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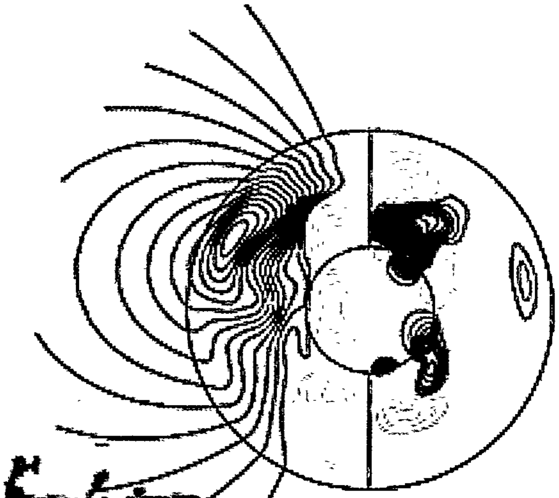
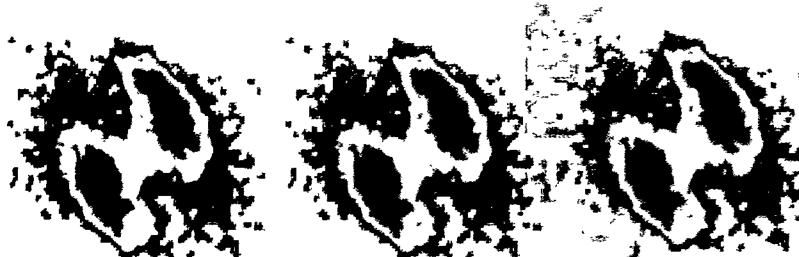
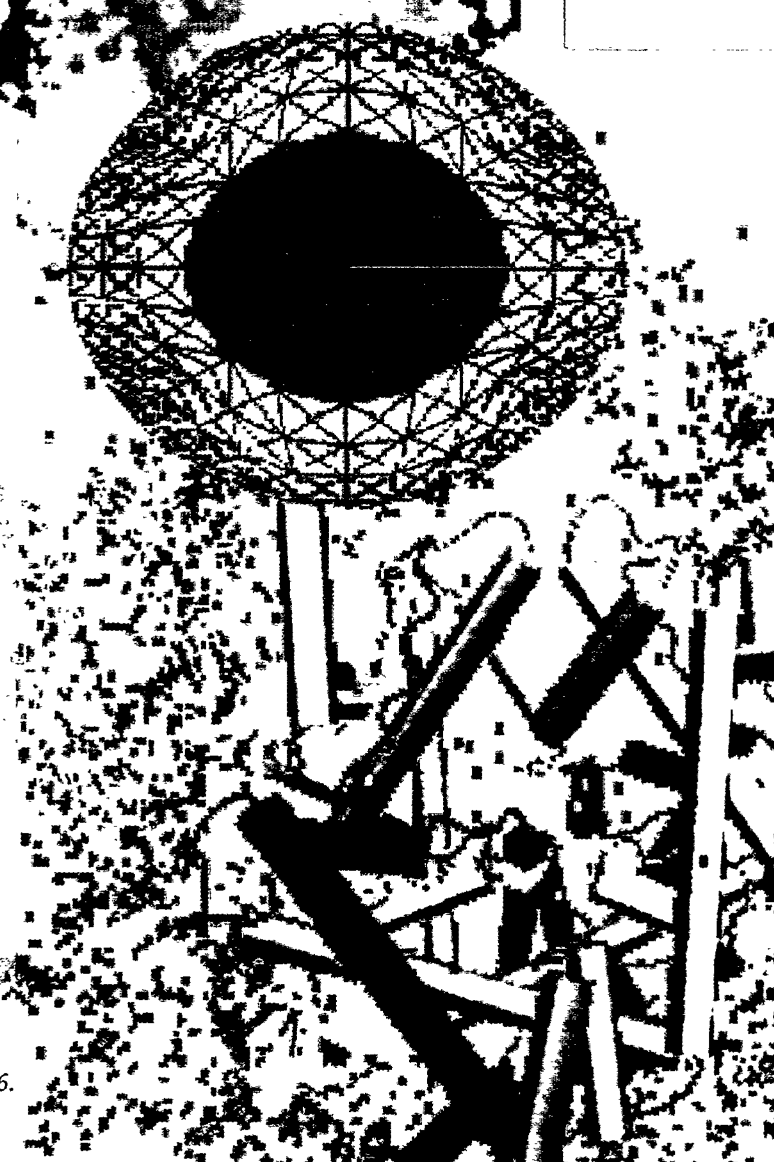
1998 LDRD



Laboratory Directed Research and Development





Los Alamos
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ABSTRACT

This is the FY 1998 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, molecular, optical, and plasma physics, 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.

Color computer simulations greatly aid many LDRD projects. The images on the front and back cover are from projects summarized in this report; the images on the section dividers are from projects described in the respective sections.

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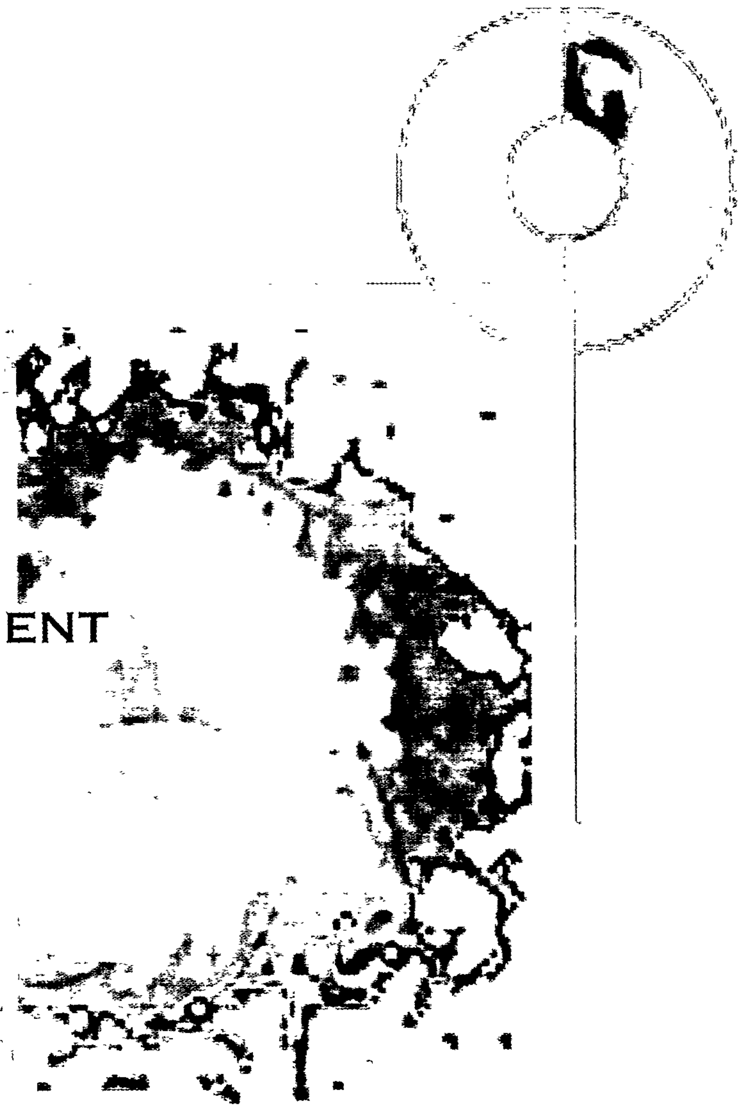

Laboratory Directed Research and Development

FY1998 Progress Report

Compiled and Edited by
John Vigil and Kyle Wheeler

Published by Los Alamos National Laboratory

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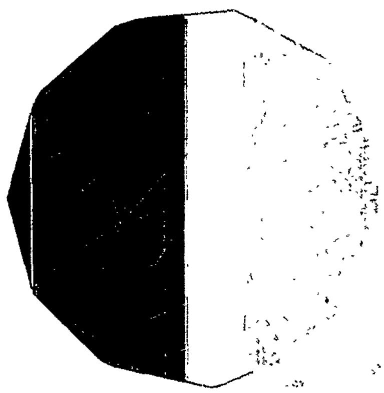

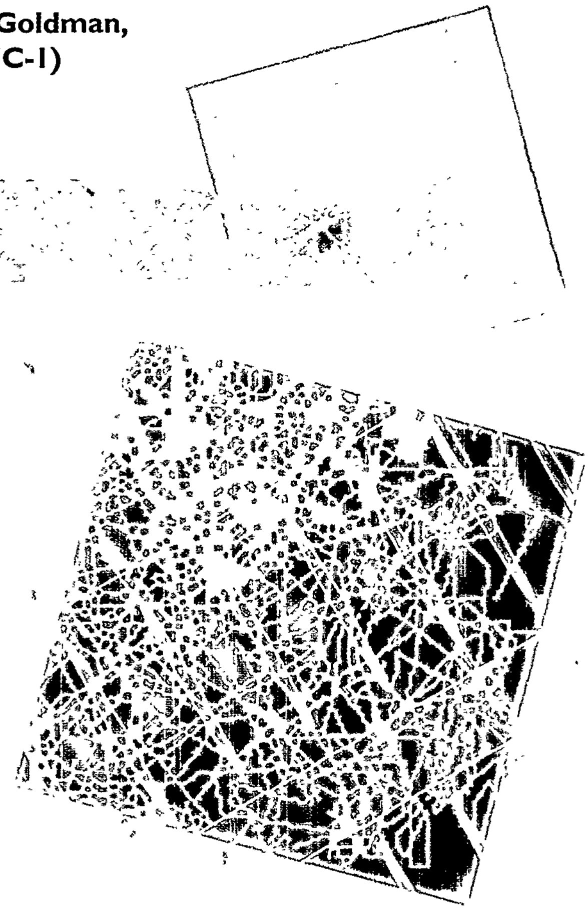

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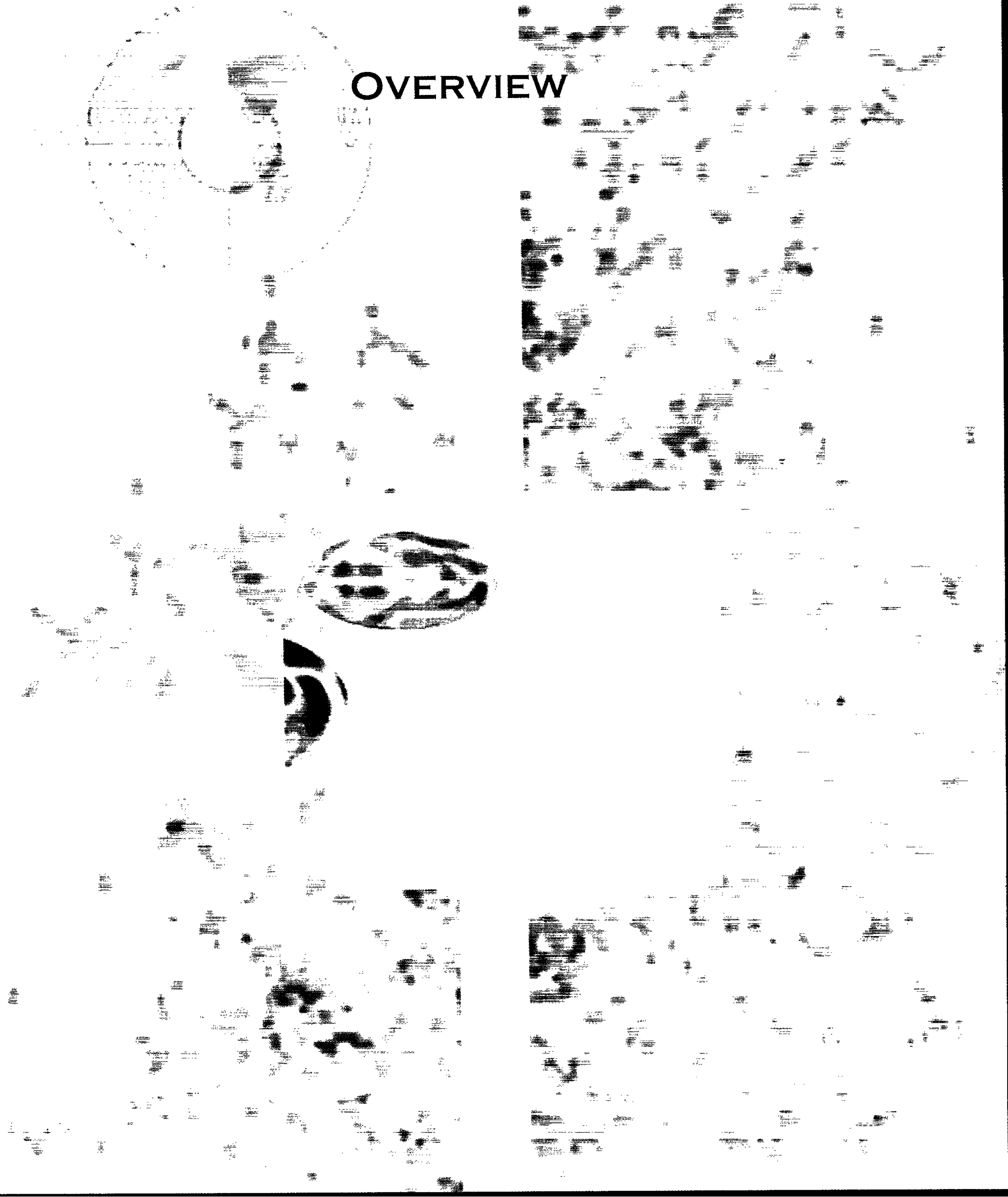
OVERVIEW



Overview

LDRD Program Overview—FY 1998

Ed Heighway and John Vigil

The Laboratory Directed Research and Development (LDRD) Program is authorized by Congress as a means for the Laboratory to maintain its scientific and technological vitality. Innovative research and development (R&D) projects are internally proposed and selected and have potential to extend the Laboratory's science and technology capabilities. There are three ways in which LDRD extends those capabilities in support of the Laboratory's missions. First, it 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 areas of expertise needed to fulfill the missions, perhaps adding multidisciplinary approaches that provide new insight. Third, it may broaden the fundamental science and technology base in areas that underpin the Laboratory's ability to execute its missions.

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. LDRD research 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," which includes four major components: (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. A breakdown of the Laboratory's FY 1998 funding by major program sponsor is shown in Fig. 1, and a table of all LDRD projects as they relate to these sponsors is provided at the end of this report.

LDRD Program Structure

LDRD is a formally managed program that imposes accountability in the processes for selecting, executing, and reporting on projects. Peers or Laboratory technical managers select all projects through competition and review. Innovation and scientific excellence are key selection criteria, and all projects must be in areas of science and technology that support the Laboratory's missions.

In FY 1998 the LDRD Program at Los Alamos had three components—Competency Development (CD), Program Development (PD), and Individual Projects (IP)—each with differing goals. The CD component invests in relatively large multidisciplinary projects that tackle significant mission-related problems from several viewpoints, often combining theory and experiment, and drawing on scientists from several technical divisions. The PD component is intended to invest in nearer-term R&D that requires proof-of-principle, feasibility, or other confidence-building demonstrations for potential program 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.

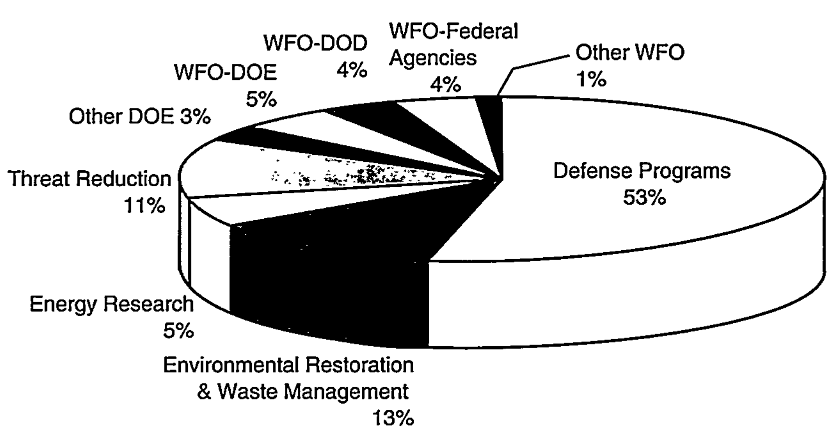


Figure 1. Funding for major programs at Los Alamos National Laboratory during FY 1998 (WFO means work for others).

In FY 1998 the LDRD Program funded 300 projects for a total expenditure of $64.3 million. This sum represented 5.6% of the Laboratory's operating budget. The funding distribution among the three LDRD components is shown in Fig. 2.

Competency Development. This component funds research related to the core 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 draw on the resources of several Laboratory divisions and have clear institutional value beyond the strategic interest of any single division. They 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 1998, 77 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 program offices are listed below:

- 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 and Technology Base and Energy Research Programs

In FY 1998, 72 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 the nine technical categories), with the highest ranked and most relevant ones being funded. In alphabetical order, the nine technical categories are as follows:

- Atomic, Molecular, Optical and Plasma Physics, Fluids and 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 1998, 151 IP projects were funded.

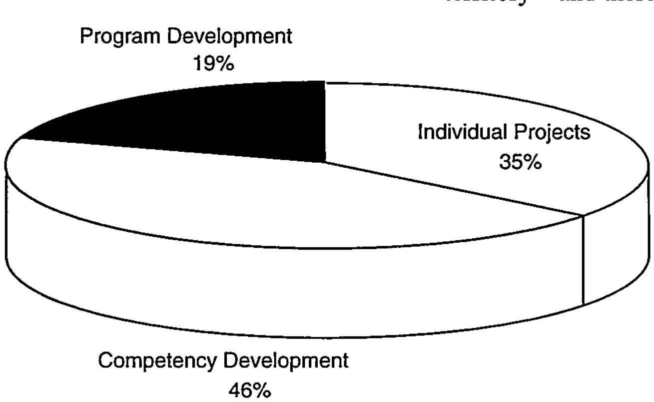


Figure 2. Distribution of FY 1998 funding among the three LDRD program components.

Quality of LDRD Research

The results of the FY 1998 LDRD Program are impressive. The program continues to produce a large volume of high-quality scientific output. LDRD-sponsored research represents approximately 6% of the Laboratory's budget, yet it continues to produce approximately 30% of the Laboratory's total output of refereed publications. Two of the Laboratory's four R&D 100 awards received in 1998 were attributable to LDRD investments. An additional testimony to the state-of-the-art nature of LDRD projects is the very significant contribution to the intellectual property of the Laboratory in FY 1998. Of the 41 patents issued to the Laboratory, fully 20 of these (49%) sprang from work either totally or partially supported by LDRD. The LDRD Program, therefore, continues to be an important vehicle for keeping the scientific staff at the forefront in their fields and keeping the Laboratory visible in the broader science community, particularly academia, from which we draw most of our recruits.

As described above, the LDRD contribution to the Laboratory's scientific publications, citations, and patents is a significant factor in the

Laboratory's ability to attract the innovators and scientific leaders of the future. The postdoctoral program remains an important vehicle for recruitment: about one-half of all postdoctoral students contribute significantly to LDRD projects and about one-fifth of all postdoctoral students ultimately become full-time employees. LDRD is an essential activity in attracting the highest-caliber scientists and engineers to support our national security missions.

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, counter-proliferation, 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.

Advanced X-Ray Radiography. Advances in the traditional x-ray approach to hydrodynamic testing are needed to diagnose a complete hydrodynamic test from many views and at many times throughout the critical phase of the implosion to create high-resolution, three-dimensional (3-D), time-dependent images of the entire event. LDRD researchers are investigating the fundamentals of algebraic reconstruction of time-dependent 3-D images from multi-view x-ray data and the technology limits for use of x-rays in meeting long-term stockpile stewardship requirements. Additional areas of investigation include diamond photocathode technology, high-current linear-induction accelerator technology, target bremsstrahlung converters, and solid-state detector technology capable of >200-MHz frame rate with frame storage capability in the thousands.

The researchers have identified thick-object scattering as the ultimate limit on x-ray hydro-radiography, and significant progress has been made in determining the minimum number of views needed for accurate reconstructions of hydrodynamic tests. Small area samples of boron-doped, hydrogen-terminated, 111-crystalline diamond have been fabricated and tested. These have good efficiencies and show no deterioration of the average bulk time-independent properties. Simulations of emittance growth resulting from high-current beam profiles interacting with nonlinear self-fields and electromagnetic fields induced in accelerator structures were achieved. Simulations on emittance growth through an entire induction accelerator structure have been started, and researchers are finding that high-current-induced wake fields couple to the outer wings of the beam current profile. They have extended hydrodynamic simulations to predict behavior of high-density plasma erupting from a bremsstrahlung target due to the intense electron-beam density. This LDRD effort has already influenced the thinking on the second axis of the Dual Axis Radiograph Hydrotest Facility under construction at Los Alamos.

High-Resolution Proton Radiography. LDRD researchers have continued to make excellent progress using high-energy protons to produce high-resolution radiographs of events that take place very rapidly in high-density materials. This new diagnostic capability allows detailed multi-image examination and verification of the behavior of components as they are being compressed during high-explosive, nonnuclear detonation, thus providing additional precision to the assessment of stockpile component safety and reliability. Proton radiography supplements x-ray radiography, which has limitations that come from source size, detector blur, and scattered background, and it has the potential to meet stockpile stewardship needs such as multiple axes for 3-D effects, multiple frames to capture time dependence, higher resolution, and better analysis and tomography.

LDRD research has been instrumental in all early concept validations including high-energy feasibility (static) experiments at the Alternating Gradient Synchrotron at Brookhaven National Laboratory, first proof-of-principle dynamic experiments at the Los Alamos Neutron Science Center (LANSCE), simulation and modeling to demonstrate basic concepts, and detector concept development. The work is clearly relevant to the next-generation Advanced Hydrotest Facility. This year LDRD researchers designed, constructed, and tested a three-lens proton radiography system in beam line C at LANSCE, and it is currently being used to perform experiments in support of a stockpile stewardship program. Quantitative measurements of density changes in exploding systems, at the several percent level, have been demonstrated.

Multiscale Science. Multiscale science refers to the analysis of problems whose dynamics are determined by the interaction of physical processes that occur on many, often widely different, scales of length and time. Such problems, which have consistently defied solution, are important throughout science and technology and are at the scientific core of nuclear weapons. The difficulty is that current computational capabilities are not adequate to allow the smallest scale (microphysical) and largest scale (macrophysical) processes to be simultaneously resolved. LDRD researchers are developing and applying rapidly emerging multiscale science to fluid/materials mixing and materials characterization, which are key issues in nuclear weapons and other applications. The specific focus is on the stockpile stewardship issue of assessing performance of a nuclear device in the absence of full-scale testing, including assessing the performance of components that have aged, been remanufactured, or no longer meet original design specifications. The objectives are to create validated, predictive models of fluid mix and material behavior under shock compression based on microphysical descriptions of the key processes and to ensure that important features of the improved models are implemented in the large-scale design and assessment codes used in the weapons program.

This project is making important progress on difficult and long-standing problems at the heart of the nuclear weapons mission. A recent accomplishment was development of a new two-phase model of fluid mix that has been used in an extensive set of full-scale numerical simulations. These simulations have confirmed a fundamental modeling assumption: a two-phase model with distinct pressures in each phase is required to correctly describe mixing zone physics. Another recent accomplishment was development of a discrete-element computer code for metal grains that includes models for contact forces at grain boundaries and provides a quantitative tool for the study of surface ejecta. Another area where important progress has been made is in the quantitative analysis of friction, based on molecular dynamics simulations. A notable result has been simulation of friction at high-velocity, dry-metal interfaces, which has revealed a transition in the friction coefficient above a critical velocity. Yet another recent accomplishment is development of a much-improved shock-front simulator that eliminates numerical diffusion, captures the shock, and handles material interfaces. The new simulator is being applied to shock front experiments at Lawrence Livermore National Laboratory.

Reaction Processes in Energetic Materials. Chemistry studies at Los Alamos and numerous other institutions have elucidated many of the elementary reaction pathways in energetic materials, especially in the gas phase. One critical fundamental piece of this problem remains to be addressed: the condensed phase chemistry. Condensed phase kinetics is widely recognized as the weakest link in our understanding of the decomposition of energetic materials. LDRD researchers are developing a competency to predict reaction processes in energetic materials—in particular, high-melting explosive (HMX)—using detailed and reduced chemical kinetics schemes with the ultimate goal of developing simple, yet rigorous, models that can eventually be used in engineering calculations for stockpile stewardship.

This year researchers have adapted a detailed chemistry code to simulate steady and unsteady burning of HMX and have developed a novel nonlinear optical technique to study the kinetics of a solid-solid phase transition that occurs in heated HMX. This technique constitutes a fundamentally new probe to study the dynamics of energetic material decomposition, ignition, and combustion, and it may have application to optical integrated devices for frequency conversion and information encoding. The researchers have designed, built, and performed initial laser ignition and deflagration experiments, and they have shown for the first time that solid-solid phase transformations occur on combustion time scales.

Science of Polymer-Based Materials and Plutonium Aging. Aging of polymeric materials subjected to various environments could be a limiting feature in the useful lifetimes of many DOE and industrial systems. LDRD researchers are developing a suite of experimental and modeling capabilities with which to understand and predict the changes in physical properties of polymer-based materials over extended periods of time. An important objective is to understand and overcome the pitfalls of standard accelerated aging studies, which are often based on empirical extrapolations from a single class of experiments and which often do not account for differences in the physical state of the polymer at different temperatures. The goal is to move toward more mechanistic methods founded on understanding of changes at the molecular level. Toward this end, the researchers are investigating Estane (an adhesive or binder material) and polydimethylsiloxane (used for adhesives, foams, or pads).

The researchers have found that the hydrolytic degradation of Estane by accelerated aging proceeds in three stages. They have assigned the vibrational spectra of Estane, particularly those bands that define the molecular interactions and the level of phase separation, through a combined experimental and modeling approach. The results are being used to characterize the physical state of the polymer at the accelerated aging temperatures compared to the state at ambient conditions. They have also developed a three-dimensional, coarse-grained bond-fluctuation code and a mesoscopic entanglement theory approach, and both have been validated with data for simple homopolymer systems. This work will ultimately support stockpile stewardship by providing new lifetime prediction techniques for polymers.

In a separate project, researchers are using a novel new technique to study aging in plutonium-239. They blend gallium-stabilized delta-phase plutonium-239 with 5% plutonium-238 to accelerate the aging process at the rate of 15 years to 1 year. A suite of sensitive, fundamental measurement techniques will be used to characterize the doped material at periodic intervals to track subtle changes as aging progresses. Samples will be stored under carefully controlled conditions to simulate realistic stockpile storage environments, accounting for self-heating and thermally sensitive aging effects. Understanding how aging manifests itself in this material will elucidate similar behavior in stockpile components. Among other accomplishments this year, researchers completed an aging chamber for use with a dry bath incubator and completed long-term tests of temperature stability.

Endoscopic Imaging and Spectroscopy of Pit Interiors. LDRD researchers are developing and demonstrating a method that will allow the inner surface of intact pits to be examined and interrogated using a fiber-optic probe inserted into the fill tube of the pit. They have fabricated a fiber-optic probe containing an illumination bundle and an imaging bundle capable of 180-degree bending motion over the appropriate length for accessing all of the interior regions of a pit. The fiber is coupled with a mechanical stage that precisely controls the rotational angle, vertical position, and the overall bending of the fiber. A color camera is used to obtain the images, and a small computer controls the motion and records the images.

The researchers have observed the weld line and machining grooves on the inner surface of a pit surrogate using this apparatus. Their efforts thus far have demonstrated the capability of imaging over 90% of the interior surface of a pit surrogate. They have also addressed the ability to spectroscopically characterize plutonium surfaces using fiber optics. In particular, they have demonstrated laser-induced breakdown spectroscopy of plutonium metal and quantitative determination of minor constituents using fiber optics. This LDRD effort is providing an innovative new capability for stockpile surveillance activities.

Electronic Structure of Plutonium and Related Compounds. This work is extending a competency of major importance to the Laboratory by enhancing the fundamental understanding of f-electron materials such as plutonium and other actinides. A better understanding of f-electron materials is needed to help address stockpile stewardship issues such as plutonium aging and phase transitions. LDRD researchers are using the capabilities of the Los Alamos National High Magnetic Field Laboratory to measure the electronic structure of plutonium and related compounds via the de Haas-van Alphen (dHvA) effect. They are currently emphasizing uranium compounds and are determining the Fermi surface topologies, as well as effective masses of the orbits as a function of magnetic field, to determine the strength of electron-electron correlations. They are using constrained, low-density approximation calculations for comparison with experiment. Because many-body phenomena involve femtosecond dynamics, they also apply ultra-fast optoelectronics to the measurement of nonequilibrium properties of correlated f- and d-electrons in condensed matter. This allows investigation of microscopic dynamical phenomena on time scales of 10^{-11} to 10^{-14} seconds. The purpose is to reveal the role of many-body interactions in 5f-, 4f-, and 3d-electron systems. The magnetic field serves as an independent control variable with which to tune the energy scale of the many-body phenomena under study.

This year researchers have observed the dHvA effect in seven uranium heavy fermions, with four of these displaying magnetic transitions in high fields. Among the many highlights reported this year is a complete transformation of the Fermi surface in UGa_3 at the transition, while UCd_{11}, the heaviest mass uranium compound ever to yield dHvA oscillations, shows no topology change at two transitions.

Quantum Cryptography. Cryptanalysis techniques and algorithms are advancing rapidly. By the start of the twenty-first century, new encryption technologies will be needed to ensure secure communications to satellites. LDRD researchers are developing quantum cryptography to provide absolutely secure encryption of communications to low-earth orbiting satellites. They are developing and demonstrating the cryptographic technology to a stage where it can be feasibly incorporated into new satellites.

This year researchers designed, constructed, and tested a quantum cryptography system that creates and transmits (using single-photon transmissions) cryptographic random numbers between sending and receiving instruments that were separated by a 1-km outdoors optical path at night. The system is based on the propagation and detection of nonorthogonal polarization states of single photons in free space at a wavelength (772 nm) for which the atmosphere has a very low attenuation. They have now demonstrated point-to-point key distribution over 0.5 km in full daylight and are planning to extend the transmission range to more than 2 km in a new system that incorporates active control to compensate for turbulence-induced beam wander. They are also developing plans for a surface-to-satellite test experiment.

Sensors for Detection of Chemical and Biological Warfare (CBW) Agents. Countering the threat of CBW has become an increasingly important national security concern. In one LDRD project, researchers are using prototype electrochemical sensors to demonstrate concepts for the sensitive detection of chemical warfare (CW) agents in the gas or aqueous solution phase. The electrodes are small and could allow formation of an array for a sensor package configured as a badge. A typical device consists of a thin sandwich of three electrodes separated by a polymer electrolyte. The active electrode is a sandwich of a gas-diffusion electrode and a thin composite catalyst layer. In the catalyst layer, chemical components that catalyze electrochemical reactions typical for the target CW agent are introduced. Researchers have made devices that respond to hydrogen cyanide gas, to a sulfur mustard analog, to an anthrax spore marker, and to a widely used simulant of CW agents. Sensitivity levels of as low as 50 ppb of hydrogen cyanide in air have been achieved with a sensor response time of a few seconds.

Researchers in another LDRD project are developing species-selective thin films for chemical sensing based on waveguide Zeeman interferometry for application to the rapid detection of CW agents. The species-selective films are based on guest–host inclusion complexes formed by bucket-shaped cyclodextrin molecules and the CW agent in question. Several different cyclodextrin molecules have been prepared and thin films formed by covalent attachment to functionalized self-assembled monolayers on surface acoustic wave (SAW) and optical waveguide transducers. Measurements have been made of the sensitivity and time response of SAW and optical transducers using both multilayer and polymer films. The sensitivity to a Sarin simulant is in the range 10–100 ppb. The waveguide Zeeman interferometry transduction approach has been demonstrated using these thin films and shown to be as sensitive as SAW devices.

In another LDRD project, researchers are developing new biosensors for bacterial protein toxins. They are using glycolipid cell membrane receptors that are optically tagged and imbedded in a fluid phospholipid bilayer membrane that mimics the cell membrane surface. They have developed three optical transduction schemes where close proximity of the tagged glycolipid receptors resulting from multivalent binding of glycolipids by cholera gives rise to fluorescence-energy quenching and resonant energy transfer. This triggered optical biosensor has been demonstrated for cholera using flow cytometry and is now being adapted to several other protein toxins.

In yet another LDRD effort, researchers are expoiting the ability of biomolecules to recognize and bind to intracellular targets with extreme selectivity. The ability of biomolecules to perform such diverse and specific molecular interactions is based on the large number of potential three-dimensional structures they can adopt. To exploit these properties, the researchers are engineering biomolecules using in vitro evolution of RNA molecules to form highly specific molecular interactions with target molecules of interest. They have created a library of starting molecules containing a high degree of molecular diversity. This initial library has been used to select for individual nucleic acid molecules that are capable of binding a recombinant version of *Bacillus anthracis* protective antigen (PA). They are currently characterizing the library of RNA molecules selected against PA and are continuing to use this library to select for RNA molecules capable of binding to other proteins.

