for hydrogen storage applications. First, $C_{60}{\cdot}H_{18.7}$ was prepared in 100-bar H_2 at 400°C, corresponding to hydrogen uptake of 2.6 wt%. Next, we studied dehydrogenation of $C_{60}{\cdot}H_{18.7}$ using thermogravimetric and powder x-ray diffraction analysis. The $C_{60}{\cdot}H_{18.7}$ molecule was found to be stable up to 430°C in argon, at which point the release of hydrogen initiated the collapse of the fullerene structure.

X-ray diffraction analysis performed on $C_{60}{\cdot}H_{18.7}$ samples dehydrogenated at 454°C, 475°C, and 600°C showed an increasing volume fraction of amorphous material in the form of randomly oriented, single-layer graphite sheets. Evolved gas analysis using gas chromatography and mass spectroscopy confirmed the presence of both H_2 and methane upon dehydrogenation, indicating decomposition of the fullerene. The remaining carbon could not be rehydrogenated. These results provide the first complete evidence for the irreversible nature of fullerene hydrogenation and for limitations imposed on the hydrogenation/dehydrogenation cycle by the limited thermal stability of the molecular crystal of fullerene.

Publications

Brosha, E.L., et al., "Fullerenes: Potential Hydrogen Storage Materials?" (submitted to *Int. J. Hydrogen Energy*).

Brosha, E.L., et al., "Thermal Stability Limitations on Hydrogenation/Dehydrogenation Cycles in Fullerenes" (submitted to *Chem. Mater.*).

Instrumentation and Diagnostics

Low-Field Magnetic Resonance Imaging of Gases

David M. Schmidt

The main effort of this project is to develop the capability to conduct magnetic resonance imaging (MRI) measurements at low or ambient magnetic field strengths. Conventional MRI detectors lose sensitivity with decreasing field strength, so we are pursuing the use of superconducting quantum interference devices (SQUIDs).

Making use of SQUID instrumentation and experience that existed in our group from other projects, we first constructed a SQUID MRI detector consisting of two high-temperature SQUIDs arranged in a gradiometer geometry. Because the noise level of this detector was too high, we next constructed a gradiometer detector using two low-temperature SQUIDs, which have a lower intrinsic noise level. Now, however, ambient electromagnetic radiation in our laboratory was the main source of noise.

We were fortunate to be able to use a shielded room that had been constructed for other SQUID applications. This room should sufficiently shield against the electromagnetic radiation so that a gradiometer geometry would not be needed. We therefore constructed a low-temperature SQUID magnetometer and observed a noise level of 30 $fT/(Hz)^{1/2}$ with this detector in the shielded room.

In addition, we constructed coils to produce the homogenous holding field at about 3 G and to produce the spin-flipping pulse of frequency <20 kHz. We also constructed electronics to control the sequence of events necessary for MRI and to record and analyze the MRI signal.

Thermal Detection of DNA and Proteins during Gel Electrophoresis

Roger Johnston

Gel electrophoresis is one of the most important techniques available to science and biotechnology. It is an electrochemical process in which colloidal particles (those with dimensions of 1 nm–1 μm) or macromolecules with a net electric charge migrate in a gel solution under the influence of an electric field. The technique can identify genetic material and is used in biological and biomedical research, polymer chemistry, forensics, law enforcement, disease control, national defense, anthropology, biometrics, genetic counseling, botany, agriculture, and animal husbandry. Gel electrophoresis is used in low-tech laboratories as well as in state-of-the-art facilities.

Recent ultrasensitive refractive-index measurements suggest that DNA fragments can be detected during gel electrophoresis using thermometry and local Joule self-heating. This new technique is simple, inexpensive, and ultrasensitive, permitting real-time detection of DNA and other proteins. In addition, the technique avoids the environmental hazards and biohazards associated with conventional detection techniques. These hazards include UV radiation, carcinogenic fluorescent DNA dyes, and radioisotopes. This project focuses on developing the instrumentation required to demonstrate the technique, performing a proof-of-principle experiment, optimizing the technique, and establishing its sensitivity.

This year we have designed and constructed a novel gel electrophoresis apparatus that will permit us to perform a proof-of-principle experiment on thermal detection next year. We have also obtained the necessary components and supplies for this demonstration.

Novel Signal Processing with Nonlinear Transmission Lines

David Reagor

Our objective is to develop time-domain signal-processing concepts in a technologically interesting regime using integrated coplanar-waveguide (CPW) devices fabricated from nonlinear dielectric materials. To reach this objective, we are building proof-of-principle devices able to perform functions of use in almost all signal-processing systems.

This year the major experimental efforts focused on developing infrastructure. We designed photolithography masks for both dispersive and nondispersive CPW meander lines with 4-, 8-, and 24-cm lengths and for impedance-modulated segments—coupled CPW lines with two inputs and two outputs. We improved processes in our clean room, fabricated CPW meander-line devices, reconfigured a time-domain measurement setup, and put together a new cryogenic system. In addition to these infrastructure efforts, we performed preliminary experimental studies of soliton formation, observed pulse sharpening caused by reduced microwave loss under bias, observed an increase in pulse propagation speed with bias, and observed the effects of tunable nonlinearity.

The theoretical efforts this year included modeling the transmission lines with discrete elements using a perturbed Toda lattice and developing perturbation theory for the modified Toda lattice to capture the essential soliton dynamics. Full numerical simulations show that the transmission line can shape an input pulse into a train of stable and predictable traveling solitons. These theoretical studies indicate that (1) the nonlinear transmission line can successfully shape an input pulse into a well-defined output pulse if the input is not dominated by high-frequency noise and (2) a simple linear *RL* input port is a useful interface for the nonlinear transmission line even when the initial pulse has significant power at high frequencies.

Optical Imaging through Turbid Media Using a Degenerate Four-Wave Mixing Correlation Time Gate

Irving Bigio

There is a significant need for a safer alternative to x-ray imaging in applications such as mammography, dentistry, and brain imaging. An optical imaging diagnostic could be used more frequently and safely than the current x-ray diagnostics. In addition, an optical diagnostic has the potential to provide functional information about the imaged tissue. However, because tissue strongly scatters light, resulting generally in diffuse or fuzzy images under normal circumstances, a method is needed for selecting and amplifying only the small number of photons that have traveled nearly unscattered through the medium. This is equivalent to detecting only those photons that have taken the straightest (shortest temporal) path through the medium.

This project calls for an optical imaging diagnostic based on the nonlinear optical technique of degenerate four-wave mixing as a very fast correlation time gate. This method generates a short, well-defined optical gate for the imaging signal, with significant advantages over competing techniques, thus ensuring that only the least-scattered photons are detected and imaged.

This year, we designed and assembled a high-pressure cesium vapor cell. Because of the high nonlinear sensitivity of the vapor, we developed this new nonlinear medium to permit the use of low-power near-infrared (NIR) diode lasers as the light source. Such diode lasers are the type of light source that any clinical application would require.

First, with a higher-power, pulsed NIR laser, we generated images of a test pattern (crosshairs) through 2 cm of scattering medium, at wavelengths appropriate for imaging in tissue, and demonstrated that the cesium vapor was working as expected. Then we purchased an appropriate diode laser and reconfigured the optical system accordingly. We generated preliminary results with this setup, and we hope to secure new future funding to demonstrate acquisition of an imaging signal with the diode laser and the cesium cell.

Publications

Findikoglu, A.T., et al., "Microwave Properties of Superconducting Yttrium-Barium-Copper Films on Buffered Polycrystalline Substrates," *Appl. Phys. Lett.* **69**, 1626 (1996).

Findikoglu, A.T., et al., "Microwave Surface Resistance of $YBa_2Cu_3O_{7-\delta}$ Films on Polycrystalline Alumina and Ni-Based Alloy Substrates with Ion-Beam-Assisted-Deposited Buffer Layers," *IEEE Trans. Appl. Supercon.* **7**, 1232 (1997).

Findikoglu, A.T., et al., "Paraelectric Thin Films for Microwave Applications," *Integ. Ferroelec.* **15**, 163 (1997).

Findikoglu, A.T., et al., "Power-Dependent Microwave Properties of Superconducting $YBa_2Cu_3O_{7-x}$ Films on Buffered Polycrystalline Substrates," *Appl. Phys. Lett.* **70**, 3293 (1997).

Magnetic Resonance Force Microscope Development

P. Chris Hammel

Magnetic resonance force microscopy (MRFM) is a microscopic imaging technique based on mechanically detecting magnetic resonance signals by measuring the force exerted on a magnetic moment by a magnetic field gradient provided by a miniature permanent magnet. Periodically modulating this force by modulating the spin magnetization alters the oscillation amplitude of a high-Q, low-spring-constant micromechanical resonator (cantilever or bridge) such as is used presently in atomic force microscopy. MRFM allows nanometer-scale imaging and characterization of materials, and it enables unprecedented detail in understanding a broad range of scientifically important systems. Our central objective is to build a tool applicable to a wide range of studies of technologically important systems, specifically layered magnetic and semiconductor systems.

We have detected, for the first time, ferromagnetic resonance (FMR) signals in thin metallic cobalt films using MRFM. The sample was approximately 40 μm^2 and 200 Å thick. This film is very close to materials used in technologically important magnetic thin-film devices. This advance is a crucial one because the cobalt FMR signal is much weaker than earlier FMR signals we had obtained in yttrium iron garnet, and because it demonstrates the applicability and utility of FMR/MRFM in the study of the technologically important magnetoelectronic devices.

We have explored the conditions necessary for obtaining magnet-on-cantilever MRFM signals, which represents an important step in developing the crucial capability of magnet-on-probe MRFM. Finally, we have observed two distinct FMR signals from two cobalt layers in a trilayer sample. This is very exciting because it indicates the potential power of MRFM as a tool for studying a broad class of magnetic materials in which juxtaposition of dissimilar materials, and thus buried interfaces, plays a crucial role.

Publications

Hammel, P.C., et al., "The Magnetic Resonance Force Microscope: A New Microscopic Probe of Magnetic Materials" (NATO Advanced Study Institute: Frontiers in Reduced Dimensional Magnetism, Crimea, Ukraine, May 27–June 3, 1997).

Zhang, Z., and P.C. Hammel, "Towards a Magnetic Resonance Force Microscope Employing a Ferromagnetic Probe Mounted on the Force Detector" (submitted to *J. Solid State NMR*).

Zhang, Z., et al., "Magnetic Resonance Force Microscope as a 3-D Probe of Magnetic and Semiconductor Materials" (Scanning Microscopy 97, Chicago, IL, May 11–15, 1997).

Zhang, Z., et al., "Sensitivity and Spatial Resolution in Magnetic Resonance Force Microscopy," *J. Appl. Phys.* **80**, 6931 (1996).

Subpicosecond Electron-Bunch Diagnostic

Bruce Carlsten

The objective of this project is to develop new, subpicosecond-bunch-length diagnostic techniques with resolutions that will scale below 0.25 ps. These techniques will be nonintercepting, simple, and inexpensive. The specific goals of our project are to (1) build a 0.50-ps streak camera for calibration purposes, (2) build and test a diagnostic based on coherent Smith-Purcell radiation, and (3) build and test a diagnostic based on a transversely deflecting radio-frequency cavity and on beam-second-moment measurements by using nonintercepting beam-position monitors.

So far, we have verified the second-moment approach using the beam-position monitors, characterized the optical transition light for the streak-tube optimization and ordered the streak tube based on these measurements, and performed additional theoretical calculations of the coherent Smith-Purcell radiation.

The major issue associated with the second-moment approach is that errors introduced in the measurement technique are magnified in the inversion of the beam-transport matrix, leading to a large uncertainty in the calculated beam moments. We experimentally verified that, by constructing the beam-transport matrix from nearly orthogonal components, the errors are not magnified and the resulting uncertainty in the calculated beam moments is low. Previous calculations of the coherent Smith-Purcell radiation were for Gaussian electron-bunch distributions. Calculations of the coherent Smith-Purcell radiation for probable electron-bunch distributions show additional frequency components, which may lead to additional information about higher-order bunch-length moments.

Femtosecond Scanning Tunneling Microscope

Antoinette Taylor

By combining scanning tunneling microscopy with ultrafast optical techniques, we have developed a novel tool to probe phenomena on atomic length and time scales. In the two previous years, we built an ultrafast scanning tunneling microscope (STM) and characterized its temporal resolution, sensitivity, and dynamic range. With this STM we gained the knowledge to develop an STM that has both cryogenic and ultrahigh-vacuum capabilities, enabling the study of a wide range of scientific problems.

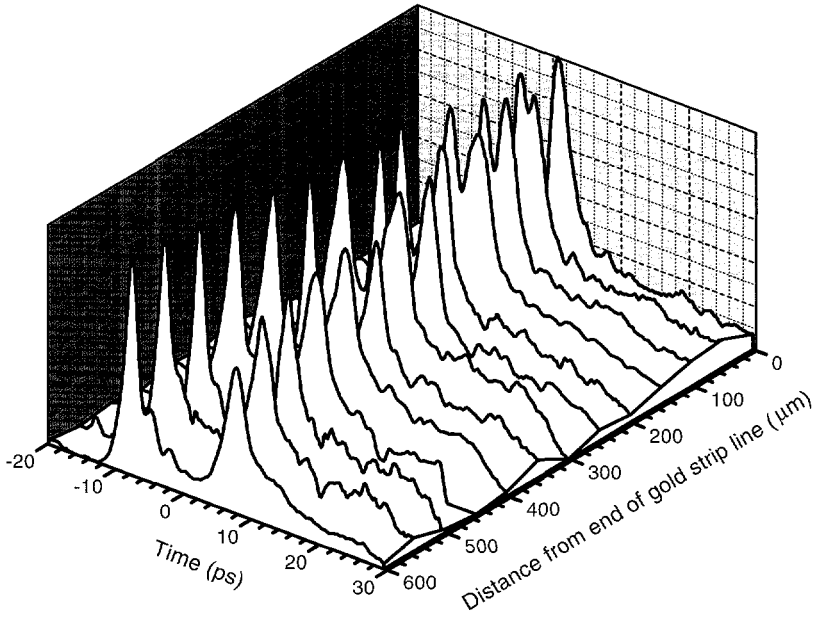
During this year we assembled and characterized a femtosecond STM with both cryogenic and ultrahigh-vacuum capabilities. We achieved a temporal resolution of 1.5 ps and a spatial resolution of 10 nm. In the first figure, we demonstrate our ability to measure spatial and temporal images of a picosecond electrical waveform propagating along a strip line using our femtosecond STM.

In addition, we developed and optimized a novel STM tip fabricated from a cleaved GaAs substrate, with a 1-mm epilayer of low-temperature-grown GaAs (LT-GaAs) deposited on the face. Since LT-GaAs has a carrier lifetime of 1 ps, photoexcitation of the tip with an ultrafast, above-band-gap pulse not only provides carriers for the tunneling current but also photoconductively gates (with picosecond resolution) the current from the tip. Our tunneling tip locates the ultrafast switch at the tip, eliminating detrimental propagation effects, and hence directly yields the waveform present on the surface (see the second figure), rather than its derivative, as is measured with other tip/switch configurations. This tip is cheap and easy to fabricate.

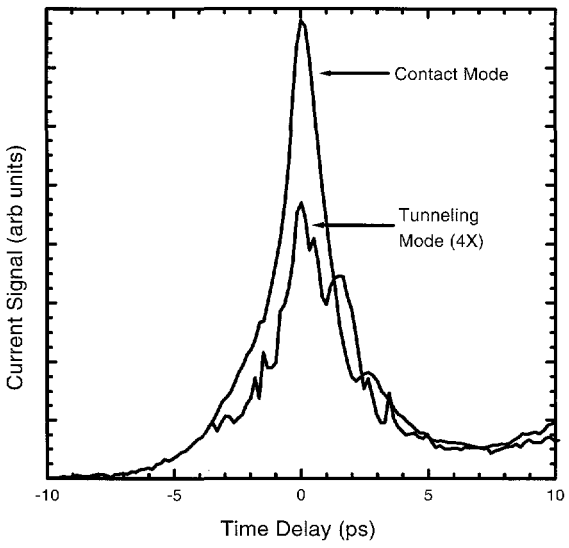
Publications

Donati, G.P., et al., "Ultrafast Scanning Tunneling Microscopy Using a Photoexcited Low-Temperature-Grown-GaAs Tip" (submitted to *Appl. Phys. Lett.*).

Taylor, A.J., et al., "Ultrafast Field Dynamics in Photoconductors," *Opt. Lett.* **22**, 715 (1997).



Temporal and spatial image of a picosecond electrical waveform propagating along a strip line. These measurements show the original picosecond pulse and its reflection off the end of the strip line as a function of distance down the strip line. As the end of the strip line is approached, the two peaks coalesce into one.



Transient current waveforms for the LT-GaAs tip in both contact and tunneling modes (total resistance = 0.3 GW).

Soliton Optical Communications

Antoinette Taylor

This project is a joint experimental and theoretical effort to investigate the fundamental physical limits of high-bit-rate propagation of nonlinear optical pulses called solitons. These pulses are being studied for use as communication links between computers. The experimental work relies on a novel diagnostic that measures the exact electric-field envelope and optical phase of ultrashort optical pulses as functions of time; such measurements will allow us to characterize the temporal evolution of optical solitons in fiber-optic waveguides. In a stretch of a purely passive transmission line, we will use this diagnostic to measure soliton propagation over distances of 1 to 10 km as a function of pulse width and interpulse separation. We will also investigate techniques that ameliorate soliton instabilities that arise at high bit rates. Finally, we will investigate combining wavelength-division multiplexing and soliton propagation at terabit per second bit rates.

An optical parametric amplifier has been set up to generate wavelength-tunable picosecond pulses at 1550 nm. We have invented and built a high-throughput multiplexer to produce variable-delay pulse trains. We have developed a cross-correlation diagnostic based on the marriage of standard frequency-resolved gating and multipulse longitudinal interferometry. With this diagnostic, low-energy soliton pulses are gated by shorter-duration, high-energy reference pulses, simultaneously yielding the intensity and phase profiles of both pulses. This two-beam, cross-correlation, phase-sensitive diagnostic will be the central experimental tool of the project. In related theoretical work, we have investigated several soliton propagation schemes that ameliorate soliton degradation.

Publications

Siders, C.W., and A.J. Taylor, "High-Energy Pulse Train and Shaped Pulse Generation" (submitted to *Opt. Lett.*).

Siders, C.W., and A.J. Taylor, "High-Energy Pulse Train and Shaped Pulse Generation Using a 100% Throughput Michelson Interferometer," in *Proceedings of Ultrafast Optics '97* (Optical Society of America, Washington, DC, 1997), p. 111.

Siders, C.W., et al., "Multi-Pulse Interferometric Frequency-Resolved Gating: Real-Time Phase-Sensitive Imaging of Ultrafast Dynamics," *OSA Topics in Optics and Photonics: Ultrafast Electronics and Optoelectronics* **14**, 157 (1997).

All-Solid-State Four-Color Laser

Timothy R. Gosnell

The goal of this project is to develop a solid-state laser that produces visible output wavelengths, including the commercially compelling blue wavelength. The basic architecture of the device consists of a single-mode optical fiber doped with Pr^{3+} and Yb^{3+} ions. When the ions are simultaneously pumped with a near-infrared laser (860 nm), complex energy-transfer processes involving multiple excited ions lead to population of a high-lying energy level of Pr^{3+}.

This year we developed a mathematical model based on measured optical parameters of Yb^{3+} and Pr^{3+}. In the model, a finite-difference numerical solver of partial differential equations performs calculations of the laser performance. The results allowed us to develop new cavity designs that include the direct deposition of dielectric mirror coatings onto the fiber ends. The coated fibers have just been returned to us from the specialty coating house and will be the subject of next year's research. We also discovered an unusual "coupled cavity" effect in the red laser that yielded an unexpected asymmetry in the output power measured from either end of the fiber. This phenomenon is as yet unexplained and is under continued study.

Publications

Gosnell, T.R., "Avalanche-Assisted Upconversion in Pr(3+)/Yb(3+)-Doped ZBLAN Glass," *Electron. Lett.* **33**, 411 (1997).

Imaging, Time-of-Flight, Ion Mass Spectrograph

Herbert Funsten

The imaging, time-of-flight (TOF) mass spectrograph uses electrostatic scanning of a monoenergetic beam of ions, a drift tube, and a position-sensitive detector to measure the TOF of an ion traversing the drift tube. The spectrograph is particularly suited to identifying ions with large masses, such as detecting chemical and biological warfare agents.

This year we designed, fabricated, and assembled a new mass filter that allows ions within a set mass range to pass to the TOF section of the mass spectrometer while rejecting all other ions. For example, the filter can reject masses 1–50, thereby enhancing measurement capabilities for heavy atomic and molecular ions such as chemical and biological agents.

High-Average-Power, Intense Ion Beam for Materials and Other Applications

Blake P. Wood

We are developing a repetitive, intense ion-beam source that uses a magnetically insulated diode with an active plasma anode. This source will allow us to produce a 250-kV, 12-kA, 1-μs pulsed beam of any gaseous species of ion. Applications of the ion beam include materials surface treatment through rapid melt and resolidification; coating production at high rates; a diagnostic neutral beam for the next generation of tokamaks; and an intense, pulsed neutron source for neutron radiography, mine detection, and medical isotope production.

During the past year, we constructed all the vacuum chambers, including those housing the diode, plasma diagnostics, and beam transport. We constructed the plasma anode and used magnetic and optical probes to characterize its operation. We constructed and characterized the anode circuit drive. We constructed and performed initial testing on the puff-valve circuit drive. We also received the thyraton drivers and controllers, the pulse receiver, and the reset transformer from Northstar Research Corp., the company that constructed them.

Geoscience, Space Science, and Astrophysics

Low-Energy Neutral Atom Imager

Herbert Funsten

Low-energy neutral atom (LENA) imaging promises to be a revolutionary technique for providing important information on the global structure and dynamics of the terrestrial magnetosphere. Neutral atoms, formed by charge exchange of plasma ions with the neutral geocoronal atoms, follow ballistic trajectories that are not influenced by electric and magnetic fields and hence can be remotely detected. Observations of energetic neutral atoms provide line-of-site integrated measurements of the magnetospheric energetic particle and plasma ion populations. Regions of the magnetosphere that may be observable using a LENA imager include the ring current, the plasma sheet, and the magnetosheath.

Previous measurements of magnetospheric plasmas involve localized, single-point measurements from which the terrestrial magnetosphere's global structure and dynamics are extremely difficult to infer. LENA imaging can provide revolutionary global information on the magnetosphere, and these measurements can provide space weather forecasting, weather-induced satellite upset diagnostics, and revolutionary insights into global magnetospheric physics.

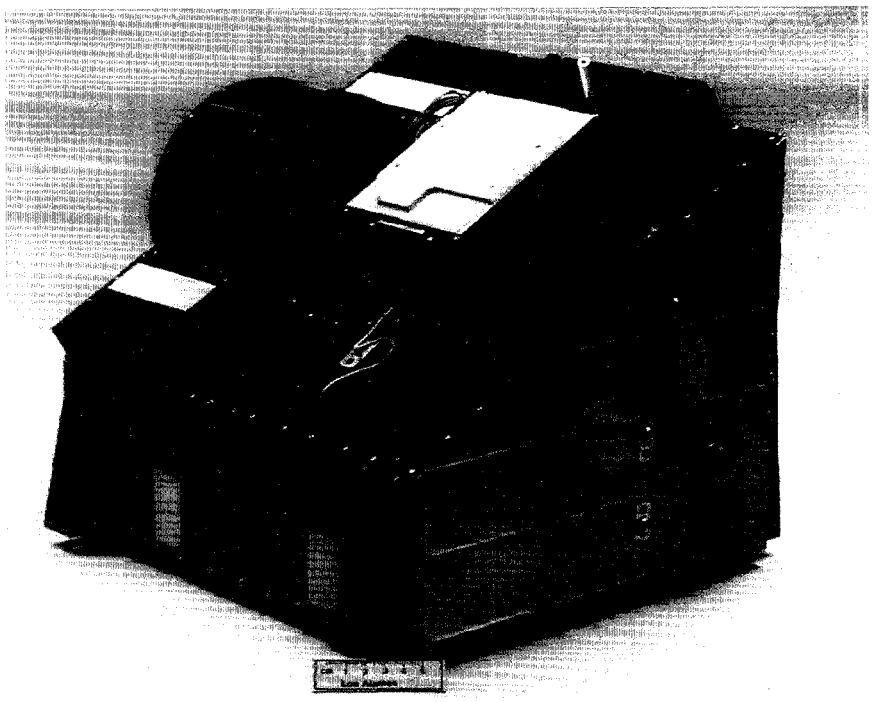
This year, a flight unit of the Los Alamos LENA imager was fabricated, assembled, tested, calibrated, mated with its data-processing unit, and delivered to our identified ride of opportunity. The imaging technique utilizes a collimator with alternately biased plates to reject ambient plasma ions, an ultrathin carbon foil to convert neutral atoms to positive ions, a hemispherical electrostatic analyzer for energy analysis of the ions, and a detector section consisting of LENA trajectory measurement and coincidence measurements. Next year, we expect to be able to turn on the imager and analyze the data it collects.

Publications

Funsten, H.O., et al., "E-Parallel-B Energy-Mass Spectrograph for Measurement of Ions and Neutral Atoms," *Rev. Sci. Instrum.* **68**, 292 (1997).

Funsten, H.O., et al., "Mechanisms for Neutral Atom Imaging," in *AGU Monograph: Measurement Techniques for Space Plasmas*, J. Borovsky, R. Pfaff, and D.T. Young, Eds. (American Geophysical Union, Washington, DC, in press).

McComas, D.J., et al., "Advances in Low Energy Neutral Atom Imaging," in *AGU Monograph: Measurement Techniques for Space Plasmas*, J. Borovsky, R. Pfaff, and D.T. Young, Eds. (American Geophysical Union, Washington, DC, in press).



The Los Alamos LENA imager weighs 5.4 kg, including power supplies and signal electronics. Neutal atoms enter the collimator (curved protrusion on trapezoid box), transit electrostatic analyzer plates (inside of trapezoid), and are detected in the detector section (rectangular box on trapezoid).

Theoretical Research on Dwarf and Classical Novae

Warren M. Sparks

The goal of this project is to understand mechanisms involved in the accretion processes in both novae and dwarf novae systems. We are particularly interested in how the accreted material from a disk or at a pole spreads and mixes over the rest of the star. Spectra taken with the Hubble Space Telescope of U Geminorum after two outbursts have been analyzed. The carbon abundance was down by 0.05 and the silicon down by 0.4 with respect to the Sun. We obtained an Einstein redshift of 80.4 km/s and hence a white dwarf mass of 1.1 solar masses. VW Hyi, a dwarf nova, has phosphorus overabundant by a factor of 900! This value, together with an overabundance of 15 for aluminum, 5 for nitrogen, and 2 for oxygen and an underabundance of 0.5 for carbon, is evidence of a thermonuclear runaway on a dwarf nova. Thus, we have found the first direct spectroscopic link between a dwarf nova and a classical nova.

We have shown that the high/low accretion states in AM Her binaries are caused by the magnetic torque of the white dwarf on the secondary. The missing boundary layer in cataclysmic variables expected from theory can be explained by accretion energy going into stellar rotation and by shear mixing spreading out the boundary layer. Using new reaction rates and OPAL opacities, we find less mass being accreted onto the white dwarf than previous models showed. Smaller amounts of aluminum-26 are produced, while the abundances of phosphorus-31 and silicon-32 increase by more than a factor of 2. The characteristics of our nova models compare favorably with observations of V1974 Cyg 1992, but the ejected mass is at least a factor of 10 less than observed.

Publications

Huang, M., et al., "Clues and Modelling for the Missing Boundary Layer in Cataclysmic Variables," *Astron. Soc. Pac. Conf. Ser.* **121**, 727 (1997).

Sion, E., et al., "Evidence of a Thermonuclear Runaway and Proton Capture Material on a White Dwarf in a Dwarf Nova" (submitted to *Astrophys. J. Lett.*).

Sion, E., et al., "Hubble Space Telescope GHRS Spectroscopy of U Geminorum during Two Outbursts," *Astrophys. J.* **483**, 907 (1997).

Sparks, W., et al., "Magnetic Torque Induced High/Low Accretion States in AM Herculis Binaries," *Astron. Soc. Pac. Conf. Ser.* **121**, 803 (1997).

Starrfield, S., et al., "Evolutionary Sequences for Nova V1974 Cygni Using New Nuclear Reaction Rates and Opacities" (submitted to *Mon. Not. R. Astron. Soc.*).

Starrfield, S., et al., "New Studies of Nuclear Decay Gamma-Rays from Novae," in *Proceedings of the 4th Compton Symposium on Gamma-Ray Astronomy and Astrophysics* (American Institute of Physics, Inc., Woodbury, NY, in press).

Sedimentary Basin Response to Strong Ground Motion in Populous Regions

Paul Johnson

An important origin of damage in modern metropolitan areas during large earthquakes is the amplification of seismic shear waves. The amplification is primarily due to local site resonances. Examples of amplification include all of the most damaging earthquakes in the past decade, including the 1994 Northridge, California, earthquake. This earthquake and others like it cost thousands of lives, mostly because of structural failure resulting from coupled shear-wave resonances, and cost tens of billions of dollars in structural damage. Consequently, understanding and mitigating the effect of the uppermost sedimentary layers on ground motion and the resulting response of engineered structures is of paramount importance and is the focus of our project. In particular, we are focusing on the long-standing and contentious debate as to whether wave amplification is reduced by nonlinear dissipative processes during large earthquakes.