Stable Polymeric Light-Emitting Devices. Defense-related devices and instruments continue to require better and higher-performance light-emitting diodes (LEDs). Polymeric LEDs have attracted tremendous scientific and technological interest because they are brighter and more colorful than liquid crystal displays, they can easily be deposited onto large-area substrates, and they offer flexibility in materials and device design. They also can be fabricated into flexible thin films and adopted into many unusual places where conventional inorganic LEDs cannot be applied. Using molecular self-assembly techniques, LDRD researchers are fabricating polymeric LEDs with the ability to control thin-film structure and composition at the molecular level.

Last year researchers focused on tuning the interface properties between the electrodes and polymeric layers, which are critical to the behavior and performance of the polymeric LEDs. They have fabricated and characterized a new type of diode device made of metal/organic monolayer/semiconductor heterostructures. The device consists of an organic barrier layer sandwiched between an aluminum metal contact and p-type silicon. Current-voltage measurements show that the turn-on voltage of the devices can be tuned by varying the structure of the organic monolayer. Unlike conventional Schottky barriers, which exhibit an exponential relationship between current and voltage, the new diodes have a power-law dependence of current on voltage that is controlled by the structure and electronic properties of the self-assembled monolayers.

Separation Science and Technology. LDRD researchers are investigating new approaches for dealing with the disposition of radioactive and other hazardous wastes that are the legacy of past nuclear weapons production, as well as approaches to minimize generation of such wastes in current operations. In one such project, researchers are advancing membrane-based separation and destruction technologies. They have evaluated polymers and polymer formulations for recovery of toxic oxyanionic metals such as chromate and arsenate from selected waste streams. They have also developed second-generation water-soluble polymeric systems for highly selective oxyanion removal and recovery and have optimized the simultaneous removal of radioactive strontium and cesium from aqueous solutions using new, nonhazardous separation agents. They have developed recyclable redox-active extractants that permit recovery of the radioactive ions in a minimal waste volume. In addition, they have produced hollow fibers and fabricated prototype hollow-fiber membrane modules for application to gas separations and the liquid-liquid extraction and recovery of actinides from process streams. They have fabricated cyclodextrin-based microporous materials that selectively absorb organic compounds in an aqueous environment; the resultant products give pure water with organics at less than 0.05 ppb. New, more efficient, membrane-based electrochemical reactors for use in organic destruction in process waste treatment have been developed. Also, advanced oxidation technologies were developed based on molecular-level materials designs that selectively remove or destroy target species.

Highlights of LDRD in Support of the Laboratory's Science and Technology Base

The above examples are but a few of the ways in which LDRD-funded research contributes directly to the science and technology required for the Laboratory's national security mission. These examples primarily explore new ways of tackling specific mission problems. Other projects, such as those highlighted below, broaden the fundamental science and technology base that underpins the Laboratory's ability to carry out its missions. All projects in the LDRD portfolio contribute to the long-term scientific and technical vitality of the Laboratory, enhance the Laboratory's ability to address future DOE missions, and foster creativity and stimulate exploration of forefront science and technology.

Dynamic Fracture of Heterogeneous Materials. LDRD researchers are investigating the fundamental aspects of the process of dynamic fracture propagation in heterogeneous materials. This work focuses on three important but poorly understood aspects of dynamic fracture for materials with a heterogeneous microstructure: the appropriateness of using a single-parameter asymptotic analysis to describe dynamic crack-tip deformation fields, the temperature rises at the tip and on the flanks of a running crack, and the constitutive modeling of damage initiation and accumulation. The complexities of the research require a multidisciplinary approach combining experimentation, theoretical modeling, and numerical simulation.

This year researchers performed an experimental characterization of the mechanical properties of the high explosive PBX-9501 (a plastic-bonded explosive) and the mock-simulant sugar material PBS-9501. They directly measured the strain field evolution during dynamic impact of PBX-9501 material using the speckle photography technique and developed a physically based, micromechanical constitutive model that describes the thermomechanical behavior of the heterogeneous materials. Additionally, they implemented the theoretical model into an explicit finite element code to simulate the experiments and to be used as a design tool that can simulate the process of dynamic fracture.

Advanced Research Capabilities for Neutron Scattering. LDRD researchers are investigating important new technologies that can potentially improve the performance and utilization of neutron spectrometers at LANSCE and elsewhere. Included are novel position-sensitive neutron detectors, new neutron beam-guide technologies, beam optics for polarized cold-neutron applications, electromechanical aspects of fast mechanical (Fermi) choppers, development of Monte Carlo simulation tools for neutron instrument design, and development of time-resolved cold-neutron radiography methods.

This year the researchers invented a new neutron beam-guide technology and built a proof-of-principle device that seems to have excellent characteristics. This type of guide can be constructed at substantially less expense than commercial guides available today. A patent is being pursued and there is commercial interest in licensing the technology. A low-cost 64-channel amplifier/discriminator suitable for use with a helium-3 ion chamber has been designed, and a two-dimensional, delay-line readout wire chamber, which works for counting rates in the 100-kHz range, has been built and tested. This technology is faster and may be cheaper than traditional resistive-wire readouts presently used for materials science applications. In neutron radiography, the researchers have demonstrated that cracks can be observed with a resolution of 25 to 50 microns.

Atomic Resolution Material Probes. LDRD researchers are developing world-class capabilities for the atomic-scale measurement and analysis of materials at the Electron Microscope Facility (EMF) and the Ion Beam Materials Laboratory (IBML) at Los Alamos. For the electron microscopes, they are developing image-processing and computational methods to realize the true atomic-scale resolution of the field-emission, scanning-transmission, and high-resolution microscopes. This work emphasizes the very high vacuum capabilities of the instruments and utilizes the Z-contrast methods of imaging atomic planes in materials such as superconductors. For the IBML, the emphasis is on the continued expansion of simultaneous, multiple analytical methods to include nuclear-reaction analysis, particle-induced x-ray emission, and elastic-recoil measurements combined with channeling in single-crystal samples.

This year researchers have made fundamental measurements of the energy-dependent cross sections of light elements (beryllium, boron, carbon, nitrogen, oxygen) with high-energy alpha particles. These measurements are essential for the quantitative subsurface analysis of light elements in the near surfaces of materials. They have measured the absolute, x-ray production cross sections for excitation with deuterons, helium-3, and alpha particles and confirmed the validity of the analytical codes for the measurement of trace elements in complex materials. Correlation with primary standards from the National Bureau of Standards has been verified, and the enhanced analytical capability is now routinely available to users of the facility. They also have added new capabilities that have led to the discovery of a number of unique incommensurate and commensurate structures in rhenium disilicide (a potential infrared detector material), textured grains in thick films of YBCO with high critical current capacity, and unusual stacking fault configurations in Laves phase alloys.

Electron Transfer in Conducting Polymers. Electron transfer in conducting polymers is a promising new field of photochemistry. In these systems, charge transfer has been demonstrated to occur in less than

200 femtoseconds 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. LDRD researchers are applying femtosecond spectroscopy to study the dynamics of the various steps involved in the charge transfer and are exploring applications of these materials by studying methods to utilize the charge transfer state to drive photovoltaic and nonlinear optical effects.

This year the researchers finished comprehensive studies of nonlinear interactions of excitons in conjugated polymers at high excitation densities, including the first observation of biexcitons. They have demonstrated universal spectroscopic features on femtosecond timescales due to photoexcitations in many different phenylene-based conjugated polymers. Their broadband, polarized, transient absorption measurements have allowed the first direct measurement of charge-transfer states in oriented polymer films where polymer chains are aligned in a common direction. They have, for the first time, extended the range of femtosecond spectroscopy in the mid-infrared range to map the low-energy, photo-induced optical transitions in these materials. They have further developed a new theory of the optical properties and energy structure of conducting polymers.

Quantum Technologies. Recent developments in the laser cooling and trapping of atoms and ions have revolutionized the ability to control and manipulate the quantum states of atomic systems. LDRD researchers are developing these laser techniques to perform fundamental experiments on multi-quantum systems that previously have not been possible. Of particular interest are experiments related to the development of a quantum computer. They are also advancing the abilities to confine, concentrate, manipulate, and detect selected radioactive (or stable) atoms using optical and magnetic traps. The researchers have demonstrated the trapping of six million atoms of radioactive rubidium-82 in a magneto-optical trap coupled to a mass separator. This represents a 100-fold improvement in the number of trapped atoms over all previous radioactive atoms trapping work.

Quantum computation has recently evolved from a subject of essentially academic interest to a field with an enormous potential to revolutionize computer science with a possible impact on national security and economic issues. A key for this evolution has been the discovery of quantum error-correcting codes and implementations of individual quantum gates. LDRD researchers are pushing back the frontiers by understanding quantum information capacity in the presence of noise and studying its implications for quantum logic circuits composed of small numbers of quantum bits. They are also using computer simulations and theory to implement quantum computations with small molecules. This year researchers created a Greenberger-Horne-Zeilinger state, which is an entangled state of three quantum bits. It is the first time ever that such state of matter has been realized in the laboratory. This work was an essential precursor to implementation of quantum error correction. They have achieved the first experimental implementations of quantum error correction and have confirmed the expected state stabilization. They performed a precise analysis of the decay behavior in alanine and a full implementation of the error-correction procedure in trichloroethylene.

High-Intensity Laser-Matter Interaction Physics. The interaction of a high-intensity laser beam with a preformed plasma is fundamental to research in high-energy density physics and of special interest for investigation of the fast-ignitor concept of laser-induced fusion. In this context, high intensity implies irradiances exceeding 10^{18} W/cm^2. The laser electric field is hundreds of volts per angstrom; electrons, oscillating at speeds greater than 85% the speed of light, are stripped away from atomic nuclei. LDRD researchers are examining the question of how or whether the laser beam penetrates into a target plasma.

This year researchers have used the picosecond capability at the Los Alamos Trident laser to demonstrate an unusual effect: diffraction-limited focusing of a green laser beam on a plasma target of density $>2.5 \times 10^{20}$ cm^{-3} with an f/1.85 off-axis parabola. The beam intensity, at 2×10^{19} W/cm^2, exceeds the threshold for ponderomotive electron cavitation in the plasma. In tunneling through the plasma, the laser is refracted such that its f-number increases to nearly f/8. This experiment suggests favorable beam confinement for the alternate laser fusion concept known as the fast ignitor.

Discrete Simulation of Nonlinear Systems. LDRD researchers are developing discrete numerical methods for modeling multiscale physics in nonlinear systems, in particular complex fluids and soft condensed matter and multiphase fluid flows in porous media. Their goal is to provide detailed insight into the physics of these nonlinear systems from the microscopic to macroscopic scale.

They have developed an event-driven, three-dimensional, hard-sphere molecular dynamics (MD) model and a dissipative-particle MD code for research on the properties of high-speed, high-concentration granular flows and the clustering kinetics of granular media during cooling. For the first time, their work has made it possible to do three-dimensional MD simulations of up to 10^6 dissipative particles. They have developed Monte Carlo techniques for construction of compact wavefunctions to describe a quantum subsystem. They have developed a new, lattice-Boltzmann scheme for simulation of multiphase flow based on distribution functions. This scheme can model the interfacial dynamics including phase segregation and surface tension. Simulations of the two-dimensional and three-dimensional Rayleigh-Taylor instabilities using this scheme yield excellent results. Finally, they have used the discrete numerical method to simulate problems in micromechanical systems and have demonstrated that this discrete method can capture the fundamental behavior of micro-channel flow, including velocity slip and nonlinear pressure drop.

Fiber-Optic Communications Using Solitons. Soliton-based optical-fiber networks have the potential of sending many gigabits per second of digital data over fiber-optic links in both defense and commercial applications. Such networks would take advantage of the intrinsic stability of soliton solutions of the nonlinear Schrodinger equation, which describes to leading order 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. LDRD researchers have discovered that such devices can be designed to actually exploit losses and dispersion 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. They are studying this wider class of behavior and are applying their results directly to the theory of fiber-optic networks. Their study ranges from the analysis of individual devices to the modeling and optimization of complete networks.

This year researchers have developed a perturbation method for studying the nonlinear partial differential equations that govern the propagation of optical-fiber pulses. Additionally, they performed numerical studies of various types of pulse transmission techniques. This led them to discover a very promising power enhancement phenomenon using a technique called sliding frequency guiding filters.

LDRD Project Summaries

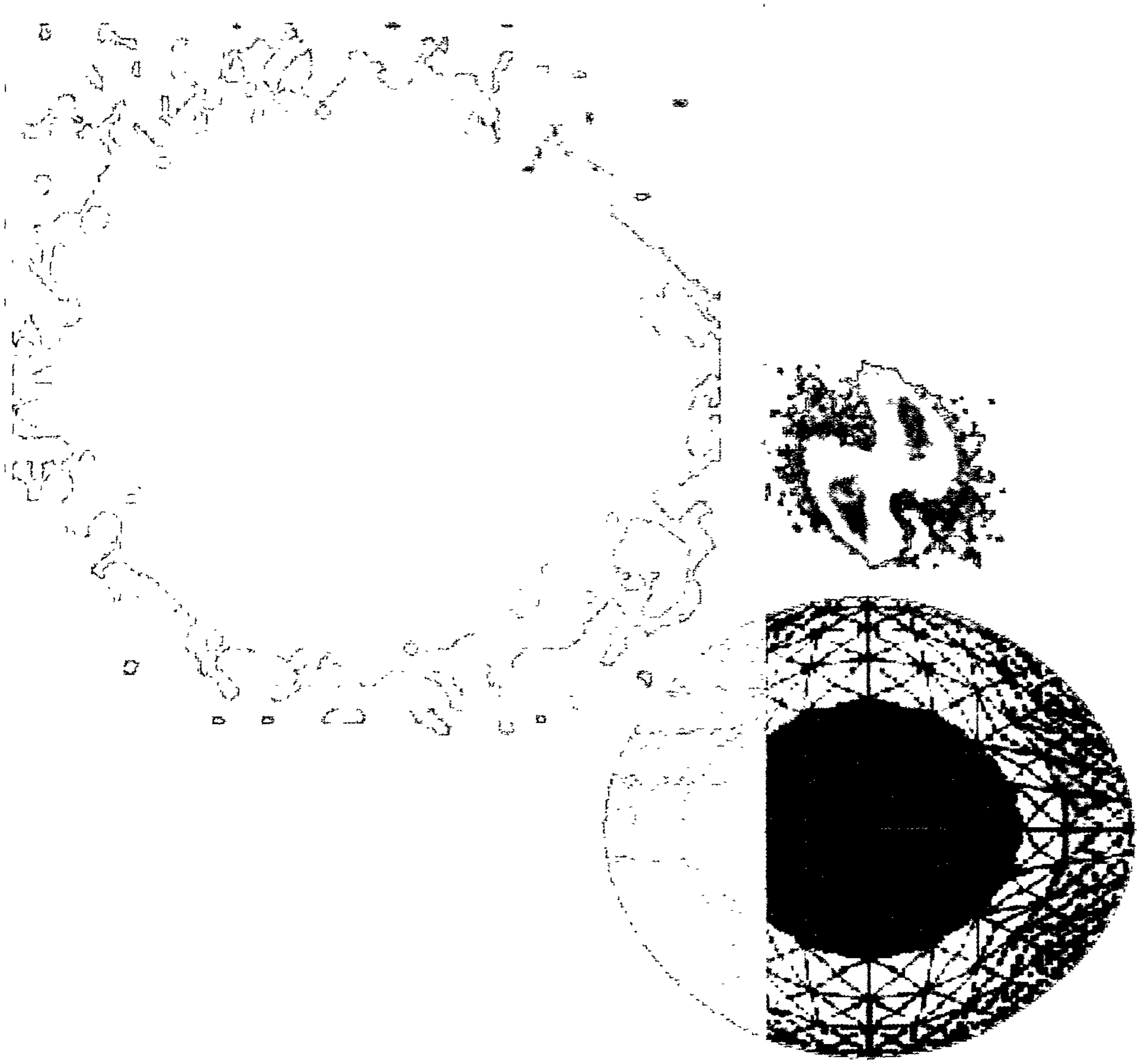
In the sections of our annual progress report that follow, project summaries are grouped by their LDRD component—CD, PD, or IP. Within each component, they are further grouped into the nine technical categories for projects, ordered as follows:

- Materials Science
- Chemistry
- Mathematics and Computational Science
- Atomic, Molecular, Optical, and Plasma Physics, Fluids, and 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.

1998
LDRD

Competency Development Projects



Nanostructured Metal-Ceramic Composites for High-Temperature Structural Applications

98802

Michael Stevens

In this project, we have studied the properties of vanadium-spinel composites with fine microstructures. Previous work has shown that the structure of equimolar spinel, $MgAl_2O_4$, is highly resistant to atomic displacements caused by radiation. In addition, spinel retains its strength in compression up to elevated temperatures. However, spinel cannot be used as a structural material because it has poor mechanical properties at room temperature, notably a fracture toughness of only about 2 $MPa(m)^{1/2}$. Vanadium also has attractive performance in irradiation environments, yet it has good toughness at ambient temperature. The goal of our research is to produce radiation-resistant vanadium-spinel composites that have a higher toughness than spinel alone.

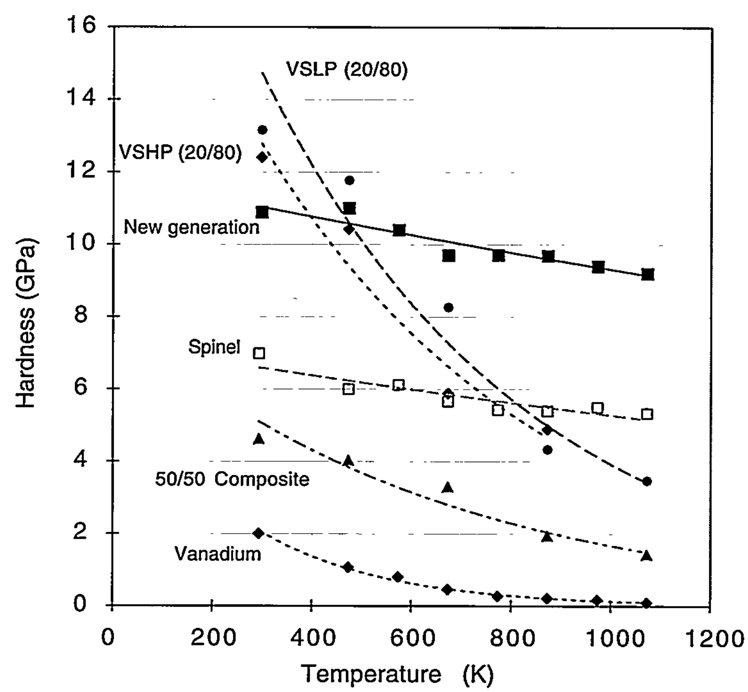
We have used a number of powder-processing routes to obtain composites with different properties: (1) ball milling mixtures of commercial vanadium and spinel powders; (2) in situ displacement reactions during ball milling, starting from V_2O_5, MgO, and elemental aluminum; and (3) same as (2) but with the addition of small amounts of reducing elements (aluminum, titanium, chromium). Following mechanical alloying, we consolidated the powders by hot isostatic pressing at 1350°C. Most of the vanadium and spinel particles in the compacts have submicrometer diameters. The compacts have a room-temperature toughness of about 4 $MPa(m)^{1/2}$. Further increases in toughness should be possible with the addition of continuous-fiber reinforcement (e.g., Al_2O_3).

The hardness of the vanadium-spinel composites strongly depends on the processing and the phase contents. Especially noteworthy is the hardness of our "new generation" sample, which does not degrade as much as that of the other composite samples, as seen in the figure.

Publications

Shen, T.D., et al., "Vanadium/ $MgAl_2O_4$ Composites for Hostile Environment Applications—Part I: Processing" (to be published in *Cer. Eng. Sci.*).

Ustundag, E., et al., "Vanadium/ $MgAl_2O_4$ Composites for Hostile Environment Applications—Part II: Properties" (to be published in *Cer. Eng. Sci.*).



Vickers hardness vs. temperature for vanadium, spinel, and several vanadium-spinel composites prepared under this LDRD project. For the composite labeled "New generation," the vanadium phase has the lowest oxygen content (<0.9 wt %), and low oxygen content correlates with the highest hardness at high temperatures. The VSHP and VSLP composites have oxygen contents of 1.7 and 1.6 wt %, respectively.

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

96603

Mark Bourke

The objectives of this project are to use neutron diffraction to study polycrystal deformation and to relate the results to micromechanical models. Residual and in situ loading strains, as well as thermal relaxation effects, all fall within the scope of our research.

Following a series of in situ deformation measurements on stainless steel, we extended the approach to beryllium both because of its institutional interest and because, as a hexagonal material, it is more complicated than a cubic material.

Complementary self-consistent modeling simulations are being performed in conjunction with Carlos Tome of the Materials Science and Technology Division at Los Alamos. During this past year, two doctoral theses, which were based largely on research performed within the context of this project, were completed. Hahn Choo's (Illinois Institute of Technology) thesis concerned neutron diffraction characterizations of dispersion-strengthened NiAl. Raj Vaidyanathan's (Massachussetts Institute of Technology) thesis addressed stress-induced phase transformations in shape memory alloys and their composites, specifically in NiTi-TiC.

Improvements to the stress rig that was extensively used in this research include (1) adapting a Labview PC program to digitally record the experimental parameters during in situ load tests, and (2) modifying tensile grips to ensure better uniaxiality during loading.

Publications

Choo, H., "Synthesis Processing and Characterization of NiAl-AlN- Al_2O_3 Composites," Ph.D. thesis, Illinois Institute of Technology (1998).

Choo, H., et al., "Thermal Residual Stresses in NiAl-AlNAl_2O_3 Composites Measured by Neutron Diffraction" (to be published in *Mater. Sci. Eng. A*).

Clausen, B., et al., "Lattice Strain Evolution in Stainless Steel" (submitted to *Mater. Sci. Eng. A*).

Daymond, M.R., et al., "Effect of Texture on HKL Dependent Intergranular Strains Measured in situ at a Pulsed Neutron Source," in *Proceedings of Fifth International Conference on Residual Stress*, T. Ericsson, et al., Eds. (Linkoping University, Linkoping, Sweden, 1998), p. 577.

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 to Fit Hexagonal Crystal Structures in the Presence of Elastic and Plastic Anisotropy" (to be published in *J. Appl. Phys.*).

Vaidyanathan, R., "Stress Induced Martensitic Phase Transformations in NiTi and NiTiTiC Composites Studied by Neutron Diffraction" (submitted to *Mater. Sci. Eng. A*).

Dynamics of Polymers at Interfaces

96605

Gregory Smith

We address fundamental questions about the behavior of polymers at interfaces: (1) What processes control the formation of an adsorbed layer on a clean surface? (2) What processes control the displacement of preadsorbed polymers? (3) Can we accurately predict the structure of nonequilibrium layers?

In FY 1997 we performed neutron reflectivity experiments to study the nonequilibrium architecture of polymer layers strongly adsorbed from the polymer melt. Hydrogenated (poly)methyl methacrylate (PMMA) was spin-coated onto quartz crystal surfaces and subsequently annealed at melt conditions. After cooling, unbound material was leached away.

This year we analyzed the neutron reflectivity data and found that the dry residual layers were indicative of a dense PMMA layer whose thickness increased with annealing time from an initial minimal value to a final equilibrium thickness. The thickness, h, was found to obey $h \sim N^{0.4\pm0.05}$, where N is the degree of polymerization. This is close to the scaling expected for a reflected random walk (RRW) immobilized by the surface ($h \sim N^{1/2}$). Data on residual adsorbed layers swollen in good solvent, deuterated benzene, indicate a strongly stretched, brushlike structure with a diffuse segment density profile, $\phi(z)$.The segment density decays as $\phi(z) \sim z^{-0.77\pm0.03}$, faster than predicted by RRW statistics, and indicates fewer long polymer loops per chain than the RRW model.

Neutron Scattering for Correlated-Electron Systems

96600

Robert Robinson

The purpose of this project has been to exploit the neutron scattering facilities at the Los Alamos Neutron Science Center in studies of correlated-electron behavior in both f-electron systems (e.g., heavy fermions) and transition-metal oxides (like the high-temperature superconducting cuprates and colossal magnetoresistance manganites). There have been two main thrusts: (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.

This year we made substantial progress in analyzing and writing up work performed in the previous year, particularly the localized magnetic excitations seen in UPdSn and UNiGe; the diffuse scattering above the Curie temperature in the two-layer manganites; and the crystal-field scheme of the quadrupolar Kondo system $PrAg_2In$. In the latter case, we showed that there is no quasi-elastic scattering, as one would expect, but there is some broadening of the first excited state. In the case of PuO_2, we determined the absolute intensity and found that our crystal-field scheme is in remarkable agreement with this intensity.

Finally, in the manganese-12 acetate clusters, which exhibit resonant mesoscopic quantum tunneling, we observed a series of six or so excitations, and these can be interpreted in terms of various m_z levels in the 21-fold degenerate (almost) spin-10 state of the molecule (see the figure). These in turn have been used to determine the Hamiltonian for the spin state of the molecule, and the Hamiltonian is crucial to understanding the quantum tunneling.

Publications

Argyriou, D.N., et al., "Two-Dimensional Ferromagnetic Spin Correlations above T_c in the Layered CMR Manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$–0.4)," *J. Appl. Phys.* **83**, 6374 (1998).

Javorsky, P., et al., "Crystal Field in ErNiAl and ErCuAl Studied by Inelastic Neutron Scattering," *J. Magn. Magn. Mater.* **186**, 373 (1998).

Kelley, T.M., et al., "Nonmagnetic Crystal-Electric-Field Ground State in the Heavy-Fermion Compound $PrInAg_2$" (to be published in *Physica B*).

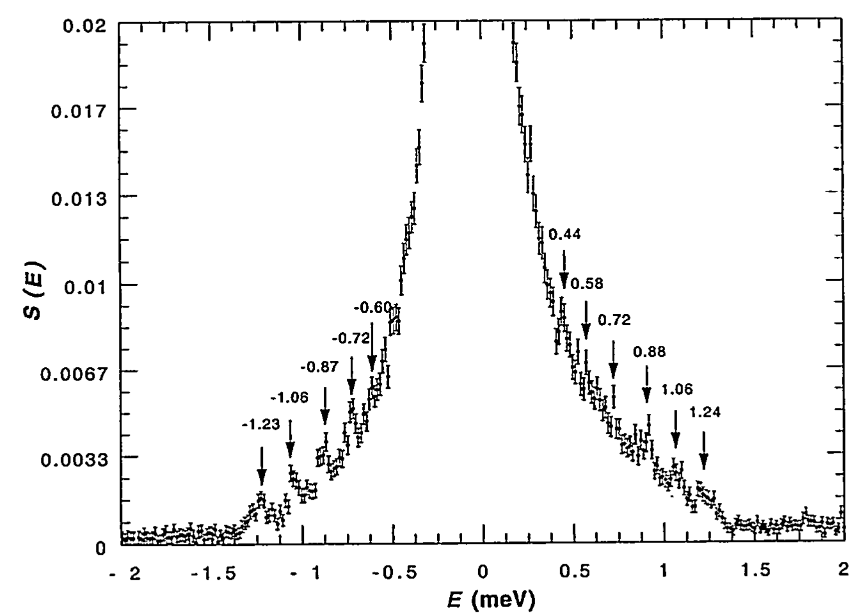
Kelley, T.M., et al., "Short-Range Spin Correlations in the CMR Material $La_{1.4}Sr_{1.6}Mn_2O_7$," *Physica B* **241**, 439 (1997).

Kern, S., et al., "Crystal-Field Transition in PuO_2" (to be published in *Phys. Rev. B*).

Nakotte, H., et al., "Localized Excitations in UPdSn," *Physica B* **241**, 675 (1997).

Robinson, R.A., et al., "Low-Energy Excitations, Symmetry Breaking and Specific Heats in YbBiPt" (to be published in *Physica B*).

Zhong, Y., et al., "Inelastic Neutron Scattering Study of Mn_{12}-Acetate" (to be published in *J. Appl. Phys.*).



Low-energy neutron spectrum from manganese-12 acetate at a sample temperature of 30 K, plotted as a function of energy transfer *E*. The arrows indicate the transitions from $m_z = -10, -9, -8$, and so on to corresponding $m_z + 1$ levels. These data were taken on the QENS spectrometer at Argonne National Laboratory.

New Initiatives in Materials Characterization, Modeling, and Synthesis

96615

Don Parkin

This project explores new techniques in characterization, constitutive modeling, and synthesis of materials. We investigated the mechanism of basal slip and basal twinning in sapphire, finding evidence that supports a model for basal slip and twinning where the slip plane cuts in half the puckered layers of aluminum in sapphire. Studies of the influence of hydrogen on the mechanical properties of alumina showed that the strength of specimens was reduced by a factor of 2 when the tests were performed in the presence of water. Alumina thin films were grown by reactive evaporation of aluminum metal in O_2 onto silicon (111) as a template for $MgAl_2O_4$. We used low-energy electron diffraction (LEED) and x-ray photoemission spectroscopy (XPS) to study the deposits in situ. The initial ordering of 1-nm films exhibited a $2\sqrt{3}\times2\sqrt{3}R30°$ diffraction pattern. The oxide growth proceeded by the Stranski-Krastanov mode.

We synthesized and investigated Laves phase alloys of niobium-chromium-titanium and found that the effect of titanium alloying on the $NbCr_2$-based C-15 structured alloys improved the fracture toughness of the material. A model of ductile spallation fracture was developed from a dynamic void-growth model, taking full account of material inertia and strain-rate sensitivity under shock-loading conditions. The results from this model are in good general agreement with previously published experimental results for ductile spallation fracture in tantalum.

To further our understanding of characterization, modeling, and synthesis, we held workshops on the following topics: probing frontiers in matter with neutrons, complex adaptive matter, damage accumulation and fracture, metal forming, plutonium, beryllium, and many-body physics.

Publications

Becker, J.D., et al., "Calculated Lattice Relaxation in Pu-Ga" (submitted to *Phys. Rev. B*).

Brooks, M.S.S., et al., "Crystal Field Excitations as Quasi-Particles" (submitted to *Nature*).

Delin, A., et al., "Cohesive Properties of the Lanthanides: Effect of Generalized Gradient Corrections and Crystal Structure" (submitted to *Phys. Rev. B*).

Heuer, A.H., et al., "Slip and Twinning Dislocations in Sapphire x-A1203" (submitted to *Philos. Mag.*).

Kotula, P.G., et al., "Defects and Site Occupancies in Nb-Cr-Ti C15 Laves Phase Alloys" (submitted to *Scripta Materialia*).

Soderlind, P., et al., "Simple Model for Complex Structures," *Phys. Rev. Lett.* **57**, 1320 (1998).

Actinide Crystal Structures with an Emphasis on Plutonium Alloys

96607

Joyce Roberts

Our purpose has been to use neutron diffraction to determine the crystal structures of three intermetallic phases that appear in actinide phase diagrams. Two are plutonium phases, zeta-phase Pu_2Th and theta-phase Pu_4Zr. In addition, we wanted to measure delta-phase Np-U in order to study the question of atomic ordering in zeta-phase Pu-U.

Our plan for this year included four tasks: (1) re-anneal Pu-Zr and Pu-Th alloys, (2) remeasure Pu-Zr and Pu-Th alloys at the Los Alamos Neutron Science Center (LANSCE), (3) prepare Np-U alloys, and (4) measure Np-U alloys at the Intense, Pulsed Neutron Source (IPNS) at Argonne National Laboratory.

The plan was not successful for two unrelated reasons. First, the change in oversight of the Chemistry and Metallurgical Research Facility and consequent reorganizations in the Nuclear Materials and Technology Division and the Materials Science and Technology Division have delayed sample preparation. Second, there was no beam at the Manuel Lujan Jr. Neutron Scattering Center at LANSCE because of delays associated with facility upgrades.

As a result, our accomplishments this year were limited to the following: (1) We re-annealed Pu-Zr and Pu-Th alloys and characterized the annealed samples by x-ray diffraction. These samples have not been measured at LANSCE, but we hope that beam time will be available for this task in the coming year. (2) Preparation of Np-U samples is proceeding slowly because of delays associated with obtaining chemical analysis of available uranium metal. (3) We have received beam time at IPNS to measure Np-U alloys, and we intend to complete these measurements in the coming year.

Microstructures of Transition Metal Oxides Characterized with Neutron and X-Ray Scattering

96602

Michael Fitzsimmons

This project has involved studies of magnetically ordered materials with reduced dimensionality, including thin films of NiMnSb and manganese, and nanocrystalline europium mono-oxide (EuO). We investigated the new triple-layer NiMnSb/V/NiMnSb on MgO(100) by means of vibrating-sample magnetometry, x-ray, and neutron reflectivity. We measured a coercive magnetic field of 23 Oe and observed a hysteresis loop similar to that of an uncoupled ferromagnet. The x-ray and neutron reflectivity data indicated that a layer system with interface roughness of 10 Å was present. A detailed analysis of these data suggests significant interdiffusion at the NiMnSb/V interfaces. Interdiffusion between the NiMnSb and vanadium layers may produce a lessening of the giant magnetoresistance effect measured in most MiMnSb-based multiple-layer systems.

Thin films of α-Mn were epitaxially grown on MgO(111) single-crystalline substrates at elevated temperatures in order to suppress the growth of metastable manganese phases previously reported. We observed the α-Mn films to have the (110) texture, consistent with body-centered-cubic (110) growth in Kurdjumov-Sacks orientation on the face-centered-cubic (111) substrate surface, with additional twinning. The structural features observed for α-Mn(110) resemble those of metastable "expanded" manganese, indicating a possible connection between the two phases.

We produced EuO using a constant volume combustion technique. Samples of EuO were ball-milled in an argon glove box to produce nanocrystalline EuO. Rietveld refinement of powder x-ray diffraction patterns were taken from samples that were ball-milled for different lengths of time. These patterns show the reduction of the grain size of the material and increased microstrain content with ball-milling time (see the accompanying figures). We measured the magnetic properties of the coarse-grained reference sample with a SQUID magnetometer and found them to be consistent with properties given in the literature for bulk EuO. Interestingly, the most heavily ball-milled material exhibited superparamagnetic behavior consistent with the small grain size of the material. The latter suggests that grain boundaries or dislocation forests inhibit the onset of long-range ferromagnetic order in nanocrystalline EuO.

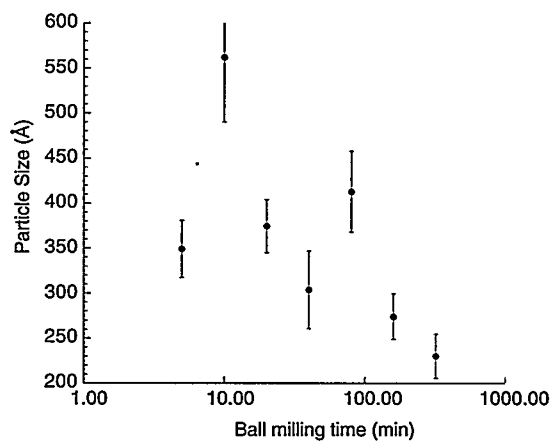
Publications

Grigorov, I.L., et al., "Observation and Analysis of Multidomain Epitaxy of α-Mn on MgO(111)" (submitted to *Phys. Rev. Lett.*).

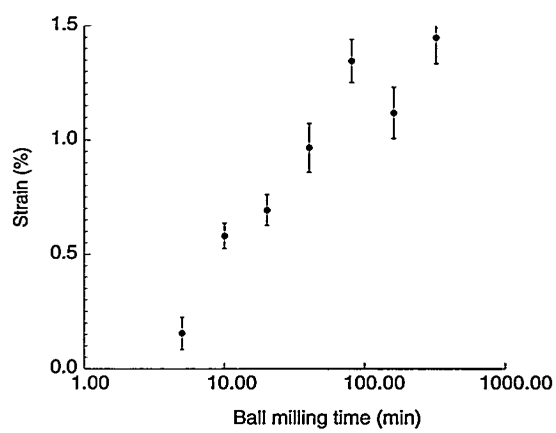
Grigorov, I.L., et al., "Structural and Magnetic Properties of Expanded Mn," *J. Appl. Phys.* **83**, 7010 (1998).

Schlomka, J.P., et al., "Structural and Magnetic Properties of Ion Beam Sputtered NiMnSb Films," *Physica B* **248**, 140 (1998).

Schlomka, J.P., et al., "Interdiffusion in NiMnSb/V/MnSb: X-Ray and Neutron Reinvestigation of Ion Beam Sputtered Trilayer Systems" (submitted to *J. Appl. Phys.*).



Particle size of ball-milled EuO.



Microstrain content in ball-milled EuO.

Neutron Scattering Studies on Shear-Induced Structure in Polymers

96604

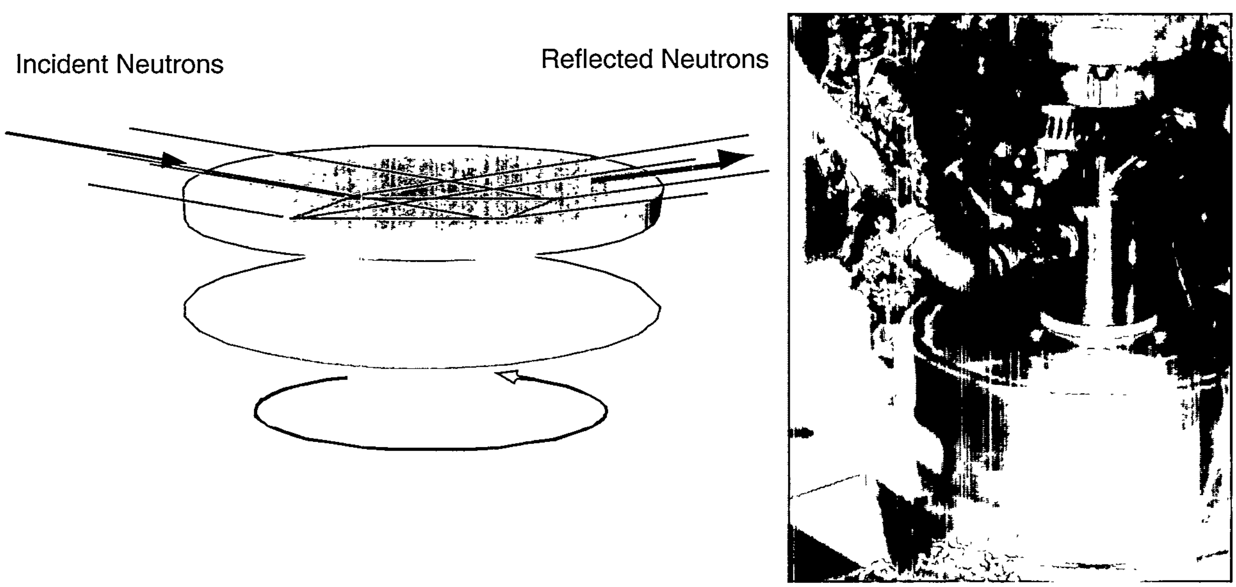
Rex Hjelm

Our objective was to develop competency in the use of neutron scattering for the study of shear-induced effects in polymers. Specifically, we used small-angle neutron scattering and neutron reflectometry to study shear-induced polymer conformation and orientation near surfaces and in the bulk. These two phenomena are fundamental to understanding shear effects in the industrial processing of polymers, and they are also of importance to the physics of polymer fluids. By simultaneously measuring structural and rheological properties of polymer solutions, we were able to correlate the properties. Such correlations have not been possible before.

This year we finished the construction of a cone-and-plate rheometer for neutron reflectometry experiments; the rheometer has a furnace to produce polymer melts at temperatures up to 200°C (see figure). Our first experiments were done on the surface structure of high- and low-density polystyrene brushes next to polystyrene melts as a function of shear rate. (Brushes are linear polymer molecules attached to the plate at one end; melts are polymers heated to become a fluid.) We were able to show swelling of the brushes into the melt and subsequent narrowing of the brush layer at low shear rates. The results of these measurements, which were made to analyze the molecular events associated with surface slippage or self-lubricating effects in polymer melts, are now being analyzed.

Publications

Hjelm, R.P., "The Structure of Fillers, Polymers and Their Interfaces in Polymer Composites using Neutron Scattering Methods," *Proceedings of the 153rd Meeting of the American Chemical Society Rubber Division, Indianapolis* (ACS Polymer Rubber Division, Acron, Ohio, 1998), Vol. 153, p. 8.



Cone-and-plate rheometer for neutron reflection studies of surface effects in polymers under shear. (Left) Schematic of the rheometer's stationary upper plate and rotating lower cone. The polymer brushes are attached to the lower surface of the plate, and the polymer melt is between the cone and polymer brush-coated plate. Neutrons are reflected from the interface between the brushes and the melt and probe the structure of the interface. (Right) View of the rheometer showing the furnace (flat cylinder at bottom) that contains the cone-and-plate assembly. Neutrons enter the oven through the oblong aluminum window on the left just above the furnace.

Texture Science and Technology

96616

Michael Stevens

The overall goal of this project is to advance R&D in texture and anisotropy at Los Alamos National Laboratory. Our specific objectives are to (1) increase the utilization of texture and anisotropy, both within and without the Laboratory's programmatic, basic, and industrial-related efforts; (2) improve our texture measurement and modeling capabilities; and (3) maintain our recognition as an international leader in these areas through basic research.

The following are many of our accomplishments from the past year. We analyzed the effect of lubrication on the evolution of microstructure and texture during rolling and recrystallization of copper and low-carbon steels. We developed a model to simulate recrystallization based on electron backscatter diffraction orientation microscopy data, and analyzed the influence of recrystallization nucleation at the grain boundaries on the recrystallization textures of aluminum alloys.

The impact of orientation pinning on the evolution of the Goss texture in iron-silicon transformer steels was analyzed. We developed an approach to simulate recrystallization textures by means of a self-consistent model and applied it to various geologic materials.

We discovered a preference for low-angle intracolony misorientation distributions in bismuth-2223 superconductors. We applied microtexture measurements in order to investigate the correlation between pitting corrosion, orientation, and grain-boundary character for variously processed nickel, stainless steel, and beryllium samples. Potential current-limiting mechanisms in coated high-temperature superconducting substrate materials were investigated.

We upgraded the software for data collection and analysis for both the orientation imaging microscopy system and the Scintag X1 x-ray goniometer system; we also upgraded the x-ray generator on the first-generation Scintag. The new capability in x-ray texture measurement and analysis is valuable for various programs and collaborations.

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

96627

P. Chris Hammel

Our objectives are twofold: (1) to gain an improved understanding of the nature of charge inhomogeneities in two-dimensional transition-metal oxides (cuprates in particular), which are responsible for local structural deviations from the average structure, and (2) to gain a better understanding of the consequences of these structures for the magnetic and electronic properties of these materials. We have performed extensive experimental studies of the magnetic properties of lightly doped lanthanum cuprate using nuclear magnetic and quadrupole resonance, muon spin resonance, and magnetic neutron scattering. These revealed that spin freezing in the AF-ordered phase is nearly universal regardless of dopant density and location and suggested that stripe dynamics may be responsible. The magnetism of more heavily doped, stripe-ordered transition-metal oxides revealed glassy behavior and an unusual sort of strong orientational disorder. Photoemission studies suggest previously unrecognized Fermi surface features.

We have explained thermal transport experiments in terms of a new, fully gapped d+id' phase. We have also developed an explanation of the results of fast optical experiments designed to investigate the breaking and reformation of Cooper pairs. We have performed an accurate calculation of the ground, excited state, and far-from-equilibrium properties of coupled electron-phonon systems, as well as a calculation of stripe formation in the high T_c superconductors. We also performed calculations that explain the unusual magnetic properties of lithium-doped lanthanum cuprate when the electron-phonon interaction is taken into account.

Publications

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Advanced Research Capabilities for Neutron Scattering

98610

Robert Robinson

This project covers a range of important new technologies that can potentially improve the performance and utilization of neutron spectrometers at the Los Alamos Neutron Science Center (LANSCE) and elsewhere. We have invented a new unpolarized neutron-beam-guide technology that is substantially cheaper than existing commercial guides. We have built a model that seems to have excellent characteristics. Our guide technology is so promising that we are pursuing a patent, and there is commercial interest in licensing the technology.

In the area of detectors, we have designed a low-cost 64-channel amplifier/discriminator suitable for use with a helium-3 ion chamber. In addition, we have built and tested a two-dimensional delay-line readout wire chamber that works for counting rates in the 100-kHz range. This technology is faster than traditional resistive-wire readouts presently used at LANSCE.

We have developed a suite of computer tools for the simulation of neutron spectrometer performance centered on the Monte Carlo neutron-instrument-simulation package, which has been made available to interested parties within and outside the Laboratory. We have made several improvements to the package including the addition of Fermi chopper elements and new neutron-source terms covering moderators at the ISIS and Intense Pulsed Neutron Source facilities (at Rutherford, England, and Argonne National Laboratory, respectively); dramatic computational speed enhancements; a new user interface for handling extra optical elements; and new debugging capabilities. We have started to collaborate on such Monte Carlo simulations with the Spallation Neutron Source project (at Oak Ridge National Laboratory), and there is a good chance that our work will form the nucleus of an international standard for such calculations/programs.

We are assessing and identifying potential industrial applications of cold-neutron radiography (see the figure). What is novel in our approach is the capability to time-gate the detector and make simultaneous radiographs at different neutron energies. In principle, this method can be combined with the measurement of Bragg-edge cutoffs along the time axis, which can give materials discrimination quite different from that in other radiography methods. So far, three imaging methods have been used: time-gated imaging, an amorphous silicon detector, and conventional x-ray film. We have already demonstrated time-gated imaging and have made high-resolution images with each of the other two methods. In particular, we observed a crack in a fire-set-case sample (supplied by Sandia National Laboratories) with a resolution between 25 and 50 μm.

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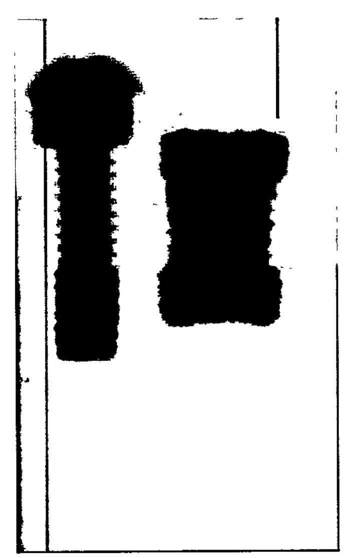

Cold-neutron radiograph of a beryllium block with a nylon screw, left, and a brass fitting containing a nylon O-ring, right. The ability to see hydrogenous objects, like the nylon screw and O-ring, inside a metal object is a specialty of cold-neutron radiography. To see them in beryllium, which is a strong scatterer itself, requires the time-resolved technique used here.

Science of Polymer-Based Materials Aging

97614

Edward Kober

We are developing combined experimental and modeling techniques that can be used to understand the complex, nonlinear aging phenomena of polymer-based materials and to reliably predict their functional lifetimes from accelerated aging experiments. Two systems of relevance to DOE and industry are being studied: Estane, a segmented polyester-polyurethane copolymer, and a silica-filled polydimethylsiloxane composite. A suite of aging environments has been established for both materials and includes controlled temperature, humidity, exposure to oxidants, and applied strain. Several experimental diagnostic techniques have also been established for both materials and include mechanical property measurements; Fourier transform infrared (FTIR) and Raman spectroscopy; matrix-assisted laser-desorption ionization mass spectroscopy; transmission electron microscopy; and small-angle neutron scattering. These experiments are interpreted with the aid of various modeling efforts at different length scales including quantum chemical calculations, molecular dynamics simulations, coarse-grained bond fluctuation models, mean field analysis based on density functional treatments, network theory, and continuum mechanics analysis.

Work this year has predominantly focused on the hydrolytic aging of Estane, which, we established, occurs by hydrolysis of the polyester links. This hydrolysis changes the level of entanglement and phase segregation of the polyester and polyurethane segments and eventually leads to a dramatic decrease in tensile strength (see first figure). The orientation of the particular chain segments could be followed by FTIR (see second figure) and the results compared with the modeling programs to develop a quantitative analysis of the polymer chain dynamics. Physically based continuum treatments are being developed from the detailed information acquired for this system.

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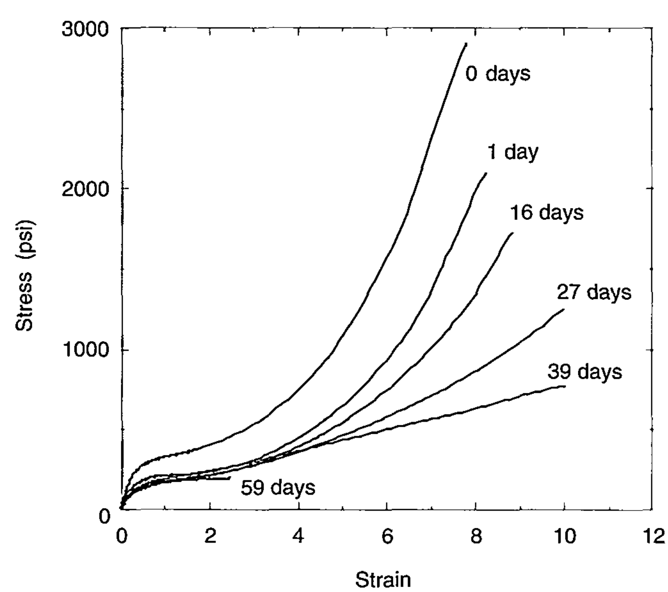
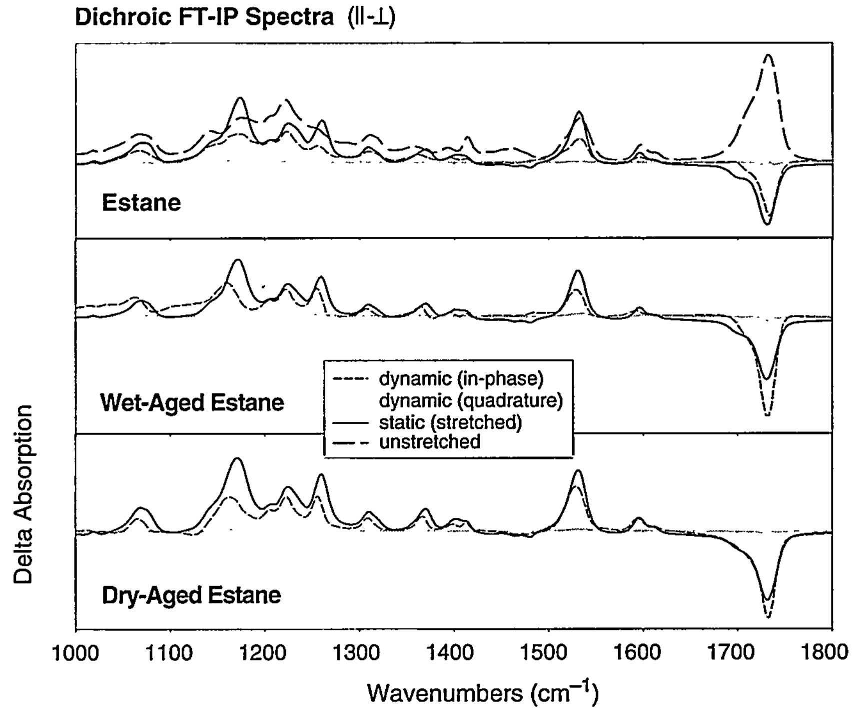


Tensile mechanical responses of Estane as a function of aging time in a 70°C, 70% relative humidity environment.There is a gradual decrease in the stress and strain at failure and a slight change in the initial modulus, until a dramatic change in properties occurs at 59 days.These curves can be fit with a second-order Mooney-Rivlin analysis, and the changes in properties correlate with changes observed with FTIR vibrational spectroscopy and other spectroscopic measurements.



Dynamic in-phase and in-quadrature FTIR responses of unaged, wet-aged (21 days, 50°C, 70% relative humidity), and dry-aged (21 days, 50°C, 0% relative humidity) Estane. Samples are prestretched and oriented by a uniaxial 500% strain and then subjected to a small, 20-Hz oscillating strain. Significant changes in the C=O stretch (1730 cm^{-1}), amide II (1530 cm^{-1}) and C-O stretch (1180, 1160, and 1070 cm^{-1}) regions provide detailed information of the polymer response to mechanical strain as a function of aging.

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Electrons in High Magnetic Fields

97611

Aloysius Arko

The goal of this project is to determine the electronic structure of highly correlated 5f materials, with special emphasis on plutonium and its compounds. Information on the electronic structure of these materials is needed to fill in gaps on the periodic table and for equation-of-state calculations. We are probing their structure through measurements of the de Haas-van Alphen (dHvA) effect and specific heat (C_p) and through fast optics measurements in high pulsed fields at the Los Alamos National High Magnetic Field Laboratory.

A facility for growing plutonium crystals has been built and is nearing final operational approval. In the interim, superb crystals of uranium heavy fermions have been grown, and we have observed the dHvA effect in seven compounds. UCd_{11} has the largest C_p gamma value among compounds in which the dHvA effect has been observed. The first figure that accompanies this summary shows measurements of the dHvA effect in UGa_3.

Our apparatus for measuring C_p in pulsed magnetic fields has been successfully tested in the Laboratory's quasi-continuous 60-T magnet (see second figure). This test represents a breakthrough in pulsed-field measurements and opens the way to collecting a whole new class of data for direct comparison with dHvA masses.

For our fast optics measurements, we have developed a technique for optical-pump/terahertz-probe spectroscopy and used it to study correlations in nonequilibrium superconductivity. This study has revealed a relaxation channel that is hidden to normal optical-pump/optical-probe experiments. In related work, polaron dynamics have been studied experimentally and successfully modeled theoretically.

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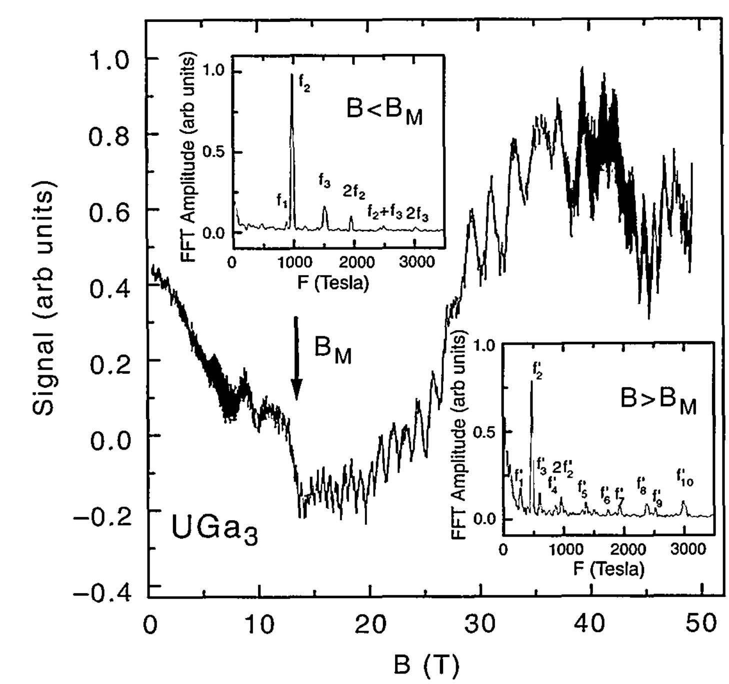

The measured inductive signal of UGa_3 versus applied magnetic field at a temperature of 0.47 K. There is a transition at a field of B_M = 12 T. There are dHvA oscillations (oscillatory magnetization versus inverse applied field) above and below B_M. The insets show the fast Fourier transform (FFT) of the data versus inverse applied field for fields above and below B_M. For B < B_M, there are 3 dHvA frequencies, labelled f1 to f3; for B > B_M, there are 10 frequencies, labelled f'1 to f'10. These results clearly indicate that there is a major Fermi surface reconstruction at B_M.

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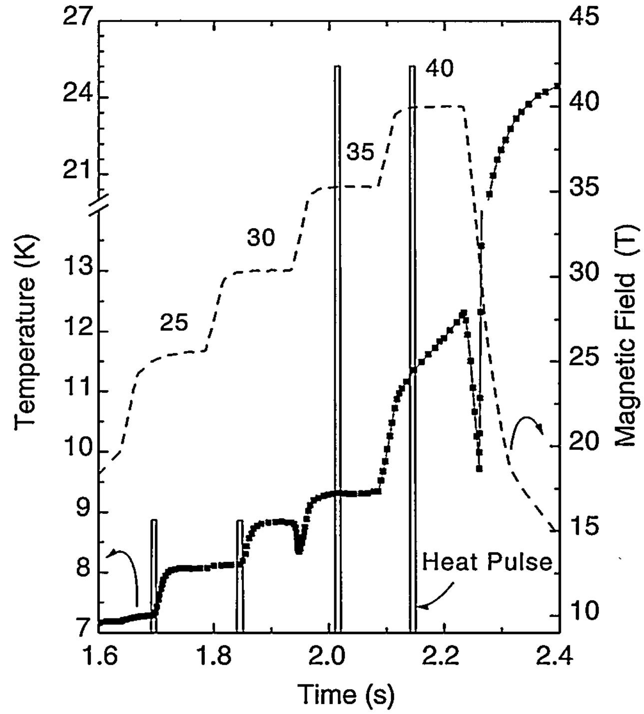

Specific heat measurements in the quasi-continuous 60-T magnet. A series of the plateaus at different magnetic fields can be produced within a single experiment. Shown here is an experiment on $YbInCu_4$ in which four plateaus—at 25, 30, 35, and 40 T, each 130 ms long (see thin dashed line with scale at right)—were created during a single magnet pulse. We measured heat capacity at each of the magnetic-field plateaus. In addition, as we increased the field from 30 to 35 T through the first-order phase boundary, the sample's temperature (lower curve with scale at left) decreased on the up-sweep (temperature dip at 1.95 s) and increased as the magnetic field decreased through 35 T on its way to down zero T (sharp temperature rise at 2.25 s). These temperature changes are a consequence of the magnetocaloric effect. Their sharpness allows direct determination of the phase diagram of $YbInCu_4$.

Fundamental Studies of Radiation Damage in Two-Phase Oxide Composites

97801

Kurt Sickafus

The goal of this project is to obtain a fundamental understanding of radiation damage in multiphase ceramic-oxide composites. These materials may find applications as rock-like nuclear-waste forms for the immobilization of high-level radioactive wastes or as nuclear-fuel forms for the incineration of plutonium and other minor actinides. Candidate materials for these purposes must be chemically durable and tolerant to physical damage from radiation exposure.

We have synthesized composites based on a radiation-resistant ceramic-matrix phase consisting of the titanate mineral geikielite, $MgTiO_3$. The next phase in our composites is an actinide host phase. We have used a cubic pyrochlore phase for this purpose (specifically 5 mol % either of $Gd_2Ti_2O_7$ or $Er_2Ti_2O_7$). We have performed ion irradiation experiments on these composites at 100 K using 350 keV Xe^{++} ions to a fluence of $5{\cdot}10^{14}$ Xe/cm^2. At the highest xenon ion fluence studied, we observed amorphization of the pyrochlore phase ($Er_2Ti_2O_7$), while the geikielite matrix phase ($MgTiO_3$) remained crystalline.

Our experiments to date indicate that the oxide constituents in our composites respond to irradiation as they do in their monolithic (single-phase) forms. These results suggest that we can design ceramic composites for application in high-radiation environments using radiation-damage data obtained from individual compounds.

Theory and Modeling of Correlated-Electron Materials

96628

Alan Bishop

We have entered 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 large 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, this project provides theory and modeling elements for 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-property relationships. These tasks are focused around three critical science issues that are recognized to be 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 the 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 and prompted extensive discussion of state-of-the-art multidisciplinary approaches to structure-function relations in modern materials. We developed and applied modeling techniques for polarons, which are charge-ordering and multiscale (glassy) dynamics in transition metal perovskite oxides. We also developed new methods for calculating transport in strongly driven polaron systems, retaining full quantum coherence. We analyzed impurity-induced and localized quasi-particle gap states in novel superconductors. We calculated metal-insulator/magnetic transitions in colossal magneto-resistance materials, including the effects of Jahn-Teller coupling and spin disorder. We proposed new crossover effects of interchain coupling in spin-Peierls systems, including spin-ladder cuprate oxides and organic charge-transfer salts. We extended data analysis techniques based on information entropy, and analyzed signatures of local structure in neutron diffuse scattering and pair-distribution functions, and extended x-ray fine structure.

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Local Correlations, Superconductivity, and Stripe Fluctuations in Strongly Correlated Systems

98803

Don Parkin

Our primary objective is to investigate the nature of superconducting states with broken time-reversal invariance. This year we focused our effort on the major issue of marginal stability of unconventional superconductors and on the role of impurities and external perturbations on the induction of the secondary components of the order parameter. We were able to show how the secondary components are generated near magnetic impurity, as in the case of nickel-doped bismuth-2212 superconductor. We also investigated the role of stripe and phase separation on the structure of superconducting order parameter in correlated systems with competing interactions.

In a separate effort we investigated the correlations between superconducting transition temperature and the *q*-width of the peak at antiferromagnetic wave vector *Q*. We found a simple linear relation between the peak width and superconducting T_c ($T_c = \hbar v^* q$) with surprisingly small velocity ($\hbar v^* = 35$ meV·Å) for the yttrium-boron-carbon-oxygen superconductor.

We also presented a number of invited talks at both foreign and domestic conferences this year.

Publications

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

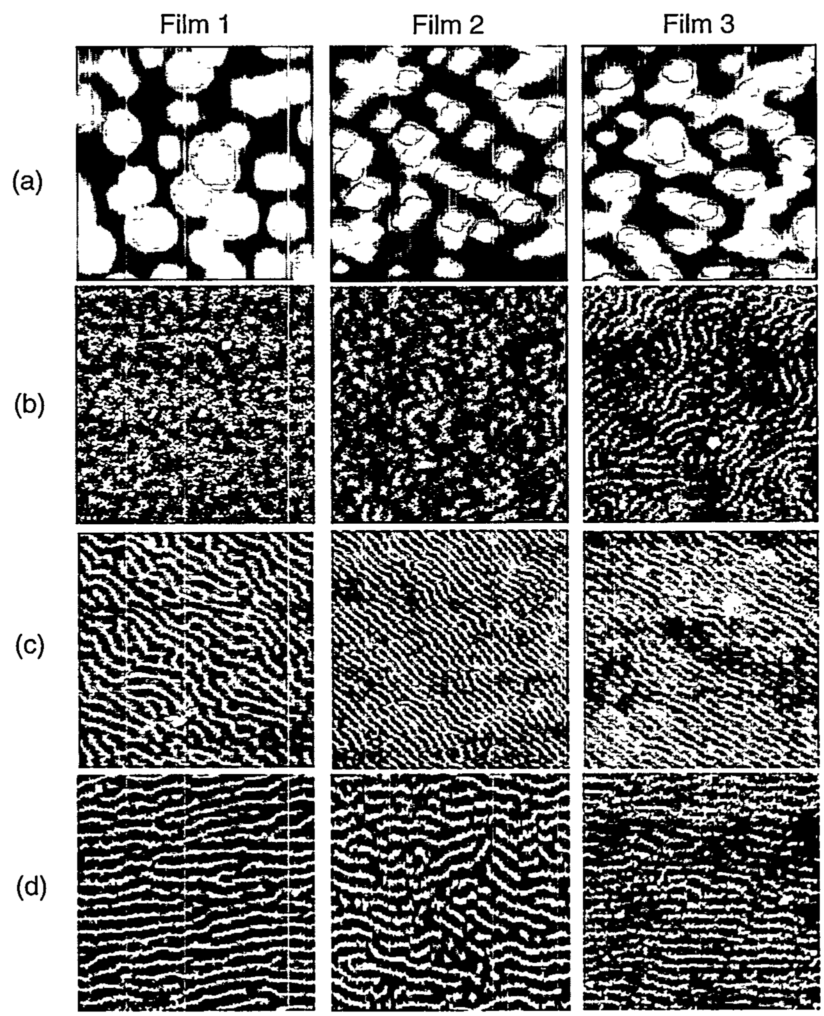
97804

Michael Stevens

Complex oxide films are increasingly important in a number of technical applications. Although the nucleation and growth of simple metal systems are reasonably well understood, this is not the case for oxide systems. The interplay of deposition variables and the role of substrate lattice mismatch have been, at best, controlled by trial-and-error methods without a complete understanding of the growth processes. By selecting the perovskites, carefully selecting a deposition matrix, and fully characterizing the films' physical properties, we hope to obtain fundamental information about the growth mechanism and resulting properties of these metal oxides. Ultimately, this method should yield valuable insight that will allow "engineered" films.

This year, we focused our efforts on two objectives: (1) to probe the relationships between growth parameters and structure, local and bulk properties, stress, and defects in strontium-doped, pulsed-laser-deposited, colossal magnetoresistive (CMR) films with potential application in the next-generation magnetic read heads and (2) to use atomic- and magnetic-force microscopies to study plastic-deformation-induced residual stress in amorphous FeSiB ribbons. For these objectives, we added a variable, external, in-plane magnetic field for in situ, field-dependent domain imaging.

For the CMR films, we were primarily interested in the role of lattice mismatch and growth processes in generating magnetic domains and defects that are expected to affect performance. Films were grown on $LaAlO_3$ (compressive) and $SrTiO_3$ (tensile) at 500°C–800°C. We observed substrate-dependent differences in transport, magnetization, and magnetic hysteresis despite indistinguishable structural differences. However, we saw magnetic maze domains for $LaAlO_3$ films associated with residual stress. These domains reverted to stripe domains in a field of ~160 Oe (see accompanying figure). We also studied the structure/magnetic property relationships of FeBSi and new multicomponent, magnetic amorphous ribbons.



Topographic and compressive (in-plane)/tensile (normal), stress-induced magnetic structure for three $La_{0.7}Sr_{0.3}MnO_3$ films grown on $LaAlO_3$ substrates at 800°C by pulsed-laser deposition: (a) 200- × 200-nm scanning tunnel microscopy images of the films' microstructure; (b) 4- × 4-μm magnetic force microscopy images of their corresponding magnetic domains; (c) the domains under a 160-Oe in-plane field applied 45° to the image horizontal; and (d) the domains after the field was removed. These materials have promise for use in soft magnetic sensor applications in which magnetic domains interfere with the response of the materials to weak magnetic fields, such as the stray field from small magnetic data bits. Understanding how the growth process creates these magnetic structures and how these structures respond to applied magnetic fields is key to developing them for these applications.