During the past year, we completed a statistical analysis of the 1994 Northridge earthquake and its aftershocks. We found that the main earthquake produced widespread nonlinearity at alluvial sites in the Los Angeles Basin. This finding will have a significant impact on predictive studies for the design and construction of buildings and infrastructure.

Publications

Field, E.H., et al., "Nonlinear Sediment Amplification During the 1994 Northridge Earthquake," *Nature* **390**, 599 (1997).

Coronal Mass Ejections in the Solar Wind

John Gosling

Coronal mass ejections, CMEs, are spectacular solar events in which large amounts of material from the solar atmosphere are ejected into interplanetary space. CMEs play a central role in the long-term evolution of the structure of the solar corona and are the prime link between solar activity and large, transient solar wind and geomagnetic disturbances. The overall goals of this project are to distinguish and understand the physical processes governing CME evolution in the solar atmosphere and in interplanetary space.

This year we tested the idea that CMEs are the prime process by which closed magnetic field lines in the solar corona are opened up to interplanetary space. Models of the coronal magnetic field, derived from measurements of the magnetic field in the solar photosphere, were used to study the rate of opening of coronal magnetic fields to interplanetary space over a long time period (16 years). We found that model predictions of increases in the amount of magnetic flux open to interplanetary space corresponded well in a statistical sense to the rate at which CMEs were ejected from the sun and detected in interplanetary space. Specific flux increase events also were reasonably well correlated with specific observed CME events, in agreement with ideas relating CMEs to the opening up of coronal magnetic fields.

Other accomplishments during the year included (1) writing one of two introductory articles to the first book ever devoted entirely to the subject of CMEs and (2) writing an overview article on the solar wind and CMEs for a book for aerospace engineers.

Publications

Gosling, J.T., "Coronal Mass Ejections: An Overview," in *Coronal Mass Ejections*, N. Crooker, J.A. Joselyn, and J. Feynman, Eds. (American Geophysical Union, Washington DC, 1997), p. 9.

Gosling, J.T., "Physical Nature of the Low-Speed Solar Wind," in *Robotic Exploration Close to the Sun: Scientific Basis*, S.R. Habbal, Ed. (American Institute of Physics, New York, 1997), p. 24.

Gosling, J.T., "The Solar Wind and the Interplanetary Magnetic Field," in *Spacecraft Orbit and Attitudes Systems, Vol. 2*, H. Koenigsmann and J. Wertz, Eds. (Kluwer, The Netherlands, in press).

Gosling, J.T., and Pete Riley, "The Acceleration of Slow Coronal Mass Ejections in the High-Speed Solar Wind," *Geophys. Res. Lett.* **23**, 2867 (1996).

Luhmann, J.G., et al., "The Relationship Between Large Scale Solar Magnetic Field Evolution and the Interplanetary Signatures of CMEs" (submitted to *J. Geophys. Res.*).

Roberts, D.A., and J.T. Gosling, "In Situ Measurement Requirements for a Solar Probe," in *Robotic Exploration Close to the Sun: Scientific Basis*, S.R. Habbal, Ed. (American Institute of Physics, New York, 1997), p. 3.

Integrated Systems Analysis Applied to Environmental Remediation

R. Wayne Hardie

Our goal in this project is to explore the feasibility of selecting, adapting, extending, and then applying Los Alamos analysis capability to the creation of an objective systems tool to aid decision-makers in their evaluation of competing options for environmental remediation. The tool would allow us to integrate technical, economic, social, and political factors into our decisions. We created the tool by investigating methods to reverse the environmental decline in the Salton Sea, a decline thought to be caused by increased salinity. Therefore, the primary objectives of the options for reversing the environmental decline were concerned with reducing the salinity of the Salton Sea. We examined four technologies as solutions to reduce the salinity: desalination, pump-in/pump-out, pump-out, and diking.

We found the desalination and the pump-in/pump-out options to be very expensive. As proposed, they would not contribute to improving the status of the Salton Sea. Two of the technologies, pump-out and diking, turned out to be feasible solutions to reverse the decline in the Salton Sea. The costs for the two options were similar: $300 to $500 million for capital costs and $2 to $5 million per year for operating and maintenance costs. The pump-out option would create a smaller sea and would cause the present shoreline to shrink. The diking option would create a high-salinity impoundment area in the sea, but would reduce the salinity of the main part of the sea and maintain the present shoreline.

Publications

Hardie, R.W., "Evaluation of Options for Remediation of the Salton Sea," Testimony before the Subcommittee on Water and Power, US House of Representatives Committee on Resources (1997).

Striation-Image Monitoring of Plasmaspheric, L-Resolved Electrodynamics (SIMPLE)

Abram R. Jacobson

This project uses very long baseline interferometry (VLBI) illuminated by satellite-borne radio beacons to synthesize images of irregularities in Earth's inner magnetosphere. These irregularities in the geoplasma are highly elongated along the geomagnetic field and cast stripe-shaped phase shadows on the interferometer plane, when considered as a radio-frequency image. The array baseline for the VLBI was hundreds of meters.

Our objective this year, which we have accomplished, was to extend the plasmasphere-imaging technique for beacons from two satellites viewed in nearly the same meridian. Since the end of December 1995, the array has continuously monitored radio beacons aboard satellites GOES-3 and ATS-3 near the longitude range 103°–107° west. We now have amassed a substantial database of dual observations and are preparing for a data-reduction effort, during the final year of the project, to extract the location of the irregularities in the plasmasphere from the dual observations. We stopped the observations on September 1, 1997; then we disassembled and packed the array.

Publications

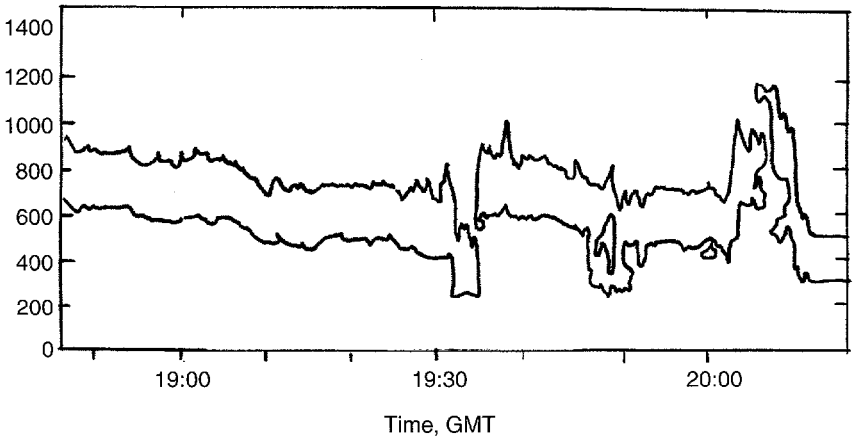
Hoogeveen, G.W., and A.R. Jacobson, "Radio Interferometer Measurements of Plasmasphere Density Structures during Geomagnetic Storms," *J. Geophys. Res.* **102**, 14177 (1997).

Data/Model Integration for Vertical Mixing in the Stable Arctic Boundary Layer

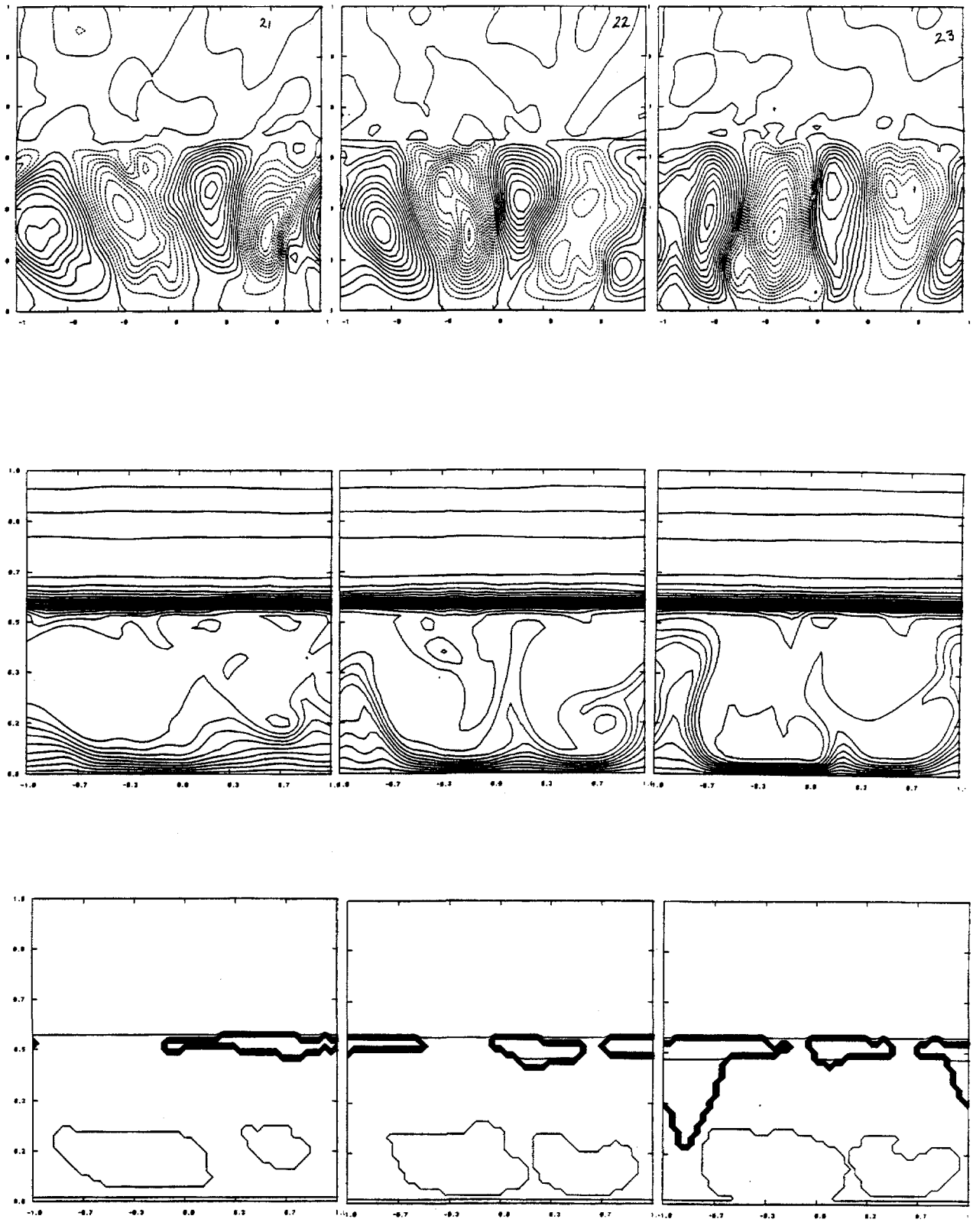
Sumner Barr

Our research focused on a signature of the lidar-observed stratus that appears to be related to mixing of deep layers of the arctic boundary layer. A segment of cloud deck separates from the quasi-continuous layer and rises or falls a few hundred meters (see first figure). We identified 25 examples of this feature in the lidar data, and many of the cases coincide with major concentration excursions in trace constituents. Such a relationship is not intuitive. The stratus layers occur at altitudes up to 1.5 km above the surface, while the trace constituents are measured near the surface. The correlation indicates a dynamical connection across deep layers of the arctic atmosphere. In mid-latitudes, where convection is a dominant mixing mechanism, mixed layers of several-kilometer depth are common, but the very stable density profiles in the Arctic suppress convection. In that environment, the coupling across the lower kilometer of the atmosphere is a surprising result that requires good dynamical models to understand. We have exercised two models and are encouraged by the results.

We performed a simulation using HIGRAD, a very high resolution dynamical model, and observed the results (second figure). The model oscillates semihourly between two modes: one is a series of roll vortices that fill the layer below the temperature inversion, while the other involves smaller-scale circulations that are less effective at mixing. The second model, BLMARC, is one-dimensional and examines turbulence, chemistry, cloud, and infrared radiation physics.



A copy of lidar imagery that displays discontinuities in stratus cloud decks. Negative perturbations, or "drop outs," occur at 19:35 and 19:50 GMT, and a "lift out" event occurs at 20:05–20:12 GMT (data from July 15, 1996).



Evolution of three parameters—vertical velocity (top), water vapor (middle), and cloud aerosol (bottom)—at 400-second intervals over about 30 minutes, taken from HIGRAD model output. The model simulates observed perturbations in cloud layer height.

Determining the Mass of the Universe

Michael Warren

The average mass density of the universe, parametrized by Omega, is the most sought after single number in cosmology. It determines whether the universe is open and forever expanding or closed and eventually collapsing. Unfortunately, after a half century of research, Omega is still uncertain by at least a factor of 5. We have the tools at hand to significantly improve the measurements of the mass of the universe. We will test the conventional approximations of Omega with state-of-the-art numerical simulations of gravitational clustering in the universe.

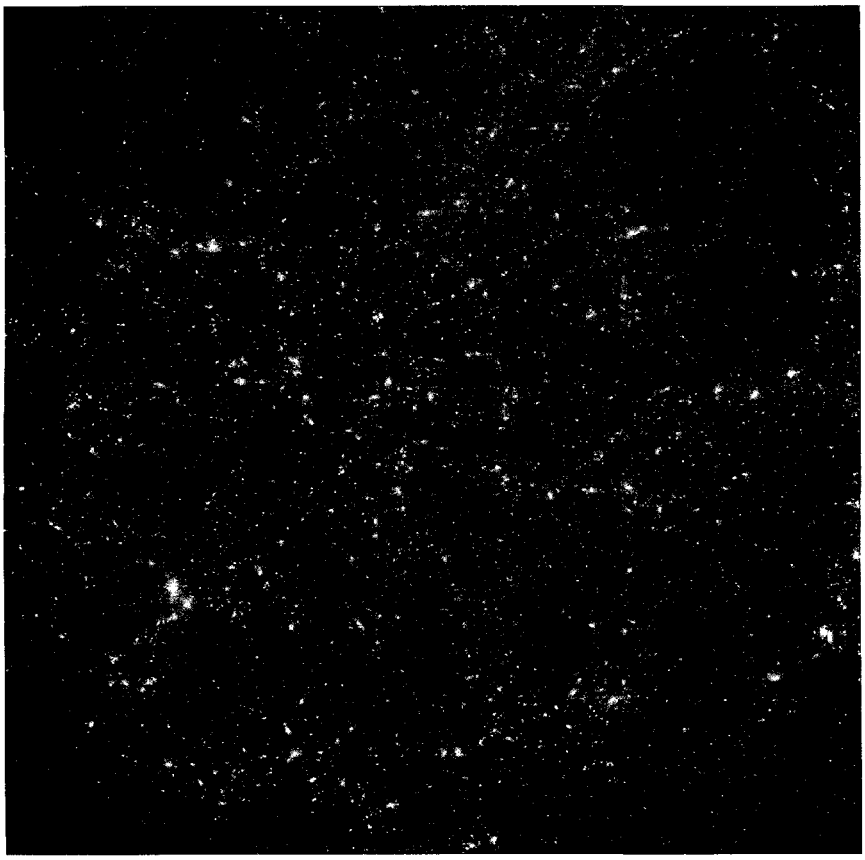
We are currently extracting Omega from the Las Campanas Redshift Survey, a catalog of more than 25,000 galaxies. The survey is the largest now available. This observational data set will be used in combination with the recently completed, 322-million-particle simulation (by far the largest gravitational *N*-body simulation ever performed) to test and refine the cosmic virial theorem and other methods of determining Omega.

We have modified our original approach toward measurements of Omega to include large-scale flows. With simulations we have optimized measurements of Omega from the statistical anisotropy in redshift surveys induced by large-scale flows. One of the simulations performed for this project (see the accompanying figure) won the 1997 IEEE Gordon Bell Prize in recognition of superior effort in practical parallel-processing research. Our tree code, which scales as $N \log N$, sustained 170 Gflops over a continuous 9.4-h period on 2048 nodes of the Accelerated Strategic Computing Initiative (ASCI) Red teraflops system, integrating the motion of 322 million mutually interacting particles for 288 time steps, while saving over 100 Gbytes of raw data.

Publications

Bromley, B.C., et al., "Estimating Omega from Galaxy Redshifts: Linear Flow Distortions and Nonlinear Clustering," *Astrophys. J. Lett.* **475**, 414 (1997).

Warren, M.S., et al., "Pentium Pro Inside: I. A Treecode at 430 Gflops on ASCI Red, II. Price/Performance of $50/Mflop on Loki and Hyglac," in *Supercomputing '97* (IEEE Computing Society, San Jose, CA, 1997).



Measuring the mass of the universe. The image shows an intermediate stage of a gravitational *N*-body simulation using 322 million particles, computed on ASCI Red. Using initial conditions derived from fundamental theories, we simulated the evolution of matter in the universe from a time shortly after the Big Bang through its nonlinear evolution to the present epoch. The illustrated region is about 400 million light years across, and the brightness of each pixel represents the logarithm of the projected particle density along the line of sight through the periodic computational volume. The particles have formed clumps that represent dark matter halos, which are the sites where galaxy formation occurs.

Heterogeneous Processing of Bromine Compounds by Atmospheric Aerosols: Relation to the Ozone Budget

Jeanne M. Robinson

Catalysis of bromine compounds on atmospheric aerosols, such as ice, solid nitric acid hydrates, and sulfuric acid aerosols, contributes to the observed ozone losses in the lower stratosphere globally and in the arctic marine boundary layer in early spring. The current lack of understanding of heterogeneous bromine chemistry on atmospheric particulates is a significant impediment to predicting future ozone levels and developing a rational policy on the use of methyl bromide.

In this project, we studied HBr interactions with porous polycrystalline ice films at stratospheric temperatures and near stratospheric pressures. Using a mass spectrometer, we measured the vapor pressures of HBr and H_2O over ice. We also simultaneously monitored transformations at the ice surface as a function of HBr exposure using a direct surface-specific probe, second harmonic generation (SHG). From our vapor pressure measurements, we constructed HBr adsorption isotherms on ice (see the figure) that are modeled by classic physical adsorption theory. Our SHG signals elucidated the phase transitions triggered at the ice surface by HBr and their effects on further HBr uptake. Comparing the results on HBr/ice with our previous work on HCl/ice gave us insight into heterogeneous halogen activation mechanisms and their impact on atmospheric chemistry.

Publications

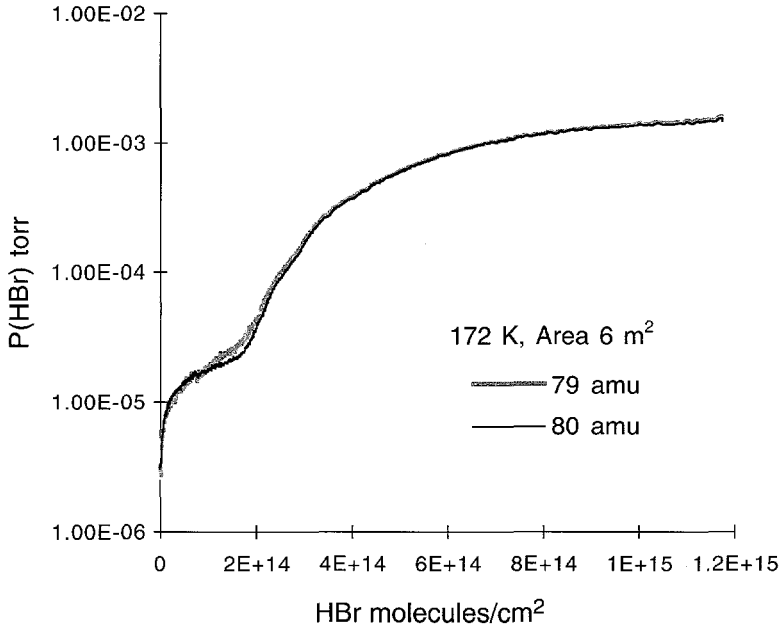
Henson, B.F., et al., "Experimental Isotherms of HCl on Water Ice under Stratospheric Conditions" (submitted to *Science*).

Henson, B.F., et al., "Measurements of Hydrogen Chloride Adsorption Isotherms on Water Ice from 180K to 200K at Near Stratospheric Pressures," *Book Abstr., 214th ACS Nat. Meet.* **214** (Part 2), 400 (1997).

Henson, B.F., et al., "Models of Hydrogen Chloride Adsorption and Chlorine Nitrate Reactivity on Simulated PSC Surfaces," *EOS Trans., Am. Geophys. Union* **77**, F95 (1996).

Henson, B.F., et al., "Porous $HNO_3(H_2O)_n$ Ice Films: Adsorption and Surface Thermodynamics Measured by Second Harmonic Generation" (submitted to *J. Phys. Chem.*).

Henson, B.F., et al., "Quantitative Measurements of Multilayer Physical Adsorption on Heterogeneous Surfaces from Nonlinear Light Scattering," *Phys. Rev. Lett.* **79**, 1531 (1997).



Adsorption isotherm of HBr on ice. A neat water-ice film was exposed to a low flow of gaseous HBr (1×10^{-8} mole/s) to ensure equilibrium. The equilibrium vapor pressure of HBr above the ice, measured by calibrated quadrupolar mass spectrometer signals at 79 amu (^{79}Br) and 80 amu ($H^{79}Br$), is shown on the vertical axis. The time integral of the gaseous HBr flow normalized by the ice surface area in molecules per centimeter squared is shown on the horizontal axis. The HBr pressure rises sharply when the ice has been coated with a monolayer of HBr (2×10^{14} molecules/cm^2).

High-Velocity Neutron Stars

Edward Fenimore

Gamma-ray bursts (GRBs) are brief, high-energy explosions that flicker for tens of seconds and that originate from either high-velocity neutron stars in our galaxy or merging neutron stars in far away galaxies. Our objective is to investigate the physics related to the global characteristics of the burst population and the detailed physics of the relativistic shells that are responsible for the production of the gamma rays.

We used the newly discovered x-ray afterglows to predict the number of GRBs that should be seen from nearby galaxies and started a search to detect them. We have excluded all neutron star and merging neutron star models that use a single release of energy to produce a relativistic expanding shell. In particular, we showed that a single relativistic shell cannot produce either the observed gaps in the emission or the average profile of emission.

We also estimated the efficiency of single relativistic shells, that is, what fraction of the shell's surface could contribute to the emission. This efficiency is extremely low (about 0.001). Since most models involving merging neutron stars require the energy to be released on a short time scale, the only currently accepted models of the neutron stars from far away galaxies are those that can release, for hundreds of seconds, energy equivalent to the rest mass of the Sun. Although this release might occur under extreme conditions, we believe the afterglows arise only from single relativistic shells that are decelerating.

We then completed models of decelerating shells and determined their ability to flicker. We compared the observed flickering in the x-ray afterglow with the models and concluded that a single relativistic shell could not explain the observations. Thus, we showed that the central source that produces GRBs must continue to release energy for as long as several days.

Publications

Duncan, R.C., and H. Li, "The Halo Beaming Model for Gamma-ray Bursts" (to be published in *Astrophys. J.*).

Fenimore, E.E., and M.C. Sumner, "Gamma-Ray Bursts: Relativistic Shells or Central Engines" (to be published in *Proc. All-sky X-ray Observations in the Next Decade*).

Fenimore, E.E., et al., "Expanding Relativistic Shells and Gamma-Ray Burst Temporal Structure," *Astrophys. J.* **473**, 998 (1996).

Fenimore, E.E., "Expanding Relativistic Shells and Limitations on Gamma-Ray Burst Temporal Structure" (to be published in *Proc. 18th Texas Symp. Relativistic Astrophys.*).

Li, H., and E.E. Fenimore, "Log Normal Distributions in Gamma-ray Burst Time Histories," *Astrophys. J. Lett.* **469**, 115 (1996).

Supermassive Black Holes and the Strong Field Limit of General Relativity

Wojciech Zurek

Los Alamos pioneered the use of analysis of x-ray line profiles to measure the angular momentum of the supermassive black holes in the center of Seyfert galaxies. To date we have the only computer code general enough and with enough resolution to examine the complex nonaxisymmetric phenomenon that certainly exists close to the innermost stable orbits in galactic centers. Although we have the first preliminary measurement of frame dragging for MCG-6-30-15, we hope that the next generation of x-ray satellites will provide a conclusive measurement of the angular momentum of these central black-hole engines.

We addressed two topics this year: The first was a statistical analysis of the differences between the broad-line x-ray emission from the vicinity of a rotating black hole and that of a Schwarzschild black hole. The second was an emissivity-independent approach to modeling the angular momentum.

We are currently using our ray-tracing code to place more-stringent limits on the inner radius of the x-ray emission. We will develop an emissivity-independent measure of angular momentum. The geometry of the inner portion of the accretion disk is no doubt complex, and if we hope to have a conclusive measure of the frame-dragging effect, it should be robust. We are also preparing to analyze more-accurate data that is forthcoming from the new generation of x-ray satellites.

Publications

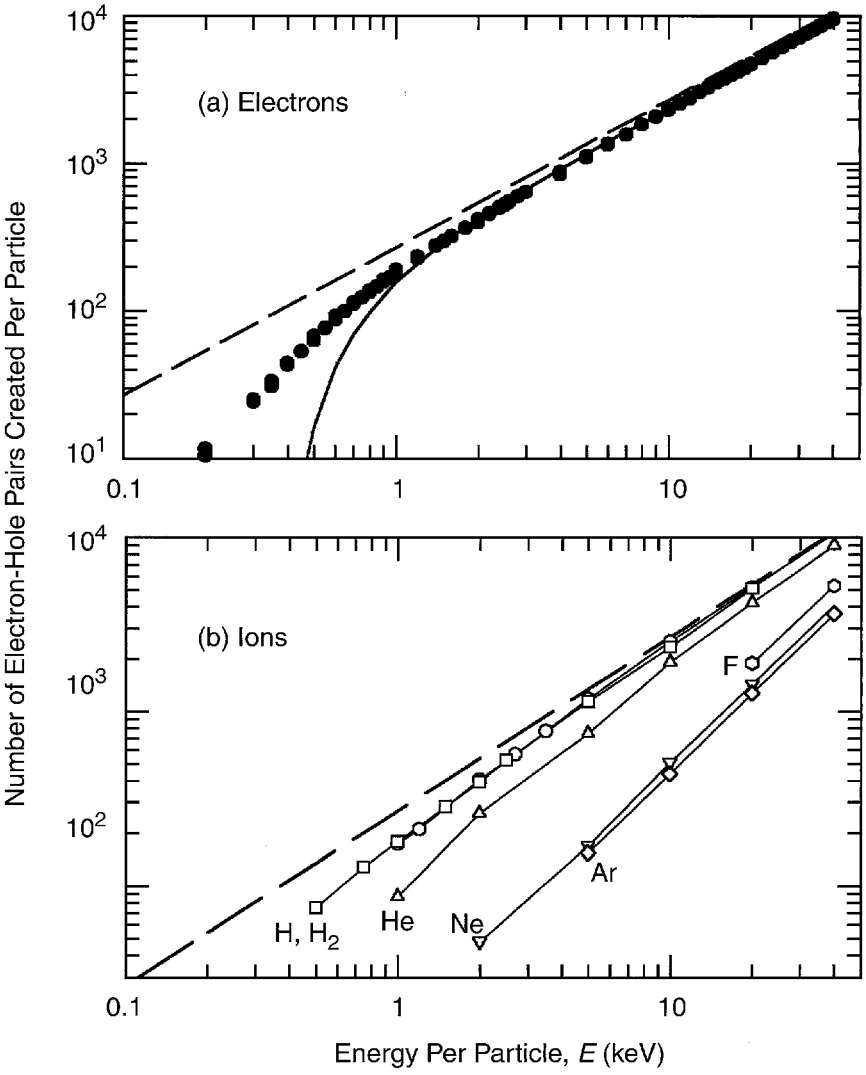
Bromley, B.C., et al., "The Inner Edge of the Accretion Disk around a Supermassive Black Hole" (to be published in *Nature*).

A New Class of Sensors for Detecting Low-Energy Particles in Space

John Gosling

The low-energy threshold (15–20 keV) for detecting ions and electrons with solid-state detectors has been caused by thick "dead layers" (inert layers of SiO_2 in the detectors). In this one-year project, we investigated the energy threshold using a new type of 100% internal-quantum-efficiency silicon photodiode with a 60-Å passivation layer and recently developed, delta-doped solid-state detectors.

Important results from our study include the following. We measured the number of electron-hole pairs created per incident ion or electron (see accompanying figure). Modeling of the electron responsivity with a Monte Carlo simulation indicated that the electron-hole pairs created in the SiO_2 passivation layer are efficiently collected. We determined that the solid-state detector is damaged by ion bombardment and quantified that damage using the responsivity, shunt resistance, and series resistance. We found that the damage increases exponentially with fluence, that the damage per incident ion increases with increasing ion mass and correlates with the production of Frenkel defects, and that the damage involves both the creation of recombination centers and a distortion of the band structure in the heavily doped near-surface region of the photodiode that together inhibit the transport of holes across the junction.



Average number of electron-hole pairs formed for (a) electron and (b) ion bombardment of a thin-window semiconductor detector. The dashed line in each panel corresponds to the ideal value based on the 3.7-eV pair-creation energy for silicon. In panel (a), deviation from a Monte Carlo simulation (solid line) indicates that electron-hole pairs created in the SiO_2 passivation layer are efficiently collected. In panel (b), deviation from the ideal for heavier ions is due to energy loss processes that do not result in ionizations in the detector.