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Understanding and Controlling Self-Assembly

96636

Basil Swanson

To synthesize advanced materials for molecular electronics, nonlinear optics, and chemical and biological sensors, we seek to characterize and model functional self-assembled monolayers (SAMs), prepare and characterize inorganic/organic hybrid hierarchical materials, and develop an in-depth understanding of the self-assembly process. First, to provide a database for developing theoretical models, we apply synthetic techniques (e.g., solution-grown and microcontact-printed self-assembly and Langmuir-assisted assembly) to SAMs and then characterize these SAMs. Second, to characterize the mechanisms of assembly, we explore hierarchical, inorganic/organic hybrid materials in which the self-assembly process for both inorganic and organic subcomponents occurs simultaneously.

This past year we (1) directly characterized structures of bimolecular chain assemblies in a self-consistent series of pillared, layered, organic-inorganic, long-chain silver thiolates, (2) directly compared models for three-dimensional (3-D) hierarchical self-assembly with 2-D self-assembly, (3) characterized mobility and dynamics of chains in 3-D self-assembled silver thiolates and polysiloxanes, (4) developed Langmuir-assisted approaches to the preparation of SAMs, (5) studied and compared defects in solution-grown versus microcontact-printed SAMs, and (6) successfully modeled thioalkanes at the gold surface.

Publications

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Synthesis and Characterization of Correlated Electron Materials

96629

Joe D. Thompson

Our objective is to understand the competition and interplay among electronic, structural, and magnetic degrees of freedom in electronically correlated d-electron (lanthanum manganite) and f-electron (cerium- and uranium-based) materials. Our focus is on three important scientific issues: (1) How does this competition control the metal-insulator transition in d-materials? (2) How can the local structure be probed? (3) What conditions induce non-Fermi-liquid behavior in f-electron systems?

From x-ray absorption measurements we have discovered a functional relationship between a local-structure distortion and the magnetization, resistivity, and ferromagnetic transition temperature in a series of three-dimensional manganites. We also established, from a study of the spin dynamics in two-dimensional and three-dimensional manganites, that dimensionality affects the spatial extent of local-structure distortions. After analyzing neutron pair-distribution data, we have shown that long-range charge ordering is lost while charge localization is preserved in a three-dimensional manganite. Through x-ray absorption and magnetic resonance studies, we have demonstrated that non-Fermi-liquid behavior arises from disorder in f-electron systems—even in material believed to be chemically/structurally ordered. We have connected disorder to phase transitions in plutonium and worked closely with a companion theoretical project to close the loop on synthesis-characterization-modeling.

Publications

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

97810

Rex Hjelm

A knowledge of membrane stability in aqueous solutions is needed in order to understand the self-assembly of membranes. On theoretical grounds, this stability is due to spontaneous curvature and bending rigidity—whether the membrane comprises stacked, infinite bilayer sheets (lamellae) or closed, finite bilayer sheets (vesicles). The effects of surface tension are also thought to come into play through the work required to change the volume and surface area. However, these ideas have not been subjected to rigorous experimental tests. Our objective is to design such experimental tests, using small-angle neutron scattering (SANS) to measure the changes in vesicle versus lamellar phase stability with composition, temperature, and pressure in mixed bile salt–phosphatidylcholine solutions in the isotropic region of the phase map.

This year we demonstrated that there are reversible temperature-driven transitions between vesicles and rodlike micelles and reversible pressure-driven transitions between vesicles and stacked lamellar phases in a pressure range predicted by the Young-LaPlace equation. However, the pressure-driven transition is not complete at elevated temperatures (>33°C) and does not occur at all if a small amount of alcohol is present. These observations suggest that the standard model of vesicle stability is not relevant to mixed solutions. We are completing a thermodynamic analysis of the data, opening the way to demonstrate a new model of vesicle stability in mixed solutions.

Publications

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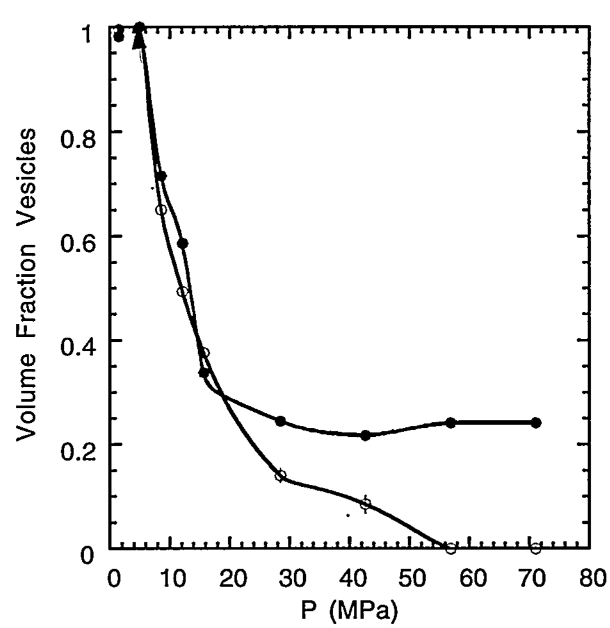

Volume fraction of vesicles with pressure and temperature. Data is for T = 15°C (open dots) and T = 33°C (solid dots), showing the temperature-related differences in the vesicle transition. Note that the transition is not complete at 33°C.

Tailoring the Interfacial Electronic Structure of Organic Electronic Materials and Devices

96637

Ian Campbell

Our objective is to learn to tailor the electronic structure of interfaces between a metal and an organic electronic material in order to control the electrical transport properties of that interface. Metal/organic interfaces are the essential active element of every organic electronic and electro-optic 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. There are a great many design options available for organic electronic materials and devices both at the molecular and mesoscopic scales. Because metal/organic interfaces are the essential active elements in every organic electronic device, the ability to tailor these interfaces to control their interfacial charge transport properties is the key to realizing the potential of organic electronic materials and devices.

There are two basic elements of the interfacial electronic structure involving organic electronic materials: (1) the relative positions of characteristic energy levels in the two constituent materials forming the interface and (2) cross sections for charged excitation transfer across the interface. In this project, we used self-assembly techniques to insert a molecular coupling layer between the metal and organic interface constituents and to investigate how the coupling layer can be used to tailor these elements of the interfacial electronic structure. For example, we manipulated the relative energy levels of the constituent materials using molecular coupling layers with electric dipoles. We carried out these investigations using a combination of experimental and theoretical methods.

Publications

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Structure-Property Relationships in Elasticity and Plasticity

97805

Michael Stevens

We are generating a comprehensive model of the mechanical response of materials, including elasticity and thermal expansion, anelasticity and viscous behavior, creep and plasticity, and eventually recovery and recrystallization. We initially base such modeling on microstructural observations but will carry it to a macroscopic scale. The end result is "average constitutive relations" at the large scale, derived from those at the small scale. Because the macroscopic modeling is based on physical fundamentals, it should be applicable beyond the range of experimental verification. Our models generate new tools for a broad range of internal and external Laboratory programs.

This year we developed an improved recrystallization and implemented it in the viscoplastic self-consistent (VPSC) polycrystal code. VPSC is a general code for predicting plastic deformation of aggregates. Using this application, we predicted deformation and recrystallization textures of calcite, showing that recrystallization alone can explain the observed textures without having to involve deformation twinning. This study resulted in a publication. We also implemented the VPSC polycrystal code in the finite element code EPIC and applied it to simulate compressive forming of zirconium, thus demonstrating the feasibility of incorporating constitutive laws based on physical mechanisms into finite element forming codes.

Publications

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Advanced Research Capabilities for Neutron Science and Technology—New Polarizers

98609

Seppo Penttila

High-powered, short-pulse neutron sources, such as the spallation source at the Los Alamos Neutron Science Center (LANSCE), require novel instrumentation to fully utilize their high instantaneous neutron fluxes. A new broad-band neutron polarizer, the optically polarized helium-3 neutron spin filter, is a technology that is well suited for these sources. The helium-3 spin filter can be used to polarize neutron beams in the large energy range up to epithermal energies where the traditional polarizers do not work.

The simple relationship between helium-3 polarization and transmitted neutron beam polarization allows for a new and reliable way of measuring and monitoring online the absolute polarization of the neutron beam to accuracies of 0.1% or below. In addition, with the time-of-flight measurement, components of the beam backgrounds such as gamma rays can be determined precisely. The well-understood beam and the accurate beam polarization open a number of new exciting opportunities in fundamental physics as well as in condensed-matter neutron scattering.

The goal of the project is to continue the development of the optically polarized helium-3 technology and to build a neutron spin filter that can be used in fundamental physics and condensed-matter scattering experiments at LANSCE. We have built helium-3 spin filters based on the rubidium spin-exchange method. During the '97–'98 LANSCE run cycle, we used the spin filter to measure the neutron beam polarization with an absolute accuracy of 0.2% in the energy range from 40 meV to 10 eV. This is one of the most accurate beam polarization measurements in the field.

We used two spin filters in an experiment at LANSCE to measure neutron spin rotation on lanthanum-139. One spin filter was used to polarize the beam, and the other one was used to detect a small rotation of the neutron spin direction. With the personnel from the Manuel Lujan Jr. Neutron Scattering Center, we have also proposed to measure the lateral correlation length of magnetic roughness in iron-chromium multilayers using the helium-3 spin filter to analyze polarization of diffusively scattered neutrons.

Publications

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Nuclear Microprobe Analysis of Impurities at Grain Boundaries

98801

Carl Maggiore

The objective of this project is to extend the capabilities of the Ion-Beam Materials Laboratory (IBML), the nuclear microprobe, and the electron microscope (EM) facility to the study of impurities at grain boundaries. Quantitative measurements of impurities and defects in materials are essential for a complete understanding of the mechanical behavior of materials under extreme conditions, and this project enhances our ability to make these important measurements.

We have extended our fundamental measurements of the energy-dependent cross sections of light elements (beryllium, boron, carbon, nitrogen, oxygen) with high-energy alpha particles. These measurements are essential for the quantitative, subsurface analysis of light elements in the near surfaces of materials.

We have also measured the absolute x-ray-production cross sections for excitation with deuterons, helium-3, and alpha particles and confirmed the validity of the analytical codes for the measurement of trace elements in complex materials. Correlation with primary standards from the National Institute of Standards and Technology has been verified, and the enhanced analytical capability is now routinely available.

Principal achievements at the EM facility this year have been (a) a new x-ray detector for the analytical EM to improve resolution, (b) a plasma cleaner for removing organic contamination from thin transmission-EM specimens, and (c) a new mechanical polishing system for initial specimen preparation. A wide variety of applications for the high-resolution EM, analytical EM, scanning transmission EM, and scanning EM have been seen. Noteworthy is the discovery of a number of unique incommensurate and commensurate structures in rhenium disilicide (a potential infrared-detector material), textured grains in thick films of yttrium-barium-copper-oxygen superconductor with high critical current capacity, and unusual stacking fault configurations in Laves phase alloys.

Controlling Function of Polar Organic Multilayers

96638

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 (such as conjugated polymers) linked to fullerenes for ultrafast photo-induced intermolecular charge transfer and (2) porphyrin donor/acceptor molecules used for directed energy transport. Our focus is on ultrafast excited-state spectroscopy with the following goals: to study the relationship between photoexcited charge transfer and molecular nonlinearities, to develop nonadiabatic dynamical models that describe electronic states following photoexcitation, and to understand the effects of orientation and molecular structure on charge and energy transfer in polar multilayer structures.

This year, we demonstrated rapid multilayer formation using ionic bonds of several molecular species, including conducting polymers, fullerenes, porphyrins, and phthalocyanines. We have designed and manufactured superlattices that show controlled charge and energy transfer among adjacent layers. Most notably, we have observed collective effects of chromophore reorganization and long-range charge-transfer quenching as multilayers are built up (see accompanying figures). We have studied the photo-induced charge and energy transfer using a variety of spectral probes, including absorption and emission spectroscopy as well as ultrafast time-resolved methods. In our modeling efforts, we studied (1) stability of bipolarons near a polymer-metal interface, (2) formation of excitons in organics in the strong electronic correlation regime, (3) charge transfer in molecularly doped polymers, and (4) enhancement of optical nonlinearity in polyenes in the presence of localized vibrational excitations (breathers) using quantum chemical techniques.

Publications

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Li, D.Q., et al., "Preparation, Characterization, and Properties of Mixed Organic and Polymeric Self-Assembled Multilayers," *J. Am. Chem. Soc.* **120**, 8797 (1998).

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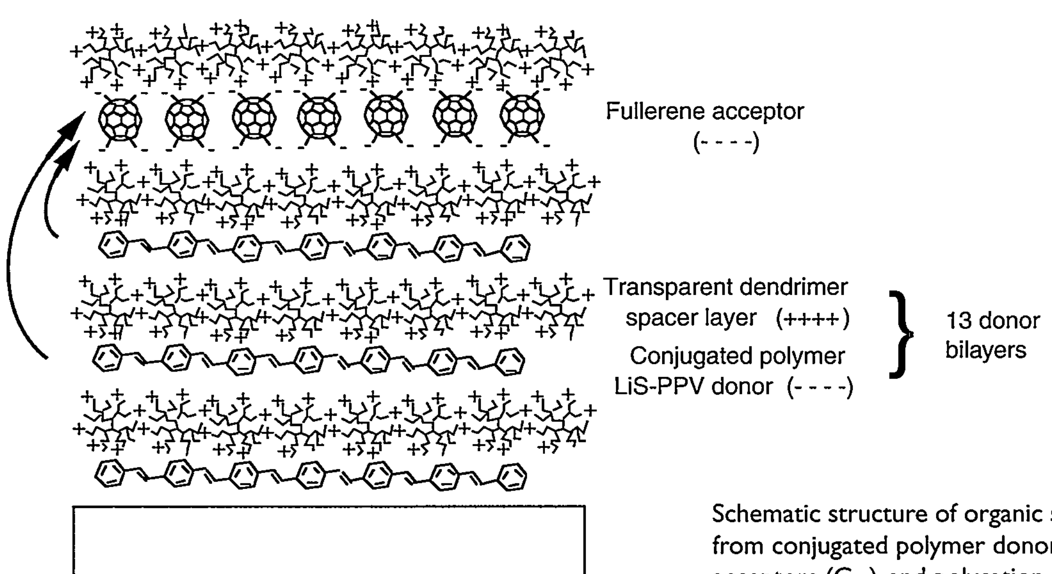
Mashl, R. J., et al., "Theoretical and Experimental Adsorption Studies of Polyelectrolytes on an Oppositely Charged Surface" (to be published in *J. Chem. Phys.*).



Schematic structure of organic superlattice built from conjugated polymer donors (LiSPPV), fullerene acceptors (C_{60}), and polycation binder layers.

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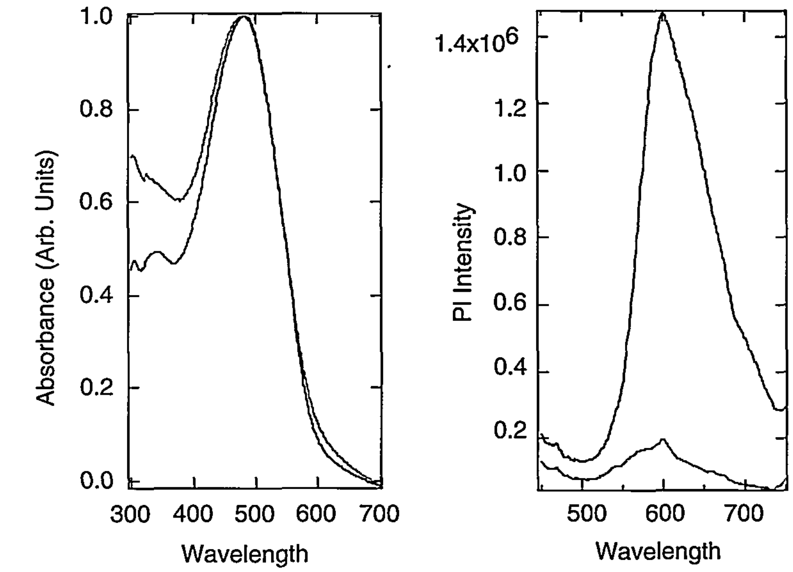
Yu, Z.G., et al., "Variational Studies of Charge Transfer in a Molecularly Doped Polymer," *SPIE Proc., Fullerenes and Photonics IV* **3145**, 303 (1997).



Demonstration of quenching of luminescence from LiSPPV/polycation bilayer as a fullerene layer is placed on top, followed by multiple (3,5,7) LiSPPV bilayers, followed by another fullerene layer.

Studies of Ultrahigh-Strength Materials

97619

J. Embury

Our objective is to explore both experimentally and theoretically the behavior of materials at stresses close to their theoretical limits. This effort involves preparing and examining ultrafine-scale structures by a variety of fabrication methods.

This year we concentrated on wire drawing of in situ composites such as copper-silver and copper-niobium. These materials can be used as high-strength conductors for fabricating the coils of high-field magnets. Some of this work was documented as a journal article and has appeared in conference proceedings. Theoretical and computational studies of copper-silver composites indicate that the mechanical response of these materials is dominated by dislocation generation at interfaces. These results are consistent with the experimental observations made with high-energy electron-microscopy imaging.

We fabricated materials by melting alloys in glass and drawing them into filaments at high temperatures using a method known as the Taylor wire technique. We have drawn copper-silver and copper microwires with this technique. We found in this system unique nanostructures, such as unusual structural twins and super-saturated solid-solution nanocrystals, and documented some of this work in a journal article. In addition, we investigated fine-scale platinum-iron multiple layers with iron stabilized in the face-centered-cubic iron form. We found very high hardness values in this system resulting from layer thickness and lattice distortions resulting from epitaxy. Some of this work was published in a conference proceedings.

Publications

Embury, J.D., and K. Han, "Conductor Materials for High Field Magnets," *Curr. Opin. Solid State Mater. Sci.* **3**, 304 (1998).

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Han, K., et al., "Fabrication Routes for High Strength High Conductivity Wires," in *Proceedings of the 7th International Conference on Megagauss Magnetic Field Generation and Related Topics, 1998* (World Scientific, River Edge, NJ, in press).

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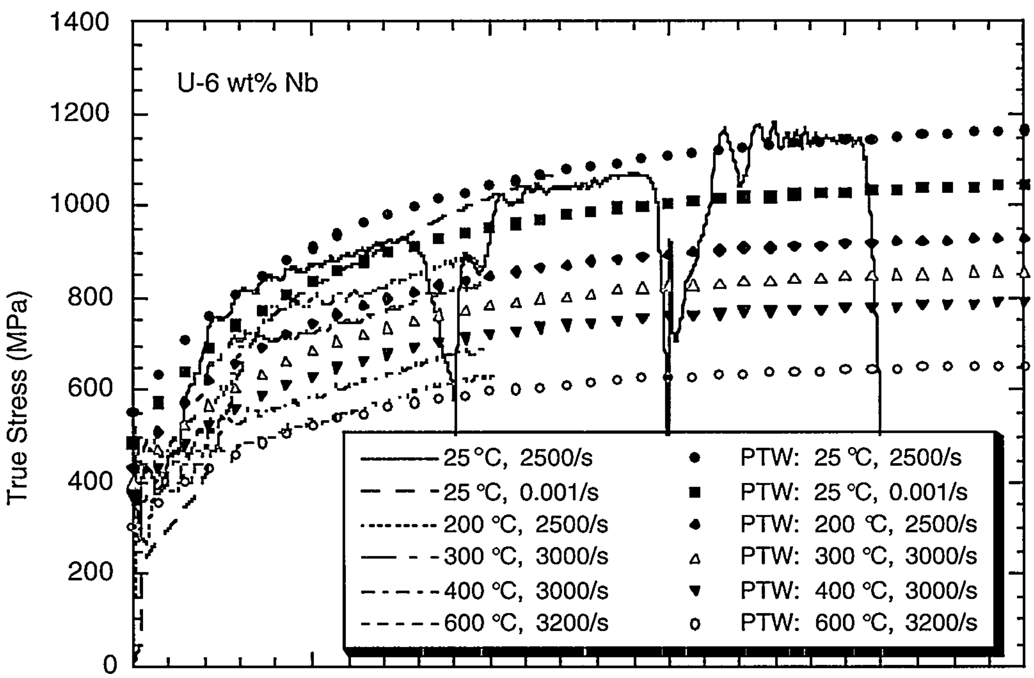
Dynamic, High-Strain Deformation of Metals: Experiments, Advanced Constitutive Modeling, and Computational Implementation

96642

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 mechanical threshold stress and Preston-Tonks-Wallace (PTW) strength models using new Hopkinson bar data at elevated temperatures and 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 three-dimensional (3-D) MD simulations of vacancy formation by a moving, jogged dissociated dislocation in copper at low temperatures. A 3-D code was developed for the computation of elastic energies during dislocation intersection. We formulated a new method to calculate the mobility of dislocations as a function of applied stress, temperature, and point-defect concentration. The constitutive behavior in U-6 wt% Nb was measured and analyzed and used as the basis for fitting the large-strain response to the PTW model (see first figure). We applied a 3-D, 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 stress propagation of stress waves, the shape and evolution of the yield surface, and hardening. Finally, we validated the modeling by geometrically comparing it to Taylor impact specimens for titanium (second figure), zirconium, and U-6 wt% Nb. We also conducted MD simulations of the defect generation behind shock fronts in face-centered-cubic crystal structure metals.



Stress-strain response of U-6 wt% Nb as a function of temperature and strain rate. These data and the material model developed are directed at providing Los Alamos with the ability to perform predictive numerical simulations of dynamical behavior by synthesizing material strength with condensed-matter theory and mesoscale physics to construct advanced constitutive models.

Publications

Chen, S.R., et al., "Mechanical Properties and Constitutive Relations for Molybdenum under High-Rate Deformation," *Molybdenum and Molybdenum Alloys,* A. Crowson, et al., Eds. (The Minerals, Metals, and Materials Society, Warrendale, PA, 1998), p. 155.

Holian, B.L., and P.S. Lomdahl, "Plasticity Induced by Shock Waves in Nonequilibrium Molecular Dynamics Simulations," *Science* **280**, 2085 (1998).

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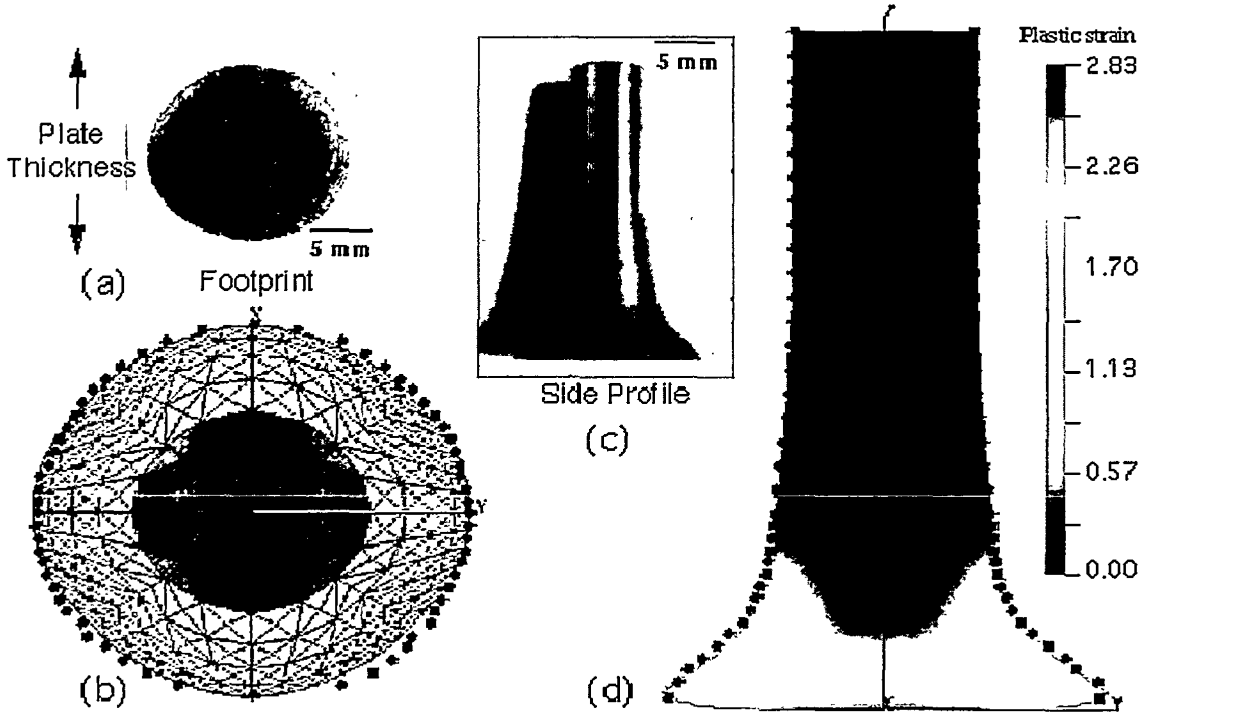

Constitutive modeling validation of the large-strain, high-strain-rate mechanical response of tantalum using a Taylor-cylinder impact test. The anisotropic nature of the mechanical behavior of the tantalum plate used in this study is seen to result in an ellipsoidal-shaped, Taylor-cylinder foot shape. This behavior is accurately captured using a rate-sensitive anisotropic constitutive model.

Chemistry and Microstructure of High-Temperature Superconductor Interfaces

96625

Quanxi Jia

Josephson junctions are the key components used in producing exceptional magnetic sensors and sophisticated circuitries that are useful in computers. The interface chemistry between a high-temperature superconductor and the normal-metal layers is one of the most important issues in the fabrication of reproducible and controllable ramp-edge superconductor/normal-metal/superconductor (SNS) Josephson junctions. Both the interface chemistry and the microstructure are controlled by engineering the superconductor electrodes and normal-metal materials to tune the performance of the device. Engineering involves investigating chemical dopants in the materials, exploring defect structures near the interface between the normal metal and the superconductor, processing parameters during device fabrication, and laying out the device design.

Last year we continued to investigate directly coupled superconducting quantum interference device (SQUID) magnetometers on $LaAlO_3$ substrates fabricated using ramp-edge SNS junctions. Silver-doped $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ were used for the electrode and the normal-metal barrier, respectively. The flux noise of the SQUID magnetometer, tested under a dc current bias at 75 K, was below $10^{-5}\ \Phi_0\,Hz^{-1/2}$ at kilohertz frequencies. The field sensitivity of the magnetometers increased with increased pick-up loop area, as expected. Importantly, the field noise of the SQUID magnetometers increased by less than a factor of 2 in the background of the earth's static magnetic field. The magnetometers were used successfully in our underground radio project that won a 1998 R&D 100 Award.

Our research has also led to identification of a new class of compounds $(Pr_yGd_{0.6-y})Ca_{0.4}Ba_{1.6}La_{0.4}Cu_3O_7$ (y = 0.4, 0.5, and 0.6) that is suitable for the normal-metal barrier in ramp-edge SNS junctions.

Publications

Jia, Q. X., et al., "Development of Ramp-Edge SNS Josephson Junctions and SQUIDs" (to be published in *Mater. Sci. Eng., B*).

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Pinning Vortices and Enhancing High-Temperature Superconductor Critical Currents

96626

Martin Maley

Maintaining low resistance in magnetic fields at high current densities is the main problem for applications of high-temperature superconductivity (HTS), requiring dense arrays of nanoscale defects to "pin" the vortices created in the magnetic fields. Amorphous columnar defects (CDs) created by fast heavy ions provide the optimum pinning of vortices in HTS. Our objective is to elucidate the mechanisms of vortex pinning by studying static and dynamic properties of vortices in the presence of CDs. This will guide us in efforts to enhance critical current densities in HTS. We have employed 1.0-GeV uranium-238 ions to create arrays of CDs in single crystals of bismuth-2212.

Our focus this year was to study the effect on the transport properties of filling the CDs with vortices. At low fields, the vortices can easily "hop" between unoccupied CDs, and the transport of vortices is expected to mimic variable-range hopping (VRH) in semiconductors. We observed that at high temperatures the vortices in irradiated bismuth-2212 enter a melted mobile state described by VRH. For the first time, we observed a reentrant behavior in the magnetoresistance: when the magnetic field approaches the matching field where densities of vortices and CDs are equal, the magnetoresistance first increases with magnetic field, then decreases, reaches a minimum, and increases at higher fields. This behavior indicates that vortices are localized on CDs even in the liquid state and that their mobility is severely reduced with the density of unoccupied sites near matching.

Publications

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Bulaevskii, L.N., et al., "Vortex Phases in Irradiated Highly Anistropic Layered Superconductors," *Phys. Rev. B* **57**, 5626 (1998).

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Comparative Investigation of Spin, Charge, and Lattice Degrees of Freedom in Three Classes of Colossal Magnetoresistive Materials

98817

Dimitri Argyriou

Our goal is to isolate the effects of local structure on the spin, charge, and lattice dynamics in three classes of colossal magnetoresistive (CMR) materials, each class possessing different degrees of local structure. To achieve this goal, we are using neutron scattering to investigate the spin and lattice degrees of freedom, and muon spin relaxation as a complementary probe of spin dynamics. We are also using both crystal chemistry and crystal structure to decouple the combined effects of spin, charge, and lattice. The first step in our project is to grow high-quality single crystals of various manganite compositions for neutron and muon spin relaxation measurements.

This year we used an optical floating-zone technique to grow high-quality single crystals of the layered CMR manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$. We are now using a variety of techniques to characterize these materials before muon or neutron experiments are carried out.

We also used neutron scattering to investigate the structure and magnetism in the layered manganite with $x = 0.3$, finding that this composition separates into an antiferromagnetic and a ferromagnetic phase. Furthermore, the structural data suggest that direction of the electron-phonon coupling is different compared to the prototypical $x = 0.4$ composition. These variations suggest changes in the stabilization of the d-orbital of the manganese ions.

Lastly, we carried out zero-field, muon-spin-resonance experiments in single crystals of $La_{1.4}Sr_{1.6}Mn_2O_7$ for $T = 5$ to 325 K. The spin-lattice relaxation rate is spatially inhomogeneous below the three-dimensional (3-D) magnetic-transition temperature T_C and anisotropic above T_C. We find evidence against 2-D spin ordering or in-plane correlations above T_C. Additionally, the very slow spin fluctuations found below T_C in cubic perovskites like $(La,Ca)MnO_3$ or $(La,Sr)MnO_3$, attributed to relatively small magneto-elastic polarons, are absent in $La_{1.4}Sr_{1.6}Mn_2O_7$. This absence suggests that the polaron size just below T_C in the layered material is significantly larger than in the 3-D perovskites, a consequence of the reduced dimensionality.

Publications

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Microstructure and Microanalysis of Materials at Atomic Resolution

97806

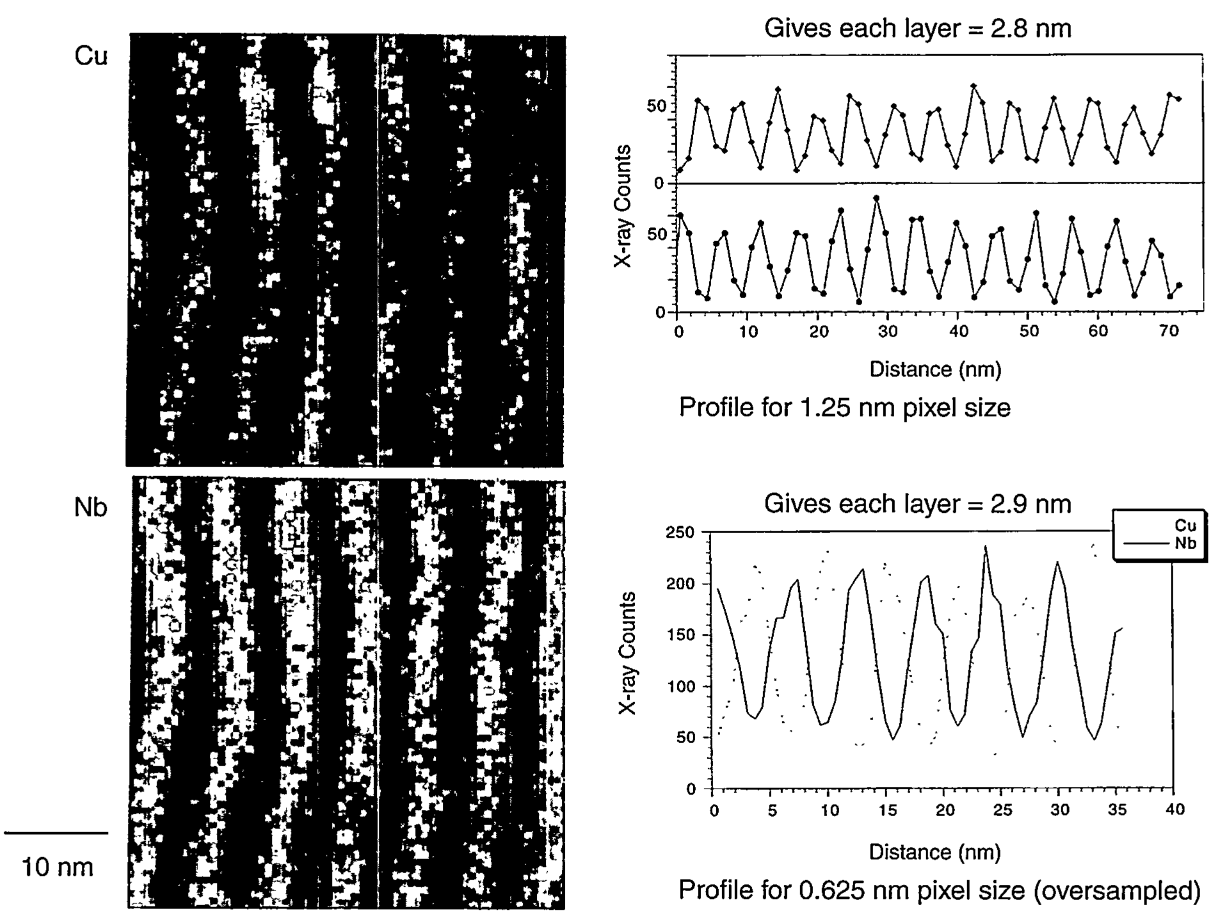
Terence Mitchell

The objective of this research is to use the most advanced techniques in transmission electron microscopy (TEM) to solve important problems in materials science. Such problems include understanding the relationship between the microstructure and current-carrying capability of high-temperature superconductors and investigating the reason for the strength of high-temperature ceramics and intermetallics. In our research, we are using high-resolution TEM to image atomic arrangements around defects to a resolution approaching 1 Å. Combining scanning TEM with a probe size of less than 10 Å with x-ray energy-dispersive spectrometry (XEDS) and electron energy-loss spectrometry (EELS) allows chemical analysis at the near-atomic level.

Many results have been obtained over the last year, as indicated by our extensive list of publications. For example, we examined cross sections of Cu/Nb multilayers. High-resolution TEM shows that, for layer thicknesses around 10 Å, the copper is bcc (body-centered cubic) rather than fcc (face-centered cubic). XEDS in combination with scanning TEM shows that the layers are atomically sharp (see accompanying figure), which means that the copper templates on the niobium (i.e., mimics the niobium's bcc structure).

To study site occupancies in ternary AB_2 Laves-phase alloys, we developed a means of locating atoms with channeling enhanced microanalysis (using XEDS and multivariate statistical analysis with delocalization correction). We have found that in $NbCr_2$+V alloys, niobium exclusively occupies the A site while chromium and vanadium occupy the B site. By contrast, for HfV_2+Nb, niobium partitions between the two sites.

Finally, with high-resolution TEM and electron diffraction, we explored a remarkable series of incommensurate and commensurate structures that



Left panels: x-ray maps showing the distribution of copper and niobium in a Cu/Nb multilayer. The dark regions indicate high concentrations of copper (top map) and niobium (bottom map). Right panels: line profiles through the Cu/Nb cross section for two different pixel (probe) sizes; the narrowness of the curves indicates that the copper and niobium boundaries are atomically sharp.

we have discovered in the $ReSi_{2-x}$ intermetallic. These structures are very important for understanding and developing the material's narrow band-gap semiconducting properties.

Publications

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Lu, P., et al., "Microstructural Characteristics of Conductive $SrRuO_3$ Thin Films Formed by Pulsed-Laser Deposition," *J. Mater. Res.* **13**, 2302 (1998).

Misra, A., et al., "Effects of Ion Irradiation on the Residual Stresses in Cr Thin Films," *Appl. Phys. Lett.* **73**, 891 (1998).

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Misra, A., et al., "Incommensurate Structures in Rhenium Disilicide," *Microsc. Microanal.* **4**, 534 (1998).

Misra, A., et al., "Structure and Mechanical Properties of Cu-X (X=Nb, Cr and Ni) Nanolayered Composites," *Scripta Mater.* **39**, 555 (1998).

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Advancing X-Ray Hydrodynamic Radiography: Advanced Cathodes

97605

Daniel Prono

For advanced hydrodynamic radiography, the spot size of the x-ray source is the most influential parameter in obtaining fine-feature resolution. The x-ray spot size is determined by the electron beam's interaction with the bremsstrahlung converter target and by the electron beam's transverse motion, current-density profile, and emittance. The beam's transverse motion is determined by how the electron beam is transported through the accelerator structure, but the beam's profile and emittance are largely controlled by the emission process from the cathode source. A photocathode (a cathode electron source whose emission is controlled by short-wavelength laser illumination that causes stimulated electron emission) is an ideal source since it is an intrinsic low-emittance source and its emitted electron current-density profile can be controlled by the laser intensity profile. Because the next-generation accelerator designs feature high currents and require large-area cathode surfaces, the challenge is to make a high quantum-efficiency (QE) cathode surface that minimizes the laser intensity and operates in a moderate vacuum environment (most photocathodes operate only in ultrahigh vacuum).

Last year we demonstrated QEs of >0.4% for diamond-film photocathodes, efficiencies that are at least 6 times greater than those of conventional photocathodes. This year we focused on solving fabrication problems associated with bonding a diamond-film interface to a silicon semiconductor surface and terminating the diamond vacuum surface with hydrogen so that it (1) has a negative electron affinity so as to achieve a low work-function emission, (2) is chemically inert so as to operate in a modest vacuum, and (3) can withstand the intense UV illumination without ionizing the hydrogen.

We have solved the bonding problem by using a metalization technique developed for the semiconductor industry to give uniform surface conductivity for its chips. The termination issue is still being addressed, but we have promising preliminary results using submonolayer deposition of various metals (e.g., zirconium) as a substitute for hydrogen termination. (Zirconium still has negative electron affinity but is also robust to ionization.) The work to develop a stable surface termination will continue next year with emphasis on uniform electron surface-barrier conduction for large-area cathodes and uniform electron conduction for long-pulse durations.

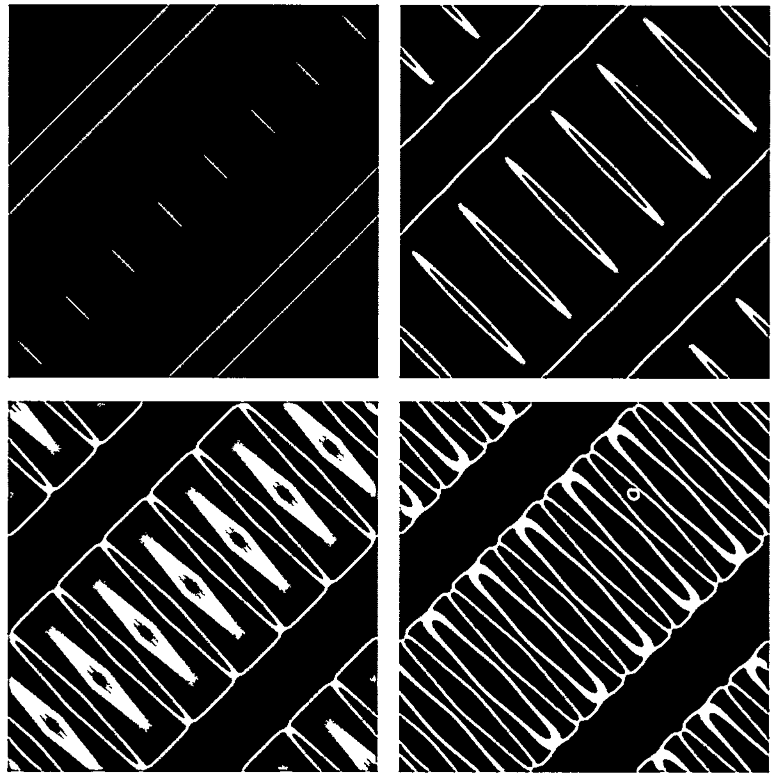
Multiscale Phenomena in Materials

97802

Alan Bishop

We are developing and supporting a technology base in nonlinear, nonequilibrium phenomena underpinning fundamental issues in condensed matter and materials science, and applying this technology to selected problems. We are focusing on those areas in the materials community for which nonlinear and nonequilibrium approaches will have decisive roles and where productive teamwork among elements of modeling, synthesis, characterization, and applications can be anticipated. This particularly includes multiscale and nonequilibrium phenomena and complex matter in and between fields of soft, hard, and biomimetic materials. Principal topics are (1) complex organic and inorganic electronic materials including hard, soft, and biomimetic materials, self-assembly processes, and photophysics; (2) microstructure and evolution in multiscale and hierarchical materials (see accompanying figure) including dynamic fracture and friction, dislocations and large-scale deformation, metastability, and inhomogeneity; and (3) equilibrium and nonequilibrium phases and phase transformations with emphasis on competing interactions, frustration, landscapes, glassy and stochastic dynamics, and nonlinear energy focusing.

This year we developed and applied large-scale molecular dynamics and induced textures models. We developed and applied fully constrained, continuum, nonlinear elasticity modeling to solid-solid phase transformations exhibiting hierarchical textures and dynamics. We applied new methods for handling long-range interactions to physical problems in polyectrolytes (adhesion, aggregation), doped transition metal oxides, and flux flow in layered superconductors. We demonstrated new mechanisms for energy localization and transduction in discrete nonlinear classical and quantum systems, and we began their applications to friction and time-resolved photoexcitation in electronic and biological materials. We also organized international conferences on "self-assembling and biomimetic materials" and "predictability," and attracted senior visitors, postdoctoral fellows, and graduate students in the area of multiscale materials science.



Stress-induced evolution of two generations of twins with two martensitic variants. Top left panel: small white lines are the initial "seeds" of one variant. Top right panel: subsequent emergence of one generation of twins along the diagonal. Lower panels: strain embryos between the initial twins lead the nucleation of a new generation of twins.

Publications

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Manipulation of Residual Stresses to Improve Material Properties

97809

Mark Bourke

The objective of this project is to study the effects of residual stresses, generated by phase changes, on material properties. Nickel spinel is used as a model system because it undergoes a significant volume shrinkage during reduction to a ceramic matrix composite of alumina and metallic nickel. Experimentally, we are monitoring the residual stress generation using x-ray diffraction (XRD) and in situ neutron diffraction (ND), while the results are complemented by finite element simulations.

Initially, XRD measurements were made on a sample geometry, designed to produce a compressive residual stress in an alumina disk by surrounding it with a spinel annulus and then reducing the nickel spinel. Because of degradation of the surface during manufacture, the XRD measurements showed substantial uncertainties. Therefore, we made spatially resolved ND measurements. Although the ND measurements employed a larger sampling volume (and thus had poorer spatial resolution), they did not suffer from the poor repeatability associated with the surface XRD measurements. Moreover, the trends of the ND results were qualitatively similar to the XRD results and amenable to simulation with the finite element method.

In a separate study of the kinetics of the reduction, we made in situ measurements of the residual stress generation in nickel spinel. In an unexpected result, it became evident that the disorder of the spinel changes during the reduction. This complicated our analysis because the change disorder causes a change in the lattice parameter, which can be misinterpreted as a mechanically induced strain. Currently, work is under way to quantify this effect, and thus to provide a route to decouple it from lattice parameter changes associated with shape incompatibility of the respective phases.

Publications

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Chemistry

A New Paradigm in Separations: Molecular Recognition Membranes

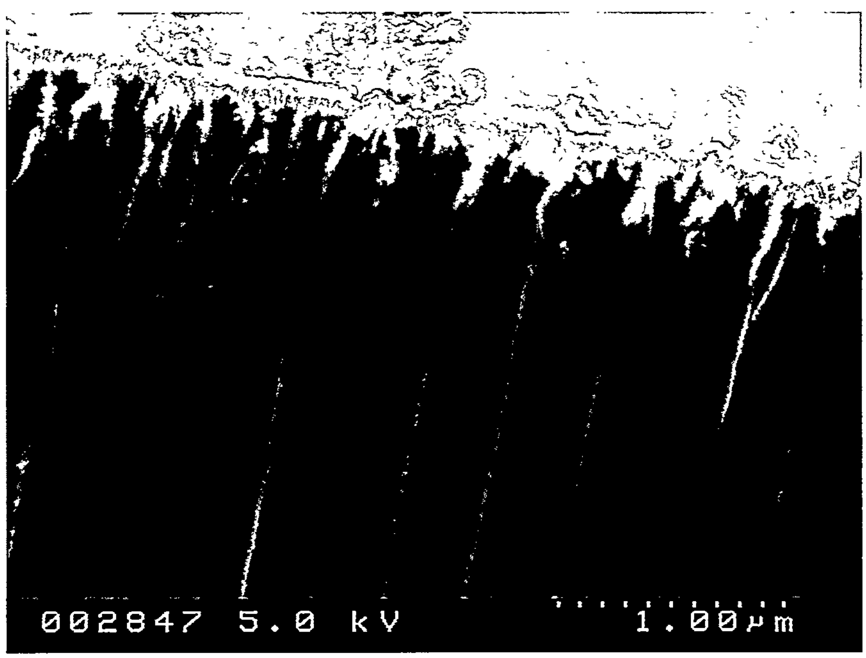
98602

Gordon Jarvinen

The objectives of this project are to understand the molecular-level mechanisms that control transport of ions and molecules through semipermeable membranes and to develop novel molecular recognition membranes. Using porous materials with well-defined structures, we are constructing arrangements of ionic and molecular receptors in the pores to gain this understanding.

Commercially available alumina membranes have served as the rigid framework structure for most of our work this year. We have measured the rates of diffusion through these membranes of salts of alkali and alkali earth-metal ions in aqueous solution. At pH values in the 5–8 range and in solutions of a single salt, the divalent ions calcium and strontium diffuse through the membrane at a rate 10–20 times slower than the alkali metal ions. With mixtures of salts, we have observed that cesium and sodium substantially speed up the transport of calcium and strontium, but lithium, potassium, and rubidium have little effect. We are using both experimental and theoretical methods to investigate this unexpected facilitation of the diffusion of the divalent cations.

In addition, we have coated the alumina membranes with gold of various thicknesses to allow further functionalization (see the figure). The gold-coated openings to the pores have been "plugged" with amine functionalized polymers that adhere only to the gold surface. We have observed that the polymers serve as a selective gate that can greatly slow diffusion of large cationic complexes. This approach of docking a preformed selective structure in the pores represents a novel and potentially efficient route to molecular recognition membranes that we are evaluating.



A scanning electron micrograph of a cross section of a gold-coated alumina membrane showing the straight nonintersecting pores in the alumina of diameters averaging 0.2 µm. The 0.05-µm-thick gold coating was deposited using an electron-beam evaporator with a 90° incidence angle. The gold deposit is visible as a fairly uniform layer on the surface of the alumina, as small clumps near the mouths of the pores, and as larger masses piled on the alumina surface.

Publications

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Energy Transfer in Molecular Solids

97807

Juergen Eckert

The goal of this research is to significantly advance our understanding of energy transfer processes and vibrational dynamics in molecular crystals of two important classes, those closely related to high explosives (HEs) and to biological model systems containing peptide hydrogen-bond chains. We are utilizing inelastic neutron scattering (INS) vibrational spectroscopy in combination with theoretical modeling of the INS spectra because they can be related to molecular interactions in a much more direct way than other spectroscopic techniques.

In HEs, it is the molecular-level details of how mechanical energy from a shock wave is transferred to chemical energy in individual molecules that must be elucidated. Virtually all microscopic models of such vibrational energy transfers require detailed knowledge of the molecular interactions to derive vibrational densities of state and energy transfer rates. We are now using *ab initio* calculations of vibrational frequencies rather than using empirical force fields and have carried out high-level calculations for a number of molecules including the high explosives TATB, RDX, and HMX. Comparison of INS spectra computed from such calculations with experiments clearly demonstrates that intermolecular interactions involving clusters of molecules must be included. Our calculations on formamide clearly give a much more reasonable representation of the observed spectrum with 34 molecules than with the isolated molecule.

For biomolecules, it is the mechanism of long-range energy transport that continues to be unresolved despite its importance in the function of proteins, for example. Our INS, structural, and theoretical studies of simple systems with peptide hydrogen bonds (formamide, L-alanine, N-methylacetamide, acetanilide) have now conclusively shown that proton transfer (along the peptide hydrogen bond) cannot be responsible for this mechanism and favor models based on polarons.

Publications

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Condensed Phase Kinetics and Reduced Reaction Mechanisms of Energetic Materials

98603

Steven Son

The objective of this project is to develop a competency to predict reaction processes in energetic materials, in particular octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX), using detailed and reduced chemical kinetics schemes. Detailed chemistry experiments and calculations are being performed to contribute to the ultimate aim of developing simple, yet rigorous, models. There are four interacting components to this project: (1) fundamental decomposition measurements, (2) detailed and reduced kinetic schemes, (3) modeling of the coupling of transport with kinetics, and (4) integrated chemical and transport experiments.

This year we made progress in each of these areas. Specifically, the following are our major accomplishments: (1) We selected paths and intermediates for study and began initial studies of these chemical pathways. (2) We collected available chemical data and explored and applied reduction methods. In particular, intrinsic low-dimensional manifold methods were explored. (3) We adapted a detailed chemistry code to simulate steady and unsteady burning of HMX. (4) We developed a novel nonlinear optical technique to study the kinetics of a solid-solid phase transition that occurs in heated HMX. This application of second-harmonic generation (SHG) to HMX and other energetic materials constitutes a fundamentally new probe in the dynamics of energetic material decomposition, ignition, and combustion and may have application to optical integrated devices for frequency conversion and information encoding. (5) We designed, built, and performed initial laser ignition and deflagration (regression rate) experiments. For the first time, solid-solid phase transformations were shown to occur on combustion time scales (see accompanying figure).

Publications

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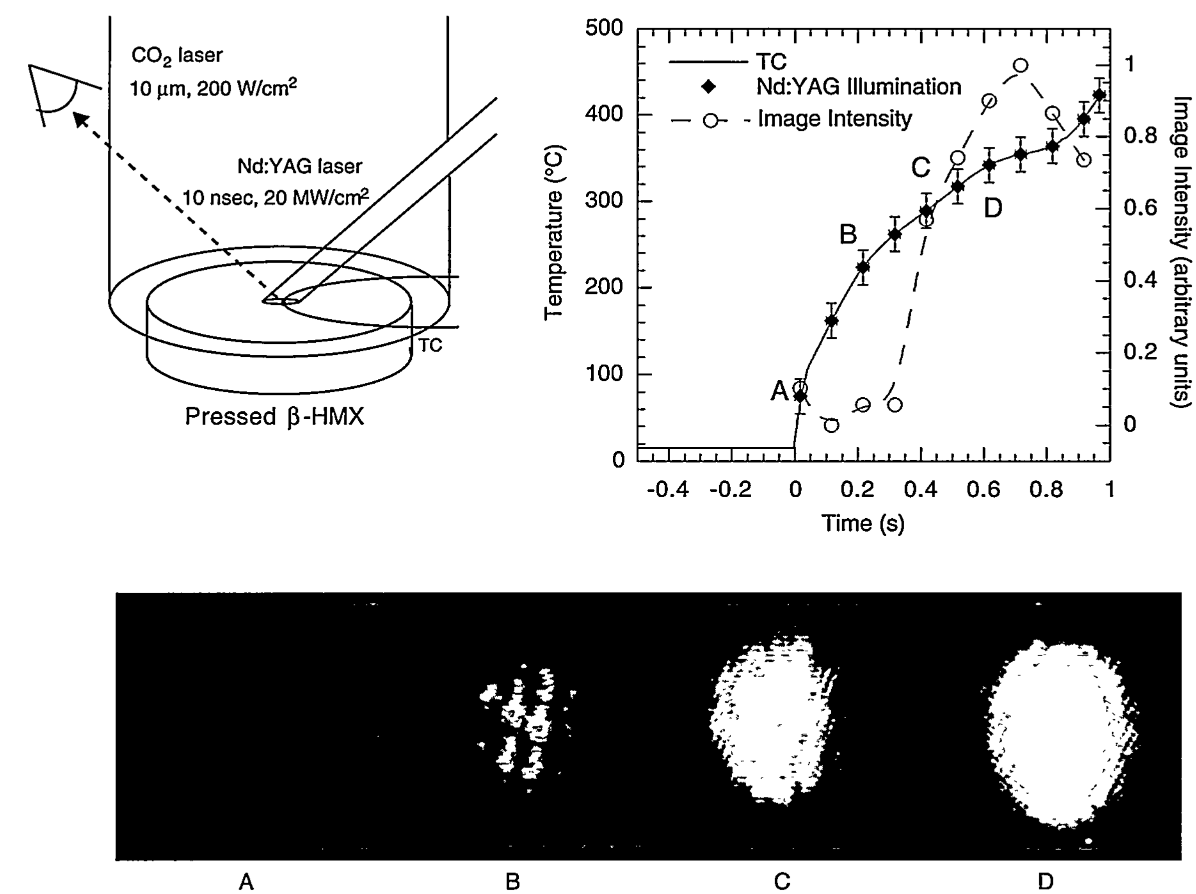

Composite figure showing a schematic of the laser ignition experiment (top left), time series of temperature rise (top right), and imaging of SHG appearance (bottom). The CO_2 laser illuminates the entire pellet surface with 1064-nm illumination, and SHG detection is over a smaller area. A thermocouple (TC) was placed under the 1064-nm Nd:YAG laser illumination spot to record the temperature as a function of time. The letters indicate the time and temperature at which the images shown were recorded. The images were captured from the video record of the 1064-nm illumination spot. This figure shows the beta-delta solid-solid phase transformation occurring in approximately 0.35 s.

Catalysis Science and Technology

97603

Kevin Ott

Our objective has been to develop new catalytic chemistry, theoretical techniques, and spectroscopic tools for the elucidation of structure and reactivity of catalysts. Our studies have been focused in six areas: solid-state nuclear magnetic resonance techniques for the elucidation of structure in zeolites containing metal clusters and metal ions either in or on the zeolite framework; theoretical techniques to describe metal ions on or in the framework of aluminophosphates and zeolites; neutron scattering techniques for the determination of structure and of adsorbed molecules within the zeolite pores; new catalytic chemistry in dense-phase carbon dioxide; new catalytic and stoichiometric chemistry of boration and diboration of unsaturated organic substrates; and synthesis, catalytic activity, and deactivation of titanium ions on the surface of mesoporous silicates.

We have made advances in all of these areas. One example is the conversion of unsaturated organic substrates such as sulfoxides, ketimines, and aldehydes with diboron compounds; the conversion, performed under mild conditions, yields a variety of useful organic intermediates. The catalytic reaction of diboron compounds with imines provides a useful mechanism for the efficient synthesis of boronic acids, which are finding applications as enzyme inhibitors. Another example of a significant advance is using our combined capabilities in theory, neutron scattering, and solid state nuclear magnetic resonance to provide more detailed knowledge of the structure and electronic structure of metal ions and clusters in the cages and frameworks of zeolites and aluminophosphates.

Publications

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Proliferation-Resistant, Low-Environmental-Impact Treatment Processes for Nuclear Waste Destruction

96635

Francesco Venneri

This research leverages the pyrochemistry developed at Argonne National Laboratory and at Los Alamos, extending it to waste transmutation applications. After several years of evaluation, we have focused on pyrochemical processes for nuclear waste treatment as best satisfying the requirements for high proliferation resistance and low environmental impact. The processes are robust, radiation resistant, and tailored for group separations rather than the isolation of single elements. In addition, they allow for recycling the process media numerous times.

Working toward pyrochemical processes that can be applied to the concept of accelerator-driven transmutation of nuclear waste (or simply ATW), we established a chemistry flow sheet for the conversion of spent commercial nuclear fuel to ATW fuel (see the accompanying figure). We performed scoping experiments, including bench-scale, solvent-anode/solvent-cathode, plutonium electrorefining experiments. For process modeling, we used models currently available in the literature to develop an initial mass balance for the ATW flow sheet. Both the front-end conversion of commercial fuel to ATW fuel and the back-end recycling of ATW fuel were considered in the system mass balance. The mass-balance results were the basis for the process chemistry flow sheet, which was presented and positively evaluated during the technical review of the ATW concept at the Massachusetts Institute of Technology.

Publications

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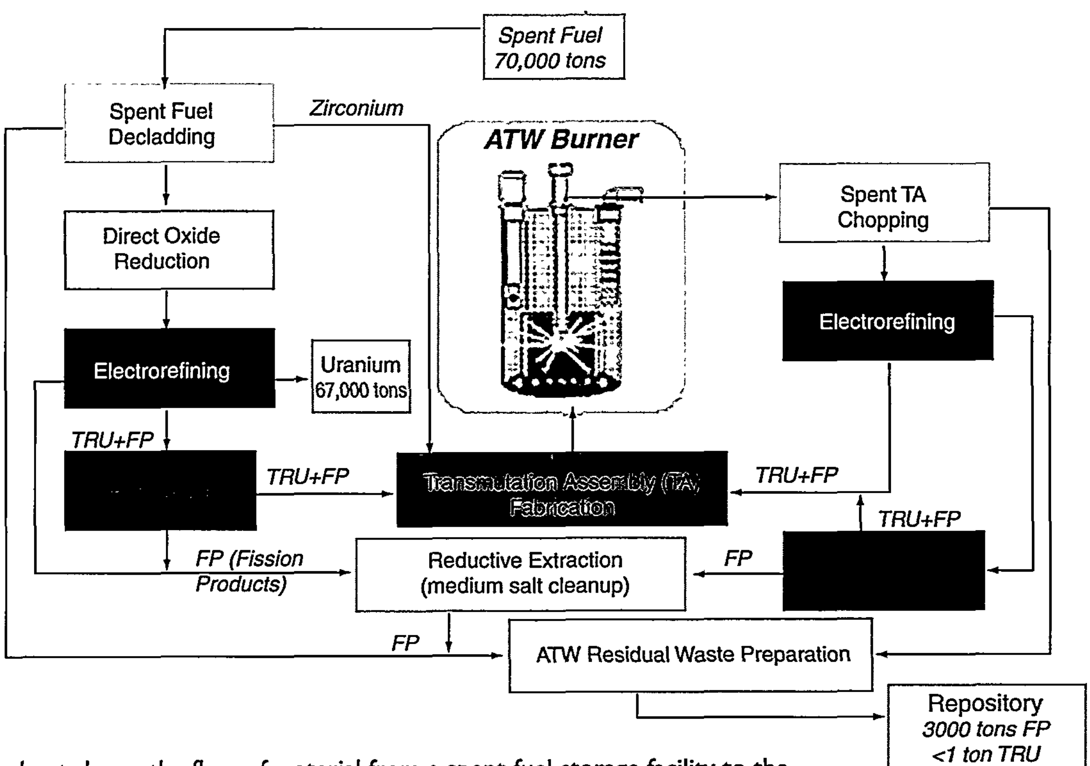

The flow sheet shows the flow of material from a spent-fuel storage facility to the repository, reflecting the ATW concept for the destruction of transuranics and selected fission products.

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

97609

David Clark

The chemistry and physics of the actinide elements have always been of prime importance at Los Alamos, providing the technical basis for process and separation chemistry and metallurgy related to the weapons mission. The goal of our project is to increase our understanding of the manifestation of actinide electronic structure on the physical and chemical properties of actinide materials by integrating chemical synthesis, characterization, spectroscopy, and theory. We have developed new methodologies for synthesis and characterization of both aqueous and nonaqueous actinide systems.

The bond strength and bond length relationships and the bonding changes needed to promote oxo ligand exchange in trans dioxo ions are of special interest. For plutonium aquo ions we determined the number of water molecules associated with oxidation states III through VI, and studied their effect on structural and spectroscopic properties. We demonstrated that crown ethers can modify the solubility of aquo complexes and that the crown ether coordination is affected by the nature of the solvent (aqueous vs. nonaqueous). Using relativistic density functional theory, we studied the electronic structure of the uranyl ion supported by aquo, hydroxide, chloride, and fluoride ligands. We studied the intramolecular conversion between cis and trans isomers in detail for hydroxo species. Overall the predicted bond lengths are in relatively good agreement with EXAFS (extended x-ray absorption fine structure) and other experimental measurements in aqueous solutions. The calculated bond distances and vibrational frequencies determined from theoretical studies compare extremely well with those measured experimentally.

We developed new descriptive chemistry for nonaqueous complexes of uranium and plutonium with aryloxide ligands, including redox and ligand exchange behavior. We also studied optical spectra of organouranium compounds to examine electronic states and to assign spectral peaks to specific electronic transitions between f-electron states. We are working to develop the theoretical tools to predict these electronic transitions.

Publications

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Self-Assembling and Photoreactive Materials
98804

Don Parkin

The focus of this project is the design and synthesis of new molecules with novel self-assembling properties and reactivity as supramolecular assemblies. In addition, we are developing novel and efficient photoredox reactions of organic and inorganic molecules based on single-electron transfer processes. We are building on a novel scheme for amplifying reactions initiated by light, ionizing radiation, or redox-active reagents by coupling tandem-chain fragmentation of donor and acceptor substrates.

During the past year our efforts have been directed toward two major studies. One effort involves developing novel and sensitive chemical radiation detectors based on radiation-induced, one-electron, redox, tandem-chain fragmentation reactions where the radiation events are effectively amplified by a combination of radical-ion redox chain reactions coupled with the strong fluorescence from the high-yield product. The results have led to a follow-up project funded by DOE that will commence in the next year. Another complementary effort has been focused on the studies of supramolecular self-assembly of aromatic-functionalized amphiphiles. The results indicate that strong, attractive noncovalent interactions between the aromatic chromophores may control the structures and strengths of the assemblies. These fundamental studies have provided a basis for understanding the mechanism of orangels formation from low-molecular-weight organic gelators in which aromatic groups have been incorporated. Thus, our investigations included the structure, dynamics, and photoswitching behavior of gels formed from cholesterol-stilbene and cholesterol-squaraine gelators. Our studies suggest that these organic gels, and new gels based on these studies, have potential as advanced materials having controllable properties and versatile applications.

Publications

Geiger, C., et al., "Orangels Resulting from Competing Self-Assembly Units in the Gelator: Structure, Dynamics and Photophysical Behavior of Gels Formed From Cholesterol-Stilbene and Cholesterol Squaraine Gelators" (submitted to *Langmuir*).

Whitten, D., et al., "Self-Assembly of Aromatic-Functionalized Amphiphiles: The Role and Consequences of Aromatic-Aromatic Noncovalent Interactions in the Building of Supramolecular and Novel Assemblies" (submitted to *J. Phys. Chem.*).

Mathematics and Computational Science

Density Function Estimation for Monte Carlo Simulations

96631

James Gubernatis

The goal of this project is to improve the efficiency of Monte Carlo simulations by using adaptive methods. We emphasized method development for models of slowly relaxing physical systems. In one application, we applied our recently developed canonical transition probability method to the standard statistical physics problem of the two-dimensional Ising ferromagnet near its critical temperature. Here, standard Metropolis-based Monte Carlo methods require long simulation times. Our new method, on the other hand, because it allows greater flexibility in adapting importance sampling weights, can produce shorter simulations times and has the added advantage of reducing the simulation variance. We documented these advantages through a series of simulations.

We also finished the development of a bivariate version of the multicanonical method and applied it to the long-standing problem of slowly relaxing processes: the random-bond Ising glass. We used the multicanonical procedure to estimate both the joint probability density of the energy and the Edwards-Anderson order parameter as opposed to estimating just the energy or order parameter density. We found that the simulation executed a random walk in this bivariate space, as illustrated in the accompanying figure. This observation means not only that we have an ergodic simulation but also that we can reduce the simulation time by several orders of magnitude. We continued our application of this new method to the Ising glass to determine more precisely the glass transition temperature and the degeneracy of the ground state. This study is in progress.

Publications

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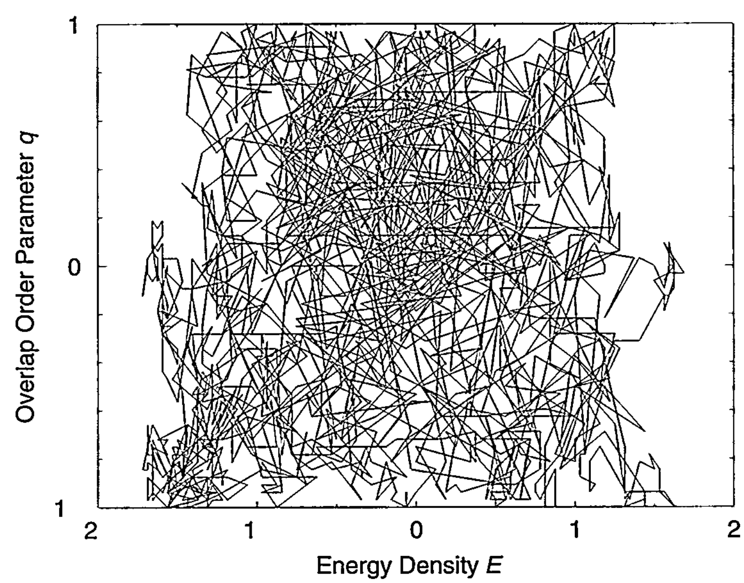
Fitzgerald, M., et al., "Canonical Transition Probabilities for Adaptive Monte Carlo" (to be published in *Europhys. Lett.).*

Gubernatis, J.E., "Multimodal Sampling Methods for Statistical Mechanics" (submitted to *Int. J. Mod. Phys. C*).

Hatano, N., and J.E. Gubernatis, "Bivariate Multicanonical Monte Carlo for the +/- J Ising Glass" (Slow Dynamics in Complex Systems Workshop, Tohwa University, Fukuoka, Japan, November 9–13, 1998).



The sequence of the values of the Edwards-Anderson order parameter *q* and the energy *E* produced by our bivariate multicanonical method for the three-dimensional Ising glass. The temperature is below the glass transition temperature. The plot illustrates the random-walk character of the Monte Carlo dynamics of our new method.