Publications

Funsten, H.O., et al., "Response of 100% Internal Quantum Efficiency Silicon Photodiodes to 200 eV to 40 keV Electrons," *IEEE Trans. Nucl. Sci.* **44**, 2561 (1997).

Ices on Titan: Laboratory Measurements that Complement the Huygens Probe

Jeanne M. Robinson

In this project, we study the physical chemistry of Saturn's moon Titan to address the heterogeneous processes influencing both its present state and past evolution. We have characterized vapor-deposited water ice films that mimic Titan's porous crust using a unique combination of complementary methods in situ.

First, we used physical adsorption to measure the surface area of the films and the distribution of the pore radii (see the first figure). The form of the isotherms is Brunauer Type IV, indicative of mesoporous solids with a pore distribution between 10 and 100 Å.

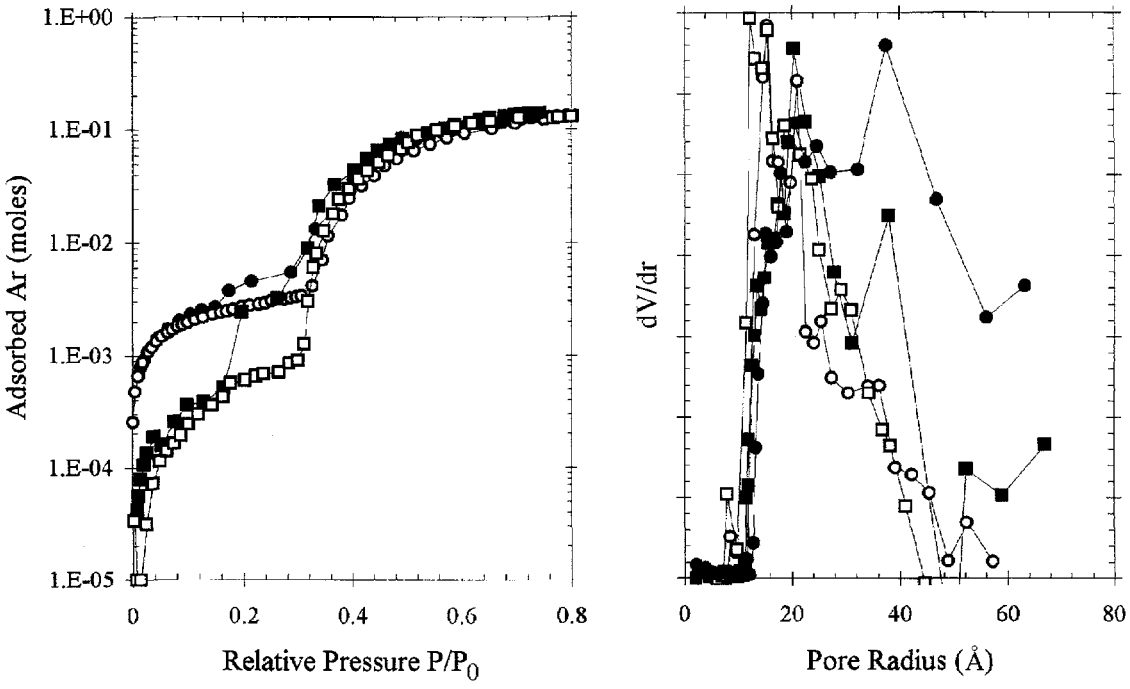
Second, we used quadrupole mass spectrometry to measure the vapor pressure of water over the ice surface during annealing (see the second figure). We observed that unannealed ice films grown at low temperature are metastable, displaying higher vapor pressure than polycrystalline ices do, and that annealing to about 200 K results in a polycrystalline ice form in equilibrium with the vapor.

Third, we used second harmonic generation spectroscopy to monitor the ice/vapor interface during annealing (see the third figure). We report the first nonlinear optical measurements of two transformations of water ice. One signature correlated with the weak glass transition observed near 130 K by calorimetry. An additional signature correlated with the amorphous-to-crystalline transition and supported the results from Brunauer-Emmett-Teller surface analysis suggesting that the amorphous ice film presents a more energetic, polarizable surface due to uncoordinated hydrogen bonds at the surface than the crystalline films do. The results show that in situ characterization is essential to evaluate the role of surface area and porosity in the uptake of gases by the porous ice.

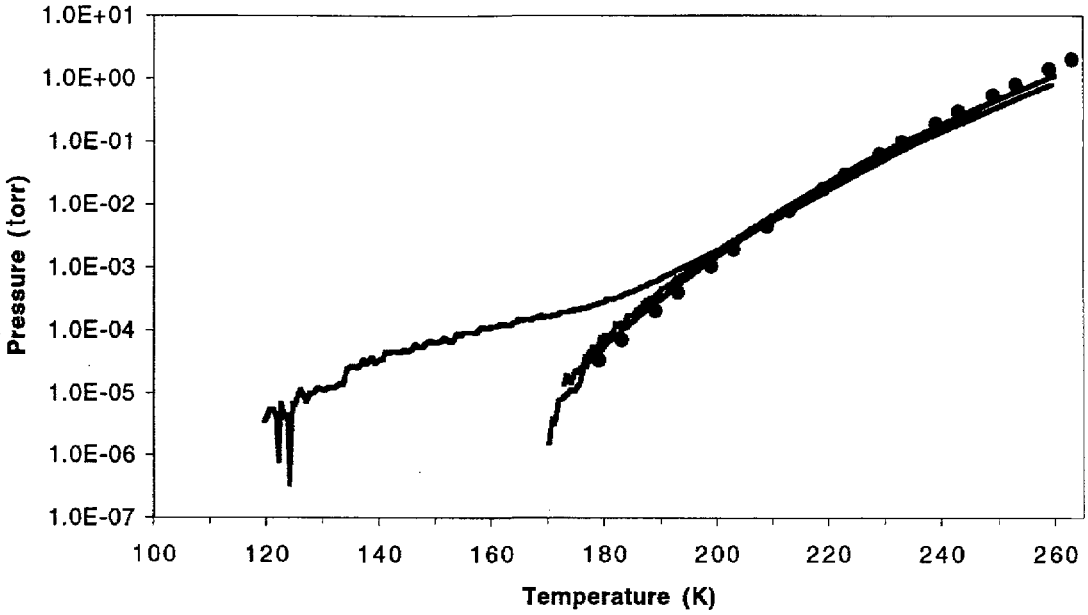
Publications

Henson, B.F., et al., "Characterization of Porous Water Ice Films by Physical Adsorption, Mass Spectrometry, and Second Harmonic Generation" (submitted to *J. Phys. Chem.*).

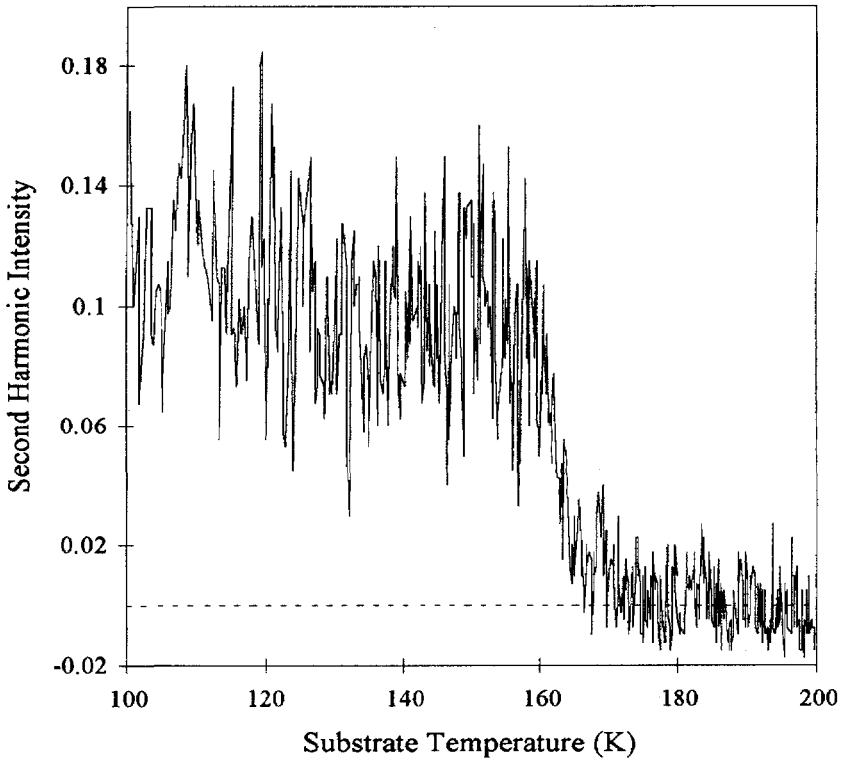
Henson, B.F., et al., "Quantitative Measurements of Multilayer Physical Adsorption on Heterogeneous Surfaces from Nonlinear Light Scattering," *Phys. Rev. Lett.* **79**, 1531 (1997).



Physical adsorption measurements of surface area and pore distributions for water ice films. The left panel shows the adsorption (open symbols) and desorption (solid symbols) isotherms for argon onto 0.5-g H_2O ice films condensed at 85 K with (squares) and without (circles) annealing to 180 K. The data are plotted as the log of the moles of gas adsorbed as a function of the pressure relative to the condensation pressure P_0. The form of the isotherms at high coverage is roughly Type IV and results from capillary condensation onto the mesoporous solid. The hysteresis at high pressure is indicative of network effects in the mesoporous matrix. The right panel shows the pore distributions calculated from the high-pressure region of the isotherms using the Kelvin equation. The data are plotted as the derivative of the volume adsorbed as a function of the pore radius.



Measurements of the vapor pressure of water over ice films as a function of temperature using mass spectrometry. The ice film was condensed at 85 K, slowly annealed to 260 K, then cooled and rewarmed. The solid circles are the known vapor pressures of water based on the Clausius-Clapeyron relationship. We used a calibration factor of 3×10^7 for the ion signal from the mass spectrometer based on pressure measurements with a capacitance manometer above about 10 mtorr. For heating above ~200 K and subsequent cooling and rewarming, the polycrystalline ice is in equilibrium with the known vapor pressures.



Second harmonic generation spectrum from a 0.5-g sample of neat water ice condensed at 90 K during annealing to 200 K. The glass transition at 130 K and the amorphous-to-crystalline transition at higher temperature were detected using this surface-sensitive technique.

Numerical Simulations of Convection Experiments and the Earth's Interior

Gary Glatzmaier

We are studying the dynamics of Earth's core by conducting three-dimensional, dynamically self-consistent computer simulations of convection and magnetic field generation and comparing the results with laboratory experiments and geophysical observations.

Our computer simulations of the Geophysical Fluid Cell Experiment (GFFC), which was flown aboard the NASA space shuttle Columbia in November 1995, have improved our understanding of how rotation and differential boundary heating can affect the structure of convection in Earth's core. Our geodynamo simulations further show how this convection maintains Earth's magnetic field.

Our model has maintained an Earth-like (simulated) magnetic field for an additional 250,000 years, and during this time the magnetic field has spontaneously undergone two more magnetic dipole reversals (see figure) that are very similar to what is seen in the Earth's paleomagnetic reversal record. This was done with a prescribed heat flux pattern on the interface between the core and the mantle that is similar to Earth's present pattern.

For comparison, we did another simulation using instead a uniform heat flux at this interface (unlike Earth's nonuniform flux). So far this simulation has not produced a magnetic reversal. This failure to produce a magnetic reversal illustrates how influential the pattern of heat flux out of the core, which is controlled by the mantle, is on the magnetodynamics of the fluid core.

Publications

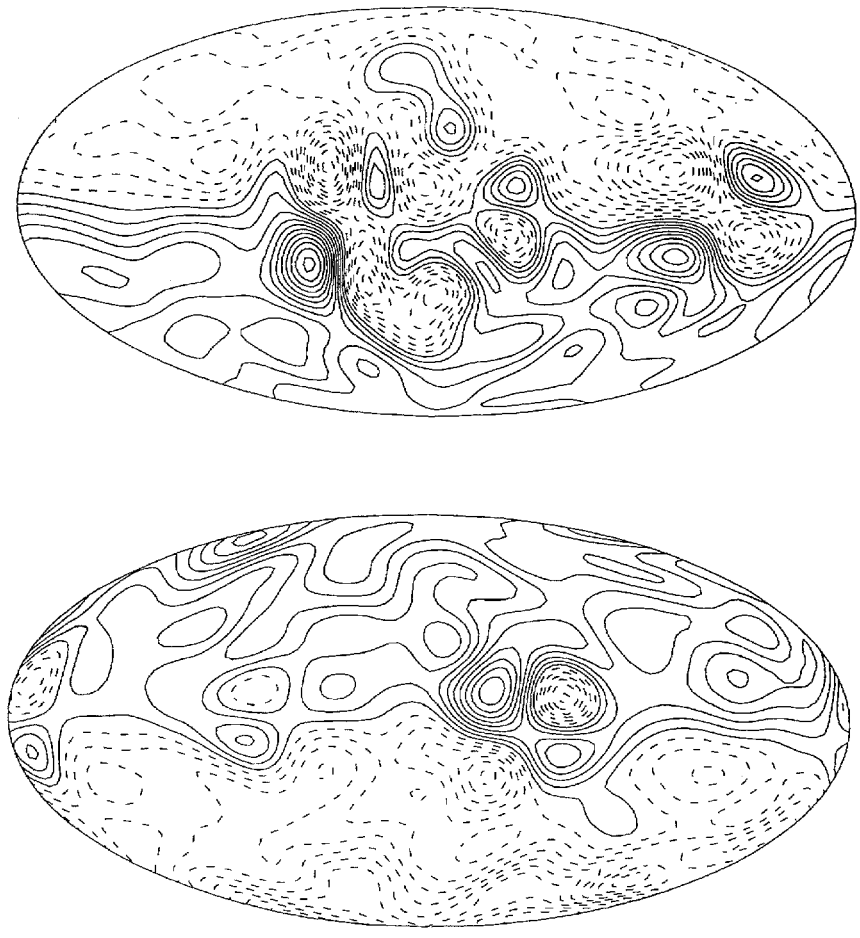
Glatzmaier, G.A., and P.H. Roberts, "An Anelastic Evolutionary Geodynamo Simulation Driven by Compositional and Thermal Convection," *Physica D* **97**, 81 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Computer Simulations of the Earth's Magnetic Field," *Geowissenschaften* **15**, 95 (1997).

Glatzmaier, G.A., and P.H. Roberts, "On the Magnetic Sounding of Planetary Interiors," *Phys. Earth Planet. Inter.* **98**, 207 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Rotation and Magnetism of Earth's Inner Core," *Science* **274**, 1887 (1996).

Glatzmaier, G.A., and P.H. Roberts, "Simulating the Geodynamo," *Contemp. Phys.* **38**, 269 (1997).



The radial component of the simulated magnetic field plotted at the top boundary of the core before (top image) and after (bottom image) the dipole reversal. The images are plotted in equal area projections, showing the entire core-mantle boundary surface; the equator runs horizontally through the middle of each image, separating the northern hemisphere (upper part) from the southern hemisphere (lower part). Solid contours represent outward directed field; broken contours, inward field.

Numerical Simulation of Explosive Volcanism and Its Effects on the Atmosphere

Gregory A. Valentine

The objective of this project is to advance the state of numerical simulation of explosive volcanic processes by adapting three-dimensional, multiphase, multicomponent computational fluid-dynamics codes to volcanic regimes and to use these models as input for regional atmospheric models of downwind ash-plume transport.

This year we did a comprehensive review of explosive volcanism modeling to date and used the results as part of the basis for a new multifield theoretical framework for describing eruption phenomena.

We began using the CFDLIB family of codes to model near-field (high-velocity, -temperature, and -particle-loading regimes) dynamics of explosive eruptions. Much of our effort went into learning the codes and their underlying theories in order to be most efficient in applying them to volcanic problems. Results to date include a model of simple gas explosions and initial work on loading explosions with ash particles.

We performed numerical experiments with the regional meteorological code RAMS to determine what, if any, modifications would be necessary for the code to be able to model intense thermal buoyancy effects, ash transport, and fallout in explosive eruption clouds as they are transported downwind. In our initial calculations for small-scale (but quite common) explosive eruptions, we determined that velocities of up to about 80 m/s can be computed without any modification to RAMS. We are exploring the adaptation of the numerical models of precipitation and condensation in rain clouds to fallout and particle coalescence in volcanic plumes.

Publications

Valentine, G.A., "Eruption Column Physics," in *Physics of Explosive Eruptions* (Elsevier Science Publishers, Amsterdam, in press).

Cosmology with Massive Neutrinos

Wojciech Zurek

Addition of massive neutrinos to the standard cold dark matter (CDM) cosmology was originally motivated by the indications that the galactic-scale structure has less power than CDM would predict. This conflict became particularly acute after the Cosmic Background Explorer established rather stringent bounds on the large-scale power. In the past year we have focused on assessing the reliability of the observational methods that have led to the indications of conflict between large and galactic scales. We have used "observational" methods (i.e., methods used by observers on the observational data to infer inherently three-dimensional information about power on various scales) on our simulations. We have exhibited several systematic biases introduced by these methods and assessed their reliability by comparing the mock observation results with simulations (where full information about the power is available). The conclusion is a significant decrease of the conflict between CDM and observational data.

The main recent accomplishment is the success of our methods in the comparison run of a large (Abell) cluster of galaxies. Our code has proved to have an excellent small-scale resolution, which allowed it to reproduce galactic-scale data better than approximately a dozen competing groups, without the necessity to include a cumbersome and model-dependent baryonic component. Using that accomplishment as a stepping stone, we expect to run large-scale simulations (10^8 particles) to model formation of structure on galactic and large scales. We shall compare observational predictions of standard CDM, mixed dark matter with neutrinos, and models with $\Omega < 1$.

Publications

Bromley, B.C., et al., "Cosmic Structure on Small Scales: Results on Cluster Cores and Redshift-Space Power Spectra," in *Mapping, Measuring and Modeling the Universe: Workshop Held in Valencia, Spain, 18–22 September 1995,* P. Coles et al., Eds. (Astronomical Society of the Pacific, San Francisco, CA, 1996), Conf. Ser. 94.

Bromley, B.C., et al., "Estimating Omega from Galaxy Redshifts: Linear Flow Distortions and Nonlinear Clustering," *Astrophys. J.* **475**, 14 (1997).

Zurek, W.H., et al., "Star-Disk Collision and the Origin of Broad Lines in Quasars," *Astrophys. J.* **470**, 652 (1996).

Evolution of Coronal Mass Ejections in the Solar Wind at Low and High Heliographic Latitudes

John Gosling

Coronal mass ejections (CMEs) are spectacular events in which large amounts of material from the solar atmosphere are ejected into the solar wind. The goal of this project is to use numerical simulations to study the evolution of CMEs in the solar wind over a wide range of heliographic latitudes.

The Ulysses spacecraft has shown that the solar wind flows much faster at high heliographic latitudes than at low latitudes. We have used a two-dimensional hydrodynamic code to study the evolution of a CME in such a latitudinally varying solar wind. We found that the CME produced radically different types of disturbances at low and high latitudes (see first figure). Our results compare favorably with observations of CME-driven disturbances made by Ulysses at a variety of latitudes.

We used a one-dimensional hydrodynamic code to explore how initial conditions affect the radial evolution of solar wind disturbances associated with CMEs that have initially high internal pressures. We found that the type of disturbance observed depends importantly on the relative motion between the CME and the ambient wind, and that our simulation results agreed well with Ulysses observations of CME-driven disturbances at high latitudes (see second figure).

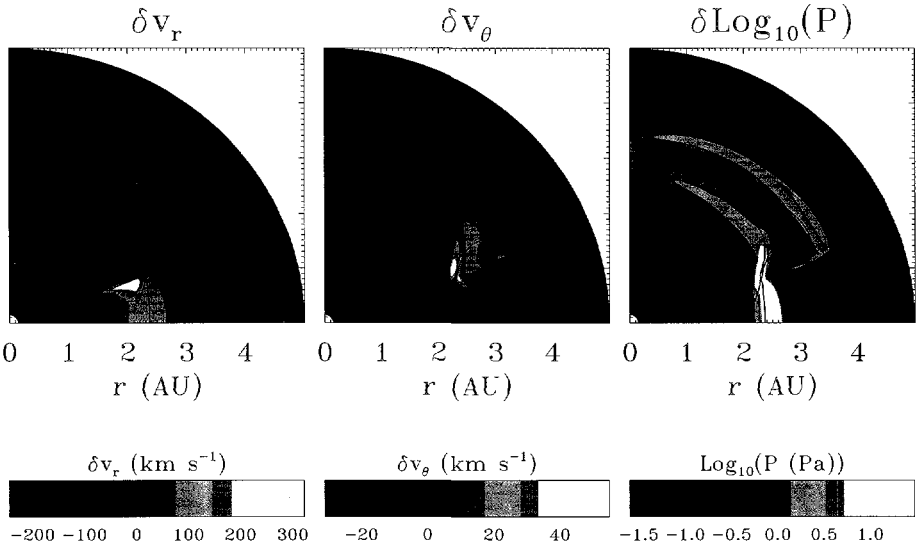
We also used the one-dimensional code to explore whether the rarefaction produced by an overexpanding CME (i.e., one whose expansion is driven by an initially high internal pressure) eventually leads to an implosion within the CME at large heliocentric distances. We found that for a wide range of initial perturbations close to the Sun, the CMEs do not implode at any heliocentric distance because of the overall reduction in pressure associated with the spherical expansion of the solar wind (see third figure).

Publications

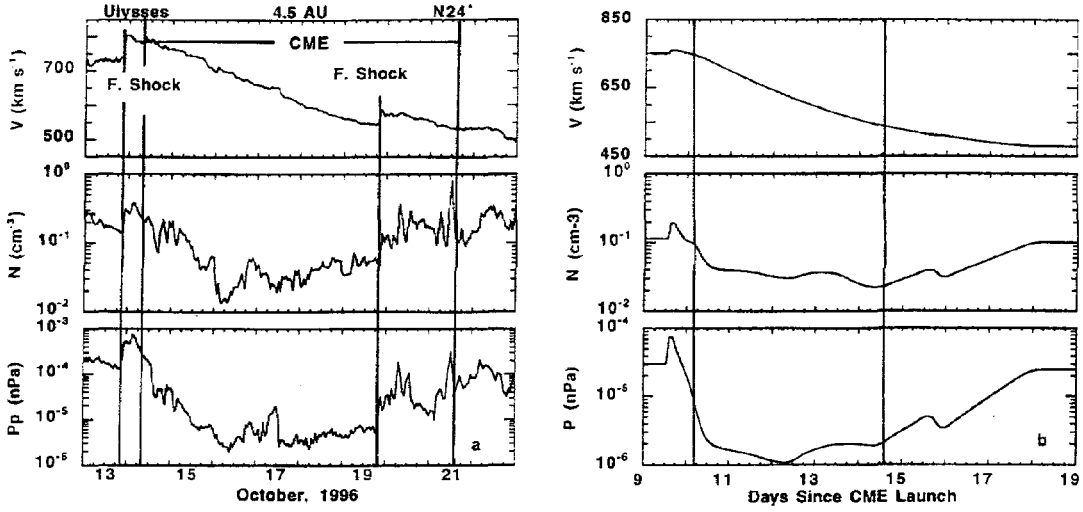
Gosling, J.T., et al., "Over-Expanding Coronal Mass Ejections at High Heliographic Latitudes: Observations and Simulations" (to be published in *J. Geophys. Res.*).

Riley, P., and J.T. Gosling, "Do Coronal Mass Ejections Implode?" (submitted to *Geophys. Res. Lett.*).

Riley, P., et al., "A Two-Dimensional Simulation of the Radial and Latitudinal Evolution of a Solar Wind Disturbance Driven by a Fast, High-Pressure Coronal Mass Ejection," *J. Geophys. Res.* **102** (14), 677 (1997).

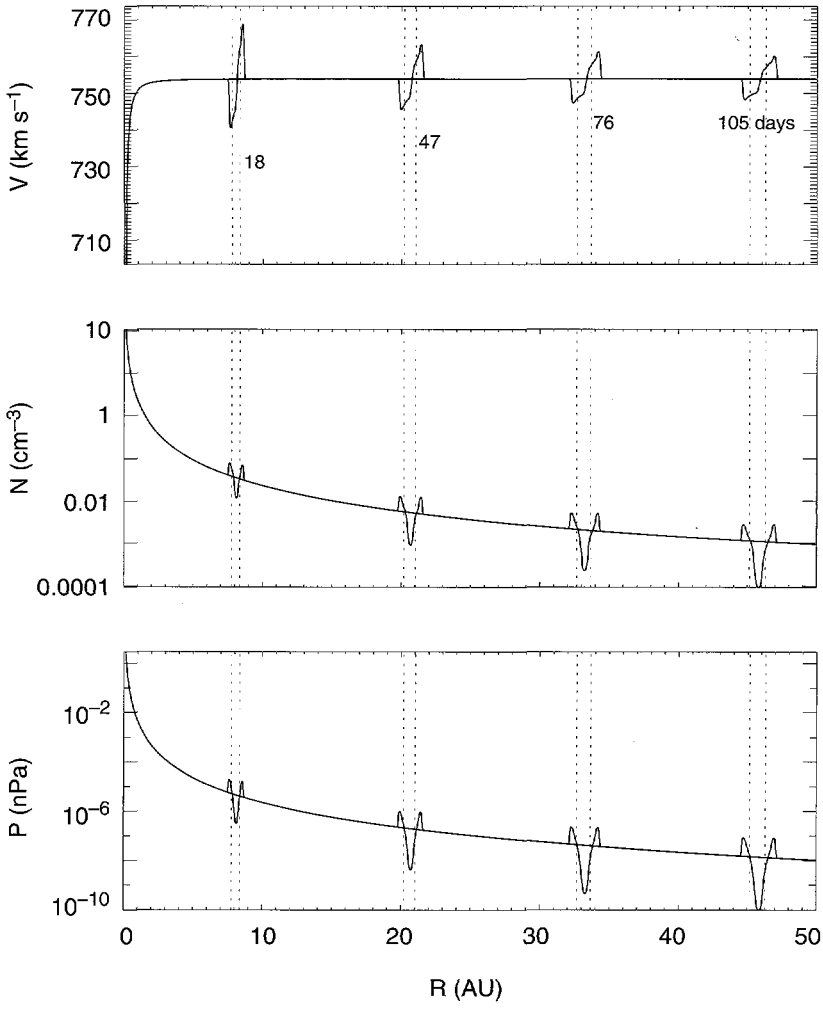


Grey-scale plots of (left to right) radial velocity, meridional velocity, and pressure in a meridional plane of the heliosphere ~7 days after the launch of a coronal mass ejection. The CME is introduced into the ambient solar wind, whose speed and density vary with latitude, as a bell-shaped pressure pulse over an interval of 10 hours; this pulse has a speed and temperature equal to that of the high-latitude wind. The plots are shown as differences between the solution at ~7 days and the initial steady-state solution. The solid line marks the boundary of the CME in the simulation. Note that the CME separates into high- and low-latitude pieces and that the disturbance is qualitatively different at the two latitudes. Radius is given in astronomical units (AU).



(a) (b)

(a) Ulysses measurements of solar wind speed, proton number density, and proton thermal pressure for a CME observed 4.5 AU from the Sun. The first vertical line marks a forward shock driven by the CME; the third vertical line marks a forward shock produced by a trailing CME. (b) Simulated profile of the CME observed by Ulysses. Vertical lines bracket the material introduced with a higher pressure close to the Sun and thus identify the CME.



Snapshots of solar wind speed, density, and pressure as a function of heliocentric distance for a disturbance driven by an over-expanding CME. The dotted lines indicate the outer edges of the expanding CME in the simulation. Note that the CME continues to expand to the outer bound of the simulation, giving no evidence of an implosion.

Close Encounters of Asteroids and Comets to Planets

Jack Hills

The well-publicized saga of Comet Shoemaker-Levy 9 illustrates the importance of tidal encounters between asteroids or comets and the planets. Comet Shoemaker-Levy 9 was first captured into a chaotic orbit about Jupiter as a result of the differential (tidal) pull of the Sun and the planet. After orbiting Jupiter for 70 years, it impacted in July 1994.

Until Shoemaker-Levy, tidal encounters were an under-appreciated feature of solar system dynamics. We are studying the physics of tidal encounters between comets–asteroids and the planets. We use a robust, few-body code to study encounters between the Sun, a planet, and asteroids (including double asteroids), and we are writing a code to study the breakup of an asteroid by the tidal field of a planet.

We believe that this work will explain a number of previously mysterious properties of asteroids whose orbits cross those of the inner planets. It will also determine the natural probability of capturing asteroids into orbits around Earth and the other inner planets by the tidal interplay between the Sun and the planet (and for Earth, that between the moon and Earth). We shall also determine the probability of asteroid capture by an exchange collision, if the asteroid is double. We believe that this mechanism allowed Mars to capture its small moons.

The permanent capture of asteroids around Earth would reduce the number of potentially dangerous Earth impactors, and it may allow us to utilize the resources of captured asteroids for construction in space.

This year we developed a spherical atmospheric model to find the probability of asteroids being captured into bound orbits around Earth in grazing encounters. We found that the critical corridor for capture is about half a scale height thick. We applied our results to the grazer that passed over New Mexico and Texas in October 1996 and then entered California 90 minutes later. We determined the probability that a small iron asteroid could hit Earth and survive in one piece after a grazing encounter. This work led to two refereed publications, a submitted paper, and three papers in meeting proceedings.

Publications

Hills, J.G., "Meteoroids Captured into Earth Orbit by Grazing Atmospheric Encounters: The New Mexico-Texas Grazer of October 3, 1996" (to be published in *Bull. Am. Astron. Soc.*).

Hills, J.G., and M.P. Goda, "Effects of Elevation on Ground Impact of Small Asteroids," *Bull. Am. Astron. Soc.* **29**, 961 (1997).

Hills, J.G., and M.P. Goda, "Meteoroids Captured into Earth Orbit by Grazing Atmospheric Encounters," *Planet. Space Sci.* **45**, 595 (1997).

Hills, J.G., and M.P. Goda, "The Largest Mass of Nickel-Iron Meteorites," *Meteorit. Planet. Sci.* **32** (4), A61 (1997).

Hills, J.G., and C.L. Mader, "Tsunami Produced by the Impacts of Small Asteroids," *Ann. N.Y. Acad. Sci.* **822**, 381 (1997).

Hills, J.G., et al., "Statistics of the Orbits of Large Bolides Detected by Surveillance Satellites" (submitted to *Astron. J.*).

Modeling Core-Collapse Supernovae in Three Dimensions

Michael S. Warren

Understanding core-collapse supernovae represents a fascinating challenge. The extreme density, temperature, and gravity at the heart of supernovae make them a natural laboratory to test ideas about physics that are otherwise difficult to investigate on Earth. We have established that the mechanism that powers supernova explosions relies on large-scale convection driven by neutrino heating. We will use the modeling results to estimate the kick velocity imparted to neutron stars at birth in these explosions.

This year we investigated the kick imparted to a neutron star when the accretion of matter is from one side and when the neutrino emission is similarly asymmetric. We modeled the mass captured by a high-velocity neutron star that collides with and passes through a companion binary star. When aimed correctly at the companion, the neutron star captures a significant mass (1%) of the companion star into a stable orbit of the neutron star, forming an accretion disk similar to our own proto-planetary system. The mass in this accretion disk is more than sufficient to later power the enigmatic soft gamma-ray repeaters and possibly later gamma-ray bursts.

We have also been modeling the early radiation flow and later accretion of matter onto the nascent neutron star. The very much larger opacity (10^5 times greater than the Compton opacity) of the iron-rich ejecta from the explosion greatly reduces the later expected accretion rate of matter by the neutron star.

High-Resolution Records of Global Climate Change

Steven Goldstein

One of the major challenges of paleoclimate research is to accurately age-date proxy records of global climate change that exist in terrestrial, ice-core, and deep-sea samples. Our objective is to apply accurate, high-resolution uranium-series chronometers for dating paleoenvironmental records stored in polar ice cores and deep-sea corals. Uranium-series dating of polar ice will contribute to resolution of the controversial chronology of Greenland ice cores, which show rapid and large climate fluctuations and may be analogues for present climate change. We will establish ventilation times for the deep ocean in the past by comparing uranium-series and carbon-14 ages for deep-sea corals.

This year we identified criteria required to obtain accurate and precise uranium-series dates for both modern and ancient deep-sea corals from the South Pacific Ocean. Data obtained for a specimen from the last glacial period represents the first precise and concordant protactinium-thorium ages for an ancient deep-sea coral. From this coral we obtained the first reliable ventilation age for the glacial oceans, ~390 years older than modern seawater at its location. This increase is consistent with other work suggesting diminished rates of mixing in the glacial Atlantic, and it indicates that deep-sea corals can be used to assess changes in oceanic mixing rates in the past. We also established the general validity of the uranium-thorium-protactinium dating method for corals. For dating polar ice, we have prepared radioactive tracers, doped synthetic ices, and evaluated improvements in chemical processing techniques for ice samples.

Publications

Edwards, R.L., et al., "Protactinium-231 Dating of Carbonates by Thermal Ionization Mass Spectrometry: Implications for Quaternary Climate Change," *Science* **276**, 782 (1997).

Goldstein, S.J., et al., "Protactinium-231 and Thorium-230 Dating of Deep Sea Corals," *EOS Trans. AGU, Fall Meeting Suppl.* **77**, 291 (1996).

Goldstein, S.J., et al., "Protactinium-231 and Thorium-230 Dating of Deep Sea Corals from the Last Glacial Period," *Proceedings of the Third Alfred O. Nier Symposium on Inorganic Mass Spectrometry*, 40 (1997).

High-Spectral-Resolution Infrared Absorption and Emission Signatures as Observed against Thermal Background Sources for Selected Molecular Species

Stephen Schmidt

The identification and measurement of concentrations of atmospheric molecular species have long been subjects of scientific and engineering research. Interest ranges from determining molecular species for the purpose of understanding basic global atmospheric processes to monitoring effluents from local pollution sources and industrial plants. One of the most successful measurement techniques is that of remotely obtaining spectral signatures characteristic of the subject molecules and then analyzing the spectra to identify particular species and, in many instances, to quantitatively determine concentrations.

The purpose of our project is twofold. First, we want to characterize in great detail and in high resolution, over the 8- to 14-μm wavelength region, the absorption/emission spectra of trace gases in time-varying plumes composed of other more abundant absorbing/emitting molecules. Second, we want to determine the alteration of these spectra by the absorption/emission and scattering characteristics of naturally present atmospheric constituents such as ozone and water vapor.

During the first year of this project, we have identified and used a line-by-line, high-resolution, spectral simulation code, FASCODE, to successfully compute sample laboratory spectra for individual molecules, an atmospheric transmission spectrum, the sky background spectrum in the vicinity of an active volcano, and preliminary spectra from a power-plant plume. These calculations included important molecular species from the high-resolution-transmission (HITRAN) molecular absorption database, thermal background sources, thermal downwelling, aerosols, and convolution with appropriate instrument functions. In a pseudoline database, we have installed the spectral-line parameters for several molecules not included in the HITRAN molecular database. We will use the pseudoline database in conjunction with HITRAN as source files for FASCODE.

Enhanced Analytical Performance of Laser-Induced Breakdown Spectroscopy

David Cremers

The main objective of this work is to enhance the analytical capabilities of laser-induced breakdown spectroscopy (LIBS). LIBS is a method of elemental analysis in which powerful laser pulses are focused on a sample to form a microplasma. Because of the high temperatures in the plasma, the sample is vaporized, and the resulting atoms are excited to emit light. By spectrally analyzing this light, we can determine the elemental composition of the sample. Determining the elemental composition is possible because each element has a unique spectral "fingerprint." LIBS is perhaps the most versatile elemental analysis method, applicable to a variety of different real-world analysis problems. Therefore, it is important to enhance the capabilities of the method as much as possible.

Methods of increasing analytical performance examined in this work include the following: (1) use of double pulses that form two closely spaced (in time) plasmas on the sample in order to increase the element signals, (2) study of the use of low laser-pulse energies for LIBS measurements because these lower energies are to be used in portable LIBS instruments, and (3) comparison of gated vs nongated detection of the plasma light.

During this past year, we have demonstrated signal enhancements of 5 to 30 times from soils and metals by using the double-pulse method, developed a model of the observed enhancement obtained by using double pulses, demonstrated that the analytical performance achievable using low laser-pulse energies (10 and 25 mJ) can match that achievable using an energy of 100 mJ, and demonstrated that time-gated detection is not necessary with LIBS.

Publications

Chinni, R.C., and D.A. Cremers, "Dependence of Analytical Results on Low Laser Pulse Energies (<100 mJ) and Timing Parameters in Laser-Induced Breakdown Spectroscopy" (submitted to *Appl. Spectrosc.*).

Pichahchy, A.E., et al., "Double-Pulse Laser-Induced Breakdown Spectroscopy for the Analysis of Solids" (submitted to *Appl. Spectrosc.*).

Balloon-Based, High-Time-Resolution Measurements of X-Ray Emissions from Lightning

David M. Suszcynsky

The objective of this project is to fly instrumented balloons through active thunderstorms to determine whether or not the runaway air-breakdown mechanism for lightning initiation is responsible for the generation of lightning discharges. This mechanism is currently the leading theory to account for unexplained x-ray flux enhancements associated with lightning discharges that have been recently measured from aircraft and balloons in thunderstorms. The results of our experiments could have important implications for understanding some of the outstanding fundamental issues associated with lightning initiation, above-cloud lightning events, and the global electric circuit and for the design of future space-based sensors to measure the electromagnetic pulse from an atmospheric nuclear detonation.

We began collaborating on balloon flights with Langmuir Laboratory/ New Mexico Institute of Mining and Technology. The laboratory provides technical expertise on lightning physics and ballooning issues and is responsible for launching the balloons.

Another accomplishment was designing and fabricating a prototype science package. The package weighs 20 lb and collects triggered, high-time-resolution, simultaneous measurements of electric-field and x-ray emissions associated with lightning strikes. Data are stored onboard and also downlinked to the ground via a radio-frequency modem.

In addition, we conducted three preliminary balloon flights to develop our ballooning skills and to test various cut-down devices that we designed and built this year. These devices will be used in next year's formal flights. Two of the three cut-down devices performed as designed, and the third provided valuable information concerning limitations of the design.

Fundamentals of Laser Ablation in the Analysis of Geological Materials

Aaron Koskelo

Laser ablation underlies many of the methods that hold promise for real-time materials recognition and rapid chemical analyses of environmental samples. Our goal is to determine the pertinent parameters that contribute to matrix effects in laser ablation of materials. We are placing particular emphasis on fundamental measurements of processes in the laser ablation of geological materials and soils.

This year we mapped the laser-induced breakdown spectroscopy (LIBS) spectra of Envirostone, an important material for waste remediation, using two ablation wavelengths, 1.06 μm and 266 nm. Light at 1.06 μm should create an ablation plume through a thermal mechanism, whereas light at 266 nm may cause ablation through a photoexcitation mechanism. However, we observed no clear difference in the LIBS spectra of the material. The lack of difference indicates that, at least for nanosecond pulses, the initial mechanism of plume formation has little effect on the resulting emission spectra in this material. We plan to investigate further the dynamics of energy flow in this material with our picosecond holography apparatus, which is nearing completion.

In addition, we continued to develop the use of correlations between analyte lines in LIBS spectra in order to examine inhomogeneity of soils and mine tailings. Using correlation matrices, we found that some Resource Conservation and Recovery Act metals correlate well with iron-containing components of the soil matrix but not with titanium components. Where an analyte resides in a material can determine how tightly it is bound and how difficult it is to remove by laser ablation. Donald Sparks of the University of Delaware has seen evidence for sequential adsorption of trace heavy metals on different sites in clays. We have begun a collaboration with him to determine whether the strength of binding on these sites results in a quantifiable change in the amount of analyte removed by laser ablation.

Publications

Koskelo, A.C., and T.K. Gamble, "Laser Ablation of Geological Materials" (1997 European Winter Conference on Plasma Spectrochemistry, Gent, Belgium, February 1997).

Regge Geometrodynamics in Support of Gravity-Wave Astronomy

Raymond Laflamme

The exciting feature of the emerging field of gravity-wave astronomy is that these waves will allow us to probe deeper into cosmic events than will any other known means. Our goal is to solve Einstein's gravitational field equations by employing the innovative and beautifully geometric approach known as Regge calculus. Regge calculus was developed at Princeton University early in the 1970s as a purely geometric approach to modeling a curved space-time geometry such as the gravity-wave emission from a two black-hole collision. We are exploiting the unique geometric properties of Regge calculus and applying this numerical technique to the analysis of astronomically relevant gravity-wave generation and its subsequent detection. Our research has direct relevance to the National Science Foundation's Laser Interferometric Gravity-Wave Observatory currently under construction.

Warner Miller and Ardian Gentle wrote and benchmarked the first (3+1)-dimensional Regge calculus code, with which we were able to accurately model the Kasner cosmological model. The code implements the first initial-data and parallel evolution in Regge calculus. In addition to the code development, Miller advanced the analytic foundations of Regge calculus by providing the first exact geometric derivation of the Regge-Hilbert action.

Using our newly developed code, we are currently setting up a simulation of a nonlinear gravitational wave. This will be benchmarked against a finite-difference code developed in part by Pablo Laguna of Pennsylvania State University. We expect to begin analyzing nonlinear gravitational wave propagation within the coming fiscal year.

Publications

Miller, W.A., "The Hilbert Action in Regge Calculus" (to be published in *Class. Quantum Grav. Lett.*).

Minerals of the Earth's Deep Interior

David Schiferl

Our goal is to gain understanding of a major geophysical issue that has lasted over two decades: the nature of the boundary between the earth's upper and lower mantle. Specifically, we want to understand the seismic velocity and density discontinuity at the boundary, which lies at a depth of 670 km and has a temperature of about 1900 K and a pressure of about 23 GPa. A phase change at this depth would represent a relatively small barrier to mantle convection through the discontinuity, but a compositional change would inhibit thermal convection throughout the mantle.

Thermoelastic equations of state of the mantle minerals provide important constraints on the chemical composition of the earth's mantle. We made measurements on two upper-mantle minerals with two different techniques, and introduced a new method of determining pressure and temperature in a large-volume pressure apparatus.

With a unique diamond-anvil cell designed for single-crystal x-ray diffraction, we determined the pressure and temperature derivatives of the bulk modulus and thermal expansion for orthoenstatite ($MgSiO_3$) at up to 1000 K and 1.5 GPa and at lower temperatures with pressures up to 4.5 GPa (see first figure). These results break previous records for single-crystal diffraction at high pressure and temperature and define a new state of the art.

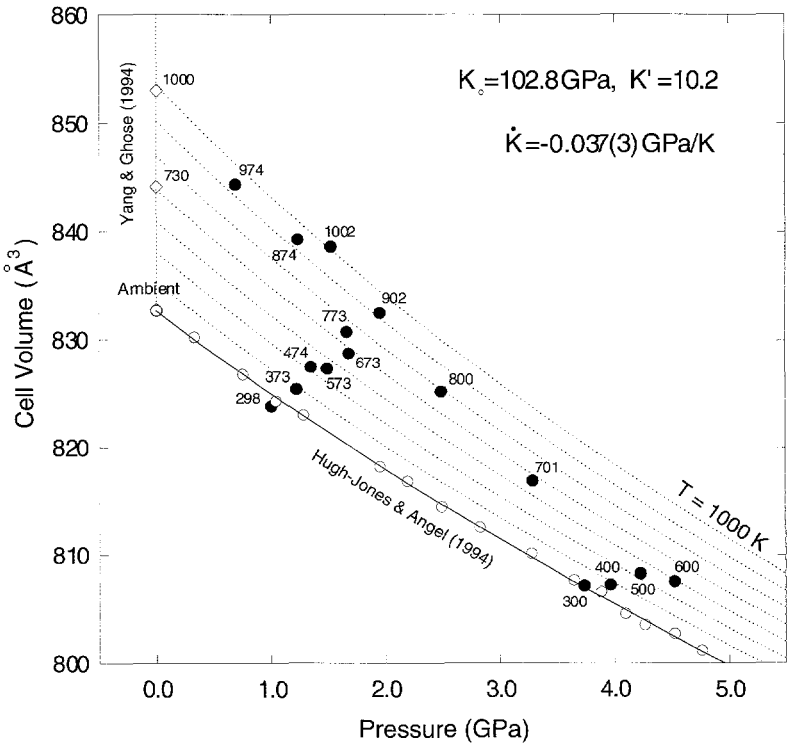
Pyroxenes, the second most abundant minerals after olivine, are of great importance in mineralogical models of the upper mantle. Jadeite ($NaAlSi_2O_6$) is a major component of the pyroxene minerals. We determined the temperature derivatives of the elastic moduli and the pressure derivative of thermal expansion of jadeite using a large-volume pressure apparatus with a synchrotron radiation source (see second figure).

Finally, we developed a new procedure to determine pressure and temperature in a large-volume pressure device. We conducted a synchrotron x-ray diffraction study on hexagonal boron-nitride (*h*BN) and NaCl at simultaneous high pressures (up to 9.0 GPa) and temperatures (300 to 1280 K). We applied a modified Rietveld profile refinement to these diffraction spectra of low symmetry and multiple phases observed in the energy-dispersive mode. Because the thermoelastic equations of state of *h*BN and NaCl are quite different, the unit cell volumes of these two materials, derived from the same diffraction pattern, can be used to derive the experimental pressure and temperature conditions. The large intersection angle of the isochoric lines of these phases in pressure and temperature space ensures a determination of pressure and temperature with satisfactory precision (see third figure).

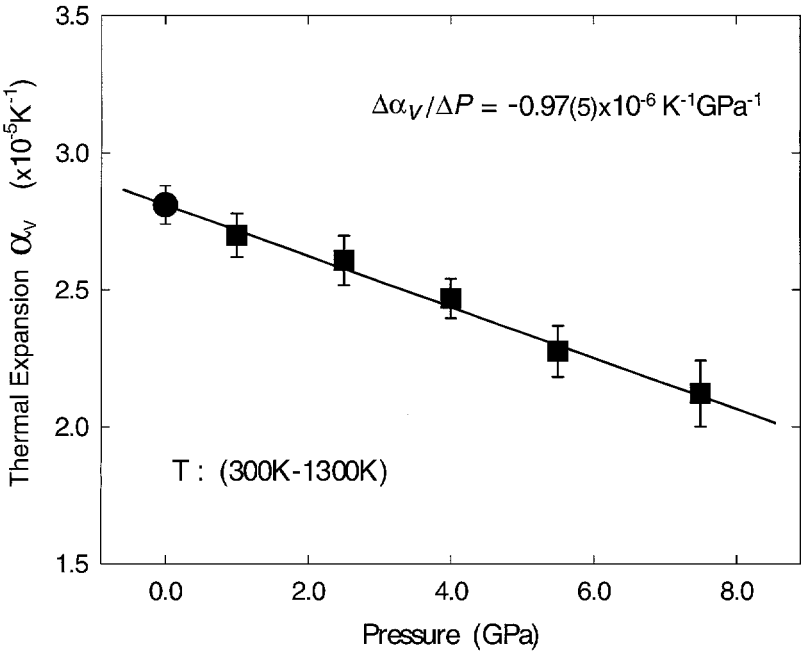
Publications

Zhao, Y., et al., "*P-V-T* Data of Hexagonal Boron-Nitride *h*BN and Determination of Pressure and Temperature Using Thermoelastic Equation of State of Multiple Phases," *High Pressure Res.* **15**, 1 (1997).

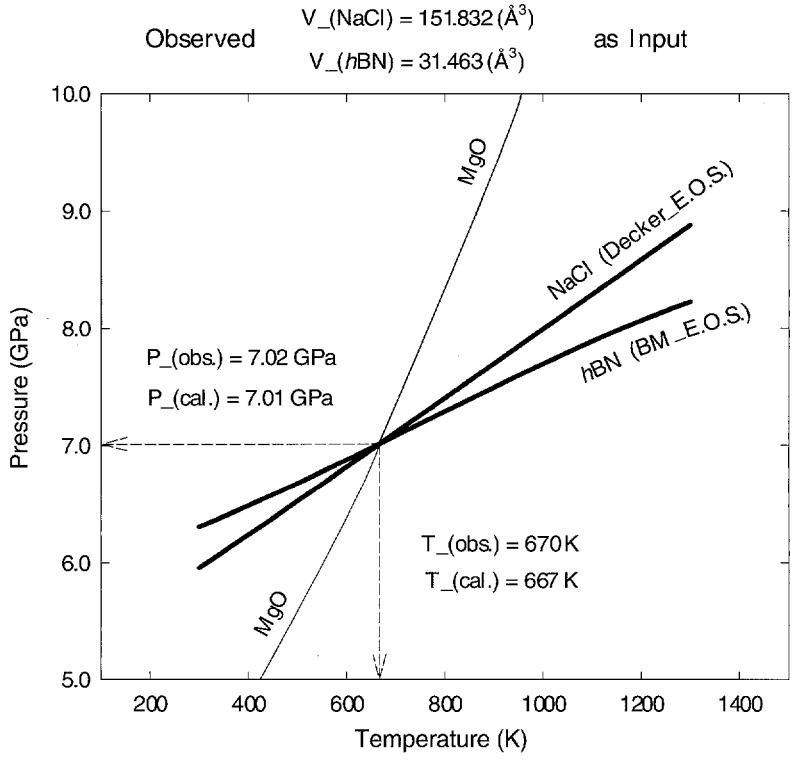
Zhao, Y., et al., "Thermoelastic Equation of State of Jadeite $NaAlSi_2O_6$: An Energy-Dispersive Rietveld Refinement Study of Low Symmetry and Multiple Phases Diffraction," *Geophys. Res. Lett.* **24**, 5 (1997).



The cell volume $V(P,T)$ of the orthoenstatite against pressure. The temperatures (K) are labeled next to the data points. The dotted lines represent the fitted isothermal compressions with temperature steps of 100 K, from 300 to 1000 K.



Thermal expansions of jadeite $NaAlSi_2O_6$ plotted as a function of pressure. The slope of the fitted straight line represents the pressure derivative of thermal expansion, $\Delta\alpha/\Delta P$. The solid circle is the regression result for thermal expansion at atmospheric pressure α_0.



Determination of pressure and temperature from the observed cell volume V_{obs}(NaCl) and V_{obs}(*h*BN) and their thermoelastic equations of state. The intersection point (P_{cal}, T_{cal}) of the isochoric lines of NaCl, *h*BN, and MgO closely matches experimental (P_{obs}, T_{obs}) conditions.

Deriving the Structure of Presupernovae and Delta Scuti Stars Using Nonradial Oscillations

Joyce A. Guzik

In this project, we modeled the evolution and pulsation of luminous blue variables (LBVs) and delta Scuti stars. The LBVs are massive stars and Type II supernova progenitors, whereas the delta Scuti variables are slightly more massive than the sun. Both types of stars pulsate in multiple radial and nonradial modes. We have improved the physics of our computer models and compared predicted and observed pulsation properties to try to better understand the internal structure of these stars. The study of stellar interiors can contribute to the understanding of fundamental physics, such as energy transport, turbulent mixing, and the equations of state and opacities of dense plasmas.

During the past year, we considered in detail the interaction between pulsation, radiation pressure, and convection for LBV models (see the first figure). We believe we have developed a consistent picture to explain the behavior of these very massive stars and the cause for their irregular "outbursts." When the luminosity-to-mass ratio of these stars is high enough, we find that pulsation causes the Eddington limit (the point at which radiation pressure pushing outward balances gravity pulling inward) to be exceeded periodically in the envelope, resulting in an outburst.

We also calculated stellar models that match, to first order, the pulsation frequencies of the delta Scuti stars FG Vir, CD-24° 7599, and the prototype delta Scuti. We explored the uniqueness of model fits, in particular the effects of uncertainties in composition, opacities, and mode identification on interpretation of the age, evolution state, and core structure of these stars (see the second figure).

Publications

Bradley, P.A., "Asteroseismological Constraints on the Structure of the ZZ Ceti Stars G117-B15A and R548" (to be published in *Astrophys. J.*).

Bradley, P.A., "Asteroseismology of DAV White Dwarf Stars" (to be published in *Baltic Astron.*).

Bradley, P.A., "Seismological Procedures for ZZ Ceti Stars and an Application to G117-B15A," in *A Half Century of Stellar Pulsation Interpretations* (Astronomical Society of the Pacific, Provo, UT, 1997).

Bradley, P.A., "Variable White Dwarf Data Tables" (to be published in *Baltic Astron.*).

Bradley, P.A., and J.A. Guzik, "Seismological Modeling of the Delta Scuti Star CD-24 7599," in *Sounding Solar and Stellar Interiors*, F. Schmider and J. Provost, Eds. (University of Nice, Nice, France, in press).

Cox, A.N., et al., "Linear Pulsations of Strange Modes in LBVs," in *Luminous Blue Variables: Massive Stars in Transition* (Astronomical Society of the Pacific, Provo, UT, 1997), Conf. Ser. 120, p. 133.

Cox, A.N., et al., "Pulsations and Outbursts of Luminous Blue Variables," in *A Half Century of Stellar Pulsation Interpretations* (Astronomical Society of the Pacific, Provo, UT, 1997).

Despain, K.M., et al., "Nonlinear Pulsation Modeling of Luminous Blue Variables," in *A Half Century of Stellar Pulsation Interpretations* (Astronomical Society of the Pacific, Provo, UT, 1997).

Guzik, J.A., "What Have We Learned about the Solar Interior from Solar Oscillations?" in *The Sun and Beyond,* Tran Thanh Van et al., Eds. (Editions Frontieres, Gif-sur-Yvette Cedex, France, 1996), p. 151.

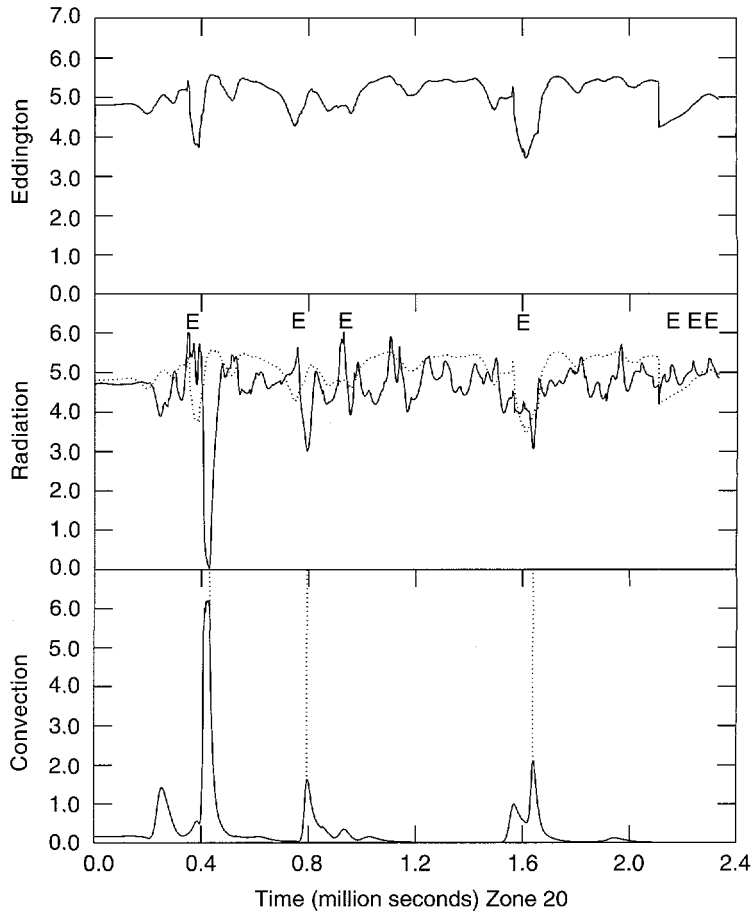
Guzik, J.A., and R.G. Deupree, "Two-Dimensional Solar Evolution Models: Evolution and Hydrodynamics," in *Sounding Solar and Stellar Interiors*, F. Schmider and J. Provost, Eds. (University of Nice, Nice, France, in press).

Guzik, J.A., and F.J. Swenson, "Seismological Comparisons of Solar Models with Element Diffusion Using the MHD, OPAL, and SIREFF Equations of State" (to be published in *Astrophys. J.*).

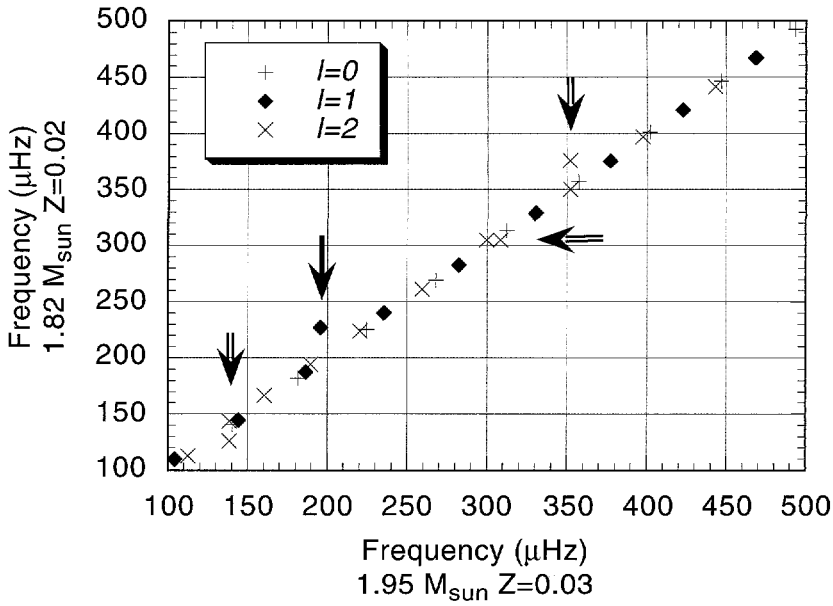
Guzik, J.A., et al., "A Nonlinear Study of Luminous Blue Variables and Possible Outbursts," in *Luminous Blue Variables: Massive Stars in Transition* (Astronomical Society of the Pacific, Provo, UT, 1997), Conf. Ser. 120, p. 138.