Probabilistic and Combinatorial Analysis for Biological Systems

98806

David Torney

This project involves research in several aspects of probabilistic and combinatorial analysis for biological systems. We are undertaking several types of combinatorial analysis motivated by new molecular-biological data. To further the mathematical and statistical analysis of biological sequences and systems, we are studying the characterization of collections of numbers that are potentially the cumulants of a probability distribution. To make inroads into the analysis of evolution, we are pursuing enumeration of evolutionary trees. We are studying the existence and construction of Hadamard matrices and projective planes, issues central to the design of large-scale biological experiments and to the analysis of sequence data. Finally, we are undertaking a detailed combinatorial and probabilistic modeling of mutation and selection of HIV strains in humans.

This year we successfully completed the construction of lattice square designs and enumeration of the constructed designs. We completed an analysis of probability distributions on sets, motivated by the analysis of biological sequences. We also began the investigation of the preliminary HIV evolution model and made progress in modeling HIV therapies.

In addition, we sponsored a workshop, "Frontiers of Combinatorics," that was attended by 16 mathematicians and mathematical biologists from around the world and 8 Los Alamos scientists. The purpose of the workshop was to evaluate new mathematical problems motivated by molecular biology and to generate dialog and in-depth understanding on this topic. We focused on experimental designs, combinatorial analysis of biological sequences, and techniques of phylogenetic analysis. To facilitate access to this new field of research, the participants authored articles to appear in a dedicated issue of *Annals of Combinatorics*. During the workshop many combinatorial problems with biological motivations, including ours, were described and some of these were actually resolved during the workshop. This workshop proved to be a valuable mechanism to establish new collaborations between Los Alamos scientists and the visitors and to continue ongoing efforts to address these problems.

Publications

Chateauneuf, M.A., et al., "Pooling, Lattice Square and Union Jack Design" (to be published in *Ann. Combinatorics*).

Colbourn, C.J., "Weakly Union-Free Maximum Packings" (to be published in *Ann. Combinatorics*).

Detours, V., and A.S. Perelson, "Explaining High Alloreactivity as a Quantitative Consequence of Affinity-Driven Thymocyte Selection" (to be published in *Proc. Natl. Acad. Sci. U.S.A.*).

D'yachkov, A.G., and V.V. Rykov, "Optimal Superimposed Codes and Designs for Renyi's Search Model" (to be published in *Ann. Combinatorics*).

Eastman, P.S., et al., "Genotypic Changes in Human Immunodeficiency Virus Type 1 Associated with Loss of Suppression of Plasma Viral RNA Levels in Subjects Treated with Ritonavir (norvir) Monotherapy," *J. Virol.* **72**, 5154 (1998).

Erdos, P.L., "Some Generalizations of Property B and the Splitting Property" (to be published in *Ann. Combinatorics*).

Macula, A.J., "Probabilistic Nonadaptive Group Testing in the Presence of Errors and DNA Library Screening" (to be published in *Ann. Combinatorics*).

Mittler, J.E., et al., "Influence of Delayed Virus Production on Viral Dynamics in AIDS Patients," *Math. Biosci.* **152**, 143 (1998).

Notermanns, D., et al., "Rate of Decline of HIV-1 Following Antiretroviral Therapy Is Related to Viral Load at Baseline and Drug Regimen," *AIDS* **12**, 1484 (1998).

Percus, A.G., and O.C. Martin, "Scaling Universalities of Kth-Nearest Neighbor Distances on Closed Manifolds," *Adv. Appl. Math.* **21**, 424 (1998).

Sinha, K., and R.K. Mitra, "Construction of Nested Balanced Block Designs, Rectangular Designs and Q-ary Codes" (to be published in *Ann. Combinatorics*).

Spouge, J.L., et al. "The Combinatorics and Extreme Value Statistics of Protein Threading" (to be published in *Ann. Combinatorics*).

Steel, M.A., and L.A. Szekely, "Inverting Random Functions" (to be published in *Ann. Combinatorics*).

Torney, D.C., "Sets Pooling" (to be published in *Ann. Combinatorics*).

Torney, D.C., and J. Wang, "Systems of Linear Congruences with Individual Moduli" (to be published in *Linear Algebra Appl.*).

Torney, D.C., et al., "The Stationary Statistical Properties of Human Coding Sequences" (submitted to *J. Mol. Biol.*).

Wan, H., and J.C. Wooten, "Axiomatic Foundations of Complexity Functions of Biological Sequences" (to be published in *Ann. Combinatorics*).

Multiscale Science for Science-Based Stockpile Stewardship

97601

Len Margolin

Our overall goal is to develop and apply the methods of multiscale science to the problems of fluids and materials mixing and materials characterization. Our specific focus is on the science-based stockpile stewardship issue of assessing the performance of a weapon with off-design, aged, or remanufactured components in the absence of full-scale testing. Our product will be the 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.

Our research is organized into three general areas. The first is evolution of interfacial fluid instabilities (Rayleigh-Taylor, Kelvin-Helmholtz, Richtmyer-Meshkov) and the resulting interpenetrative mix of materials. We have developed new closures for two-pressure, two-phase flow and studied the nature of the boundary conditions at the edges of the mixing layer, which are based on drag and buoyancy laws. We have validated our front-tracking code by comparing simulations of Richtmyer-Meshkov instability growth with other codes, with data, and with a new nonlinear analytic theory. We have taken moments of the spectral equations that describe the distribution of fluctuating energy across the entire spectrum of length scales, rederiving the usual transport equations plus one new extension—a nonlocal transport of length scale from the middle of the mixing layer to beyond its outer edges. This year we completed a characteristic analysis of our hybrid multifluid turbulent-mix model. We have continued our development of a model for crenulative instability and have applied it to the spherical convergence of an incompressible fluid with a nonhomogeneous distribution of kinematic viscosity.

The second research area is prediction of material properties (effective elastic moduli, yield strength, friction coefficient) based on microstructure. We have extended our study of the spallation process in ductile metals, based on void growth and including the effects of damage clustering and void linking, to uranium and tantalum, and our results were compared with experimental data. We have continued our molecular dynamics studies of interfacial friction. In addition, we are studying the intermixing of light and heavy constituents in a hot, dense plasma environment and have begun looking at the penetration of fast charged particles into plasmas.

The third research area is formation and subsequent transport of surface ejecta from a shocked metal/gas interface. We have continued development of a distinct element model for metals by implementing contact forces (Hertzian and frictional) and bonding into a smooth particle hydrodynamics code. To test the implementation, we have simulated shock-wave propagation and plate impact experiments. We are beginning to develop laws for the interaction between the solid particles (interacting through contact forces) and fluid particles (interacting through potential forces), leading ultimately to a discrete model for particle suspensions.

Publications

Cheng, B., et al., "Boundary Conditions for a Two-Pressure Two-Phase Flow Model" (submitted to *J. Phys. Fluids*).

Hammerberg, J.E., et al., "Nonlinear Dynamics and the Problem of Slip at Material Interfaces" (to be published in *Physica D*).

Holmes, R.L., et al., "Richtmyer-Meshkov Instability Growth: Experiment, Simulation, and Theory" (to be published in *J. Fluid Mech.*).

Margolin, L.G., et al., "Large Eddy Simulations of Convective Boundary Layers Using Nonoscillatory Differencing" (submitted to *Physica D*).

Mikulla, R.P., et al., "High-Speed Friction at the Atomistic Scale: Dislocation Generation and Motion in 2D and 3D" (to be published in *Mater. Res. Soc. Bull.*).

Rivas, J.M., et al., "Quantification of Ductile Fracture in Spalled Tantalum," in *Electron Microscopy 1998*, H.A.C. Benavides and M.J. Yacaman, Eds. (Institute of Physics Publishing, Philadelphia, PA, 1998), Vol. 1, p. 169.

Roeder, J., et al., "Multichain Frenkel-Kontorova Model for Interfacial Slip," *Phys. Rev. B* 57, 2759 (1998).

Romero, C.A., et al., "Crenulative Turbulence in a Converging Nonhomogeneous Material" (submitted to *Phys. Fluids*).

Thissell, W.R., et al., "Damage Evolution and Clustering in Shock Loaded Tantalum" (International Metallurgical Society '98, Ottawa, Canada, July 25–29, 1998).

Thissell, W.R., et al., "Quantitative Damage Evolution in Tantalum under Spallation Conditions" (International Conference on Shock Waves in Condensed Matter, St. Petersburg, Russia, July 12–17, 1998).

Ulitsky, M., et al., "Testing a Random Phase Approximation for Bounded Turbulent Flow" (submitted to *Phys. Rev. E*).

Wilson, P.N., et al., "Spectral Nonequilibrium in a Turbulent Mixing Layer" (submitted to *Phys. Fluids*).

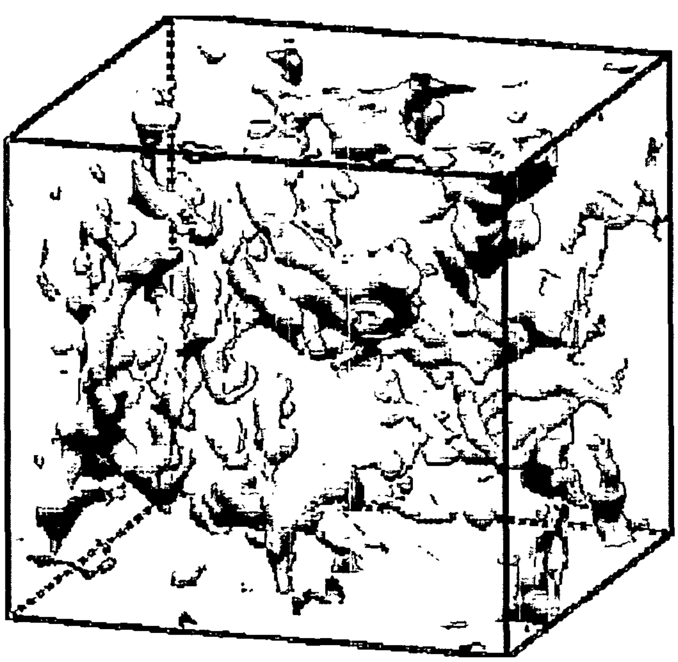
Discrete Simulation of Nonlinear Systems

98805

Shi-Yi Chen

We are developing discrete numerical methods for modeling multiscale physics in nonlinear systems. The methods include the kinetic lattice-Boltzmann method, dissipative particle dynamics, and molecular dynamics (MD). In particular, we will study complex fluids and soft condensed matter (granular materials, vesicles, colloids, and nanotribology) and multiphase fluid flows in porous media, using the discrete methods. We will compare the simulation results with theoretical predictions from cycle expansions and renormalized perturbation schemes. The tool box developed in this project will increase our simulation capabilities and provide detailed insight into physics from the microscopic scale to the macroscopic scale.

In addition to organizing workshops and hosting visitors that help us explore the state-of-the-art in simulation of nonlinear systems, we have made some important progress in several areas. We have developed (1) an event-driven, three-dimensional, hard-sphere MD model and message-passing-interface, dissipative-particle MD code for research on the properties of high-speed, high-concentration granular flows and the clustering kinetics of granular media during cooling (see accompanying figure); (2) Monte Carlo techniques for construction of compact wave functions to describe a quantum subsystem; and (3) a new lattice-Boltzmann scheme for simulation of multiphase flow based on distribution functions. Simulations of the two-dimensional and three-dimensional Rayleigh-Taylor instabilities yield excellent results. The discrete numerical method has been used for simulating problems in micro-electrical-mechanical systems.



Isosurface of the particle density in a granular media from a molecular dynamics simulation. The tubelike structure is apparent.

Publications

Bender, C.M., and S. Boettcher, "Quasi-Exactly Solvable Quartic Potential," *J. Phys. A* **31**, 273 (1998).

Bender, C.M., and S. Boettcher, "Real Spectra in Non-Hermitian Hamiltonians Having PT Symmetry," *Phys Rev. Lett.* **80**, 5243 (1998).

Bikaki, A., et al., "Energy Relaxation in Discrete Nonlinear Lattices" (submitted to *Phys. Rev. E*).

Boettcher, S., "Aging Exponents in Self-Organized Criticality," *Phys. Rev. E* **56**, 6456 (1997),

Cao, N., et al., "Statistics and Structures of Pressure in Isotropic Turbulence" (submitted to *Phys. Fluids*).

Chen, S., and Robert Kraichnan, "Simulations of a Randomly Advected Passive Scalar Field" (submitted to *Phys. Fluids*).

Chen, S., et al., "Clustering Kinetics of Granular Media in Three Dimensions" (submitted to *Phys. Rev. Lett.*).

Chen, S., et al., "A Connection between the Camassa-Holm Equations and Turbulent Flows in Channels and Pipes" (submitted to *Phys. Fluids*).

Chen, S., et al., "Recent Advances in Lattice Boltzmann Methods," in *Proceedings of the National Congress of Applied Mechanics* (Cambridge University Press, New York, in press).

Gomez, M.A., and L.R. Pratt, "Construction of Simulation Wavefunctions for Aqueous Species: D30+" (to be published in *J. Chem. Phys.*).

He, G., et al., "Calculations of Longitudinal and Transverse Velocity Structure Functions Using a Vortex Model of Isotropic Turbulence" (submitted to *Phys. Fluids*).

He, G., et al., "Scalings of Dissipation and Enstrophy Induced by Random Strained Vortices in Fluid Turbulence" (submitted to *Phys. Rev. Lett.*).

Hou, T.Y., et al., "Effect of Finite Computational Domain on Turbulence Scaling Law in Both Physical and Spectral Spaces" (submitted to *Phys. Rev. E*).

Korolov, L., "Transport by Time Dependent Stationary Flow" (submitted to *Commun. Math. Phys.*).

Nelkin, M., and S. Chen, "The Scaling of Pressure in Isotropic Turbulence," *Phys. Fluids* **10**, 2119 (1998).

Nie, X., et al., "Lattice-Bolzmann Simulations of Fluid Flows in Mems" (submitted to *Phys. Fluids*).

Paczuski, M., and S. Boettcher, "Avalanches and Waves in the Abelian Sandpile Model," *Phys. Rev. E* **56**, 3745 (1997).

Percus, A., and Oliver Martin, "Scaling Universalities of K TH-Nearest Neighbor Distance on Closed Manifolds," *Adv. Appl. Math.* **21**, 424 (1998).

Rose, H.A., and S. Ghosal, "Nonlinear Theory of Power Transfer between Multiple Crossed Laser Beams in a Flowing Plasma," *Phys. Plasmas* **5**, 1461 (1998).

Seta, T., et al., "Lattice Boltzmann Method for Two-Phase Flows" (submitted to *Phys. Fluids*).

Straub, S., "Dynamics of Rapid Granular Flows: Results from Molecular Dynamics Simulations" (Workshop on Granular Materials: Statics, Excitations and Dynamics, University of New Mexico, Albuquerque, NM, June 25–26, 1998).

Straub, S., and G. Valentine, "3-D Modelling of Rapid Granular Flows and Its Application to High-Concentration Pyroclastic Flows" (Society of Economic Paleontologists and Mineralogists and Geological Society of London Conference on Sediment Transport and Deposition by Particulate Gravity Currents, University of Leeds, UK, September 7–9, 1998).

Nuclear Futures and Scenario Building

96644

Edward Arthur

We are developing state-of-the art technical tools to assess conditions and issues related to achieving and maintaining nuclear stability in the future. In the final year of this project, we have built upon results from previous years efforts to expand the project's efforts in three principal areas: (1) assessment of conditions surrounding the future role of nuclear energy globally, (2) definition and testing of a proposed nuclear fuel cycle architecture whose aim is to manage plutonium and other nuclear materials much better in the future, and (3) development and initial testing of models to assess security issues associated with nuclear weapon stockpiles held by current and potential nuclear weapons powers. In the latter area, we extended conventional deterrence theory to examine the effects not only of existing stockpiles but also of "latent" national capabilities to produce nuclear weapons, particularly routes that lead to nuclear materials production. This latter development provides a link back to previous efforts of the project associated with identifying and assessing proliferation risks associated with current and future civilian nuclear fuel cycles.

In the first area, this year we assessed the sensitivity of nuclear energy demand shares (on a global and regional basis) in terms of assumptions concerning future efforts to manage and mitigate the buildup of atmospheric carbon dioxide.

In the second area, results centered around the creation and testing of a proposed nuclear fuel cycle architecture and associated technologies, which would effect much greater barriers to materials diversion and would create waste forms having lesser environmental impact. A feature of this architecture is the goal of maximizing security associated with plutonium and other nuclear materials, which is achieved via a combination of secure interim storage and implementation of "burner" technology to deal with key radionuclides before they are disposed of permanently.

In the third area, major results involved tests of conditions that stabilize (and others that destabilize) effects associated with nuclear weapons inventories, particularly as such inventories are driven toward lower numbers. Analysis results showed that reintroduction of multiple warhead launchers had a significant near-term destabilizing impact. Time dependence was introduced into the stability model, and initial testing began of stability indices impacts and sensitivity. This work set the stage for explicit incorporation of latency effects, especially in the area of capabilities for nuclear weapons materials creation.

Publications

Peterson, P.F., "Relative Proliferation Attractiveness of Reprocessing Waste and Spent Fuel: Implications for Long-Term Safeguards Technical Requirements" (International Symposium on Energy Futures, Honolulu, Hawaii, March 1998).

Theoretical Foundation for Adaptive Monte Carlo
96633

Richard Picard

The Monte Carlo method is an important tool for analyzing many complex problems, especially in the nuclear sciences, for 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 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 been shown to provide exponential convergence. The goal of this project is to obtain exponential convergence under less restrictive conditions.

In our third year, we completed a major paper on exponential convergence for continuous-state spaces. Until now, no theoretical foundation existed for the simulation results displaying such behavior that have appeared in the literature. This work extends our earlier efforts for discrete problems.

Our work related to statistical physics involved the direct use of the Monte Carlo transition dynamics in the Metropolis algorithm. This approach, based on canonical transition probabilities, also allows for extrapolation of results to system temperatures other than that of the simulation. When we coupled this procedure with importance sampling, we obtained substantial efficiency gains over standard Boltzmann methods. In an application to magnetization susceptibility in a ferromagnet, for example, computational efficiency is improved 25% when using the same underlying simulation data.

Publications

Baggerly, K., et al., "Exponential Convergence for Adaptive Importance Sampling of Markov Chains" (submitted to *J. Appl. Probability*).

Fitzgerald, M., "Canonical Transitions in Adaptive Monte Carlo" (American Statistical Association Spring Research Conference, Santa Fe, NM, June 3–5, 1998).

Fitzgerald, M., et al., "Towards Fully Adaptive Monte Carlo" (submitted to *Europhys. Lett.*).

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

Picard, R., "Adaptive Learning Algorithms for Particle Transport Simulation" (American Statistical Association Spring Research Conference, Santa Fe, NM, June 3–5, 1998).

Silver, R., "Adaptive Non-Boltzmann Monte Carlo" (Vanderbilt Workshop on Condensed Matter Theories, Nashville, TN, June 1–5, 1998).

Advancing X-Ray Hydrodynamic Radiography: Radiography Chain Model
97604

Daniel Prono

The goal of advanced hydrodynamic radiography is to be able to quantify specific details of the implosion dynamics of a device undergoing hydrodynamic testing. To validate the large codes that predict hydrodynamic behavior, we must precisely measure and determine the three-dimensional temporal evolution [3-D(t)] of irregular shapes and mass distributions. There are three main challenges that make such measurements difficult: (1) to resolve in sufficient detail the exact 3-D(t) shapes and edges requires many views or, with a limited number of views, highly sophisticated reconstruction algorithms; (2) to quantify the mass distributions requires an even greater number of views and yet-more-advanced reconstruction algorithms that model physical parameters, such as equations of state and x-ray interaction cross sections, for many materials; and (3) both of the above requirements are strongly compromised by the fact that x-rays scatter when passing through the thick primary systems, and thus when we interrogate with many axes, the "scatter background" swamps the direct signal on each axis.

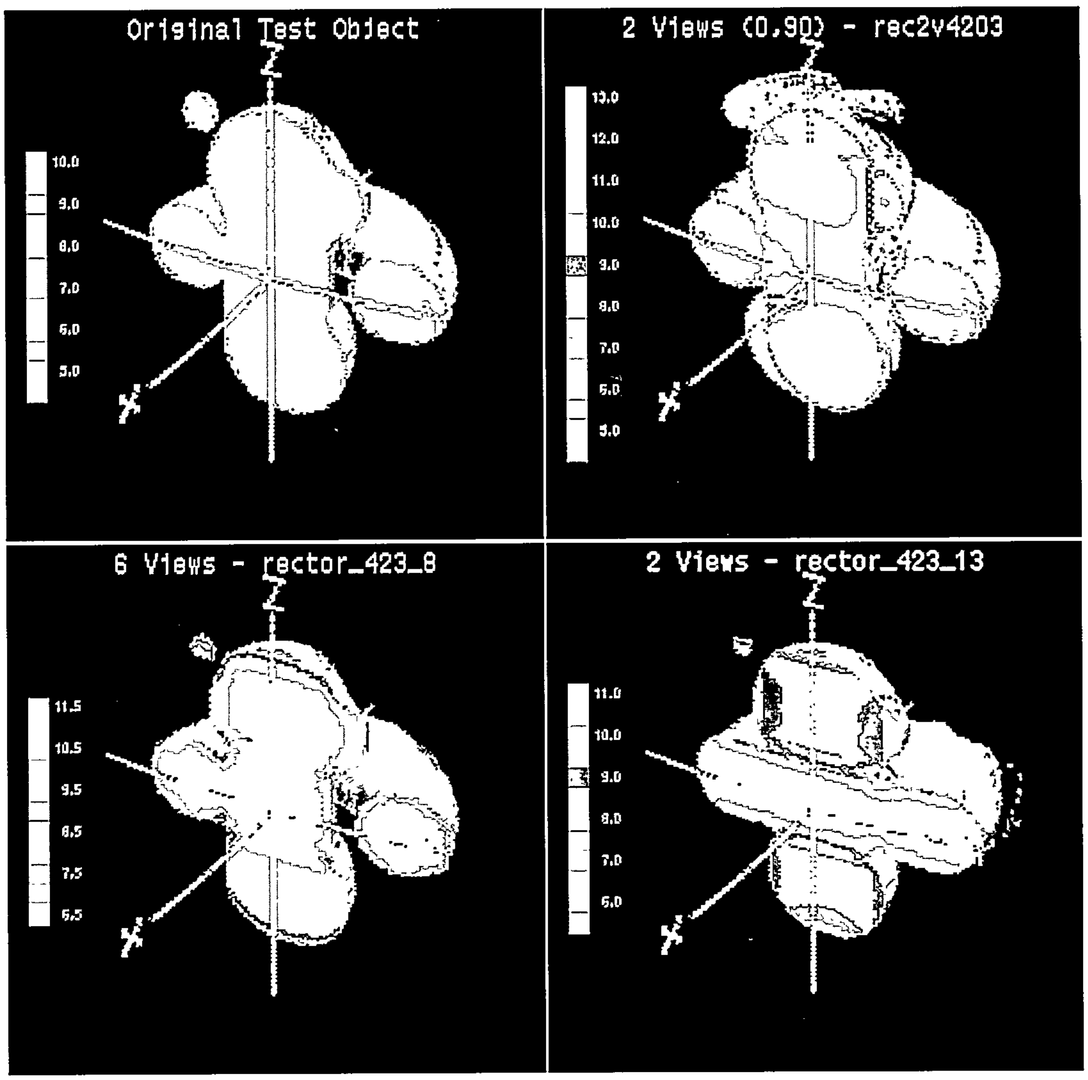
We have made substantial progress in meeting each of these three challenges. On the scatter-background issue, we have performed detailed computer simulations of experimental configurations and then validated those simulations with experimental measurements. Corrections have been made to the simulation codes so that agreement to within a few percent has been achieved. With these validated "scatter codes," we have begun designing x-ray scatter "filters" that will enable multiaxis interrogations and actually cause the direct signal received by the detector on one axis to be relatively free of background scatter.

On the advanced reconstruction algorithms, we have developed techniques that duplicate synthetic axisymmetric objects to a few percent fidelity and have demonstrated that, with assumptions of symmetry, only one or two viewing axes are required to give the necessary feature resolution. However, on nonsymmetric objects or when the 3-D(t) shapes are irregular, the minimum number of views required for fine-feature resolution converges slowly to a large number (<16 axes). The accompanying figure shows how reconstruction quality improves as more views are added or when increasingly sophisticated algorithms are applied. We are currently doing numerical testing on asymmetric synthetic objects to determine what contraints can be used in the reconstruction algorithms that would lower the requisite number of viewing axes.

On the issue of being able to quantify mass distribution, our efforts have concentrated on being able to resolve the difference between mass distribution (mixing) and 3-D irregular shapes. Here, our best results have come from developing momentum conservation algorithms. The concept is that when taking many time-sequenced images along many different axes, we can analyze the time sequences for mass-velocity vectors and thus determine if the vectors are consistent with the unknown feature being an irregular 3-D shape or a mass distribution.

Publications

Mathews, A.R,. et al., "Analysis of 3D Hydrodynamics Calculations" (Nuclear Explosives Design Physics Conference, Livermore, CA, October 1998).



Comparison of reconstructions for an almost axisymmetric test object achieved with different algorithms and number of views. The scale, originally in color, corresponds to density. (Top left) Original constant-density object, which was generated by distorting portions of an axisymmetric object and adding small 3-D features. (Top right) Two-view annular harmonic decomposition (AHD) reconstruction. (Bottom) Six-view (left) and two-view (right) reconstructions using the algebraic reconstruction technique (ART) Adding more views reduces density variations for the reconstructed object. With six views, a third of the reconstructed object's volume has a constant density of the correct value. Combining AHD and ART further improves reconstruction quality.

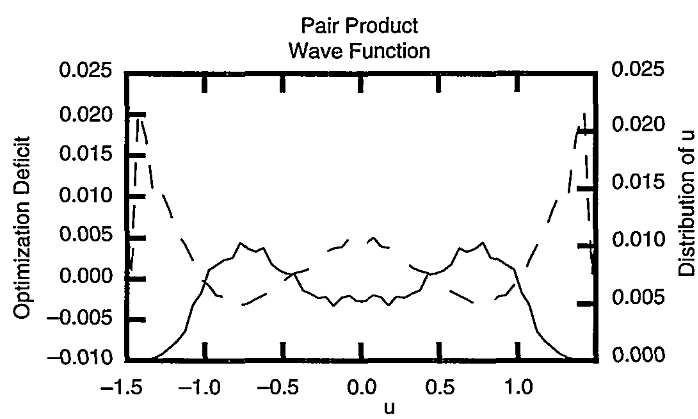
Novel Monte Carlo Algorithms for Statistical Mechanics

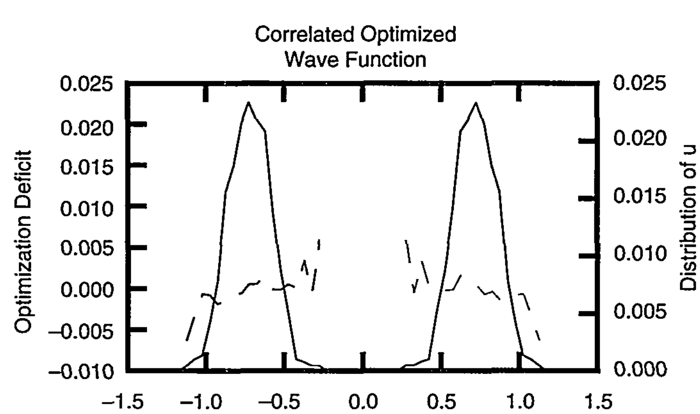
96632

James Gubernatis

To advance the computation of molecular structures, we have developed Monte Carlo procedures for the adaptive improvement of quantum mechanical wave functions. First, using the Metropolis Monte Carlo method, we obtained pair-product vibrational wave functions for hydronium from the asymptotic high-temperature many-body density matrix. Then, since the variational theorem permits identification of wave-function features with significant potential for further optimization, a subsequent Variational Monte Carlo simulation yielded significant improvement in the accuracy of energy estimates. The resulting wave function will be used in an approximation to the density matrix of a quantum molecule in a bath of classical molecules. The accompanying figure illustrates the optimization achieved. The top graph in the figure shows the starting point; the bottom, the optimized result. The deficit is a measure of error. An optimal reduction of error occurs when the deficit is small for both small and large values of *u*, a parameter that is a measure of three-body correlations.

We also studied methods to improve the accuracy of Monte Carlo solutions to eigenvalue problems. The Monte Carlo method was combined with perturbation theory, and we used multiple histories of random walks to produce estimates of the average dominant eigenvalues and their (small) differences. We successfully applied the method to a problem involving parity-violating interactions whose strengths are a million times smaller than those of the strong interaction. Such new methods will be useful in a variety of other problems, both classical and quantum, including the nuclear criticality problem.





An illustration of the adaptive optimization of the wave function for hydronium by Monte Carlo methods. The top graph is the starting point; the bottom, the result. The deficit, a measure of error, is the dashed line. The solid line is the probability density represented by the wave function.

Publications

Carlson, J., "Quantum Monte Carlo Methods and Few-Body Nuclei" (to be published in *Few-Body Systems*).

Carlson, J., "Quantum Monte Carlo Methods in Nuclear Physics," in *Quantum Monte Carlo Methods in Physics and Chemistry,* M.P. Nightingale and C. Umrigar, Eds. (Kluwer Academic Publishers, Netherlands, 1999), p. 287.

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Evolutionary Computation

97621

Chris Barrett

We are developing a mathematical basis for a theory of computer simulation of dynamical systems. This theory will lead to a practical engineering science of simulation applicable to sociotechnical 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.

During this year we established a new class of formal dynamical systems, sequential dynamical systems (SDS), that represent simulation in a general mathematical framework. Analysis of SDS allows understanding of computer simulation in a way that transcends particular application domains and is essential for the rigorous use of simulation in the variety of circumstances demanded by the Laboratory's mission. As a new area of applied mathematics, SDS will be established as a topic for graduate study at New Mexico State University's mathematics department in 1999, with associated collaboration with the University of California at Berkeley. This will provide expanded research and a talent pool in this important new area. SDS will form the applied theoretical core of future program development for simulation science in Los Alamos's Technology and Safety Assessment Division.

SDS are arbitrarily high-dimensional, dynamical systems composed of local mappings. Each local mapping updates the state of one simulated entity, which is "data-local." Our studies have focused on the orderings in which local mappings are updated. In practical terms the question is how many dynamical systems are produced (given a software object library) by varying the update order. Understanding isomorphic classes of systems generated by the simulation is essential for representation, design, and validation of simulations. The concept of neutrality (different update orderings producing isomorphic systems) is deeply related to mutation stability of biomolecules. Order dependence and transitivity of compositions of simulated entities are closely related to such issues as voting and consumer-demand paradoxes and the relationships between simulations of physical, biological, and socioeconomic systems.

Publications

Barrett, C.L., and C.M. Reidys, "Elements of a Theory of Computer Simulation I: Sequential CA Over Random Graphs," *Appl. Math. Comput.* **2**, 241 (1998).

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Self-Organization and Pattern Formation

96611

Robert Ecke

The formation of regular structures is important in science and technology and recently much excitement has focused on self-organizing and self-assembling systems. Instead of building structures by hand, as in microelectronic lithography, one would design or discover 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, defects in rotating thermal convection, replicating spot dynamics in reaction-diffusion systems, and spatiotemporal dynamics of calcium waves (as models of biological self-organization).

This past year we built soap tunnels and demonstrated thickness, velocity, and vorticity measurements in two-dimensional hydrodynamic systems (see the accompanying figure). We also computed the dynamics of defects and their effect on spatiotemporal chaos for some generic models, and applied these concepts of extensive chaos to experimental data using Karhunen-Loeve decomposition techniques. We cosponsored a nonlinear science conference at Los Alamos, which included topics on dynamical systems.

Finally, we developed a method for synchronizing two identical chaotic systems in any of its coordinates, with potential applications in communication, encryption, and nonlinear control. We also developed a novel technique for computing the effects of arbitrary noise on general dynamical systems.

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Nonequilibrium Science: Assessment, Control, and Prediction

98605

Emil Mottola

The goals of our project are to (1) create a new and more fundamental understanding of the common physical principles underlying the complex real-time dynamics of nonequilibrium phase transformations and multiscale phenomena, and (2) develop practical methods for dealing with them.

We have studied the square-triangle transition and developed a Landau-Ginzburg-type free energy in two dimensions (2-D) in order to model and understand the three-dimensional (3-D) alpha (bcc) to epsilon (hcp) phase transition in iron, which occurs at 13 GPa. We have developed both 2-D and 3-D elastic codes, with full elastic compatibility and coupling to stress, to study this phase transition using the time-dependent Ginzburg-Landau approach. We have begun studying the equilibrium thermodynamics and structure of solid and liquid copper, as modeled with the embedded atom potential of Art Voter. We have used Monte Carlo simulations to determine the equation of state, as well as the liquid-state correlation functions. The absolute free energy of the liquid, which is needed to calculate the solid-liquid phase boundary, has been obtained by coupling constant integration from a reference fluid defined by a $1/r^{12}$ pair potential.