Guzik, J.A., et al., "Using Solar Oscillations to Constrain the Equation of State and Low-Temperature Opacities," in *The Sun and Beyond,* Tran Thanh Van et al., Eds. (Editions Frontieres, Gif-sur-Yvette Cedex, France, 1996), p. 163.

Templeton, M.R., et al., "A New Pulsation Spectrum and Asteroseismology of Delta Scuti," *Astron. J.* **114**, 1592 (1997).



Eddington, radiative, and convective luminosity (10^{39} erg/s) versus time of a zone in the envelope of a 47.28-solar mass, 16,980-K LBV model. The convective luminosity (bottom panel) is out of phase with the radiative luminosity (middle panel), allowing the radiative luminosity to exceed the Eddington limit (top panel and dotted line in middle panel) at points labeled "E."



Predicted frequencies for two candidate models of the delta Scuti star FG Vir, plotted against each other. The two models have a different mass fraction of elements heavier than hydrogen and helium and consequently have different interior structures and ages. However, these two models would appear identical to an observer, except for small differences in predicted frequencies of the nonradial (l = 1 and 2) modes due to the internal structure differences. The single-stem arrow indicates a fairly large shift between models for one l = 1 mode frequency; the double-stem arrows indicate an additional closely spaced l = 2 frequency present in one model that is not present in the other. Neither model matches all of the 21 observed frequencies of FG Vir.

Micro/Macroscale Coupling in Magnetospheric Plasmas

S. Peter Gary

Microscale plasma processes can significantly affect the large-scale dynamics of a plasma. Computer models of large-scale space plasmas cannot directly model kinetic processes and require algorithms to represent small-scale effects. We have recently developed the first scaling relation for the upper bound of the hot proton anisotropy in magnetospheric plasmas and have used it as an algorithm in a computer model of the magnetotail to demonstrate that it qualitatively affects the predictions of that model. We propose to use plasma theory, hybrid simulations, and spacecraft data analysis to develop and refine new scaling relations for the consequences of small-scale instabilities in the magnetosphere.

During the past year we have used both one- and two-dimensional hybrid simulations to examine the competition between the electromagnetic proton-cyclotron anisotropy instability and the mirror instability in collisionless plasmas with bi-Maxwellian proton velocity distributions. We found that the former growing mode dominates the fluctuations and wave-particle scattering if the proton parallel beta is less than about five, but that the two instabilities both contribute to scattering at larger values of beta.

We confirmed that the upper bound for proton temperature anisotropy that we derived earlier remains a valid constraint, and we obtained a new upper bound for magnetic fluctuation energy densities from these instabilities as a function of the proton parallel beta. We also showed that the temperature anisotropy upper bound is an appropriate constraint for proton anisotropies observed in the terrestrial magnetosheath.

Publications

Gary, S.P., et al., "Proton Temperature Anisotropy Upper Bound," *J. Geophys. Res.* **102** (1997).

Nuclear and Particle Physics

Helium-3 Magnetometry for a Neutron EDM Measurement

Martin Cooper

The ideas for a new experiment to improve the sensitivity of measurement of the neutron's electric dipole moment (EDM) by three orders of magnitude (to around 10^{-28} e·cm) have been worked out in the literature. This project investigates several areas related to the helium-3 magnetometry required to establish the technical feasibility of the experiment: the density distribution, the diffusion time, and the polarization lifetime of helium-3, which can be obtained in a low-temperature bath of helium-4.

The project has three phases: (1) development of a tomographic method for determining the position of the helium-3; (2) measurement of the helium-3 density distribution and diffusion time in a 0.5-K bath of helium-4; and (3) measurement of the polarization lifetime of the helium-3 in the bath. This summary reports progress made in the project's first phase.

We used a cold-neutron beam to establish that tomography is a viable method for determining the distribution of a dilute mixture of helium-3 in helium-4. The tomographic method involved first scanning the neutron beam across a gas cell filled with 2.5 atm of helium-4 mixed with 7% xenon and 1% helium-3 and then observing the scintillation light produced in the helium-4 by the reaction products from n(^{3}He,T)p. The cross section of the cell differed in a known way from a perfect cylinder, and we clearly observed the deviation, which we can calculate. The results indicate that deviations from a uniform distribution will be detectable for concentrations of helium-3 as low as 10^{-8} in liquid helium-4, near the range of interest to the full experiment. Additionally, we determined that each of our photomultiplier tubes saw a mean of 1.5 photoelectrons. This value highlighted the importance of the optics design of the cryogenics experiments for project phases 2 and 3. We also began establishing a cryogenics laboratory for carrying out the latter phases of the project.

Publications

Bangert, P.D., et al., "Enhancement of Superthermal-Ultracold Neutron Production by Trapping Cold Neutrons" (to be published in *Nucl. Instrum. Methods Phys. Res. A*).

Neutrino Physics at Fermilab

Geoffrey Mills

The Liquid Scintillator Neutrino Detector (LSND), which is currently located at the Los Alamos Neutron Science Center, has found evidence for the violation of lepton number and the oscillation of different neutrino species among themselves. The measurement of neutrino-oscillation parameters is one of the paramount problems in nuclear particle physics today. The goal of this project is to research the technology necessary to precisely measure the oscillation parameters of the LSND effect.

Understanding how to produce intense, pure beams of neutrinos is one of two major objectives of this work. This first objective has required us to study neutrino production in a focused secondary-particle beam. We have developed a simulation of the neutrino-production system for this purpose. As a starting point we have used the parameters of the 8-GeV booster accelerator at Fermi National Accelerator Laboratory (Fermilab), which can provide 5×10^{12} 8-GeV protons at a repetition rate of 15 Hz. Designing the focusing system of the secondary pion beam (a beam that decays into neutrinos) has been our primary thrust. We are optimizing the configuration of a pulsed magnetic horn system to focus the beam.

The second major objective of this project is to detect higher-energy neutrinos and reject serious backgrounds such as π^0 decays. We are extrapolating our experience gained at LSND to a larger system and believe we can achieve the required detection efficiencies and background rejection. We have uncovered critical issues such as the precise time distribution of light from relativistic particles in pure mineral-oil fluid. We will address this issue in the near future by a series of measurements at the cyclotron at Texas A&M University.

Instantons and Duality in Strongly Coupled Quantum Theories

Michael Mattis

A long-standing goal of quantum field theory has been to understand analytically (as opposed to on the lattice) the important properties of strongly coupled gauge theories. Because of the celebrated recent work of Seiberg and Witten, we have gained this understanding, albeit only in a highly restricted class of models. The strong-coupling properties include the mechanism for confinement, as well as the spectrum of stable states. These theoretical advances rely on the interplay of two important assumptions: (1) supersymmetry, which relates fermions and bosons as components of a common underlying "superfield," and (2) duality, which exchanges the strong- and weak-coupling regimes of the theory, and at the same time, swaps the roles of the electric and magnetic fields.

We propose to access the same physics in a complementary (and completely novel) way by using a direct, dynamical, nonperturbative calculation of the instanton series. Instantons are the Euclidean-space "particles" associated with tunneling between distinct ground states in the theory. In a significant preliminary result, we have recently calculated all 1-instanton and 2-instanton effects in the Seiberg-Witten model, independently confirming the relevant parts of their solution. We regard these calculations as an "existence proof" for the feasibility of our work, the principal goals of which are (1) extension to n-instanton effects for all $n > 2$, and (2) extension of instanton methods to other strongly coupled quantum-field theories that more closely resemble quantum chromodynamics (QCD). This second goal is particularly important, as instantons generically exist even in theories (for example, QCD itself) that admit neither duality nor supersymmetry.

Using these instanton techniques, we hope ultimately to develop a compelling physical picture of the vacuum in strongly coupled gauge theories. Most importantly, we hope to shed light on the long-unsolved problem of confinement in QCD.

Publications

Dorey, N., et al., "Instantons, Higher-Derivative Terms, and Nonrenormalization Theorems in Supersymmetric Gauge Theories" (to be published in *Phys. Lett. B*).

Dorey, N., et al., "Instantons, Three-Dimensional Gauge Theory, and the Atiyah Hitchin Manifold" (to be published in *Nucl. Phys. B*).

Dorey, N., et al., "On Mass-Deformed N = 4 Supersymmetric Yang-Mills Theory," *Phys. Lett. B* **396**, 141 (1997).

Dorey, N., et al., "Multi-Instantons, Three Dimensional Gauge Theory, and the Gauss-Bonnet-Chern Theorem" (to be published in *Phys. Lett. B*).

Dorey, N., et al., "N = 2 Supersymmetric QCD with 4 Flavors," *Nucl. Phys. B* **492**, 607 (1997).

Dorey, N., et al., "Supersymmetry and the Multi-Instanton Measure II from N = 4 to N = 0" (to be published in *Nucl. Phys. B*).

A GaAs Detector for Dark Matter and Solar Neutrino Research

Thomas J. Bowles

A GaAs detector can offer substantial advantages over other technologies in searches for weakly interacting massive particle (WIMP) dark matter and in high-resolution measurements of the solar neutrino spectrum. GaAs may provide backgrounds lower than those possible in other detectors, along with a low-energy threshold and very good energy resolution at room temperature. The issues that our work can address are at the forefront of modern physics.

In the short term, our efforts will be directed at searching for the cold dark matter that apparently makes up 90% of the mass in the universe. In the long term, it may prove feasible to address the question of neutrino mass using a large GaAs solar neutrino detector. GaAs detectors, if proven feasible, are also very likely to have applications in many other fields, including medicine, satellite-based detectors, and environmental monitoring.

Under special conditions, we produced 1-kg GaAs crystals that have electronic properties close to the theoretical maximum. We now have in place techniques for measuring all of the required characteristics of the crystals to determine their performance. We also initiated efforts to fabricate detectors from these crystals.

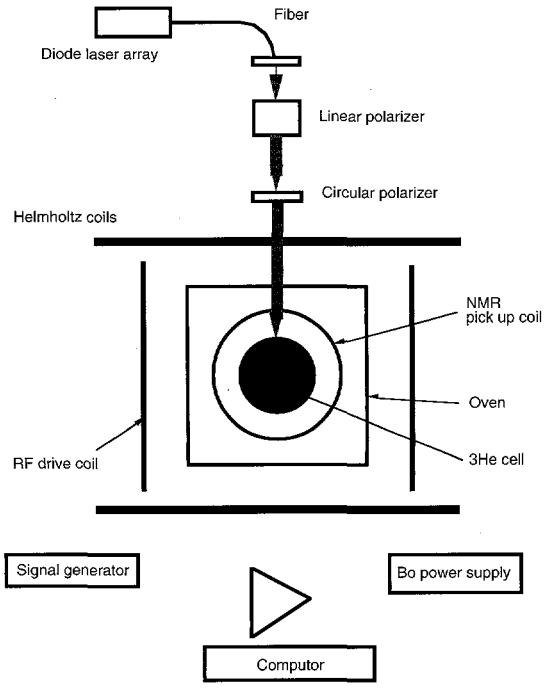
In addition, we are pursuing the possible use of an existing large array of SiLi detectors from Russia that would allow us to carry out a sensitive search for low-mass WIMPs at the same time as we are developing the counting and background-suppression techniques that will be required for a large array of GaAs crystals.

Time-Reversal Tests in Polarized Neutron Reactions

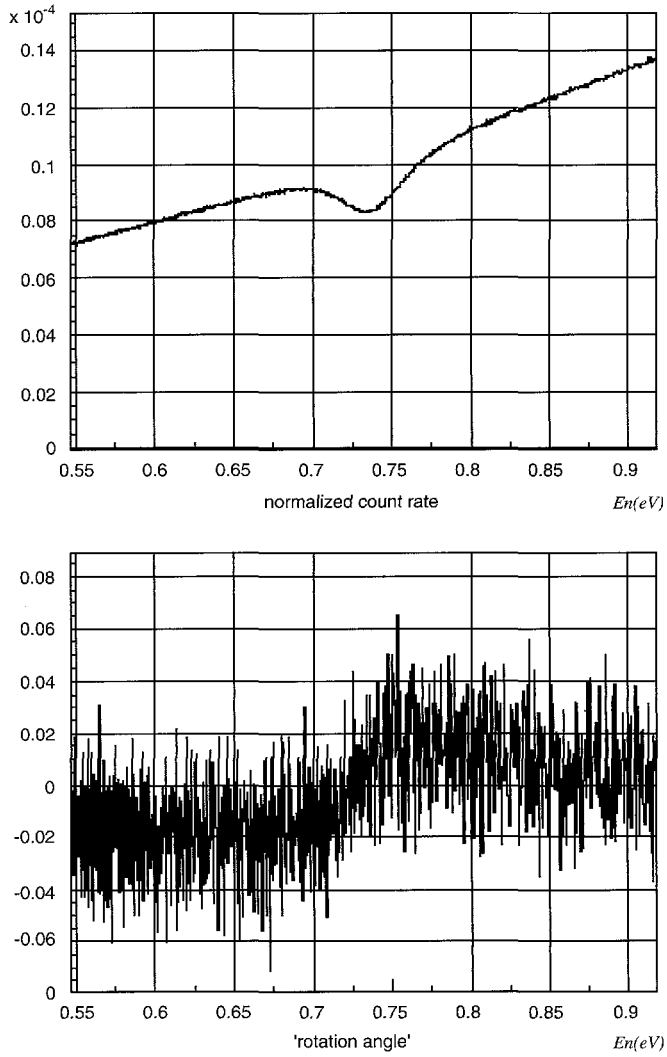
J. David Bowman

In recent years researchers have studied the nuclear weak interaction in the compound nucleus in light of parity violation. Nuclear structure strongly enhances the observed effects of parity violation. The predictions are that the interaction of polarized neutrons with polarized nuclear targets also could be used in sensitive tests of time-reversal violation because of the nuclear enhancements.

We have designed experiments to search for time-reversal violation in neutron/nucleus interactions. We have also developed techniques to polarize neutrons with laser-polarized helium-3 gas targets. This year we performed two experiments at the Manuel Lujan Jr. Neutron Scattering Center using the polarized helium-3 neutron spin filter (see the first figure): an absolute neutron beam polarization measurement with an accuracy of 0.2%–0.3% and a neutron spin-rotation measurement on a lanthanum-139 sample (see the second figure). Demonstration of this accuracy leads to the possibility of several important measurements of fundamental symmetries in nuclear physics. Tests of time-reversal invariance in neutron scattering and the measurement of angular correlations in neutron beta decay are limited by the accuracy of the knowledge of neutron polarization. This work has involved collaboration with groups from Kyoto University and the National Laboratory for High-Energy Physics in Japan.



Schematic of the helium-3 spin filter. Shown are the laser, optics, holding-field coils, radio-frequency coils for nuclear magnetic resonance (NMR) measurement of helium-3 polarization, oven, pickup coils for NMR, and signal-processing electronics.



(Upper) Time-of-flight spectrum of the 0.734-eV *p*-wave resonance of lanthanum-139. (Lower) Dispersion curve of neutron spin rotation from the on-line data of the experiment.

Publications

Bowman, J.D., and Y. Masuda, "Time Reversal Tests in Neutron-Spin Optics" (to be published in *Phys. Rev. C*).

Penttila, S.I., et al., "An Absolute Neutron Beam Polarization Measurement with High Accuracy Using Optically Polarized ^{3}He" (to be published in *Nucl. Instrum. Methods*).

The QCD Phase Transition in Relativistic Heavy-Ion Collisions

Frederick Cooper

We have developed practical numerical methods to describe and study the nonequilibrium phase transition of nuclear matter following ultrarelativistic heavy-ion collisions based on quantum chromodynamics (QCD). Our approach, which relies on a systematic $1/N$ expansion, will allow us to make specific predictions for thermalization time scales, energy-momentum flow, lepton pair production, and correlations in the nonequilibrium evolution of the quark-gluon plasma produced in the central rapidity region of the collision event.

We have made progress this year in understanding several aspects of the nonequilibrium phase transition. We have studied the linear s model, which is the effective-field theory of the chiral phase transition, and we have performed numerical simulations of the chiral phase transition for both a longitudinal as well as a radial expansion into the vacuum. Since one of the thrusts of the PHENIX detector to be installed at the Relativistic Heavy Ion Collider at Brookhaven National Laboratory (BNL) is the detection of dileptons, we have developed the formalism for calculating dileptons starting from Schwinger's closed-time-path formalism. We presented our predictions for the dilepton spectrum at the Japanese Institute of Physical and Chemical Research (RIKEN)-BNL conference on September 23.

We have also made progress in developing a transport theory for the production phase of the quark-gluon plasma. By careful analysis of an exactly solvable model, we have found the correct single-particle distribution function that allows one to determine the correct form of the particle production rate in a transport approach to the time evolution of the plasma. We also studied the scattering corrections to the mean field approximation in a simple quantum mechanics problem by simulating the integro-differential equations obtained from the $1/N$ expansion.

Publications

Cooper, F., "From Landau's Hydrodynamical Model to Field Theory Models of Multiparticle Production," available from the LANL E-Print archive at *http://xxx.lanl.gov* as document hep-ph 9701203.

Cooper, F., et al., "Anomalous Transverse Distribution of Pions as a Signal for the Production of DCCs," *Phys. Rev. C* **54**, 3298 (1996).

Cooper, F., et al., "Non-Equilibrium Dynamics of Symmetry Breaking in $\lambda\,\phi^4$ Field Theory," *Phys. Rev. D* **55**, 6471 (1997).

Cooper, F., et al., "Order 1/N Corrections to the Time Dependent Hartree Approximation for a System of N+1 Oscillators," *Phys. Rev. D* **56**, 5400 (1997).

Cooper, F., et al., "Time Evolution of the Chiral Phase Transition During a Spherical Expansion," *Phys. Rev. D* **54**, 2213 (1996).

Kluger, Y., et al., "Dileptons from Disoriented Chiral Condensates" (to be published in *Phys. Rev. C*).

Search for Cosmic Antimatter with Milagrito

Cyrus Hoffman

An important question in cosmology is the following: Why is the universe apparently composed only of matter? The limits on the existence of antimatter beyond our galaxy are quite poor. Antiprotons are expected to be produced by the interaction of high-energy cosmic rays with material in our galaxy. This interaction should result in approximately 0.001 antiprotons per proton in the cosmic-ray flux at earth at energies from 1 to 100 GeV. The detection of a higher cosmic antiproton flux could provide evidence for the existence of antimatter galaxies.

As part of this project, we are searching for energetic cosmic antiprotons by studying their deflection in the earth's magnetic field and using the shadow of the moon as observed with cosmic rays. The shadow due to 1-TeV protons should be deflected approximately 30 mr to the west; antiprotons would produce a shadow displaced to the east. The project will use data from the Milagrito air-shower detector.

Milagrito was installed in the fall of 1996. We have already completed the data acquisition code and started data taking in February 1997. Work for the remainder of the past year has concentrated on calibrating the timing and pulse-height response of the photomultiplier tubes and on writing and testing reconstruction algorithms. We will start looking at data to optimize the angular resolution.

Publications

Berley, D., et al., "First Light in the Milagrito Detector," in *Proceedings —XXV International Cosmic Ray Conference,* M.S. Potgieter, et al., Eds. (Potchefstroom University, Potchefstroom, South Africa, 1997), Vol. 5, p. 201.

Solar-Neutrino Physics

Andrew Hime

To date, four independent experiments have measured a solar-neutrino flux that falls substantially below the predictions of the standard solar model (SSM). Taken together, these results provide a tantalizing hint that a resolution of this so-called "solar-neutrino problem" resides in new physics such as neutrino oscillations.

Our project involves a collaboration with the Sudbury Neutrino Observatory (SNO), a 1000-tonne, heavy-water, Cerenkov detector presently under construction in Sudbury, Canada. SNO will be capable of measuring independently both the charged-current (CC) interactions of electron neutrinos and the neutral-current (NC) interaction of all active neutrino species, thus offering a model-independent resolution of the solar-neutrino problem. Our efforts focus on the design and construction of an ultralow-background array of discrete neutral-current detectors (NCDs) and the methods for extracting the NC signal.

This past year saw the complete installation and shakedown of the gas-handling system and electronics used to fill and test detectors during the production phase. In addition, we have refined a detailed set of gas-handling and detector-diagnostic procedures to ensure the reliable performance of the counters before we ship them to Sudbury.

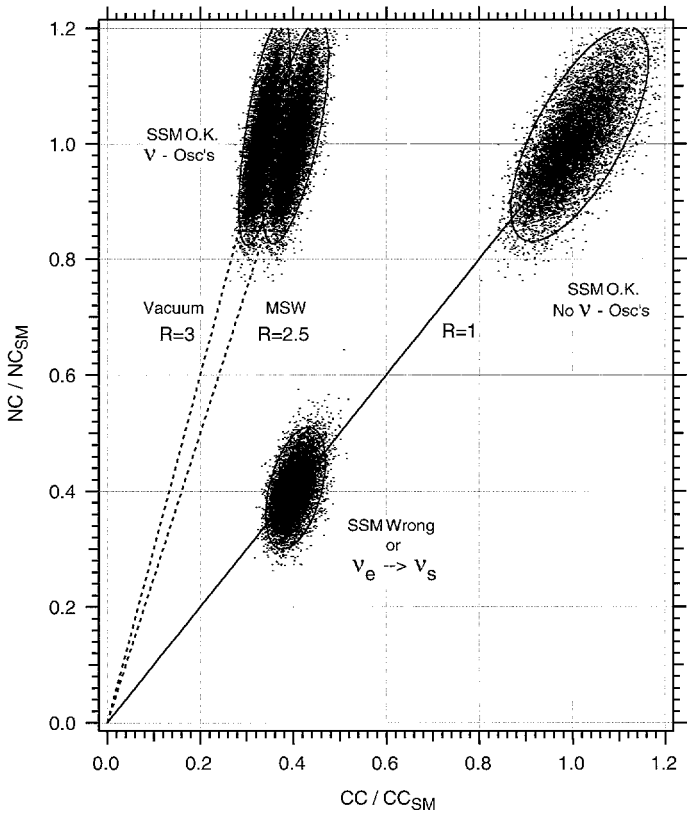
Extracting the NC signal from the NCD array in SNO relies on a robust means for extracting the neutron-capture signal in the presence of backgrounds. We have developed a novel technique for extracting the signal on an event-by-event basis using digitized waveform analysis; we have developed this technique in our local test bench as well as at our low-background counting facility at the Waste Isolation Pilot Plant. The results of this R&D, together with numerous radioassay measurements of detector components, indicate that the NCD array can meet the stringent radiopurity constraints imposed by SNO and that the NC signal can be extracted in a robust and accurate fashion.

We have also led the NCD collaboration in defining the overall program for extracting physics from the NCD array. We have developed algorithms to extract the signal in the presence of the all-important photodisintegration background, and we have developed techniques to use the discrete NCD geometry for separating the solar-neutrino signal from these backgrounds. The result of this analysis is depicted in the accompanying figure, which shows that SNO is capable of differentiating the various possible solutions to the solar-neutrino problem.

Publications

Hime, A., "Exorcizing Ghosts—In Pursuit of the Missing Solar Neutrinos," in *Los Alamos Science* (Los Alamos National Laboratory, Los Alamos, NM, 1997).

Hime, A., "Neutral-Current Detectors for the Sudbury Neutrino Observatory," in *Proceedings of the 4th International Solar Neutrino Conference* (World Scientific, Singapore, in press).



Simulation of NC and CC signal sensitivity for one year of data from SNO operating with the NCD array deployed. The NC-to-CC ratio *R* is defined by the slope in the NC-CC plane and is independent of SSM predictions. Three distinct regions are displayed based on different solutions to the solar-neutrino problem. These regions are clearly separable in SNO at the 95% confidence level. The ratio changes for the different neutrino-oscillation scenarios, but we can differentiate between the two scenarios from a detailed analysis of the shape of the CC spectrum.

Neutrinos and Theory of Weak Interactions

Joseph Carlson

The objective of this project is to perform theoretical research relevant to Laboratory studies of weak interaction physics. These studies include the Physics (P) Division work in neutrino physics, rare decays, fundamental symmetry tests, and nuclear and nucleon weak interactions as well as the efforts of Laboratory experimenters to interpret data from their consultations at Fermi National Accelerator Laboratory. Accomplishments during the past year include the following:

- We found that only weak-isospin doublet leptoquarks contribute to the T-violating D-coefficient in beta decay and then only if two such leptoquarks mix. We found that the new experimental limit on muonium to antimuonium conversion from the Paul Scherrer Institute implies an upper limit on the $\mu^+ \rightarrow e^+ + \nu_e + \nu_x$ branching ratios that is an order of magnitude below the oscillation probability obtained in the Liquid Scintillator Neutrino Detector (LSND) experiment here at Los Alamos.
- We calculated neutrino-carbon elastic and inelastic cross sections and related processes to check on a California Institute of Technology claim that the measured LSND neutrino-carbon cross section must be erroneous. Our results support the possibility that the LSND measurement is correct, within statistical errors. We exposed a fallacy in the literature regarding the nature and detection of neutrino (and other) oscillations.
- We showed that Λ–Σ coupling leading to an s-wave π^+nn three-body decay can provide a satisfactory explanation of the branching ratio and the resulting π^+ decay spectrum in the decay of L hypernuclei.
- For an experiment at the Los Alamos Neutron Science Center (LANSCE) in which a proton + neutron goes to a deuteron + photon, we showed that the spatial resolution of a typical detector should be adequate to prevent significant contamination of the parity-nonconserving asymmetry by the parity-conserving asymmetry, unless the former should prove to be an order of magnitude smaller than expected. Our theoretical estimate of the parity-conserving asymmetry appears to be sufficiently model-independent that this assymetry observable can serve as a valid polarization monitor.
- We worked with experimentalists in P Division to develop medium-energy hypernuclear experiments to run on the Alternating Gradient Synchrotron at Brookhaven National Laboratory. We also worked with P Division members on the neutron + proton bremsstrahlung effort and on the few-body parity-nonconservation scattering and reaction experiments at LANSCE.

Publications

Ahluwalia, D.V., and T. Goldman, "Interplay of Non-Relativistic and Relativistic Effects in Neutrino Oscillations," *Phys. Rev. D* **56**, 1698 (1997).

Csoto, A., et al., "Parity Conserving Gamma Asymmetry in Neutron-Proton Radiative Capture," *Phys. Rev. C* **56**, 631 (1997).

Goldman, T., et al., "MSW Without Matter," *DPF'96 Proceedings* (World Scientific, Singapore, in press); also available from the LANL E-Print archive at *http://xxx.lanl.gov* as document hep-ph/9608287.

Herczeg, P., "Aspects of CP-Violation" (Workshop on the State of Physics at the End of the 20th Century—P. Carruthers 61st Birthday, Santa Fe, NM, October 26–29, 1996).

Herczeg, P., "Muon Decays in Extensions of the Standard Model," in *Proceedings of the Sixth Conference on the Intersections of Particle and Nuclear Physics*, T. William Donnelly, Ed. (American Institute of Physics, New York, in press).

Herczeg, P., "The Nature of Neutrinos in Muon Decay and Physics Beyond the Standard Model," in *Los Alamos Science,* N. Cooper, Ed. (Los Alamos National Laboratory, Los Alamos, NM, 1997).

Herczeg, P., "Neutron Electric Dipole Moment Measurements and Physics Beyond the Standard Model" (Workshop on a New Experiment for Measuring the Neutron's Electric Dipole Moment, Los Alamos, NM, 1997).

Herczeg, P., "Time Reversal Violation in Nuclear Processes" (Workshop on Physics Related to Nuclear Orientation at TRIUMF-ISAC, Vancouver, Canada, April 25, 1997).

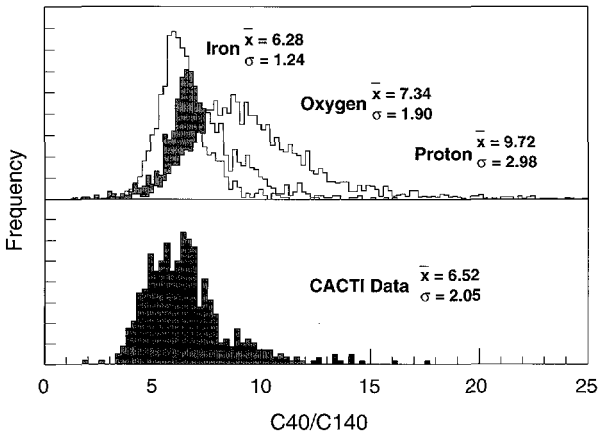
Herczeg, P., and I.B. Khriplovich, "Time-Reversal Violation in Beta Decay in the Standard Model," *Phys. Rev. D* **56**, 80 (1997).

Lowe, J., et al., "No Lambda Oscillations," *Phys. Lett. B* **384**, 288 (1996).

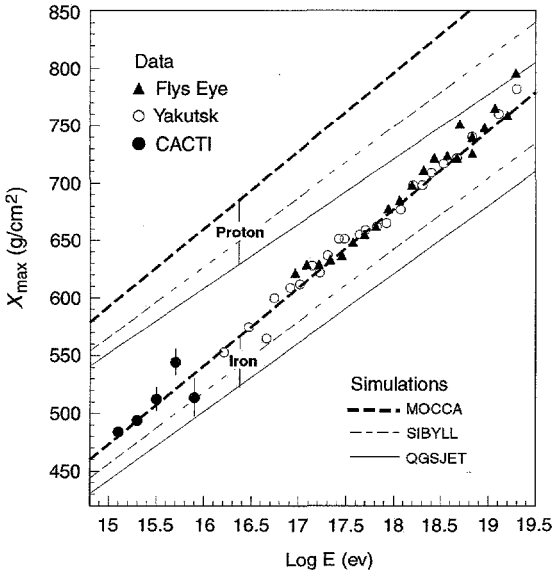
Determination and Study of Cosmic-Ray Composition above 100 TeV

Constantine Sinnis

Cosmic rays were first observed over 100 years ago, yet we still do not know their origin. The nuclear composition of cosmic rays is one of the best-known methods of constraining the origin of cosmic rays. The goal of this project was to determine cosmic-ray composition at energies around 100 TeV. At this energy regime, the cosmic-ray spectrum steepens. This feature, known as the "knee," may be caused by a change in the acceleration mechanism or a change in the galactic confinement of cosmic rays. A measurement of the composition around this feature would help answer this question.