We have developed and implemented numerical algorithms for solving the problem of a domain boundary self-consistently coupled to its own phonon excitations. This is the necessary first step to the corresponding time-dependent problem of a moving interface coupled to phonons. We have studied, by both models and numerical methods, vortex lattice melting in superconductors in a variety of experimentally accessible situations. We have implemented stochastic partial differential equations to study vortex dynamics in a noisy environment, and initiated a study of the precise quantitative control of systematic uncertainties in the presently available algorithms for stochastic equations. Finally, we have studied the Markov (stochastic) limit of particle-creation processes in quantum field theory by an appropriate averaging over fast microscopic degrees of freedom to characterize their effect on slow degrees of freedom in coarse-graining approximations.

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Adaptive Monte Carlo Methods for Radiation Transport

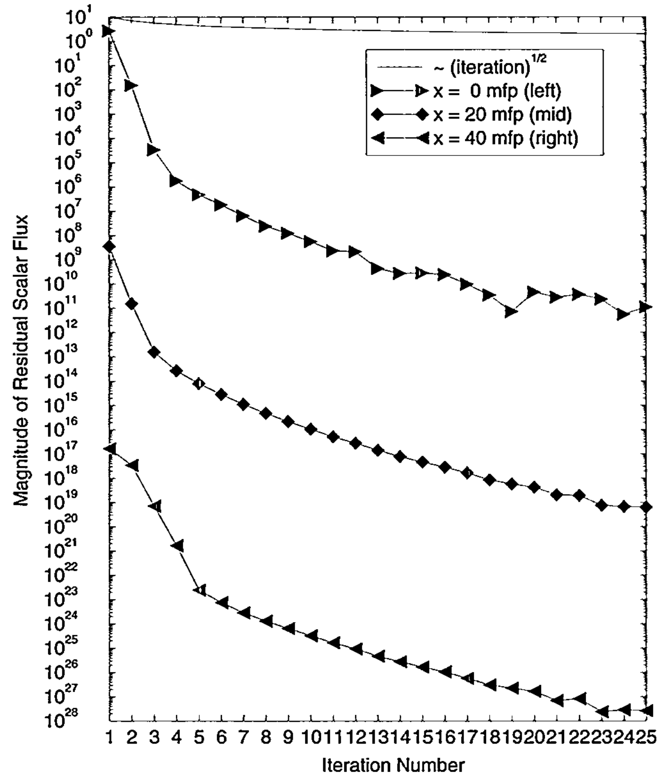
96630

Thomas Booth

In this project we seek to obtain exponential convergence for Monte Carlo particle transport calculations, instead of the slow inverse square root of the computer time dictated by the central limit theorem of statistics. This theorem applies to independent and identically distributed (IID) samples; both the reduced-source and the zero-variance approaches violate this IID assumption and thus are not bound by the theorem. The reduced-source method samples a source at the n+1st iteration that is the difference between the true solution and the estimated solution at the nth iteration. The zero-variance biasing method biases the transport kernels on the n+1st iteration based on the importance function learned on the nth iteration.

The test problem is a slab of thickness T in the x-direction and covering all of y and z. The cosine of the angle between the particle direction and the x-axis is μ. The source particles enter at $x = 0$, and the desired estimate is the fraction of particles penetrating at $x = T$. The scattering is isotropic.

The first figure shows the exponential decrease in residual error in the scalar flux (at three positions) with the reduced-source approach. The second figure shows the exponential error decrease with the zero-variance biasing approach. The importance function is truncated after four terms for the solid lines and after eight terms for the dashed lines. Ten calculations for each truncation order show the dependence on random-number seed. The exponential rate is independent of the truncation order.



The reduced-source approach. The convergence behavior of the magnitude of the residual scalar flux at the left, middle, and right of a 40-mfp (mean-free-path) slab. The incident source was $S(0,\mu) = \mu e^{-\mu^2}$ and the collision survival probability was $c = 0.5$ with isotropic scattering.

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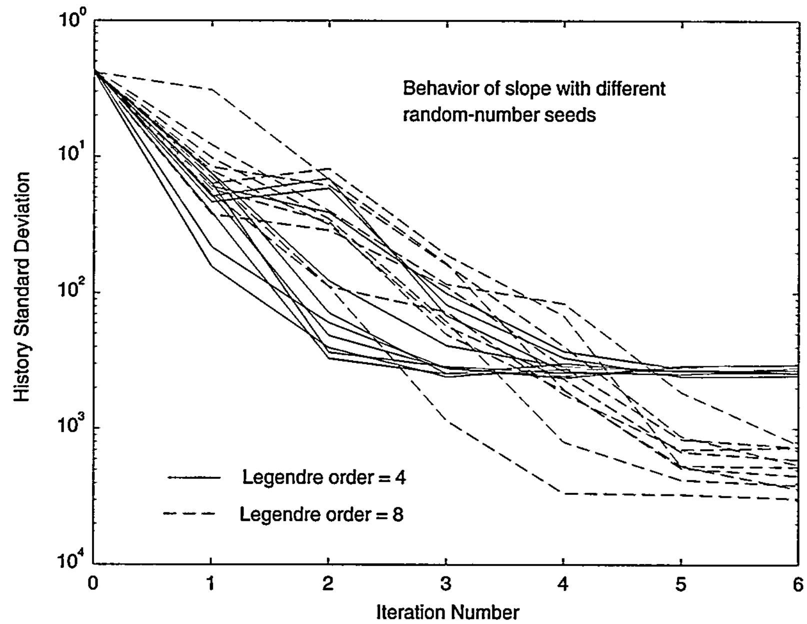

The zero-variance biasing approach. The standard deviation of the individual history scores for two levels of approximations for a $T=1$ mfp slab. The incident source was $S(0,\mu) = 1$, and the collision survival probability was $c = 0.5$ with isotropic scattering.

Applications of Nonlinear Stochastic Dynamics

96613

Darryl Holm

The objectives of this project are to (1) extend the scientific community's technology base relevant to fundamental problems arising in applications of nonlinearity in science and (2) apply this extended technology in those problems of special interest for the Laboratory's technological base. Our numerical simulations and experiments have focused on nonlinear and stochastic processes and have provided important insights into nonlinear science, while our theoretical analyses of nonlinear processes have helped advance our understanding of the scientific principles underlying the control of complex behavior in nonlinear systems with strong spatial and temporal internal variability.

This year the development of a new Euler-Poincaré formulation of ideal continuum dynamics with applications to turbulent flows in pipes and channels at high Reynolds numbers; the use of this new formulation to develop multisymplectic numerical algorithms for long-time, accurate simulations of nonlinear dynamics; the analysis and simulation of dispersion-managed solitons, which produce "power enhanced solitons" by varying the optical fiber dispersion periodically; and the analysis and simulation of stochastic phi-fourth kink dynamics.

As part of our scientific communication and interactions with a larger community, we organized two weekly seminar series, "Stochastic Differential Equations" and "Statistical Solutions of the Navier-Stokes Equations"; organized two international conferences, "Singularities in Nonlinear Physics, Mathematics and Engineering, " January 1998, and "Nonequilibrium Dynamics," April 1998; and ran a weekly working group, "Geophysical Modeling."

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Atomic, Molecular, Optical, and Plasma Physics, Fluids, and Beams

Liquid Lead and Lead/Bismuth Technology for Use in Subcritical Systems Applied to Nuclear Waste Destruction

96634

Francesco Venneri

The objective of this project is to acquire knowledge of lead and lead/bismuth eutectic (LBE) technology for spallation-neutron-source and nuclear-coolant applications in systems for the accelerator transmutation of nuclear waste.

This year we constructed and successfully operated an LBE test loop (see the accompanying figure). The loop can transport LBE at over 5 m/s in 2-in. (internal diameter) pipes, providing a realistic environment for thermohydraulic and material studies. It is the first of its kind in over 35 years outside Russia, where LBE coolant technology was developed for submarine reactors. Working with our Russian partners at Obninsk (the Institute of Physics and Power Engineering, or IPPE), we have acquired the basic knowledge of the corrosion control techniques employed in LBE coolant systems. This technique, through the control of the thermodynamic activities of the oxygen content, can prevent corrosion of structural materials in LBE and remove solid admixture contamination of the coolant.

We have also examined the polonium hazards associated with using LBE as a nuclear coolant. Polonium is produced in irradiated bismuth. We investigated the polonium release mechanisms during normal conditions and accidents and found that the polonium problem can be controlled and should not pose major radiological hazards.

An LBE spallation target for installation at the Los Alamos Neutron Science Center is under construction at IPPE. The final statement of work with milestones for the next three years was established; the engineering design of the target was completed; and detailed thermohydraulics and neutronics calculations are currently being performed.

Publications

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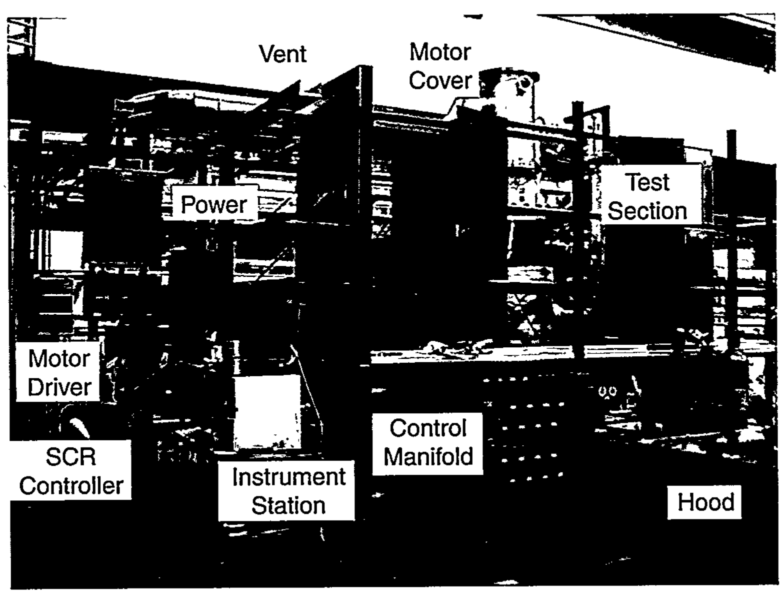

The LBE flow test loop before it was fully assembled.

Advancing X-Ray Hydrodynamic Radiography: Multipulse Converter Development

97607

Daniel Prono

A crucial performance parameter for advanced x-ray radiography systems is very high resolution for distinguishing fine features during a hydrodynamic test. The quality of resolution depends on how closely the x-ray source resembles an ideal point source. Analysis and simulations of how various finite-sized x-ray sources influence resolution have shown that if we can maintain an x-ray source size of less than 1-mm diameter, then we can resolve fine features.

The dilemma is that the x-ray source is produced by the impact of a small-diameter electron beam onto a high-atomic-number converter with an energy fluence that is more than sufficient to create a rapidly expanding plume from the 1-mm-diameter spot. The plasma plume quickly expands many centimeters from the converter surface, with the result that later incoming electron-beam pulses can be disrupted by interactions with the plume. A key feature of the expanding plasma plume is therefore the time evolution of its velocity and density contours.

By applying Saha and other kinetic thermal-equilibrium equations, we have extended the normal hydrodynamic calculations, which span particle densities from 10^{22} to $10^{19}/cm^3$, down 7 orders of magnitude to ionized densities of $10^{12}/cm^3$. To test and correct our predictive algorithms, we are deploying several interferometers that will allow determination of line densities (density times path length) to $10^{12}/cm^3$ (see accompanying figure). These interferometers will be used at various axial positions in front of the target (converter) surfaces so that both axial and radial velocity/density contours can be measured as a function of input-beam parameters and target material composition. Since interferometry integrates densities along the interferometer's line of sight, radial velocity and density contours will be based on assumptions of azimuthal symmetry and mass/momentum conservation.

Numerical simulations indicate (1) that the plasma plume is nearly isotropic and that the leading plasma edge has a plasma density of $\sim 10^{14}/cm^3$ with expansion velocities asymptotic to ~4 cm/ms; (2) that the bulk of the atomically neutral ejecta has a density of $\sim 10^{16}/cm^3$ with expansion velocities asymptotic to ~1.5 cm/ms; and (3) that the partially ionized target melt has a neutral density of $10^{19}/cm^3$ with a 1% ionized plasma density of $10^{17}/cm^3$ and expansion velocities asymptotic to ~0.5 cm/ms. Additionally, simulations indicate that a radial shock is launched at speeds of ~0.3 cm/ms in the target material itself.

The final phase of this research is to perform the interferometry experiments that will validate and probably correct these simulations of the plasma-plume characteristics and to empirically evaluate techniques that will slow the plume expansion so that multiple incoming electron-beam pulses will not have their spot size disrupted.

Publications

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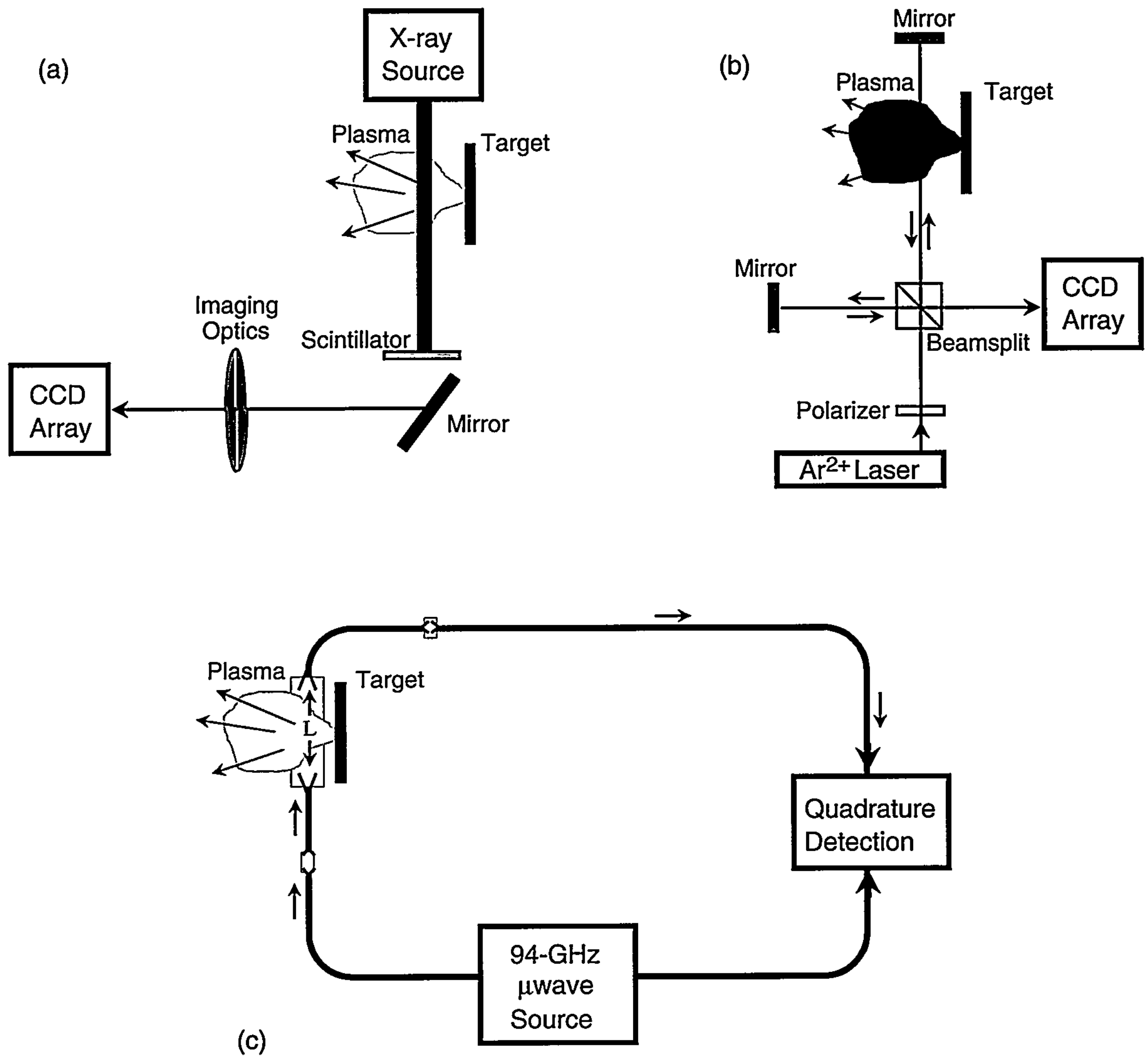

Schematics of three experimental configurations being used to diagnose the temporal and spatial evolution of the plasma plume's density and velocity. An x-ray shadowgraph technique (a) measures the expansion characteristics of atomically neutral hydrodynamic ejecta. Two other interferometers measure the expansion characteristics of the ionized portion of the plasma plume. An argon laser interferometer (b) measures plasma densities in the range of 10^{15} to $10^{21}/cm^3$; a 94-GHz microwave interferometer (c) measures plasma densities in the range of 10^{10} to $10^{14}/cm^3$. In a series of experiments, the interrogating beam of x-rays, laser light, or microwaves is moved progressively farther from the target. From the resulting sets of sequential measurements, we can quantify the temporal and spatial expansion characteristics of the entire range of densities encountered. Results are then compared with predictive simulation codes.

Advancing X-Ray Hydrodynamic Radiography: Multipulse Accelerator Cores and Injectors

97606

Daniel Prono

This project involves theoretical studies of advanced induction accelerators focused on understanding emittance growth mechanisms for the next generation of high-brightness, high-current induction linacs. We have studied two classes of emittance growth mechanisms, both of which can degrade the electron-beam emittance within these accelerators. The first class involves a parametric instability between beam density nonuniformities and differential betatron motion. We found that the growth rate depends on a scaling parameter, $[2I(K^{1/2})]/[A\,Z']$, where I is the beam current, K is the effective betatron wavelength, A is the Alfen current, and Z' is the axial gradient in beam energy. If this scaling parameter is less than unity, the emittance growth is logarithmic; if it is about unity, the growth is linear; and if it is greater than unity, the growth is exponential (see accompanying figure).

The second class of emittance growth is due to geometric effects. The dominant effect comes from conversion of the beam's kinetic energy to potential energy and vice versa as the beam converges and expands (modulation of the beam profile). Even though the radial force is linear, this energy conversion leads to a nonlinear radial equation of motion and thus to an emittance growth. This conversion was observed to account for the long-term emittance growth in accelerator designs for the Laboratory's Dual-Axis Radiographic Hydrodynamic Test (DARHT) facility.

The initial decrease in emittance of the beam as it exits the electron gun for these accelerator designs is due to a nonlinearity in electrostatic focusing from the anode. The nonlinearity from the convergence of the beam is in the opposite direction, leading to a long-term emittance increase as the beam is focused downstream. Also, the short-period emittance oscillations in these designs are coherent transverse plasma oscillations. We studied how these oscillations could affect emittance measurements (based on the conventional solenoid focal-length scan) and found that they caused significant errors in emittance measurements for beams in the space-charge-dominated regime.

Publications

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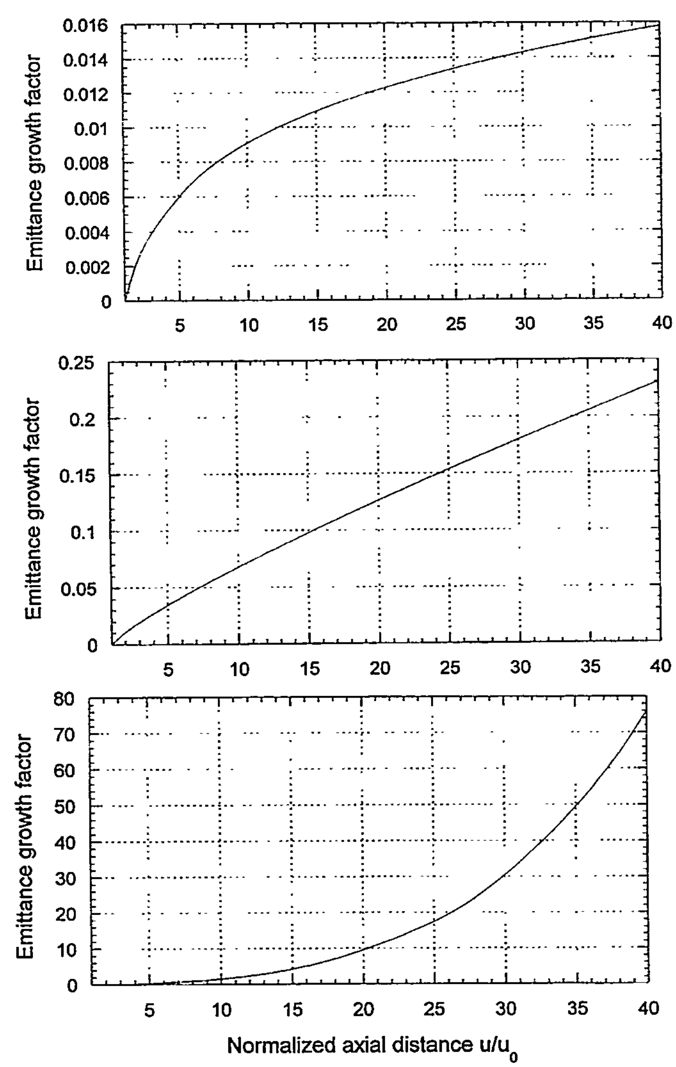


Influence of the scaling parameter $[2I(K^{1/2})]/[A\,Z']$ on emittance growth rate: (top) parameter has a value of 0.5, (middle) parameter is 2, and (bottom) parameter is 5.

Instrumentation and Diagnostics

Advancing X-Ray Hydrodynamic Radiography: Multipulse X-Ray Detectors

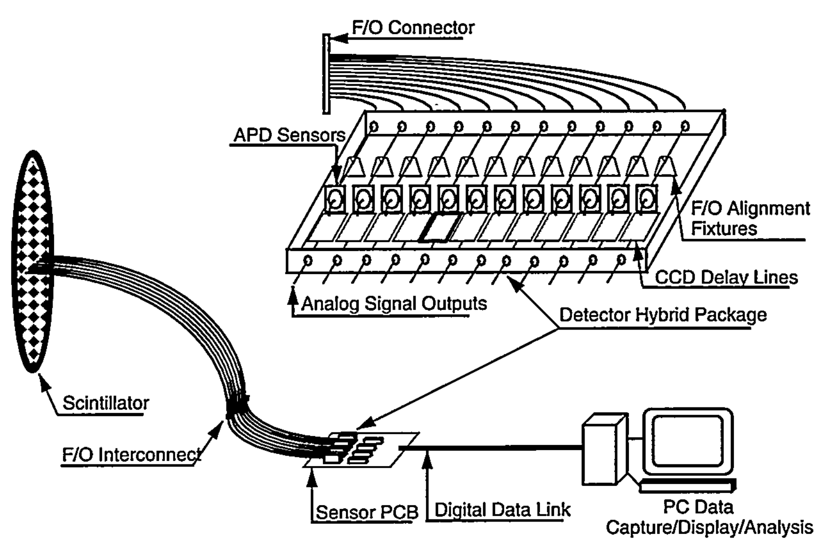
97608

Daniel Prono

Advanced radiography has the goal of validating the time-dependent details of computer codes that predict implosion dynamics. To accomplish this goal, radiographic images of dynamic tests should be taken with a framing rate that is significantly faster than the velocity of the event being recorded. The time duration of each frame and the number of frames being recorded pose additional constraints on the radiographic system. Taken together, these parameters are very difficult to satisfy with current imaging systems. We are therefore developing x-ray detectors that will be able to record images at rates up to 200 MHz. In addition, approximately 1000 frames of data will be taken during an event. The timing and duration of each frame will potentially be independently controlled. This independence will give maximum flexibility for interfacing to important time points for model verification.

Our x-ray detectors are designed to accept visible photons from a large pixelated scintillator through a fiber-optic light guide. The 20- × 20-cm^2 scintillator is made up of 1-mm^2 pixels. In the complete system, each pixel to be recorded will be coupled by a fiber-optic link to a detector channel. Unimportant areas of the image can be left unsampled, while important areas can be oversampled, resulting in higher resolution and/or improved statistics through averaging the data.

For an initial demonstration, we are developing a 40-channel system (see schematic). Detectors are packaged in groups of 12 in a cooled hybrid package. Each package contains 10 detectors that are active and 2 that are reserved as backup sensors, resulting in 4 hybrid packages for the 40-channel system. The fibers from the scintillator mate in groups of 10 or 12 to the fibers that are pigtailed from each hybrid package, with the mating done through linear, fiber-optic (F/O) array interconnects. Inside a hybrid package, each detector has an avalanche photodiode (APD), a charge-coupled device (CCD), and a CCD delay line within its integrated circuit (IC) such that each IC can store ~1000 samples of event data. After all data have been recorded, the delay-line CCD is clocked to transfer the data to a 16-bit analog-to-digital converter (ADC). The digitized data are then recorded and displayed by commercially available systems.



Experimental setup of the advanced, multipulse radiographic imaging system.

Geoscience, Space Science, and Astrophysics

Urban Security

97616

Grant Heiken

All cities, regardless of size, have problems related to security, energy, water, nutrition, economics, and the environment that make them vulnerable to the hazards of natural events, such as a hurricane, or the unnatural event of a terrorist attack. For cities to be safe and sustainable, we must implement long-range urban planning and risk assessment tools and not rely on reactive decision-making. The tools must be based on an accurate assessment of the interrelationships among the many complex processes that occur in the urban environment.

Understanding urban systems demands multidisciplinary approaches that account for physical processes, economic and social factors, and nonlinear feedback across a broad range of scales and disparate process phenomena. We are using as components of an urban modeling system the many state-of-the-art models developed by the strong research programs in the defense, environmental, and computational arenas at Los Alamos. These include programs in transportation, air quality, groundwater transport, energy distribution, network theory, communications, synthetic population modeling, natural hazards, and risk assessment. We are building on these and other modeling tools, modifying them for urban settings, and linking them together as an integrated simulation system that takes advantage of our high-performance computing platforms. One modeling system is illustrated in the accompanying figure.

Publications

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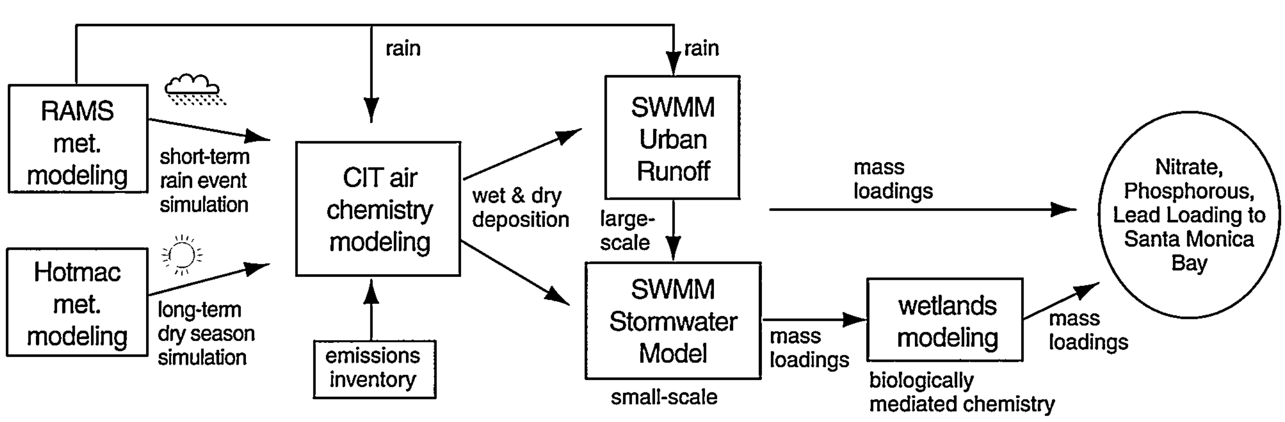
Valentine, G.A., "Damage to Structures from Pyroclastic Flows and Surges, Inferred from Nuclear Weapons Effects" (to be published in *J. Volcanol. Geotherm. Res.*).



Modeling system for following pollutants through air-water pathways in an urban environment. With some modifications, the fate of pollutants from other sources could be modeled as well, for example, accidental releases of toxic agents, heavy metals from brake pads, or noxious vapors from waste.

Earth Materials and Earth Dynamics

97814

Thomas Shankland

We are using complementary laboratory experiments and theoretical studies to describe processes responsible for the earth's structure, composition, and internal activity. Because fluids are important in nearly every geological process and are of great concern in waste isolation and water quality issues, we instituted a new thrust to examine rock-fluid interactions. Complex feedback relationships affect fluid flow in the presence of temperature and compositional gradients. We are discovering that, because of the cracks and pores through which fluids flow, rocks display a surprising variety of nonlinear elastic effects. Nonlinear attributes of rock affect earthquake slip, reservoir subsidence, seismic wave propagation and attenuation, stress fatigue damage, and hydraulic fracturing.

Using a combination of a Los Alamos three-dimensional (3-D) convection code and deformation-induced textures based on measurements at the Los Alamos Neutron Science Center, we have advanced the hypothesis that the elastic anisotropy measured for the earth's inner core can be attributed to convection. Other measurements of minerals in the clinopyroxene structure have permitted the first solid calculation of the elastic properties of clinopyroxene, which constitutes at least a third of the region in the depth range of 200–500 km. Closer to the surface, our two-dimensional (2-D) and now 3-D calculations of coupled heat and mass transfer in the vicinity of magma intrusions have illustrated strikingly both metamorphic processes and hydrocarbon formation in the earth's crust. As applied to crustal rocks, resonant ultrasound spectroscopy (RUS) extracts the elastic tensor of a sample from measured resonance frequencies. Assuming isotropy, we have found that RUS provides reliable results for hard materials such as basalt, primarily because a large number of resonances can be accurately determined.

Publications

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Lutterotti, L., et al., "Combined Texture and Structure Analysis of Deformed Limestone from Time-of-Flight Neutron Diffraction Spectra," *J. Appl. Phys.* **81**, 594 (1997).

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Wiryana, S., et al., "The Equation of State of Water to 200°C and 3.5 GPa: Model Potentials and the Experimental Pressure Scale," *Earth Planet. Sci. Lett.* **163**, 123 (1998).

Zhao, Y., et al., "Thermal Equation of State of Monoclinic Pyroxene: $CaMgSi_2O_6$ Diopside," *Review of High Pressure Science & Technology* **7**, 25 (1998).

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).

Lithospheric Processes

97815

W. Scott Baldridge

We are examining problems involving the thermal structure of the lithosphere (the crust and upper mantle of the earth), the thickness of the lithosphere, melt formation within and beneath the lithosphere, residence time of magmas in the lithosphere, and processes by which the lithosphere is thinned and ruptured during rifting. We are using a variety of seismic and geochemical techniques, each selected to provide distinct information. We expect to construct a better model of upper mantle velocity structure in different regions of the earth, to enhance our understanding of magma generation and extraction in different tectonic environments, and to improve our ability to understand and model fluid transport processes on several scales.

Our major goals last year were to use seismic data to characterize the structure of the lithosphere in selected regions, to understand magmatism and magma chamber processes, and to numerically model cooling of igneous units. In the Tarim Basin of China, waveform modeling of broad-band seismic data allowed us to infer a crustal thickness of 50–55 km, which is approximately the same thickness as that of the adjacent and topographically higher Tien Shan region. We do not understand why these contrasting regions have similar crustal thicknesses. For the southern Basin and Range in the western United States, we determined velocity anisotropy to document upper mantle strain history. Significant differences in regional anisotropy were observed.

We continued to identify, analyze, and trace layers of volcanic ash in the Ethiopian Rift to establish a regional volcanic history for the last 5 million years. In northeastern Spain, new neodymium and strontium isotopic analyses document that continental lithosphere was thinned more than 10 million years ago. We are uncertain whether thinning is related to the central European rift system or to collisional processes. To understand the role of crustal contamination in magmas, we continue in the difficult task of analyzing and interpreting disequilibria in uranium and thorium decay series (see figure). Finally, we have adapted computer codes to model the heat and mass transport of cooling tuff sheets.

Publications

Keating, G.N., and G.A. Valentine, "Proximal Stratigraphy and Syn-Eruptive Faulting in Rhyolitic Grants Ridge Tuff, New Mexico, USA," *Journal of Volcanology and Geothermal Research* **81**, 37 (1998).

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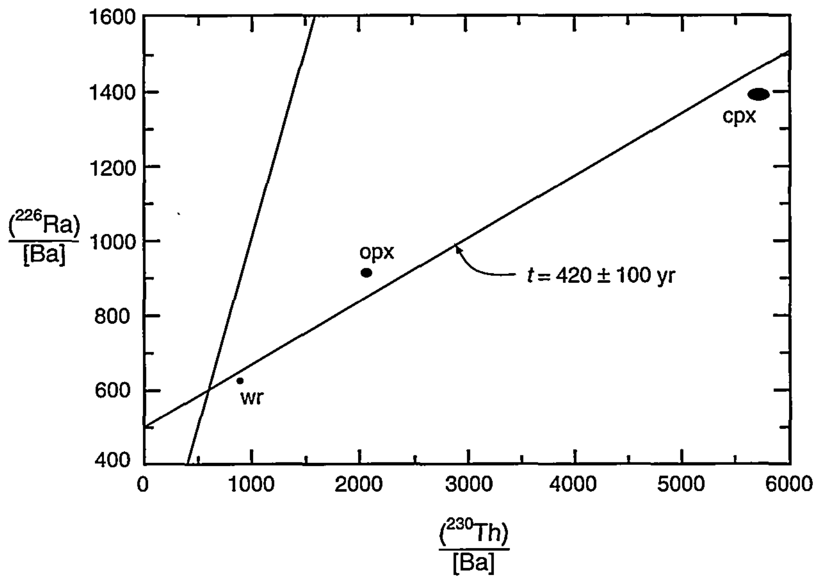


Radium-thorium isochron diagram for a xenolith brought to the surface in the 1800 A.D. eruption of the Hualalai volcano in Hawaii. A line with slope of 1, representing the radioactive equilibrium between ^{230}Th and ^{226}Ra, is shown for reference. The points labeled cpx, opx, and wr represent, respectively, clinopyroxene, orthopyroxene, and whole rock. The size of the symbols is equivalent to the measurement error. The line labeled $t = 420 \pm 100$ yr is an isochron. The fact that the analyses do not lie on the line could indicate that the minerals from the melt crystallized over a time period exceeding 100 years, or that radium, barium, or thorium underwent postcrystallization mobility.