The lower graph shows the measured distribution (over all of the data collected) of the intensity of light 40 m and 140 m from the EAS axis. This quantity is related to the height-of-shower maximum (X_{max}). The upper graph shows the expected distribution for iron, oxygen, and proton cosmic rays. Comparison of the upper and lower graphs shows that although the average composition is consistent with an iron composition, the width of the measured distribution indicates that there is an admixture of lighter nuclei in the cosmic rays.



The average X_{max} as a function of energy as measured by this experiment. Also shown are the results of several simulations and experiments performed at higher energies. The data indicate that the average composition is heavy (ironlike) and does not change over the energy range of the experiment.

The cosmic-ray composition has been directly measured up to about 50 TeV, but above this energy the flux is too low for direct techniques. We have developed a new technique to determine the primary cosmic-ray composition. When a high-energy cosmic ray enters the earth's atmosphere, it dissipates energy via particle creation —an extensive air shower (EAS) develops. An EAS is a swarm of particles that form a rough pancake. The number of particles in the pancake grows until the average energy per particle is about 80 MeV. The altitude at which this occurs is known as the shower maximum (X_{max}). The technique we developed determines X_{max} by measuring the lateral distribution of the Cerenkov light emitted by the EASs. The experiment, known as CACTI, collected data for 6 months.

This year we analyzed the data that we had taken during the previous two years. Most of our analysis centered on understanding and removing systematic errors and on understanding the absolute response of the instrument. Our results, depicted in the accompanying figures, indicate a mixed composition (predominantly iron) with no evidence of a change across the knee. This result argues against an extragalactic source of cosmic rays. We presented our results at the International Cosmic Ray Conference in Durban, South Africa. In addition, a student is completing a thesis on this work, and we are preparing a paper that we will submit to a refereed journal.

Publications

Paling, S., et al., "Results from the CACTI Experiment: Air-Cerenkov and Particle Measurements of TeV Air Showers at Los Alamos," in *Proceedings XXV International Cosmic Ray Conference*, M.S. Potgieter et al., Eds. (Potchefstroom University, Potchefstroom, South Africa, 1997), Vol. 5, p. 253.

Testing the Standard Model Using Bottom Quarks

Rajan Gupta

Both experimental and theoretical particle physicists are extensively probing bound states involving heavy quarks and are generating a wealth of new data. Their results may contain the first clues to a possible breakdown of the standard model theory at very short distances because these systems provide the cleanest ways of fixing some of the unknown parameters of the standard model. However, researchers need theoretical input to interpret these data. This need motivates our project, which involves a comprehensive study of standard model phenomenology at the hadronic scale.

This year we made a detailed analysis of the calculation of light quark masses using lattice quantum chromodynamics (QCD). Results for the average of up and down quark masses are $(m_u + m_d)/2 = 2.7 \pm 0.6$ MeV and for the strange quark, $m_s = 68 \pm 19$ MeV. These results are roughly half of the previously accepted values. The new results significantly affect the size of charge-conjugation and parity (CP) violation in kaon decays.

We reanalyzed previous extractions of quark masses using QCD sum rules, and we showed four potential sources of errors in previous analyses and estimated the size of corrections. Putting all the factors together, we showed that estimates of light quark masses could easily be smaller by a factor of 2.

In addition, we calculated the matrix elements of 4-fermion operators that arise in the weak decays of kaons and D and B mesons. We presented final results for the kaon B parameters that contribute to both direct and indirect CP violation in kaon decays and for operators that arise in the calculations of B meson lifetimes.

Publications

Bhattacharya, T., et al., "The Extraction of Light Quark Masses from Sum Rule Analyses of Axial and Vector Current Ward Identities" (submitted to *Phys. Rev. D*).

Gupta, R., and T. Bhattacharya, "Light Quark Masses from Lattice QCD," *Phys. Rev. D* **55**, 7203 (1997).

Gupta, R., et al., "Matrix Elements of 4-Fermion Operators and B-Parameters with Wilson Fermions," *Phys. Rev. D* **56**, 4036 (1997).

Kilcup, G., et al., "Staggered Fermion Matrix Elements Using Smeared Operators" (to be published in *Phys. Rev. D*).

A Continuous Watch of the Northern Sky above 40 TeV with the CYGNUS Array

Cyrus Hoffman

As part of this research, we are searching for very high energy transient emissions from astrophysical objects in the northern sky. Potential objects of interest include gamma-ray bursts, active galaxies and quasars, supernovae, variable and flare stars, and comets and asteroids, all of which are some of the most exciting objects in the cosmos.

Similar studies of high-energy transient emissions made to date have shown very exciting results. For example, a chance observation with an air Cerenkov telescope detected a large flare in a very high energy emission from Markarian 501, an active galaxy. A worldwide campaign this summer found that this object emits very high energy gamma rays that vary in intensity by nearly an order of magnitude over time scales of hours. This finding is remarkable considering that the central engine providing the power for the emission is believed to be a supermassive (about a billion-solar-mass) black hole. The study of transient astrophysical events is difficult because it requires that the telescopes be dedicated to that sole purpose. The exciting observations of Markarian 501 are a fortuitous accident; most of the sky is not monitored for transient phenomena.

This past year, we collected data with the Milagrito detector, which is a prototype water Cerenkov detector located at Fenton Hill. We are honing reconstruction algorithms and will shortly analyze the data for transient emissions. In particular, we will monitor the activity from Markarian 501, especially during the periods in which the air Cerenkov telescopes were unable to observe this object.

Publications

D. Berley, et al., "Results from the Milagrissimo Air Shower Detector," in *Proceedings XXV International Cosmic Ray Conference*, M.S. Potgieter et al., Eds. (Potchefstroom University, Potchefstroom, South Africa, 1997), Vol. 3, p. 285.

A Search for Superradiant Emission States in Nuclear Isomer Crystals

Robert Rundberg

Our objective was to verify the stimulated emission of gamma rays from ^{125m}Te, as claimed by Russian scientists. The reported cross section for stimulated emission was sufficiently large to allow gain in a single-pass gamma-ray laser.

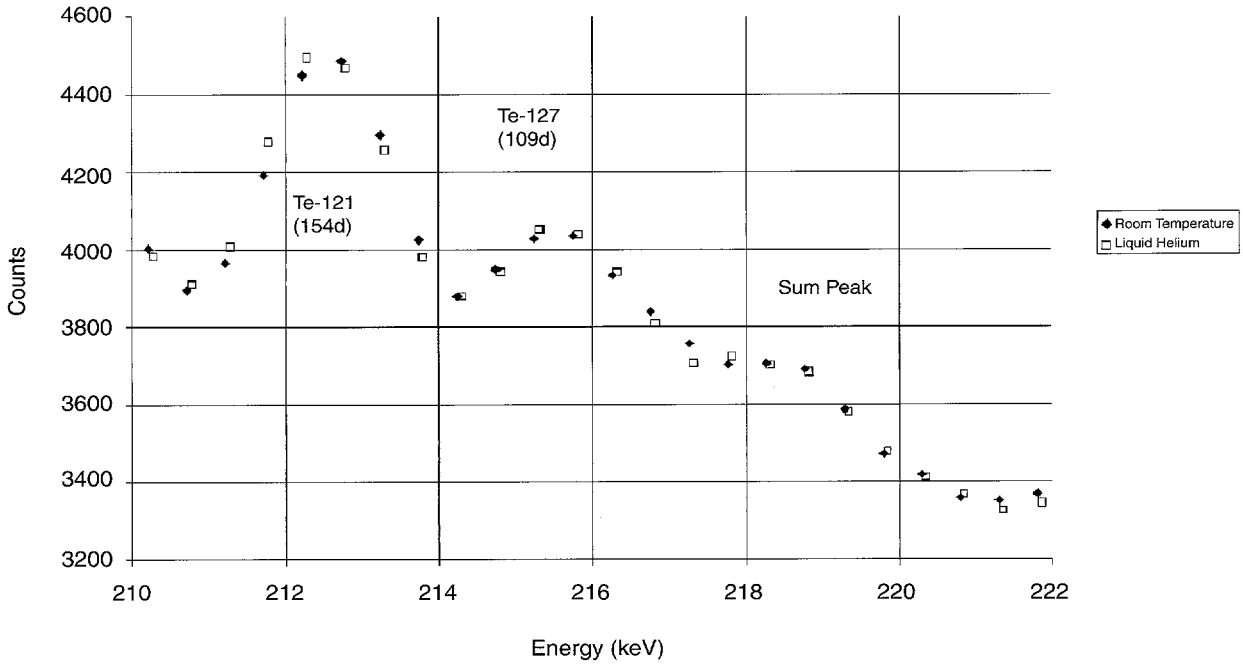
The stimulated emission of gamma rays from a nuclear isomer is expected to result in colinear photons and, therefore, should be observable as a sum peak in the gamma-ray spectrum. Skorobogatov and Dzevitskii reported a greatly enhanced sum peak (218.56 keV) when a sample of beryllium telluride containing ^{125m}Te was cooled from room temperature to near-liquid-helium temperatures. We have repeated their experiment and have observed no increase in the sum peak above accidental summing (see accompanying figure). The upper limit for the stimulated-emission cross section based on the three-standard-deviation statistical error is 1.1×10^{-20} cm^2. This result is nearly one order of magnitude lower than the cross section reported by the Russian scientists. Our cross section would not allow gain in a single-pass gamma-ray laser. Our results support the position of Baldwin and Solem rather than that of Kamenov.

We plan to continue searching for evidence of superradiant emission of gamma rays from ^{93m}Nb. This nucleus has a two-level isomeric transition and, therefore, does not suffer the difficulties inherent in three-level nuclear isomers.

Publications

Rundberg, R.S., et al., "Cross Section for the Stimulated Emission of 109.28 keV Gamma Rays from ^{125m}Te" (Induced Gamma Emission Meeting IGE97, Preton, Romania, August 14–15, 1997).

Wilhelmy, J.B., et al., "Search for Superradiant Emission from ^{125m}Te" (APS Meeting DNP97, Whistler, British Columbia, October 5–8, 1997).



Gamma-ray spectra in the region of the sum peak (218.56 keV) taken at room and liquid-helium temperatures.

Observables in Relativistic Heavy-Ion Collisions

John P. Sullivan

The objective of this project is to improve our understanding of the space-time history of the excited matter formed in nuclear collisions above 10 GeV per nucleon. We studied data from the experiments at the heavy-ion accelerators at Brookhaven National Laboratory and CERN. The goal of the experiments is to produce a state of matter called the quark-gluon plasma (QGP), which is expected to exist when matter is excited to a temperature high enough to deconfine the quarks and gluons that are normally confined within individual particles. This state of matter is believed to have existed for the first 10 μs (approximately) after the Big Bang.

We studied the sensitivity of experimental observables to determine whether a QGP was formed. We used a combination of a hydrodynamic model (Hylander) and a realistic, expanding source model, which parameterized the properties of the excited system. The expanding source model gave the correct answer when applied to the output of the hydrodynamic model—in which the correct results are known. This result gives us confidence in the results obtained by applying the model to real data.

We found that kaons stop interacting with the system sooner than pions and that kaon correlation measurements determine the size of the system better than pion correlations. In addition, we found excellent agreement between the data and our hydrodynamic model calculations when we assumed that a QGP had been formed, although other explanations can also explain the data available so far.

Publications

Chapman, S., and J.R. Nix, "Freezeout Properties in Relativistic Heavy Ion Collisions," *Bull. Am. Chem. Soc. Abstr. Pap.* **213**, 91 (1997).

Ornik, U., et al., "The Effects of Bose-Condensates on Single Inclusive Spectra and Bose-Einstein Correlations," *Phys. Rev. C* **56**, 1 (1996).

Ornik, U., et al., "Hydrodynamical Analysis of Symmetric Nucleus-Nucleus Collisions at CERN/SPS Energies," *Phys. Rev. C* **54**, 1381 (1996).

Schlei, B.R., "Extracting the Equation of State of Nuclear Matter through Hydrodynamical Analysis," *Acta Physica Hung.* **5**, 1 (1997).

Schlei, B.R. "Space-Time Extensions from Space-Time Densities and Bose-Einstein Correlations," *Phys. Rev. C* **55**, 954 (1997).

Schlei, B.R., and D. Strottman, "Hydrodynamical Analysis of Symmetric Nucleus-Nucleus Collisions at SPS Energies," in *Proc. Workshop on Nuclear Physics at the Turn of the Millennium* (World Scientific, Singapore, 1996), p. 458.

Schlei, B.R., and D. Strottman, "Probing the Equation of State of Nuclear Matter with Relativistic Hydrodynamics," *Proc. ACS Conference, Las Vegas, Nevada* **1**, 1 (1997).

Schlei, B.R., and N. Xu, "$m_{\perp}$ Dependence of Bose-Einstein Correlation Radii," *Phys. Rev. C* **54**, 2155 (1996).

Schlei, B.R., et al., "Effects of the Equation of State of Nuclear Matter in 158*A* GeV Pb+Pb Collisions," *Bull. Am. Phys. Soc.* **X**, 1 (1997).

Schlei, B.R., et al., "On the Equation of State of Nuclear Matter in 158*A* GeV Pb+Pb Collisions" (submitted to *Phys. Rev. Lett.*).

Schlei, B.R., et al., "The Linear Correlation Coefficient vs. the New Cross Term in Bose-Einstein Correlations" (submitted to *Phys. Lett.*).

Schlei, B.R., et al., "Particle Spectra and Bose-Einstein Correlations for Pb+Pb at SPS Energies from Relativistic Hydrodynamics," *Proc. Quark Matter '96 Conf.* **1**, 1 (1996).

Production and Trapping of Ultracold Neutrons in Superfluid Helium

Geoffrey Greene

Other researchers have proposed that the production and storage of high densities of ultracold neutrons can be used for fundamental research in nuclear physics and for performing sensitive measurements of condensed matter. One particular suggested application of this technique involves the trapping of neutrons in a magnetic field within the superfluid helium volume. Neutron decays within the trap volume are then detected by the scintillation light produced in the liquid helium.

We have constructed a cryostat and magnetic trap as well as a proof-of-principle light detection system. We installed this system on a cold-neutron beam line at the Cold Neutron Research Facility at the National Institute of Standards and Technology. Our preliminary results indicate the clear detection of helium scintillation light from the detection vessel.

Exploring and Testing the Standard Model and Beyond

Geoffrey West

The goal of this project was to extend and develop the predictions of the standard model of particle physics in several different directions. The focus of our work includes various aspects of the strong nuclear interactions in quantum chromodynamics (QCD), electroweak interactions, and the origin of baryon asymmetry in the universe, as well as gravitational physics.

We have extended our work on the unification of the concepts of scaling and fractals with the renormalization group (previously applied to give a fractal description of the nucleon in a generalized QCD parton model) to biophysical systems such as the cardiovascular and respiratory systems. We showed how duality in supersymmetric QCD follows from nonpertubative instanton configurations. We estimated the masses of light quarks using lattice QCD. Our estimates are considerably smaller than the values commonly used in phenomenology and have important implications for the charge-parity-violating parameters. We continued to explore the interface of quantum and classical chaos based on our discovery of semiquantum chaos, and we continued our work on the conformal anomaly, proving the effects of the theory.

Within the standard model itself, most of the research will continue to be oriented toward attempts to understand the QCD sector which governs the interaction of quarks and gluons. A major objective is to understand the structure of hadrons, specifically the nucleon and pion, as built out of quarks. We will also investigate questions associated with possible new states of matter (e.g., the quark-gluon plasma) and their possible consequence in the development and evolution of the universe. In the electroweak sector, we will investigate the origin of mass and the possibility of observing proton decay. At the very frontier of our knowledge is the question of unifying all the basic forces of nature of which the standard model is just one component. In this regard, we are examining possibilities based on superstring models in order to be able to incorporate quantum mechanics into Einstein's theory of relativity.

Publications

Anglin, J., and S. Habib, "Classical Dynamics for Linear Systems: The Case of Quantum Brownian Motion," *Mod. Phys. A* **11**, 2655 (1996).

Cooper, F., "Nonequilibrium Time Evolution Problems and Schwinger's Closed-time Path Formalism," in *Proceedings of the 23rd Coral Gables Conference in Coral Gables, Florida* (Plenum Press, New York, 1996), p. 312.

Cooper, F., et al., "Anomalous Transverse Distribution of Pions as a Signal for the Production of Disoriented Chiral," *Phys. Rev. C* **54**, 3298 (1997).

Cooper, F., et al., "Nonequilibrium Dynamics of Symmetry Breaking in Lambda Phi 4 Field Theory," *Phys. Rev. D* **55**, 6471 (1997).

Cooper, F., et al., "Spherically Symmetric Random Walks I. Representation in Terms of Orthogonal Polynormals," *Phys. Rev. E* **54**, 100 (1996).

Cooper, F., et al, "Time Evolution of the Chiral Phase Transition during a Spherical Expansion," *Phys. Rev. D* **54**, 2213 (1996).

Dorey, N., et al., "Multi-Instanton Calculus in N = 2 Supersymmetric Gauge Theory," *Phys. Rev. D* **54**, 2921 (1996).

Habib, S., et al., "Winding Transitions at Finite Energy and Temperature: An O(3) Model" (to be published in *Phys. Rev. D*).

Holzscheiter, M.H., et al., "Are Antiprotons Forever?" *Phys. Lett. A* **214**, 279 (1996).

Nieto, M.M., "Analytic Description of the Motion of a Trapped Ion in an Even or Odd Squeezed State," *Phys. Lett. A* **219**, 180 (1996).

Nieto, M.M., "Functional Forms for the Squeeze and the Time-Displacement Operators," *Quantum Semiclass. Opt.* **8**, 1061 (1996).

Nieto, M.M., "Quantum Interference from Kaons to Neutrinos (with Quantum Beats in between)," *Hyperfine Interactions* **100**, 193 (1996).

Nieto, M.M., and Daboul, J., "Exact E = 0, Quantum Solutions for General Power-Law Potentials," *Mod. Phys. A* **11**, 3801 (1996).

Nieto, M.M., et al., "Antimatter Gravity and Antihydrogen Production," in *New Frontiers in Hadronic Mechanics, Part III* (Hadronic Press, Palm Harbor, FL, 1996), p. 319.

Weak Interaction Measurements with Optically Trapped Radioactive Atoms

David Vieira

The main goal of this work is to undertake high-precision tests of electroweak interactions in atomic nuclei. We are currently concentrating on a high-precision beta-asymmetry measurement of trapped and highly polarized ^{82}Rb atoms. Such a measurement will search for the existence of right-handed interactions—testing the maximal parity-violating nature of the electroweak interaction in a pure Gamow-Teller transition at a new level of precision. The key technology needed to undertake these measurements is efficient optical and magnetic trapping of radioactive species.

Good progress was made in developing an efficient atom trap for radioactive ^{82}Rb by coupling a magnetooptical trap to a mass separator (see accompanying figure). In particular, we have been successful in (1) preparing radiochemically separated samples of ^{82}Sr→^{82}Rb, (2) operating and improving the performance of the surface ionization source of the mass separator, (3) investigating the implantation and atomic release of ^{82}Rb from a variety of inductively heated foils, (4) improving the trapping cell and laser system, and (5) developing ultrasensitive fluorescence detection schemes that are based on photon counting and phase-sensitive detection techniques. We are very close to demonstrating the first trapping of ^{82}Rb. In parallel with this work, we have been modeling and designing a time-orbiting-potential, pure magnetic trap and testing various scintillation counters for the confinement and beta-decay detection of polarized ^{82}Rb.



Photograph of the magnetooptical trap with an internal yttrium-coated foil into which a mass-separated ^{82}Rb ion beam is implanted using a mass separator. After a suitable accumulation period (~150 s), the foil is inductively heated to release ^{82}Rb as atoms. These atoms are then trapped with circularly polarized laser beams that enter through each surface of the trapping cell. Fluorescence photons are counted to detect the trapped atoms.

Publications

Guckert, R., et al., "Coupling an Optical Trap to a Mass Separator," *Nucl. Instrum. Methods Phys. Res. B* **126**, 383 (1997).

Chiral Symmetry in Finite Nuclei

David Madland

We are calculating the properties of finite nuclei over a wide mass range by using quantum chromodynamics (QCD) scales and chiral Lagrangians together with dimensional-power-counting techniques to constrain the various in-medium, N-body nuclear forces that we test. We have found good evidence that QCD and chiral symmetry apply to finite nuclei, but the evidence at this time is only partly compelling. The goal is to construct a Lagrangian whose coupling constants are not only all natural, but whose predictive power is superior to our original Nikolaus, Hoch, Madland (NHM) Lagrangian.

We have found three chi-square minima for the NHM Lagrangian: (1) the original set of coupling constants in which six of the nine constants are natural and the sum of two of the remaining three constants is natural; (2) a second set in which seven of the nine constants are natural, the sum of the remaining two is natural, and the predictive power is not as good as the original set; and (3) a third set with properties similar to the second set but with somewhat worse predictive power. However, if the coupling constants for the quartic terms in the scalar and vector densities are constrained to be identical, then all nine of the coupling constants are natural, but the predictive power is worse than for the second set.

We then introduced additional isospin dependence via a cubic isovector-scalar term, which led to ten coupling constants. With this new Lagrangian, we obtained results entirely analogous to (1), (2), and (3) above but with little improvement in predictive power. We conclude that additional terms (axial vector, tensor, other isospin dependence) may be required. A consequence of our work has been the demonstration of the origin of (approximate) pseudo-spin symmetry in nuclei in terms of the properties of the lower components of our Dirac wave functions.

Publications

Friar, J.L., "Scales: From Deinonychus to Dysprosium" (Physics Dept. Colloquium, New Mexico State University, Las Cruces, NM, February 1997).

Friar, J.L., "Scales: From Deinonychus to Dysprosium" (Physics Dept. Colloquium, University of North Texas, Denton, TX, November 1996).

Ginocchio, J.N., and D.G. Madland, "Pseudo-Spin Symmetry and Relativistic Single-Nucleon Wave Functions" (to be published in *Phys. Rev. C*).

Madland, D.G., "QCD Scales and Chiral Symmetry in Finite Nuclei," in *Proceedings of the International Conference on Nuclear Data for Science and Technology, Trieste, Italy, May 1997* (Italian Physical Society, Bologna, Italy, in press).

Theoretical Studies of Allosteric Changes and DNA Binding of a cAMP-Dependent Transcription Activation Protein

Angel E. Garcia

The *E. coli* cAMP receptor protein (CRP) plays an important role in mediating transcription activation of several genes in enteric bacteria. CRP activation is a complex process involving the binding of cAMP and the interaction of CRP with DNA and RNA polymerase. Details of the mechanism by which cAMP affects allosteric control over CRP activity remain unclear.

We study the structure and dynamics of CRP by molecular dynamics simulations. These studies will provide theoretical structures for CRP in different binding states of cAMP and DNA and provide information from which currently obscure elements of the cAMP-mediated activation pathway of CRP can be predicted. These predictions will provide the rationale for future experimentation designed specifically to evaluate the theory-based cAMP-mediated pathway to CRP activation.

Molecular dynamics simulation of CRP:$(cAMP)_{n=1,2}$ in a noncrystalline environment has been conducted. We found that one tyrosine (Tyr) amino acid, Tyr-99, makes an important contact with cAMP. In vitro experiments have revealed that Y99F and Y99A mutations affect the transcription activation. Mutants of all Tyr residues in CRP to fluorinated Tyr have been produced to further test the role of Y99 in the allosteric changes.

We have developed a multistate Gaussian model for the distribution of electrostatic solvation energies of a solute in liquid water. We studied the potential of mean force (PMF) of glycosidic angle rotation of cAMP in aqueous solution. We found that the *syn* conformation is the most stable in aqueous solution by 0.4 kcal/mol over the *anti* conformation. These two conformations are separated by 1.0- and 1.6-kcal/mol energy barriers. We further developed a PMF approach to study biomolecular hydration. Hydrophobic hydration is now well described by this method. The hydration of CRP-DNA complexes has been studied.

Publications

Garcia, A.E., et al., "Hydration of an Alpha-Helical Peptide: Comparison of Theory and Molecular Dynamics Simulation," *Proteins: Structure, Function and Genetics* **27**, 471 (1997).

Garcia, A.E., et al., "Multi-Basin Dynamics of a Protein in Its Crystalline Environment," *Physica D* **107**, 225 (1997).

Garcia, A.E., et al., "Variations on a Theme by Debye and Waller: From Crystals to Proteins," *Proteins: Structure, Function and Genetics* **29**, 153 (1997).

Garde, S., et al., "Hydration of Biological Macromolecules: From Small Solutes to Proteins and Nuclei Acids" (Material Research Society Symposium on Statistical Mechanics on Biological Systems, Boston, MA, December 7, 1996).

Garde, S., et al., "Origin of Entropy Convergence in Hydrophobic Hydration and Protein Folding," *Phys. Rev. Lett.* **77**, 4966 (1996).

Hummer, G., et al., "Electrostatic Potentials and Free Energies of Solvation of Polar and Charged Molecules," *J. Phys. Chem.* **101**, 3017 (1997).

Hummer, G., et al., "Ion Sizes and Finite-Size Corrections for Ionic Solvation Free Energies," *J. Chem. Phys.* **107**, 2275 (1997).

Hummer, G., et al., "Multistate Gaussian Model for Electrostatic Solvation Free Energies," *J. Am. Chem. Soc.* **119**, 8523 (1997).

Hummer, G., et al., "The Pressure Dependence of Hydrophobic Interactions is Consistent with the Observed Pressure Denaturation of Proteins" (to be published in *Proc. Natl. Acad. Sci. U.S.A.*).

Hummer, G., et al., "A Statistical Mechanical Description of Biomolecular Hydration," *Faraday Discussions* **103**, 203 (1996).

Structure and Function of Nucleosomes Positioned on Repeated DNA Sequences Associated with Human Diseases

E. Morton Bradbury

This project has the following specific aims: (1) determination of unusual DNA structures formed by polynucleotides containing triplet microsatellite and insulin minisatellite sequences; (2) development of an in vitro replication assay to demonstrate the presence of these unusual DNA structures during replication; and (3) reconstitution of nucleosomes with DNA containing CTG/CCG triplet repeats as positioning sequences (C = cytosine, T = thymine, and G = guanine). The successful completion of these specific aims will help us to understand the structural basis of length polymorphism exhibited by various microsatellites and minisatellites in the human genome.

Using two-dimensional nuclear magnetic resonance, we have determined the high-resolution structure of the intramolecularly folded C-rich strand of the insulin minisatellite. We performed a series of in vitro replication assays to demonstrate that the GAA/TTC triplet repeats (A = adenine) associated with Friedrich's ataxia form triplexes during replication. The stability of these triplexes is significantly enhanced for disease phenotypes with repeat number exceeding 200. We have identified (CTG)8 as the strongest nucleosome positioning sequence, and we are currently performing structural studies on this well-positioned nucleosome.

We have identified DNA sequence motifs containing trinucleotide repeats that show higher affinity for nucleosomes. We have constructed a library of DNA fragments isolated from nucleosome dimers and have screened this library for sequences containing simple sequence repeats. Using this approach, we have identified several sequences with trinucleotide repeat regions. We then tested nucleosome monomer-length fragments of these sequences for their ability to position nucleosomes. We also tested several previously characterized sequences with similar sequence motifs, including the Huntington's disease gene. Using reconstitution and a gel-mobility-shift assay, we have identified a sequence containing a stretch of 8 CTG trinucleotide repeats that positions nucleosomes to a single position. We have also shown that several other sequences with longer stretches of CTG repeats do not position nucleosomes to a single position. This information can greatly contribute to our understanding of the role of DNA sequence in nucleosome positioning.

Publications

Catasti, P., et al., "The C-Rich Strand of the Insulin Minisatellite Folds Back to Form an Intercalated C-C Base Paired Structure," *J. Mol. Biol.* **272**, 369 (1997).

Mariappa, S.V.S., et al., "Hairpin and Junction Structures of Fragile X Triplets," *Proc. Ninth Conversation Biol. Struct. Dynam.* **2**, 105 (1996).

Sum-Frequency Generation Studies of Membrane Transport Phenomena

R. Brian Dyer

The objective of this work is to study the transport of protons and ions across biological membranes. Vectorial pumping of ions, generally carried out by membrane-bound proteins, is one of the most fundamental processes in living organisms. This process is critical for energy transduction in respiration and photosynthesis and for a wide variety of cellular signal-transduction events. Because of the difficulty in obtaining structural and dynamic information about both membrane-bound proteins and membranes, a molecular-level understanding of proton and ion pumping has remained elusive.

We have developed sum-frequency generation (SFG) spectroscopy for the study of membrane phenomena. SFG is a nonlinear spectroscopic technique that is uniquely sensitive to interfaces and has demonstrated structural specificity. Our approach uses a femtosecond titanium-sapphire laser to parametrically generate broadband IR pulses throughout the mid-IR using the nonlinear optical materials ß-barium borate and $AgGaS_2$. The IR pulses are combined in a nonlinear SFG process at the membrane/water interface and are detected by a multichannel SFG instrument, which we also developed, that uses a spectrograph and charge-coupled device to detect the broadband, up-converted IR light.

We have used SFG and conventional vibrational spectroscopic approaches to study proton transport processes in cytochrome *c* oxidase. A key finding has been the identification of vibrational modes associated with proton labile groups, including a glutamic acid near the redox-active binuclear center and structural waters. These groups are sensitive to the ligation and redox states of the metal

centers and, hence, are ideal candidates for coupling redox energy to proton transport processes.

Publications

Causgrove, T.P., and R.B. Dyer, "Picosecond Structural Dynamics of Myoglobin Following Photolysis of CO," *J. Phys. Chem.* **100**, 3273 (1996).

Kim, Y., et al., "Cyanide Binding and Active Site Structure in Heme-Copper Oxidases: Normal Coordinate Analysis of Iron-Cyanide Vibrations of a_3^{2+}CN- Complexes of Cytochromes ba_3 and aa_3," (to be published in *Biospectroscopy*).

Puustinen, A., et al., "Fourier Transform Infrared Evidence for Connectivity Between Cu_B and Glutamic Acid 286 in Cytochrome bo_3 from *E. Coli*," *Biochem.* **36**, 13195 (1997).

Riistama, S., et al., "Bound Water in the Proton Translocation Mechanism of the Heme-Copper Oxidases," *FEBS Lett.* **414**, 275 (1997).

Shreve, A.P., and R.B. Dyer, "A Versatile Ultrafast Laser System for Time-Resolved Infrared and Sum-Frequency Generation Spectroscopies" (submitted to *Rev. Sci. Instrum.*)

Shreve, A.P., et al., "Sum-Frequency Generation Studies of Self-Assembled Systems Using Multi-Channel Detection," *Abstr. Pap. Am. Chem. Soc.* **214**, 298 (1997).

Advanced Telemedicine Development

David W. Forslund

A distributed electronic medical record is of fundamental importance to the improvement of health care in this nation. Our goal is to develop a database access system, called TeleMed, that allows access to electronic medical records at almost any location in the world, something that was considered impossible in the past. This year we developed distributed-object interfaces to, and implementations of, the primary components of the Essential Medical Data Set to allow physicians to simultaneously access and interact with object or relational databases in an intuitive manner. We also included support for multimedia data, such as images, in a scalable way (see accompanying figure).

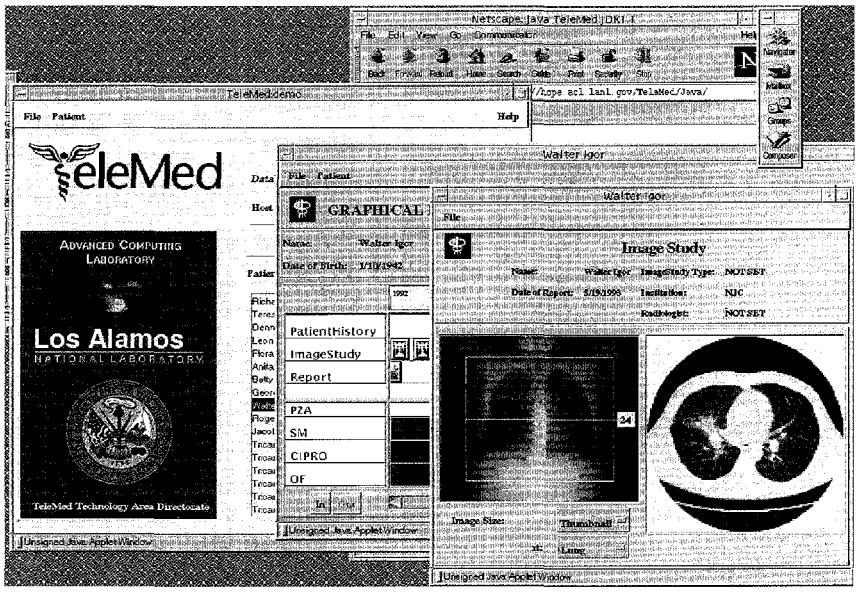
We developed the TeleMed system entirely in Java; it uses the Common Object Request Broker Architecture from the Object Management Group (OMG) for communicating among the clients and servers. The patient identification process conforms to the evolving specification being defined by the OMG.

Our research this year has had substantial impact on the design of TeleMed by making it more scalable and portable. We have outlined the basic elements of the required security and have devised a mechanism for handling immunization records over a wide area, but have not yet implemented them.

Publications

Forslund, D.W., and J.L. Cook, "The Importance of Java and CORBA in Medicine," in *Journal of the American Medical Informatics Association, 1997 Annual Fall Symposium*, D.R. Maysis, Ed. (Hanley and Belfus, Inc., Philadelpha, PA, 1997), p. 364.

Kilman, D.G., and D.W. Forslund, "An International Collaboratory Based on Virtual Patient Records," *Commun. ACM* **40**, 110 (1997).



The Netscape Communicator 4.0 browser can be used to view the entire TeleMed client, showing the graphical patient record and the management of computed tomography (CT) images.

Genome Instability

David Chen

Persistent genomic instability has been observed in the progeny of mammalian cells exposed to ionizing radiation. This instability is manifested as delayed expression of lethal mutations, accumulation of coincidental mutations, and elevated, nonclonal karyotypic heterogeneity. The molecular mechanisms underlying radiation-induced, heritable genome instability are not known. In the yeast *S. cerevisiae*, there is strong evidence that genes of the RAD52 epistasis group, which are essential for repair of double-strand breaks in DNA and homologous recombination, are involved in maintaining the global integrity of the genome. Recently, human homologs of the RAD51 and RAD52 genes in the yeast have been cloned.

The hypothesis we proposed to test is that hhRAD51 and hhRAD52 have roles in maintaining genome integrity, such that loss of expression of these genes will exacerbate radiation-induced, heritable genome instability. To test this hypothesis, we have transferred a human chromosome 4 into the Chinese hamster cell mutant that is deficient in a RAD51 homolog gene to determine whether the human chromosome 4 will be unstable in the RAD51-deficient background.

To understand the roles of hhRAD51 and hhRAD52 in repair of double-strand breaks and in homologous recombination and genome instability in human cells, we plan to construct homozygote knockouts of these genes in the human HT1080 cell line. We have developed human genomic clones of RAD51 and RAD52 and determined the genomic structure of these two genes. We have also constructed gene-targeting vectors and transfected them to the human HT1080 cells. We are in the processing of identifying both RAD51 and RAD52 heterozygote cell lines.

As an alternative to gene knockouts (or in conjunction with a single-allele knockout), we are testing our hypothesis by using a mutant cell line from a Chinese hamster that is deficient in one of the RAD51 homologs. We have transferred human chromosome 4 into a Chinese hamster mutant cell line that is deficient in the RAD51 gene. As shown in the accompanying figure, we have confirmed a single human chromosome 4 in the background of Chinese hamster cells. The human chromosome 4 will be used as a marker to establish a relationship between chromosome instability and the function of the RAD51 gene. This is a viable approach since we had previously shown that a human chromosome is unstable in a double-strand-break-deficient hamster cell line. If human chromosome 4, which served as a marker chromosome, is unstable in the hamster cell line, this implies that the RAD51 gene is involved in maintaining chromosome stability in mammalian cells.



This figure shows a Chinese hamster cell line that contains a single human chromosome 4 (white). The spontaneous and radiation-induced chromosome instability of the human chromosome 4 will be determined.

Publications

Chen, F., et al., "Cell Cycle Specific Expression of the Human Homologs of the Yeast DSB Repair Proteins RAD52" (to be published in *Mutat. Res.*).

Gadbois, D.S., et al., "Alterations in the Progression of Cells through the Cell Cycle after Exposure to Alpha Particles of Gamma Rays," *Radiat. Res.* **146**, 414 (1996).

Development of a Human Artificial Chromosome

Norman Doggett

The major objective of this proposal is to develop a mammalian artificial chromosome (MAC). In the first year of this research, we focused on cloning a human centromere for incorporation into a MAC. To achieve this goal, we have been pursuing two approaches: (1) directed cloning of centromere fragments from pulsed-field gel-electrophoresis gels and (2) transformation-associated recombination (TAR) cloning of a human centromere. For pulsed-field gel isolation, we have identified enzyme and gel conditions that result in purification of a single 1.7-million-base-pair restriction fragment containing the entire centromere region from human chromosome 16. Purification yields from these gels have thus far been too low for successful cloning.

To achieve TAR cloning of the chromosome 16 centromere, we have designed TAR hook sequences from euchromatic DNA on the short arm of the chromosome flanking the alphoid centromere repeats. Our collaborators determined this sequence from a bacterial-artificial-chromosome clone that had been isolated with markers we developed from this region. The second TAR hook is designed from the satellite II sequence on the long arm that is adjacent to the centromere. TAR cloning works by designing a vector with hook sequences flanking the region that is to be cloned. This vector is mixed with total genomic DNA and introduced into yeast cells, which perform homologous recombination between the hook and target sequence to create a recombinant clone of the target. The TAR cloning experiments are in progress and should yield clones for future analysis.

Engineering the Specificity of xylR, a Bacterial Protein that Detects Chemicals in the Environment

Thomas Terwilliger

Microorganisms have sensitive systems for detecting trace amounts of organic compounds that are based on the ability of certain regulator proteins to bind these compounds. Binding of a compound to a regulator protein initiates a cell's response by activating the pathway to degrade that compound into nontoxic metabolites useful to the cell. If we were able to manipulate the specificity of such a detector protein, we could produce microorganisms that are better able to detect and degrade organic pollutants such as trichloroethylene (TCE).

The presence of TCE in groundwater and soil is one of the most widespread and serious pollution problems in the United States today. Native bacteria that degrade TCE have not been very effective for bioremediation because the detection and expression systems for this compound are not very sensitive or well regulated. The xylR protein is particularly well suited as a target for engineering novel binding specificity because (1) it is a part of a well-characterized system, (2) it has a modular domain structure, and (3) if its specificity is altered, it would have substantial utility as a biosensor and in bioremediation. This project has three parts: mutagenesis of the gene encoding the xylR toluene-binding domain, determination of the structure of this domain, and selection of variants of the xylR protein specific for TCE.

We have continued the development of a system for the mutagenesis and selection of xylR proteins. We cloned the gene encoding the xylR protein into plasmids, and the resulting protein is functional in *E. coli*. The powerful mutagenesis and selection procedures already developed for *E. coli* plasmids can be applied to the xylR system. For efficient mutagenesis and selection of TCE-detecting variants of the xylR protein, we constructed a plasmid that contains the natural xylR gene from *P. putida* under control of an inducible production promoter. Our major development during the past year has been to use the fluorescent green protein as a marker for activity of the xylR gene. Using this protein, we can use flow cytometry to separate bacteria that are activated by specific compounds from those that are not.

Identification of Proteins that Associate with UBL1, a DNA Damage Repair Regulating Protein

David Chen

UBL1 is a ubiquitin-like protein that our group first discovered. It associates with a centromere protein (MIF2) involved in mitosis, the UBC9 protein involved in cell cycle control, and Rad51 and Rad52 proteins, which are involved in DNA repair. Based on these observations, we proposed that UBL1 might regulate DNA repair and cell cycle control. To fully understand the mechanism of UBL1-mediated cellular processes, it is important to characterize the UBL1 conjugation process. Our goal is to identify both the enzymes that catalyze the conjugation reaction and the substrates being conjugated to UBL1.

A two-hybrid system is based on the fact that Gal-4 protein has two independent functional domains: (1) the N-terminus 147 amino acids that specifically bind to an upstream activation sequence (UAS) of the Gal-1 promoter and are called Gal-4 DNA binding domain (Gal4-DB); (2) the C-terminus amino acids 768–881 that are involved in transcription activation and are called Gal-4 DNA activation domain (Gal4-DA). When Gal4-DB binds to the UAS of Gal-1, the Gal4-DA comes to the proximity of Gal4-DB, which in turn activates the transcription of the gene that is controlled by Gal4-DB.

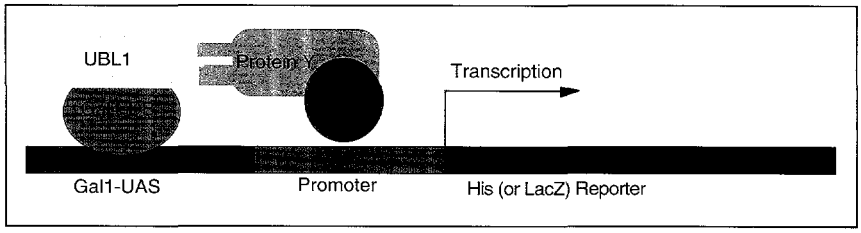
In the yeast two-hybrid approach (see figure), the Gal4-DB was fused to UBL1 in a Trp+ vector, such as pGBT9; the other protein (Protein Y) was fused to Gal4-DA in a vector bearing Leu+, such as pGAD424. These two fusion constructs were co-transfected into the host strain (SFY526 or HF7c) by selecting in Leu– and Trp– media. If UBL1 and Protein Y interact, they will mimic the function of a native Gal-4 protein; therefore, a reporter gene will be expressed. When the reporter gene is *His*, the yeast should be able to grow in a *His*– selection medium. When the reporter gene is *LacZ*, the yeast should have *LacZ* activity to turn colonies into blue color in the presence of X-Gal. Utilizing this system, we found that UBL1 interacts with UBC9 and several novel proteins. Currently, we are characterizing these new proteins.

Publications

Shen, Z., et al., "Associations of UBE21 with Rad52, UBL1, p53, and Rad51 Proteins in a Yeast Two-Hybrid System," *Genomics* **37**, 183 (1996).

Shen, Z., et al., "UBL-1, a Human Ubiquitin-like Protein that Associates with Human Rad51/Rad52 Proteins," *Genomics* **36**, 271 (1996).

Shen, Z., et al., "UBL1 Mediated Protein Conjugation Modules DNA Homologous Recombination" (submitted to *Mol. Cell. Biol.*).



A yeast two-hybrid system to identify proteins that associate with UBL1.

Optimization and Monitoring of Hollow-Fiber Bioreactors

James P. Freyer

Improving our understanding of the transport of molecules within three-dimensional masses of cells will result in several important applications. For example, supply of nutrients and removal of metabolic waste products within tumors have important implications for both drug delivery and basic therapeutic response. Current designs for the production of pharmaceuticals from mammalian cells are relatively inefficient in supplying the cells with adequate nutrients and removing the desired products from the waste stream. Much of our lack of understanding of the basic transport properties of tissues and cell cultures stems from a lack of experimental techniques for measuring microenvironmental factors within living systems.

Our original goals for this research project were (1) to develop novel nuclear magnetic resonance (NMR) methods for imaging transport and cellular physiology within hollow-fiber bioreactors (HFBs) used for culturing cells at very high densities and (2) to use those methods in optimizing the design of HFBs. Because of signal-to-noise limitations inherent in NMR measurements of traditional HFBs, however, the direction of our work has changed. The results of our new work are good: we have developed a completely

novel system for culturing cells in a three-dimensional matrix.

Work in the past year has focused on understanding the basic biology of this system. We have developed methods and specialized apparatus for the reproducible recovery of intact cells found at precise distances from the nutrient supply. The new system shows a similar dependence of cell proliferation and viability on distance from the supply membrane as do multicellular spheroids or HFBs, except that we can control the actual distance by varying the concentration of the cells.

Work is in progress to refine the NMR imaging parameters so that concentrations of critical metabolites can be measured as a function of distance from the supply membrane. In addition to the biological and NMR work with the new cell culture system, the collaborations on this project have resulted in the development of novel NMR techniques and mathematical models for measuring and predicting the transport of molecules through porous membranes. Significant progress has also been made in the design of specialized hollow fibers for gas separations.

Publications

Kunz-Schughart, L.A., and J.P. Freyer, "Adaptation of an Automated Selective Dissociation Procedure to Two Novel Spheroid Types," *In Vitro Cell. Dev. Biol.* **33**, 73 (1997).

Kunz-Schughart, L.A., et al., "Mitochondrial Distribution and Activity in Oncogene-Transformed Rat Fibroblasts Isolated from Multicellular Spheroids," *Am. J. Physiol.* **273**, C1487 (1997).

LaRue, K.E., et al., "Differential Regulation of Cyclin Dependent Kinase Inhibitors in Monolayer and Spheroid Cultures of Tumorigenic and Nontumorigenic Fibroblasts" (submitted to *Cancer Res.*).

Mattes, B.R., et al., "Formation of Conductive Polyaniline Fibers Derived from Highly Concentrated Emeraldine Base Solutions," *Synth. Met.* **84**, 45 (1997).

Mattes, B.R., et al., "Temperature and Pressure Dependent Gas Transport through Polyaniline Membranes" (to be published in *Int. J. Thermophys.*).

Pelligrino, J., et al., "Gas Sorption in Polyaniline. 1. Emeraldine Base," *Macromolecules* **29**, 4985 (1996).

Stegman, L.D, et al., "In Vivo ^{31}P MRS Evaluation of Ganciclovir Toxicity in Glioma Tumors Stably Expressing the Herpes Simplex Thymidine Kinase Gene," *NMR in Biomedicine* **9**, 364 (1997).

Winokur, M.J., and B.R. Mattes, "Determination of the Local Molecular Structure in Amorphous Polyaniline," *Phys. Rev. B* **54**, 18 (1996).

Winokur, M.J., and B.R. Mattes, "Differential Anomalous Scattering Studies of Amorphous HBr-Doped Polyaniline," *Synth. Met.* **84**, 725 (1997).

Protein Motions that Determine the Efficiency of Photosynthesis

Joel Berendzen

Our overall goal is to experimentally determine structures of photosynthetic reaction centers in dark- and light-adapted states and then to apply theoretical methods in order to understand the changes and their effects on reaction rates. The photosynthetic reaction center from purple bacteria is a standard model system for studying the remarkably efficient process of photosynthesis. In collaboration with the groups of Hartmut Michel (Max Planck Institute, Hamburg) and Ilme Schlichting (Max Planck Institute, Dortmund), we are measuring atomic motions that take place during the process of charge separation. In conjunction with these measurements, we are doing the theoretical calculations that allow us to interpret the results.

This year we made progress in both the theoretical and experimental components of this project. At this time, the experimental data are not yet advanced to the point that they are suitable for the basis of theoretical calculations, although the two components remain in close contact and are advancing in parallel.

We made three synchrotron runs to test crystal-freezing conditions. These conditions continue to be a problem because we are unable to freeze the standard (trigonal) form of crystals of reaction centers from *Rhodobacter sphaeroides* while retaining the high degree of crystalline order required. We began refining the data collected on two states of tetragonal crystals, the dark-adapted state and the state illuminated after freezing. The data in these states are good to 2.3 Å. We continued developing the microspectrophotometer, laser apparatus, and refinement methods needed to characterize the light-adapted state. We are applying the refinement methods to systems other than reaction centers. We developed a "multistate Gaussian theory" that will allow us to incorporate the structural inhomogeneity of proteins into charging-free energy calculations. We investigated the role of water in proton transfer processes within proteins. We used potential-of-mean-force calculations, Fourier transform infrared spectroscopy, and mutation studies.

Predictive Models for Transcriptional Enhancers

Goutam Gupta

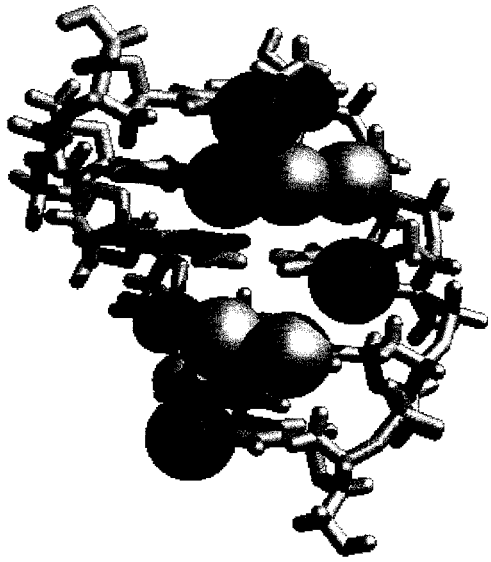
We intend to combine theory (at Los Alamos) and experiments (at the University of California, Berkeley) to accomplish the following aims: (1) development of computer algorithms (based on the principles of pattern recognition and specificity of DNA-protein recognition) for predicting the enhancers in even-skipped, dorsal hairy genes of Drosophila and other well-characterized enhancers in the eukaryotic genes and (2) use of these computer algorithms for theoretical predictions of the enhancers in the bithorax complex (BX-C) and Antenna pedia complex (AntpX-C) and for experimental verification by transformation assays.

Our molecular-modeling method enables us to assess the efficacy of groove-specific interactions in the Watson-Crick duplex of a given DNA sequence. This mechanism of binding agrees closely with the experimental data on the catabolite activator protein (CAP)-DNA complex. Our method offers a simple set of requirements a transcription element should satisfy to perform its regulatory role. The figure shows (at the top) the sequence bias at various positions of a canonical 8-base-pair-long enhancer; it also shows (at the bottom) various groove-specific interactions.

We are currently developing the molecular modeling and pattern recognition algorithms to predict the enhancers in BX-C and AntpX-C before verifying them by transformation assays.

Base occurance in enhancer pattern

	1	2	3	4	5	6	7	8
a	12	8	12	16	45	33	39	19
t	8	55	25	25	20	22	22	14
g	49	18	51	41	19	16	20	45
c	31	19	12	18	16	29	19	22



The sequence and structural patterns for protein recognition. The H-bond acceptors are plotted as white spheres, the H-bond donors are plotted as dark gray spheres, and the hydrophobic contacts are plotted as light gray spheres. We plotted the rest of the DNA molecule using the stick model.

Identification and Characterization of a Human DNA Double-Strand Break Repair Complex

David Chen

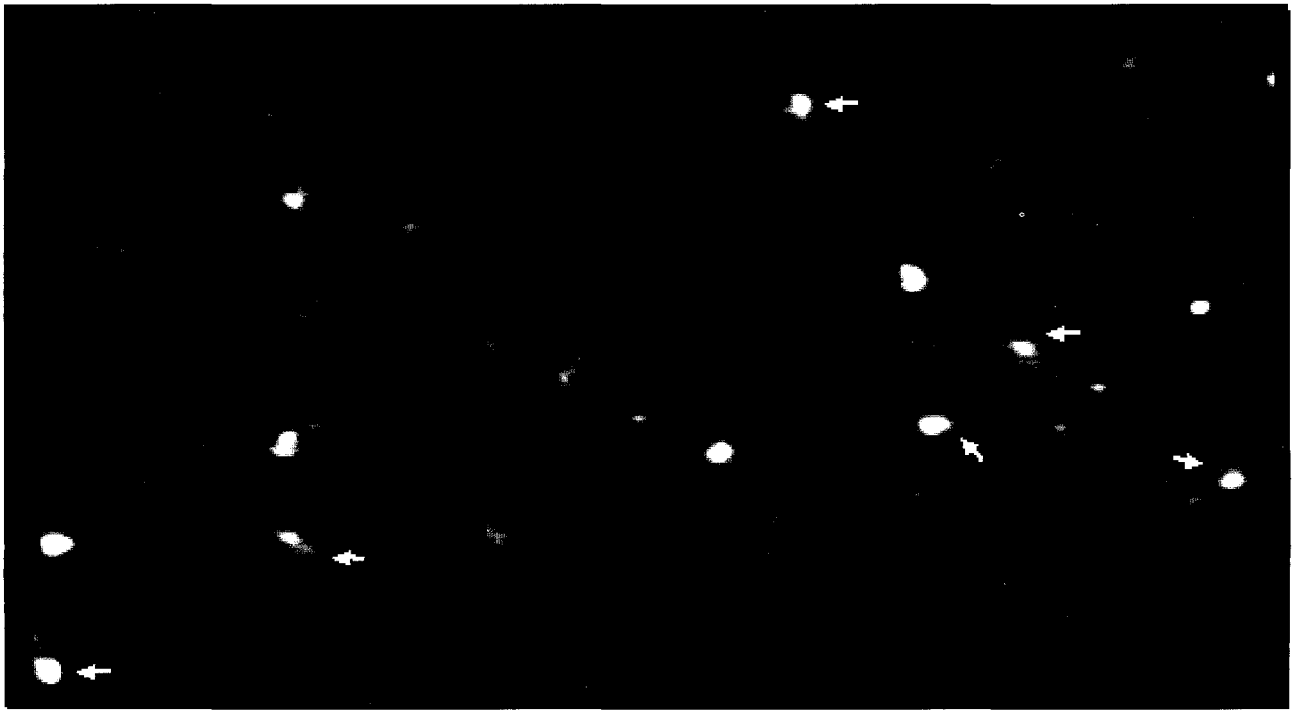
Our objective is to use atomic force microscopy to characterize the assembly and structure of the macromolecular assemblies involved in DNA repair. The studies have provided a better understanding of the structure, function, and dynamics of protein-DNA complexes formed by the DNA-dependent protein kinase, an enzyme required for DNA double-strand break repair in mammalian cells.

The atomic force microscope (AFM) is a high-resolution technique that offers several advantages over more traditional microscopic approaches. With its ability to image unstained and uncoated protein-nucleic acid complexes in air, aqueous buffers, or organic solvents, the AFM is uniquely suited for biological studies and provides a means by which macromolecular interactions can be visualized under native conditions. In particular, the AFM is useful for the analysis of DNA-protein interactions, DNA conformational changes, and structural studies of large proteins or multiprotein assemblies.

We have successfully used the AFM to characterize the DNA-dependent protein kinase (DNA-PK) and the complexes it forms with DNA (see figure). These studies demonstrate that the DNA-PK complex is capable of holding the ends of DNA together in vitro, suggesting that the protein may be responsible for assuring correct rejoining of the broken chromosomes that result from exposure to ionizing radiation. Thus, our studies have revealed important clues to the biological role of DNA-PK and demonstrate that the AFM will provide an indispensable tool for the characterization of the macromolecular complexes involved in DNA repair.

Publications

Cary, R.B., et al., "DNA Looping by Ku and the DNA-Dependent Protein Kinase," *Proc. Natl. Acad. Sci. U.S.A.* **94**, 4267 (1997).



DNA-PK assembles on the ends of DNA. AFM studies demonstrate that the end-bound DNA-PK complexes (white arrows) can self-associate to hold the ends of DNA molecules together (black arrows).

Numerical Simulations of Biochemical Self-Organization: Calcium Wave Propagation and Microtubule Growth

John Pearson

We are constructing and analyzing mathematical models of several biochemical processes. There are two primary focuses to this work: the nonlinear dynamics of calcium uptake and release by intracellular membrane compartments and the dynamics and organization of microtubules. Such models help to provide an understanding of the complex spatiotemporal dynamics involved in these processes.

This past year we investigated the role of the cell membrane in the propagation of the fertilization wave in xenopus eggs. The geometric features of the experimentally observed wave front were indicative of an influx of Ca^{2+} across the cell membrane. This influx was tested directly by removing the Ca^{2+} from the extracellular medium so that no influx was possible. The wave front was unaffected. Thus the geometry of the wave front indicates the existence of a boundary layer in the vicinity of the cell membrane. Numerical simulations are able to match the experimental wave profile only if there is a higher concentration of inosital trisphosphate near the cell wall. This interplay between numerical methods and experimental observations provides a deeper understanding into the spatiotemporal dynamics of the fertilization signal. Further work in this area should help us understand how processes such as the breakdown of the nuclear envelope are triggered by the calcium signal.

Discussions at a workshop organized by our LDRD team led to a new understanding of the role of the discreteness of the release sites. We created and analyzed a skeletal model of calcium-induced calcium release (see the accompanying figures). This model operates on an all-or-none basis: calcium is released only when the local concentration exceeds a threshold. This model helps us to understand analytically the transition from a discrete to a continuous distribution of release sites. Realistic values of the diffusion coefficients, release rates, and site spacings correspond to the poorly understood transition zone that separates the continuous realm from the discrete.

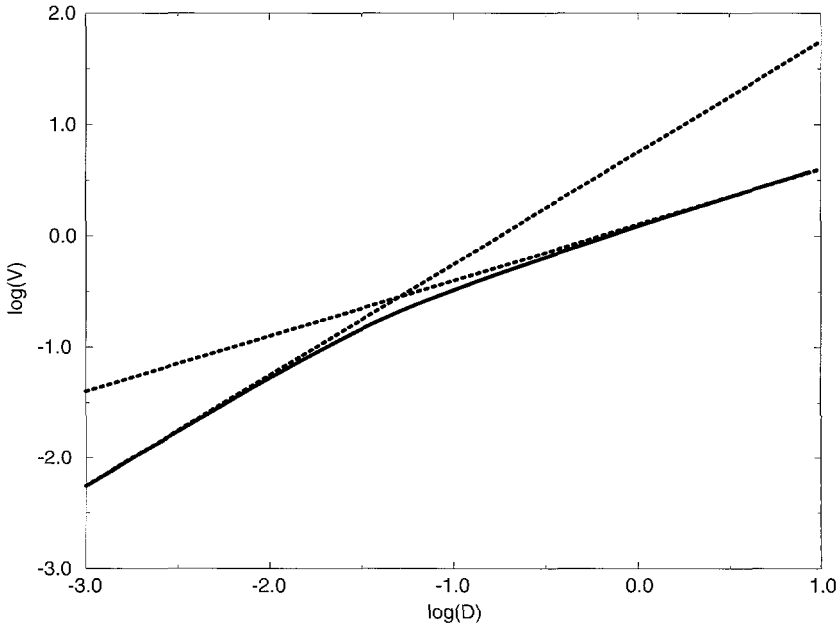
We have developed and submitted for publication a new integration scheme for stiff differential equations such as those that arise in chemical kinetics. We are preparing several other articles for publication that include models of microtubule dynamics.

Publications

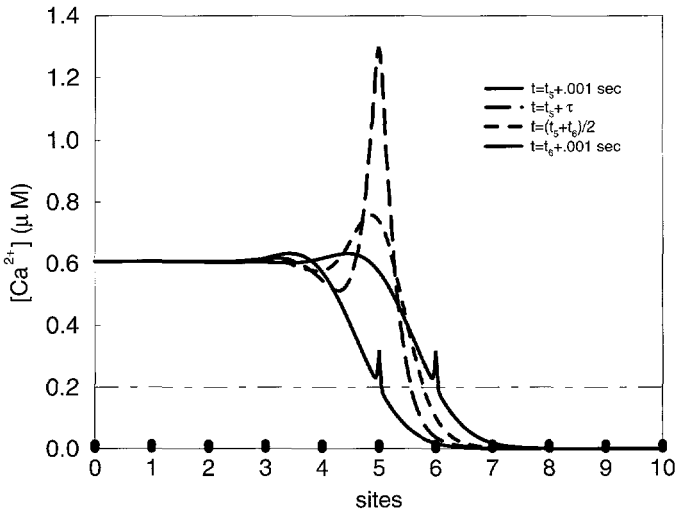
Brydon, D., and J.E. Pearson, "Solving Stiff Differential Equations with the Method of Patches" (submitted to *J. Comput. Phys.*).

Keizer, J., et al., "Saltatory Propagation of Ca^{2+} Waves by Ca^{2+} Sparks" (submitted to *Proc. Natl. Acad. Sci. U.S.A.*).

Wagner, J., et al., "Simulation of the Fertilization Ca^{2+} Wave in Xenopus Laevis Eggs" (to be published in *J. Cell Calcium*).



Skeletal model of calcium-induced calcium release: log-log plot of the velocity of the the calcium wave as a function of the diffusion coefficient, D. Dotted curves indicate the diffusion coefficient's effect on slope: for small D (the discrete limit), velocity scales linearly with D; for large D (the continuum limit), velocity scales with the square root of D.



The skeletal model of calcium-induced calcium release consists of release sites that are embedded in the membrane compartment. The sites are separated by a distance of 3.87 μm. When the concentration at a site reaches a threshold value (0.2 μM, indicated by the horizontal dashed line), the site begins to release calcium. Each site releases a total of 3.5 $\times$ 10^{-20} moles of calcium during a time τ = 0.05 s. The calcium released diffuses between sites. The figure shows the calcium concentration as a function of position at four different times. The solid curve with the short spike above site 5 corresponds to a time 0.001 s after site 5 reaches threshold. The long-dash curve corresponds to the moment that site 5 stops releasing calcium. The short-dash curve corresponds to the time midway between that at which sites 5 and 6 reach threshold. Finally, the solid curve with the short spike above site 6 corresponds to a time 0.001 s after site 6 reaches threshold. This figure illustrates the model for when the discrete nature of the release sites dominates the behavior, which occurs in immature xenopus eggs. By contrast, during the fertilization wave in mature xenopus eggs, the wave is continuous and the discrete nature of the release sites cannot be resolved.

The Role of Low-Frequency Collective Modes in Biochemical Function: Ligand Binding and Cooperativity in Calcium-Binding Proteins

Jill Trewhella

The focus of this project, low-frequency modes involving the concerted motion of groups of atoms, is a topic that has been widely discussed as a means for direct control of biochemical processes. However, the control of a biochemical process via these types of motions has yet to be clearly demonstrated. Our approach to achieving this end involves using a unique combination of isotope labeling with nuclear magnetic resonance (NMR) relaxation and vibrational spectroscopy to probe dynamic fluctuations within individual domains of the Ca^{2+} binding of the protein calmodulin. The experiments are aimed at obtaining quantitative information on the dynamic fluctuations speculated to be important in target enzyme binding and Ca^{2+} binding and on the manner in which dynamic fluctuations influence Ca^{2+} binding by giving rise to differences between individual Ca^{2+}-binding sites and domains.

In this first year of our research, we made progress in preparing the samples needed for the spectroscopy experiments, and we began the NMR experiments. We constructed and used a bacterial expression system to produce the N- and C-terminal domains of calmodulin, each of which binds two calcium ions. We successfully purified the N-terminal domain with a combined ^{15}N and 75% ^{2}H labeling pattern to be able to assign the NMR spectrum. We also prepared the N-terminal domain with an alternate ^{13}C labeling pattern needed for the relaxation measurements. We achieved this labeling pattern using the labeled glycerol we had synthesized and an *E. coli* strain we had constructed that bears metabolic mutations required to generate the desired labeling pattern. We have acquired NMR spectra on the N-terminal fragment to verify the ^{13}C labeling pattern and to make spectral assignments and determine structural constraints.

Determining the Role of the Telomere on the Severity of Birth Defects

Michael Altherr

There are a variety of birth defects associated with the loss of genetic material near the ends of chromosomes. At least one-third of human chromosomes can have terminal deletions that result in live births but that lead to significant clinical abnormalities. Terminal deletions can arise by a variety of mechanisms, including the clastogenic effects of ionizing radiation. It is clear that chromosome stability is severely impaired if the terminal structure, the telomere, is missing. In fact, complete chromosomal loss (a condition incompatible with life in humans) has been observed in experimental organisms when the telomere is removed or disrupted. The telomere is a unique chromosomal element that has been demonstrated to generate novel structures, to initiate DNA replication at the ends of chromosomes, and to affect the expression of genes in its proximity.

The goal of this project was to evaluate the effect of the telomere on gene expression in human cells derived from patients with a birth defect resulting from a terminal deletion. We have utilized cell lines derived from patients with a serious birth defect called the Wolf Hirschhorn syndrome (WHS). One of the hallmark diagnostic criteria for WHS is the deletion of the terminal segment of human chromosome 4, a condition referred to as $4p^-$. In the course of these studies, we have developed a series of landmark cosmid probes (see first diagram) that are currently being employed by researchers around the world to evaluate suspected WHS patients. Cosmids are relatively small segments (45,000 base pairs) of DNA derived primarily from the human genome but capable of being propagated in bacteria to generate sufficient mass for subsequent studies. Random cosmid clones organized into arrays of DNA segments representing the position of the clones in the genome are referred to as contigs. We have also identified a series of novel genes (see second diagram), characterized their pattern of expression, and generated a variety of reagents to further these studies. Although the primary product of gene expression is RNA, this product is represented by complementary DNA (cDNA) clones derived from cellular RNA.

In an effort to define genes whose expression might be altered by a change in the position of the telomere, we utilized cell lines to determine the smallest region of overlap that would, therefore, define the critical region (summarized by the patient cell lines represented in the lower half of the first diagram). The WHS critical region (WHSCR) is defined by this smallest region of overlap and is likely to contain genes critical to normal human development. Using the technique of fluorescent in situ hybridization (FISH) to define the smallest region of overlap, we analyzed cell lines derived from WHS patients. We used a small group of cosmids, the landmark set, in the primary analysis. Once we had defined the approximate breakpoint, we could identify the specific genomic segment containing the deletion breakpoint by using the individual members of the contig between the landmark clones (called second-tier cosmids). We are studying genes identified in this segment using a variety of molecular genetic techniques to determine the physiological role of these genes in human biology. Finally, we hope that these studies will then allow us to evaluate how the alteration in telomeric position modulates the expression of these genes.

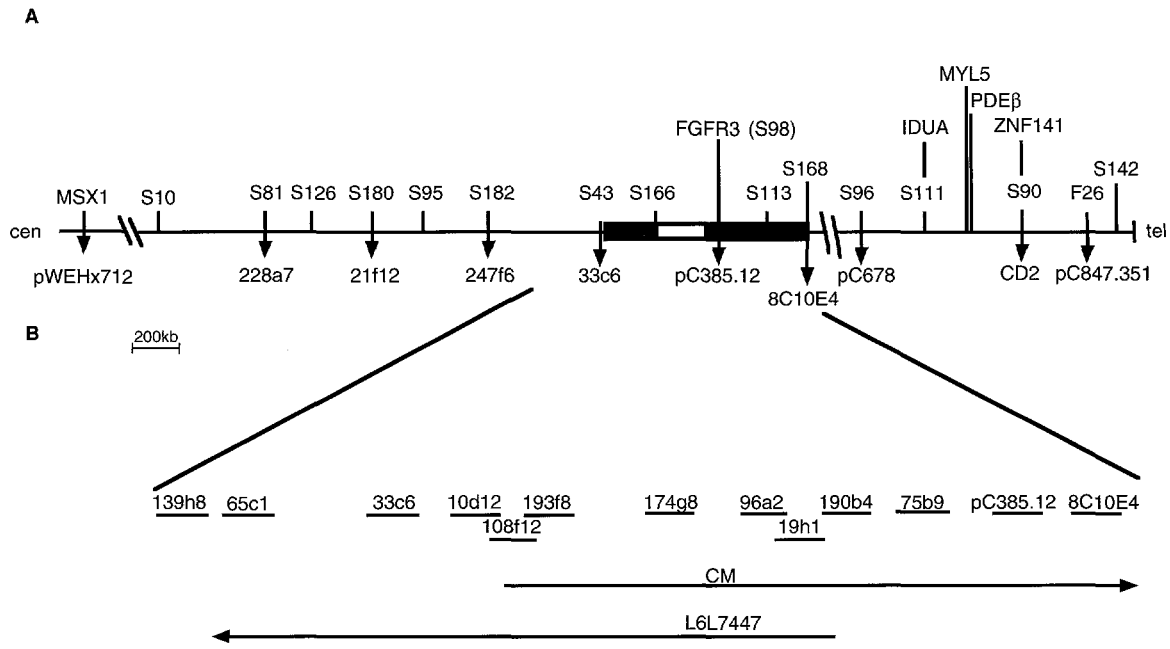
Publications

Altherr, M.R., et al., "Delimiting the Wolf-Hirschhorn Syndrome Critical Region to 750 Kilobase Pairs," *Am. J. Med. Genet.* **71**, 47 (1997).

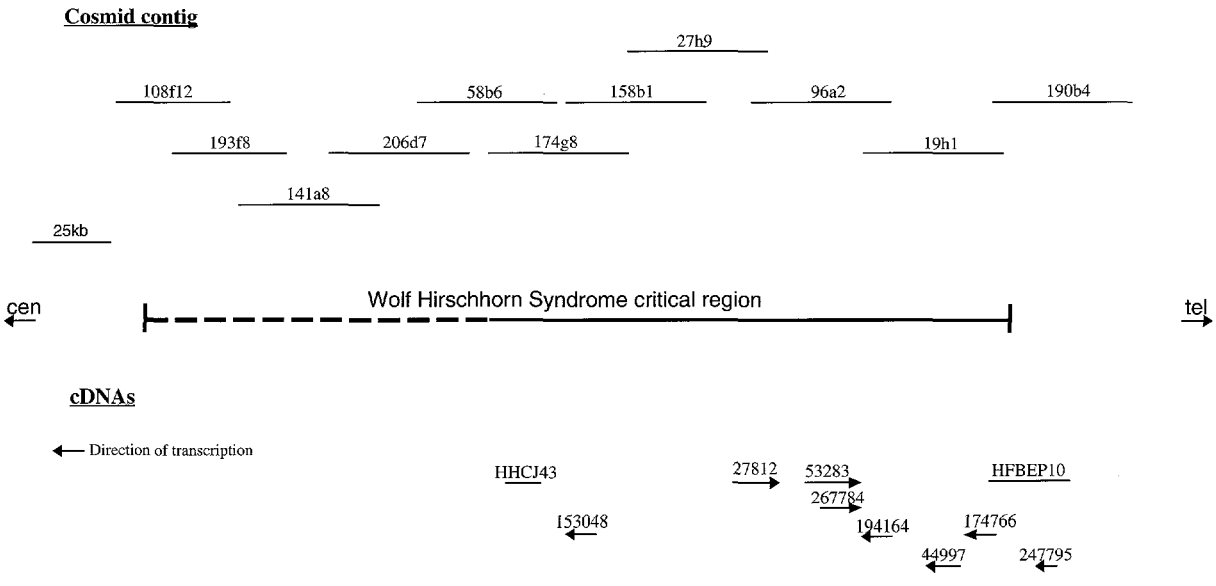
Perez-Castro, A.V., et al., "Genomic Organization of the Human Fibroblast Growth Factor Receptor 3 (*FGFR3*) Gene and Comparative Sequence Analysis with the Mouse *fgfr3* Gene," *Genomics* **41**, 10 (1997).

Wright, T.J., et al., "A Transcript Map of the Newly Defined 165 kb Wolf-Hirschhorn Syndrome Critical Region," *Hum. Mol. Genet.* **6**, 317 (1997).

Wright, T.J., et al., "Wolf-Hirschhorn and Pitt-Rogers-Danks Syndromes Caused by Overlapping 4p Deletions" (to be published in *Am. J. Med. Genet.*).



(a) Physical map of 4p16, the distal segment of the short arm of human chromosome 4. The genetic loci utilized in this study and in previous studies are shown above the line, and the probes used in this study are shown below the line. The distance from locus D4S81 ("S81" on the figure) to the telomere ("tel") is approximately 4.5 megabases. The genetic distance between loci MSX1 and D4S10 ("S10") is 3 centiMorgan, and the gap in the cloned contig between loci D4S168 ("S168") and D4S96 ("S96") is approximately 60 kilobases. The segment between the pairs of diagonal lines represents a contig of overlapping clones. The WHSCR defined by previous studies is shown by a solid box, and the new WHSCR defined by patients included in our studies is shown by an open box (within solid box). The cross-hatched portion of the open box is the area that can be excluded when considering the $4p^-$ patient. (b) Second-tier cosmids used in our FISH analysis. The probes used in the second round of the FISH analysis are shown. The results of the analysis of patient cell lines defining the WHSCR are shown. The lowest two lines indicate the position of the deletion, and the arrows indicate the direction in which the deletion continues; CM continues toward the telomere, and LGL7447 continues toward the centromere.



Transcription map of the WHSCR. A cosmid contig of the WHSCR and the region immediately flanking it are shown. The broken line near the center of the diagram represents the region that can be excluded when considering the $4p^-$ patient. The solid line indicates the position of the new WHSCR. The position and direction of transcription, where known, of cDNAs is shown below the WHSCR.

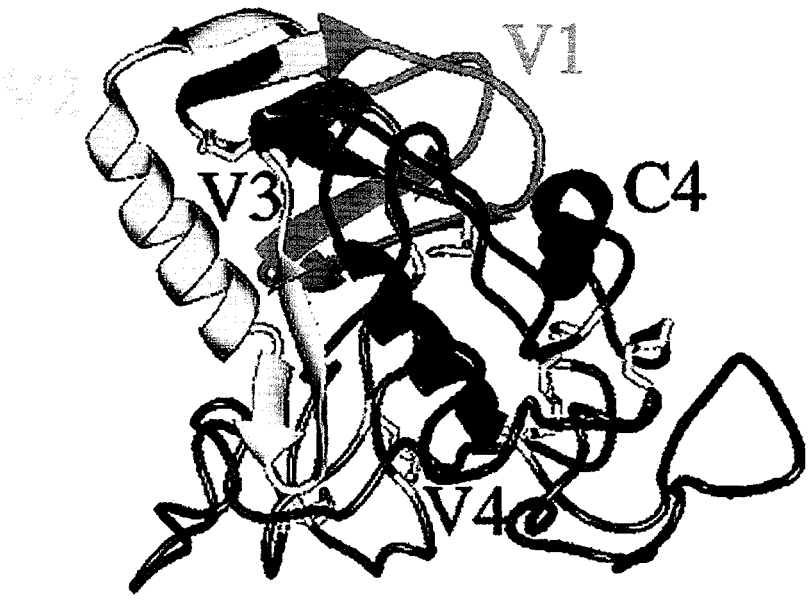
Structure and Thermodynamics of Surface Recognition

Goutam Gupta

We are exploring the design of HIV-1 antigens that might ultimately become vaccines. The interaction of the HIV-1 surface glycoprotein, gp120, with the host T cell as the macrophage defines the first step in the pathogenesis of the virus. Therefore, identifying the sites on gp120 that participate in this interaction is crucial to developing protective immunity against the virus. We found that gp120 contains several constant (C) and variable (V) loops interdigitated as C1-V1-V2-C2-V3-C3-V4-C4-V5-C5. Previously, we focused on developing HIV-1 antigens based on the V3 loop that participates in fusion and tropism.

We extended those efforts to designing HIV-1 antigens that contain two noncontiguous loops, such as V3 and C4. If the V3 and C4 loops in these HIV-1 antigens maintain the same structures and interactions that are present in the native gp120, they have potential as effective HIV-1 vaccines.

The first step in this method is to characterize the structures and interactions of various loops in the native gp120. We successfully developed a molecular modeling method for arriving at a model of the native gp120 (see the figure). The model is in agreement with (a) the surface exposure of various eptioptes obtained from immunochemical assays and (b) the loop-loop interaction data obtained from fusion, replication competency, and tropism assays. We believe that this gp120 model will help us design bispecific HIV-1 antigens that contain two gp120 loops.



Ribbon diagram of the gp120 model that contains the V1-V2-C2-V3-C3-V4-C4 segment. This view shows all the loops in an uncluttered way. The C4 loop (the primary attachment site for the CD4 receptor on T cells and macrophages) interacts with the V3 loop even though more than 100 amino acids separate them. In our bispecific HIV-1 antigen, we connect the V3 and C4 by a synthetic linker of 10 amino acids so that both the loops maintain their native structures and interactions.

Substrate-Dependent Cell Cycle Disturbances in Response to Ionizing Radiation

Donna Gadbois

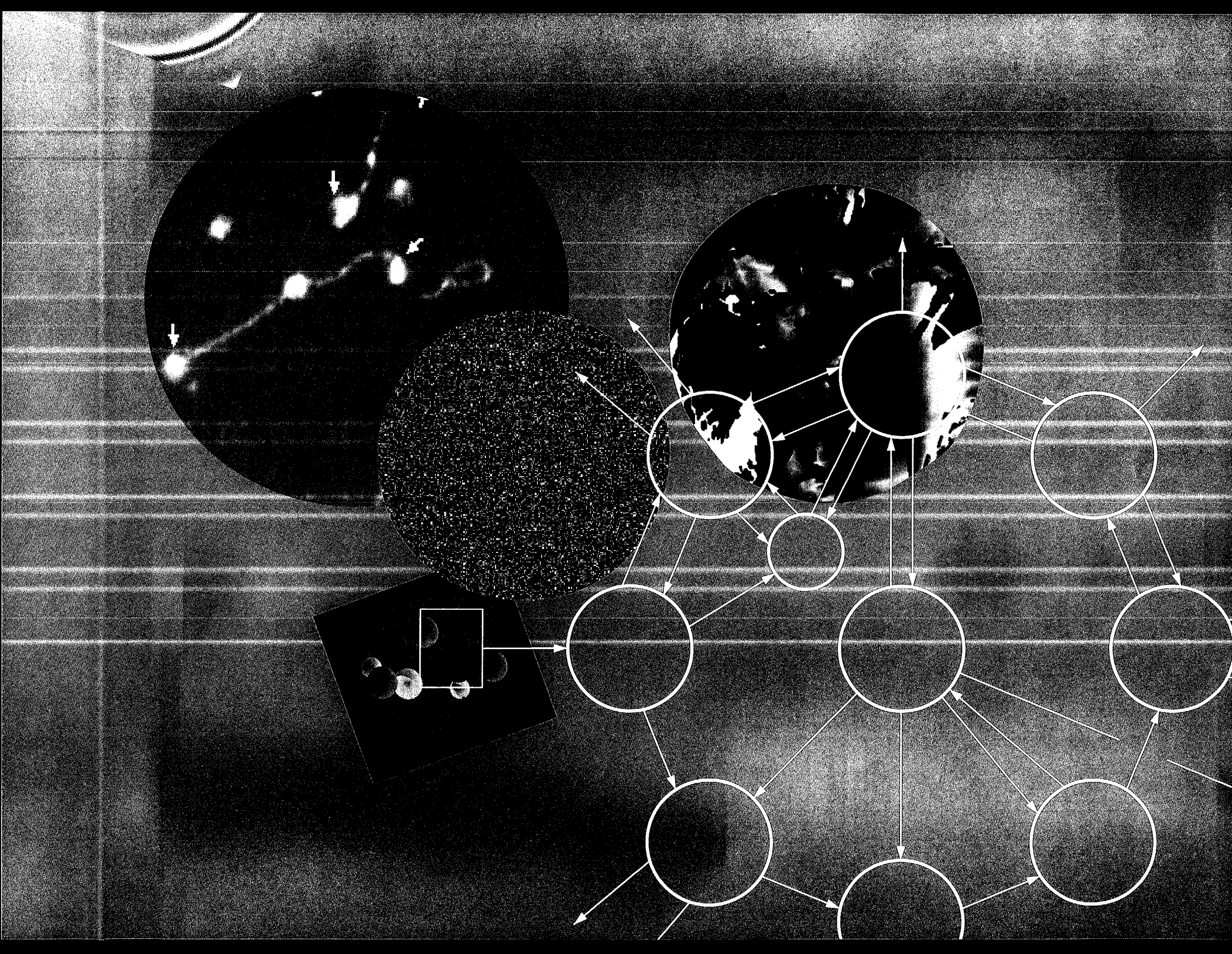
Radiation causes normal cells to delay at specific phases of the cell cycle in order to repair damaged DNA, thus preventing gene alterations that can lead to cancer. Historically, radiation cell-cycle delays in anchorage-dependent fibroblasts have been studied in cells maintained on polystyrene culture plates, even though polystyrene markedly deviates from the extracellular matrix to which such cells are juxtaposed in vivo. The goal of our project is to study the effect of the extracellular matrix on radiation cell-cycle delays.

The mammalian cell cycle is divided into the G1, G2, S, and M phases. The G1 and G2 phases are those in which proliferation control mechanisms are in place, the S phase is the DNA synthesis period, and the M phase is the one during which cells divide. We have shown that, when cells are maintained on polystyrene culture plates, radiation causes G1 and G2 phase delays in fibroblasts and that virtually all cells ultimately arrest in G1 and fail to reenter the cell cycle. We have shown that the terminal radiation arrest is related to the polystyrene substrate and that the radiation arrest can be reversed in a manner suggestive of the involvement of altered cell surface to receptor interactions with the extracellular substrate.

We have also shown that the extent of the radiation delays is markedly influenced by the type of substrate on which the cells are maintained. This finding may suggest that previous cell-cycle analyses made with radiation-exposed, anchorage-dependent cells cultured conventionally have led to erroneous conclusions that are not predictive of responses in vivo. Further, our findings suggest that a mechanistic understanding of cell-substrate interactions, particularly fibroblast-polystyrene interactions, could lead to new therapeutic measures for abating the proliferative pulmonary fibrotic response induced by treating lung tumors with radiation.

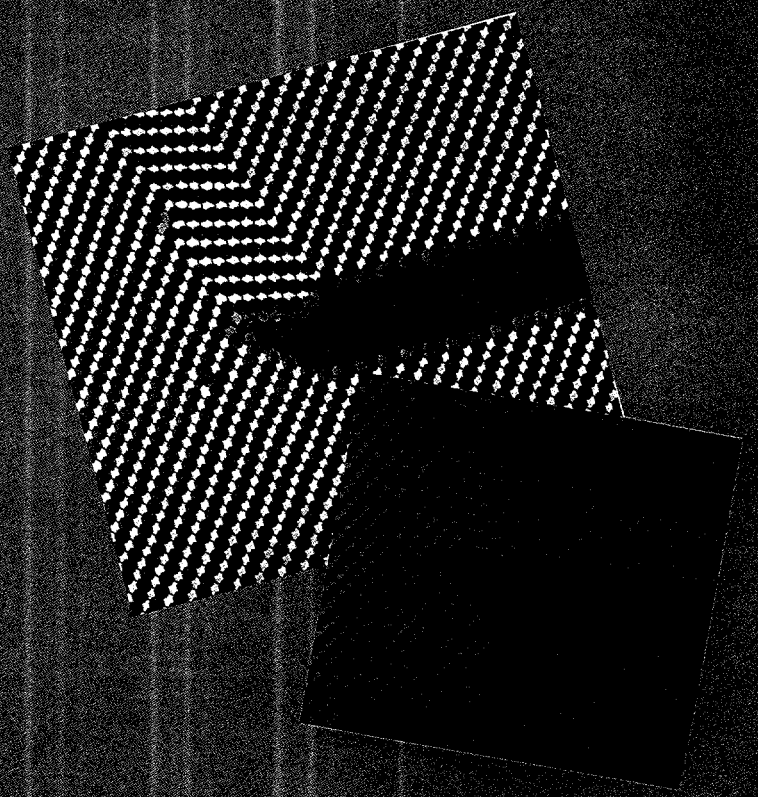
Publications

Gadbois, D.M., et al., "Control of Radiation-Induced G1 Arrest by Cell-Substratum Interactions," *Cancer Res.* **57**, 1151 (1997).



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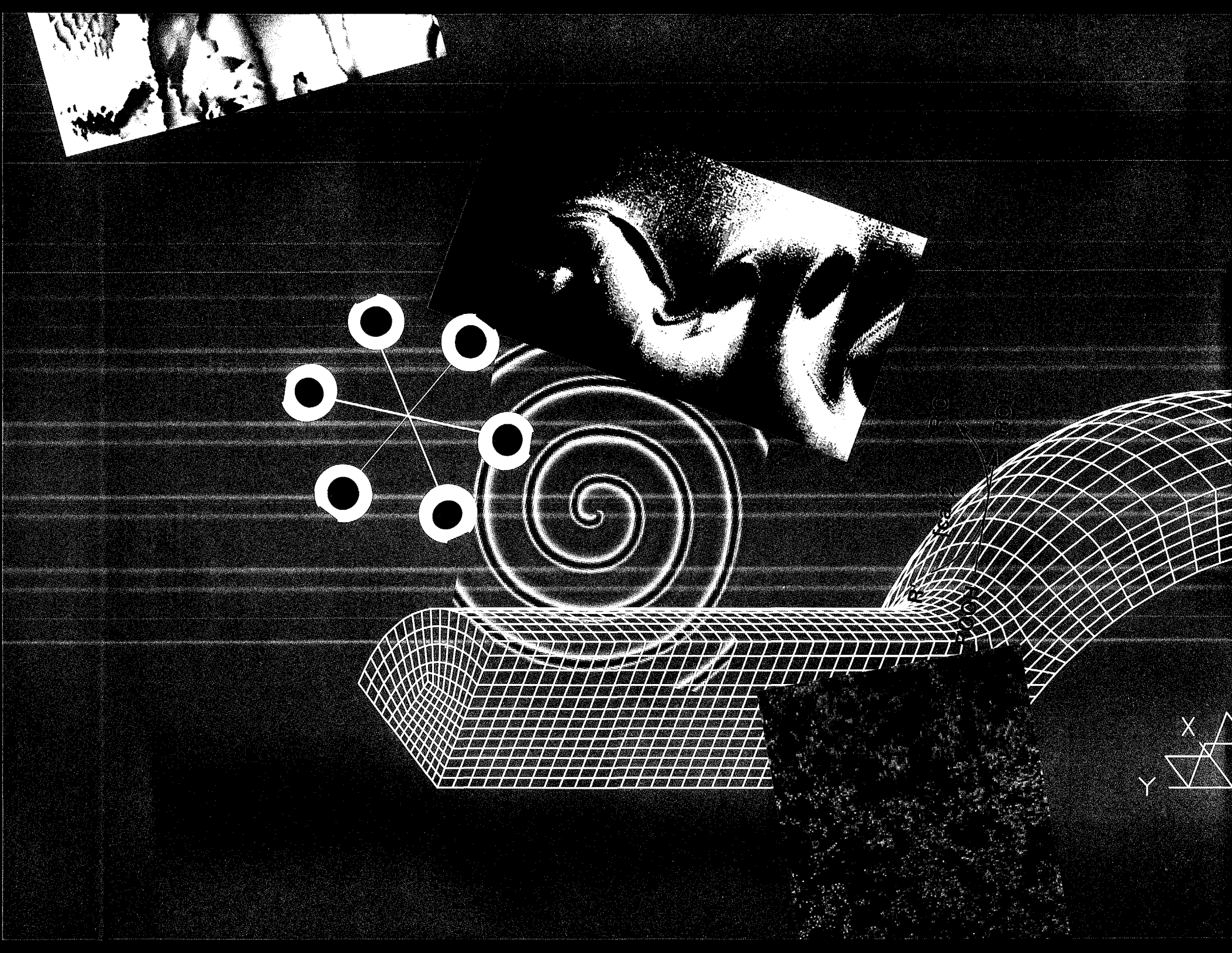
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