Remote-Sensing Science Thrust

96622

Siegfried Gerstl

Our objective is to develop an integrated Laboratory competency in remote-sensing science. The technical goal is to develop fundamental new technology capabilities that can be applied to science and technology problems that pertain to the nonproliferation of nuclear, biological, and chemical weapons of mass destruction or to environmental security for the purposes of threat reduction.

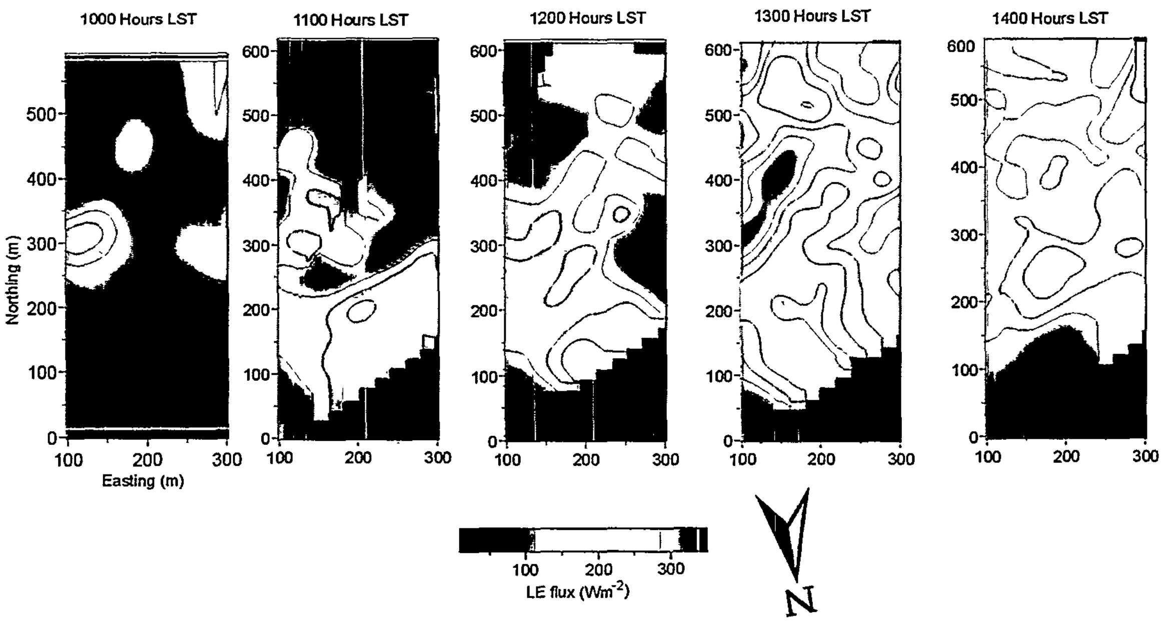
We are developing both active and passive remote-sensing technologies with ground- and space-based sensors that detect chemical plumes. We are also attempting to understand the atmospheric boundary layer and to develop computational tools and new remote-sensing techniques. The specific technologies we are working with include Raman and wide-angle imaging lidars, a Fourier transform infrared spectrometer (FTIR), an imaging spatial heterodyne spectrometer (ISHS), and a microwave interferometer.

Last year we demonstrated proof of our concepts and technology with measurements made by two of our new remote-sensing instruments at the international Semi-Arid Land-Surface-Atmosphere Mountain Experiment. Using the microwave interferometer, we acquired unique measurements of water-vapor heterogeneities in the atmospheric boundary layer (up to an altitude of about 2 km), which is an unprecedented quantification of the spatial and temporal fluctuations of water-vapor concentrations in the convective boundary layer, leading to structure function specifications.

With the Raman lidar instrument, we achieved high-resolution (1- to 2-m) measurements of water-vapor concentrations much closer to the ground and up to about 50 m from distances of several kilometers. These data allow for the first time the quantification of evapotranspiration processes above extended vegetated surfaces (see figure) and thus become important tools for regional water management. For this development, we were honored with a new external research grant from the U.S. Department of Interior's Bureau of Reclamation.

We deployed our newly developed, modified FTIR at two actively degassing volcanoes, the Popocatepetl in Mexico and Mt. Aetna in Sicily, to remotely and quantitatively measure the volcanic gas composition at ranges of up to tens of kilometers. These measurements confirmed and demonstrated the spectroscopic detection of SO_2, H_2O, and HCl at ranges of up to 6 km with good agreement from direct sampling. In the Popocatepetl data, we monitored variations of SO_2, SiF_4, HCl, and HF and extended the range for sky-background passive infrared techniques to more than 17 km.

We completed the development of another new remote-sensing instrument, the ISHS, and it is starting to acquire test data. Preliminary results appear so promising that the DOE Office of Nonproliferation has granted us a new follow-on project to study the applicability of the ISHS to detect



The patterns displayed on the five maps represent the evapotranspiration over a complex vegetated canopy during the critical midday periods when water-vapor fluxes are highest. The structures observed on the images represent the behavior of the vegetation in respect to the varying environmental conditions associated with the atmosphere, soils, and water.

gaseous effluents from chemical processing plants from very long distances.

Publications

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Low-Luminosity, Compact Stellar Objects and the Size of the Universe

97811

Richard Epstein

Our group examined high-energy astrophysical systems using a combined experimental and theoretical approach. We operated a preliminary test version of our new gamma-ray observatory, Milagro, to check its design and data-acquisition capabilities; we developed critical components for a vibration-free, optical cryocooler for space-based infrared and gamma-ray observations; and we computed the range of stellar encounters and the composition of stellar remnants in our galaxy. The objective of the latter activity is to determine whether stellar encounters are a viable mechanism to explain the paucity of red supergiants in the very center of the galaxy and to see whether primordial black holes formed in the early universe are a significant constituent of the dark matter in the universe.

This year we installed and operated the Milagro subsystem, and we wrote and debugged the on-line code. This subsystem, with 225 photomultiplier tubes, was installed in September and has recorded over 3 billion events. For the optical cryocooler tasks, we assessed the potential of three materials by measuring their absorption and emission spectra at several temperatures. We found that one material, BIGaZYT glass, could be twice as efficient as the currently used material. We investigated the likelihood that gravitational microlensing observations in the galactic halo are due to black holes or neutron stars or both. We showed that chemical abundance measurements allow only 40% of the microlensing events to be from compact stellar remnants.

Publications

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Lei, G., et al., "Spectroscopic Evaluation of Yb3+-Doped Glasses for Optical Refrigeration" (to be published in *IEEE J. Quantum Electron.).*

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Solar Terrestrial Coupling through Space Plasma Processes

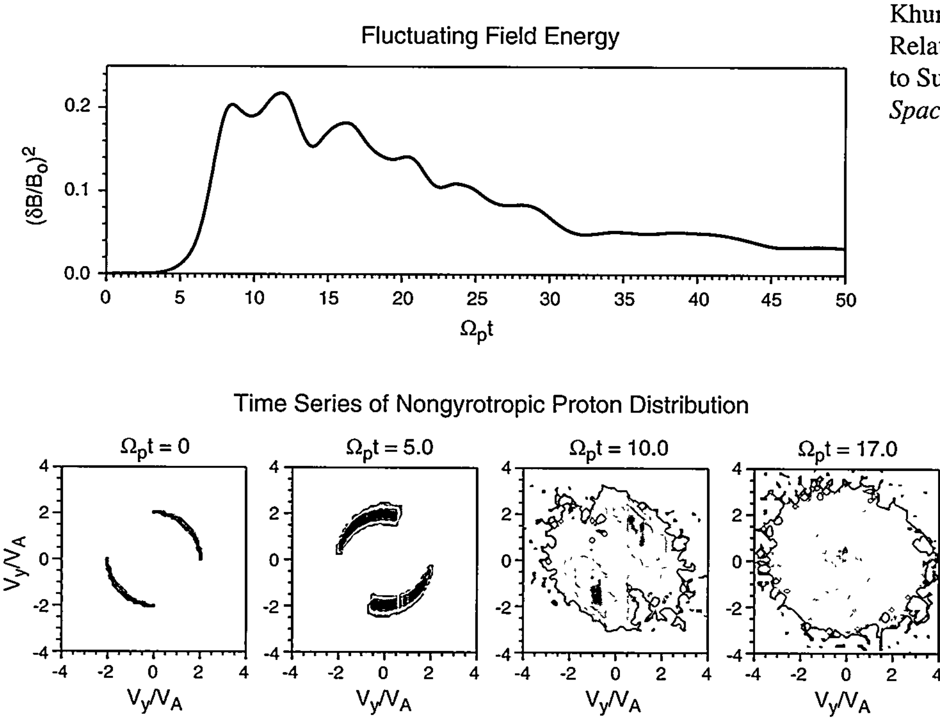
97803

Joachim Birn

This project combines efforts in space data analysis with theory and computer simulations to advance the understanding of solar-terrestrial interaction through space plasma processes. Charged particles energized by solar wind-magnetosphere interaction and micro/macroscale instabilities in the magnetosphere can strongly affect Earth's atmosphere, ionosphere, and magnetosphere, where programmatic and scientific satellites reside. This study focuses on plasma and field processes relevant for the underlying solar-terrestrial coupling processes.

The continued research concerns primarily the regions where different plasma populations interact with each other, causing interconnected magnetic field structures called flux ropes. Simulations of the bow shock and the magnetopause have shed further light on the nature of surface waves and their contribution to ion diffusion in this boundary region. In these simulations a curved bow shock and its associated foreshock and a turbulent magnetosheath are formed. We also performed simulations of tearing instabilities at the magnetopause, which is responsible for the transport of particles and magnetic flux across this surface. We found that in sharp contrast to previous nonlinear theories, the multimode tearing islands can grow to large amplitudes, resulting in large transport of magnetic flux and mixing of the magnetosheath and magnetospheric plasmas.

We have further studied the role of magnetic flux ropes in the transport of plasma in Earth's long magnetic tail and the consequences of the electromagnetic proton-anisotropy instability in the terrestrial magnetosphere. Results (see accompanying figure) show that the enhanced fluctuations from this instability have a significant effect on the ion distributions in the inner magnetosphere. While these fluctuations preserve the initial character of a hot proton distribution, they also rapidly drive a hot distribution that is initially nongyrotropic toward the gyrotropic condition.



Results from simulations of electromagnetic proton-anisotropy instability.

Publications

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Coupled Environmental Modeling

98604

C.L. Winter

We are developing high-resolution computer models of coupled environmental systems. Coupled systems consist of discrete physical domains (for instance, the atmosphere or land surfaces) that interact nonlinearly and operate at different time and space scales.

The study of coupled systems will lead to a new generation of modeling tools that can be used to predict the evolution of environmental phenomena. It will position Los Alamos to address a wide variety of future environmental security issues ranging from (1) global challenges, such as carbon and water cycles; (2) regional problems, such as fresh water supply and agriculture; and (3) local threats, such as wildfire and flooding.

Coupled environmental systems are also natural analogs of weapons systems, which comprise diverse physical elements (hydrodynamics and radiative transport that evolve at different rates and over different volumes, for example). In particular, we are emphasizing the importance of interdomain exchanges of mass and energy. These have not previously been represented with the level of detail required to capture environmental couplings because adequate computational resources and physical models have been unavailable. Similar problems arise in computational approaches to weapons stewardship.

During the past year we accomplished significant tasks in all aspects of this project. To support both fire and regional water modeling, we helped develop and install a parallelized version of the Regional Atmospheric Modeling System (RAMS) code on the ASCI Blue Mountain machine. Regarding fire modeling specifically, we also integrated an empirical fire behavior model (BEHAVE) with the RAMS mesoscale climate model to simulate wildfires in Colorado, California, and the Kennedy Space Center in Florida. We developed the FIRETEC model of fire behavior based on full turbulence physics and the HighGrad weather code for dealing with high-pressure gradients found near boundaries of forest fires. From the FIRETEC and HighGrad codes, we then began integrating a high-resolution fire/weather model. This model includes a diffusive radiation/heat transfer scheme capable of dealing with optically thin or thick media. Regarding regional water resource modeling, we installed land surface, groundwater, mesoscale climate and river network models on the ASCI Blue Mountain machine. We also developed a method for scaling mesoscale climate simulations and data to land-surface scales. Then we integrated mesoscale climate and land-surface models on the ASCI machine using the down-scaling method. Because high-resolution regional water models demand large volumes of data, we developed methods for managing and mining large, heterogeneous databases.

Publications

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Elements of Water Resources and Urban Pollution

97812

Charles Keller

Global environmental security (the protection of our natural resources) is one of the Laboratory's tactical goals, and establishing a strong capability in water resources and air quality is called for in the Earth and Environmental Science Division's strategic plan. The basic research that we are doing in this multitask project supports both goals. This effort has been substantially enhanced by collaborations with University of California faculty, postdoctoral staff, and graduate students.

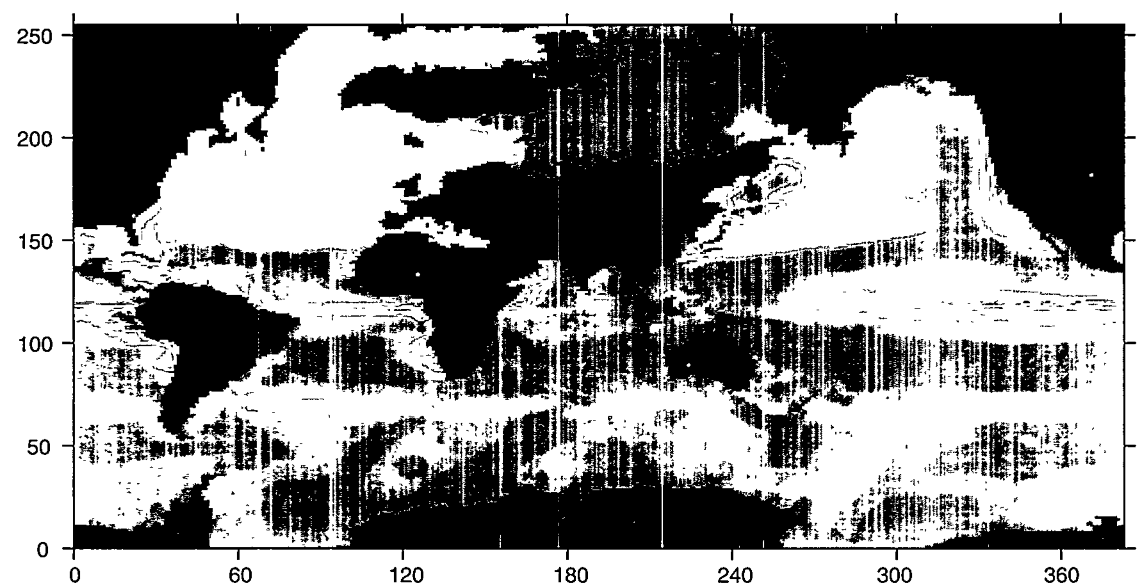
This year we made progress in several areas. We assembled a coupled suite of marine ecology, chemistry, and ocean transport models and used them to simulate the general features of the oceanic nutrient upwelling and phytoplanktonic blooms that occur seasonally. For the first time, we extended this simulation to cover the entire earth, starting from the Pacific Basin (see figure). We have also used these coupled codes to simulate fertilization of ocean phytoplankton that could occur if airborne nitrates were generated from East Asian pollution and carried downwind.

When we simulated the well-observed 1997 California flood with and without coupling the regional ocean code to the atmospheric one, we successfully showed that the ocean nearest a region plays a crucial role in its coastal precipitation. However, when we attempted to simulate the 1998 El Niño precipitation in California by using the same approach, we were not as successful because the resulting large-scale spatial boundary conditions could not resolve important details of the flow. The results on our third simulation were encouraging; they demonstrated the summer monsoon precipitation in the southwest United States.

Our numerical simulations of ocean currents in the western Pacific clearly showed a difference when the shallow part of the ocean located over the continental shelf was included. Its interaction with western boundary currents generated complicated solutions that occurred both regularly and irregularly within two distinct regions.

Laboratory measurements (in stratospheric conditions) of heterogeneous reactions on ice surfaces have shown that the uptake of hydrogen chloride is significantly lower than is commonly thought, which calls into question the significance of hydrogen chloride in chlorine-activation and ozone-depletion reactions.

We have also bridged the gap between our atmospheric precipitation code and subsurface fluid transport code with the SPLASH (simulator for processes and landscapes, surface/subsurface hydrology) code, which follows precipitation in its subsequent runoff, stream flow, ground seepage, and evapo/transpiration. This gives us a new capability to simulate the entire hydrological cycle and its impact on water resources.



Surface phytoplankton distribution throughout the global oceans is simulated by a coupled suite of marine ecology, chemistry, and ocean transport models that we introduced into the Los Alamos ocean circulation code, POP (Parallel Ocean Program). The map projection is distorted at high northern latitudes because the mesh pole has been shifted into the boreal land mass. Some major patterns that can be seen are the spring-summer bloom apparent at high northern latitudes and equatorial upwelling and growth interaction with tropical eddy structures.

Publications

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Algorithm Development for Ocean Models

97808

Len Margolin

We are applying technology developed by several Los Alamos programs, ranging from weapons physics to mesoscale atmospherics and general turbulence modeling, toward improving numerical models used for global ocean simulations. These improvements address numerical stability, computational efficiency, and enhanced physical realizability and are targeted toward the two ocean models currently supported at Los Alamos: the Parallel Ocean Program (POP) and the Miami Isopycnic Coordinate Ocean Model (MICOM). These two models are used in the study of ocean circulation and long-term climate change. Our project involves collaborations with university staff in disciplines such as applied mathematics, oceanography, and engineering turbulence.

During this past year we have started to apply our global atmospheric model to simulating the idealized Held-Suarez climate, which has become a standard for validating global atmospheric models. Our new, forward-in-time methods perform well, reproducing the standard results accurately and efficiently. We have generalized the formalism of "compatible differencing," originally designed for Lagrangian hydrodynamics schemes, to include Eulerian schemes in one dimension. The new methods ensure the consistency of the various physical fluxes by preserving the analytic relations between the fluxes. At the same time, they are computationally more efficient than other methods because they do not require separate use of expensive nonlinear advection schemes for each variable.

We have also begun exploring the suitability of using our nonoscillatory advection scheme MPDATA as an implicit turbulence model without any explicit subgrid-scale (SGS) model. In simulations of a convective boundary layer, we have demonstrated that MPDATA can reproduce the results of a sophisticated Smagorinski SGS and that the results compare well with those from experimental data and with those from data obtained by other models. The advantage of using our model is that, because it has no undetermined constants, our model is more generally applicable to different problems without requiring tuning. We are continuing to develop a mathematical basis for these results, including investigating the relationship of our "modified equations" to the newly proposed Camassa-Holm equations.

Publications

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Smolarkiewicz, P.K., et al., "Forward-in-Time Differencing for Fluids: Nonhydrostatic Modeling of Fluid Motions on a Sphere," in *Proc. ECMWF Seminar on Recent Develop. in Numer. Methods for Atmospheric Modelling* (ECMWF, Reading, United Kingdom, in press).

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Theoretical and Observational Studies of the Earth's Mantle and Core

97813

Gary Glatzmaier

We are studying convection of heat and composition in the earth's mantle and core, which determines the thermal, chemical, and magnetic structures of the earth's deep interior. This convection also affects the structure and evolution of the environment at the earth's surface by forming mountains and ocean basins and causing volcanoes and earthquakes.

Our three-dimensional (3-D) models of mantle convection now include more realistic representations of tectonic plates that allow the plates to self-consistently evolve. These models have been used to study how distinct geochemical reservoirs in the earth's mantle have developed. Other simulations have been combined with seismic imaging of the earth's mantle to improve our understanding of its structure and evolution. For example, a 119-million-year mantle-convection simulation, which ends at the present epoch and is guided by imposed tectonic plate geometries and motions based on the actual plate reconstructions, has produced very good agreement with today's seismic tomography of the earth's mantle.

We have also continued our 3-D numerical simulations of the earth's core, including local simulations that investigate the effects of anisotropic turbulence; global simulations that test the sensitivity of different model parameters; and analysis of our simulated geomagnetic field compared to the paleomagnetic record. The accompanying figure shows an example of a magnetic reversal that spontaneously occurred in one of our 250,000-year simulations; the reversal event took only 6,000 years.

Publications

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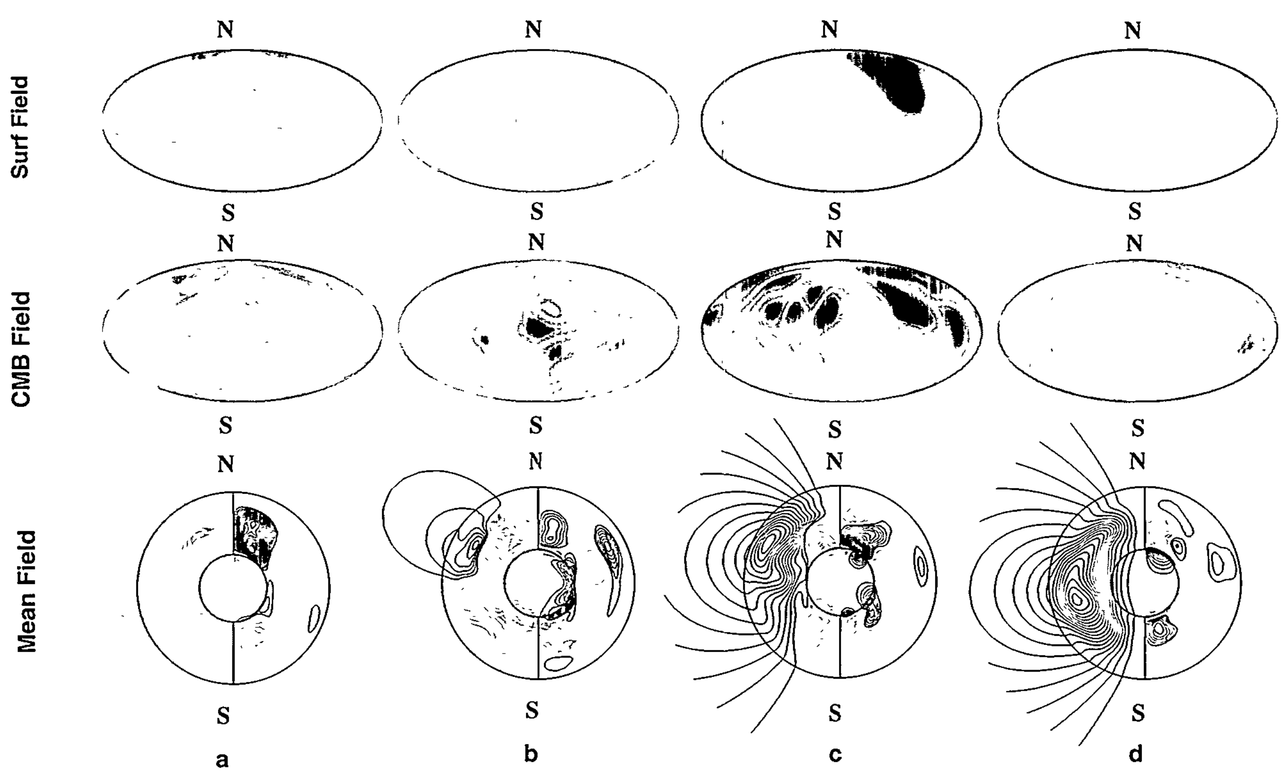

A sequence of snapshots of the radial component of the field at the surface (top row) and at the core-mantle boundary (middle row) and of the longitudinally averaged field through the interior (bottom row). In the bottom row, the dark gray lines represent the outwardly directed field and the light gray lines represent the inwardly directed field. The columns a through d are snapshots made at 3,000-year intervals around a field reversal.

Nuclear and Particle Physics

An Ultracold-Neutron Source

98607

Thomas Bowles

Our objective is to improve the performance of the rotor source of ultracold neutrons (UCNs) operating at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) such that the flux produced is sufficient to begin a program of fundamental physics using this source. This source creates UCNs by Doppler-shifted Bragg scattering of cold neutrons from a moving reflector. The reflector is a package of mica crystals.

Our previous work indicated that improving the mica scatterer would significantly improve UCN production. The existing phlogopite mica crystals contained significant amounts of hydrogen, which causes scattering of neutrons and limits the thickness (and hence the reflectivity) of the crystal package.

This year we identified a source of an artificial fluorinated phlogopite material and obtained sufficient material to construct a new crystal package. When we measured the neutron scattering from this material, we discovered a large variation of reflectivity. We therefore made scattering measurements on each individual piece of mica in order to choose the best sample from which to construct the new crystal package.

Analysis of previous data showed a significant difference in the rotor timing with respect to the neutron beam as measured at the shaft of the drive motor compared with that measured at the end of the crystal package. We identified the motor coupling as a source of the timing variations. We have redesigned this coupling and constructed the improved design.

Our new crystal package and redesigned motor coupling have not yet been tested in the neutron beam at the Los Alamos Neutron Science Center because the facility has been down for major upgrades. We have beam time scheduled in early calendar year 1999 to test the effect of these system improvements we have made, as well as the improvement we expect from the increased neutron flux at the MLNSC.

Publications

Seestrom, S.J., et al., "Recent UCN Source Developments at Los Alamos" (Sixth International Seminar on the Interaction of Neutrons with Nuclei, Dubna, Russia, May 13–16, 1998).

Advanced Nuclear Measurements Science

98601

Mark Pickrell

The purpose of this project is to develop the underlying science that will enable measurements of fissile material in the next decade. The research focuses on difficult-to-measure items that have not been amenable to measurement in the past. Specifically, we address materials that are heavily shielded, have little or no passive signal, or are encased in a matrix with strongly interfering radionuclides.

The project consists of seven components that are designed to provide, in an interactive fashion, all of the necessary measurement science to construct the fissile material assay systems that will be needed for the next decade. These seven components are (1) a study of worldwide safeguards systems and sensitivity study; (2) an investigation of data-fusion and data-mining techniques for nondestructive assay data; (3) the development of room-temperature, medium-resolution, CdZnTe gamma-ray detectors;(4) the development of fast, scintillator-based neutron detectors; (5) the development of a quantitative use of prompt-gamma, neutron-activation analysis; (6) the development of the delayed-neutron re-interrogation method for measuring fissile material in highly shielded configurations; and (7) the development of the next generation of compensated, fiber-optic calorimeters. Each of these tasks provides an essential component of a comprehensive measurement ensemble.

We achieved significant progress this last fiscal year. We completed a conceptual design for an advanced calorimeter capable of measuring (in record times) nominal quantities of highly enriched uranium (see first figure). We also designed, built, and tested advanced CdZnTe detectors. Moreover, we have demonstrated the capability of making isotopic measurements with these devices; this has never been possible with room-temperature devices. Finally, we were able to demonstrate considerable gamma-ray rejection with the fast, scintillator-based neutron detectors (see second figure).

Publications

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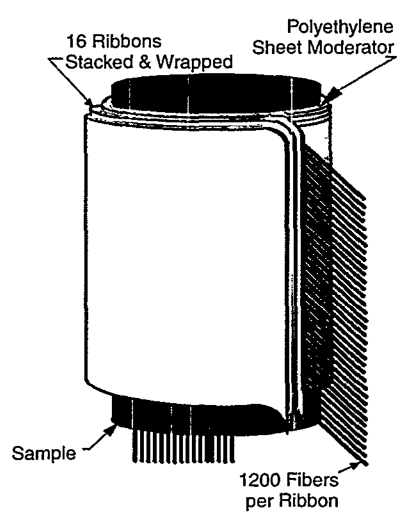

Schematic of a fiber-optic-based calorimeter system showing the fiber-optic, temperature-sensing windings.

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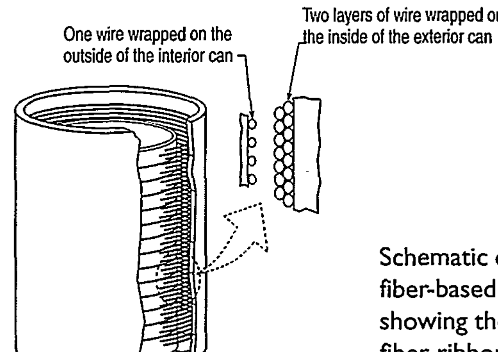
Heger, A.S., et al., "Use of Neutron-Capture Plastic Fibers for Nondestructive Assay" (The 39th Annual Meeting of the Institute of Nuclear Management, Naples, FL, July 26–30, 1998).

Mayo, D.R., et al., "Boron Loaded Scintillating Fibers as Nondestructive Assay Tools," *Bull. Am. Phys. Soc.* **43**, 1090 (1998).

Mayo, D.R., et al., "A Fingerprinting Detector for Special Nuclear Material Based on Optically Coupled BGO/BC454 Scintillators for Special Nuclear Materials with a Mixed Neutron/Gamma-Ray Signal" (The 39th Annual Meeting of the Institute of Nuclear Management, Naples, FL, July 26–30, 1998).



Schematic of a scintillating, fiber-based neutron counter showing the scintillating fiber ribbon.

Miller, M.C., et al., "Neutron Detection and Applications Using a BC454/BGO Array" (1998 Symposium on Radiation Measurements and Applications, Ann Arbor, MI, May 11–14, 1998).

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Advanced Techniques for Producing, Polarizing, and Storing Ultracold Neutrons (UCNs)

98606

Susan Seestrom

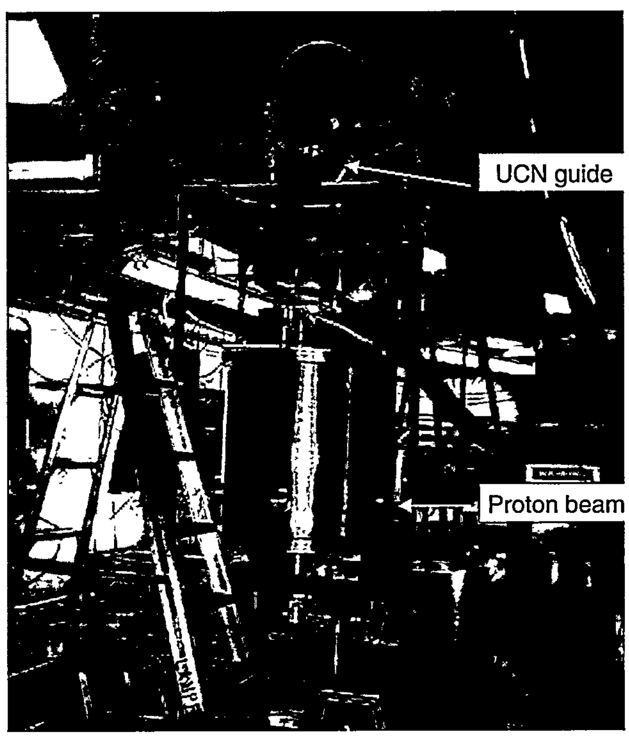
The objective of this project is to develop an advanced source of ultracold neutrons (UCNs) that are produced by a cryogenic converter. This year we took previously developed models for the production of UCNs in cryogenic materials and applied them to specific experimental geometries. We identified the possibility of developing a standalone UCN source in which neutrons are produced by a proton beam and moderated to a cold spectrum, which is then used to produce UCNs. Such a source is ideally suited to the Los Alamos Neutron Science Center (LANSCE) and holds the potential for being the world's highest-intensity UCN source.

We designed and constructed two systems to measure UCN production, one for use in a cold neutron beam at the Hahn-Meitner Institute (HMI) in Berlin and one for use in a proton beam at LANSCE (see figure). At HMI we measured UCN production per neutron. At LANSCE we measured both cold-neutron and UCN production per proton. Comparison of our data with calculations verifies our model of UCN production in frozen deuterium but indicates that UCNs are being lost by absorption or upscattering in the windows of the system. On the basis of these measurements, we have redesigned the system, which is now being prepared for testing.

Publications

Seestrom, S.J., et al., "Proposed Experiment to Measure the Neutron Spin-Electron Correlation in Polarized Neutron Beta Decay with Ultra Cold Neutrons" (Sixth International Seminar on the Interaction of Neutrons with Nuclei, Dubna, Russia, May 1998).

Seestrom, S.J., et al., "Recent UCN Source Developments at Los Alamos" (Sixth International Seminar on the Interaction of Neutrons with Nuclei, Dubna, Russia, May 1998).



Experimental system for measuring UCN production at LANSCE. The arrow on the proton beam label points to the connecting flange for the beam. An unshielded portion of the UCN guide's stainless steel tube can be seen at the top of the photo; the tube terminates in the shielded UCN detector.

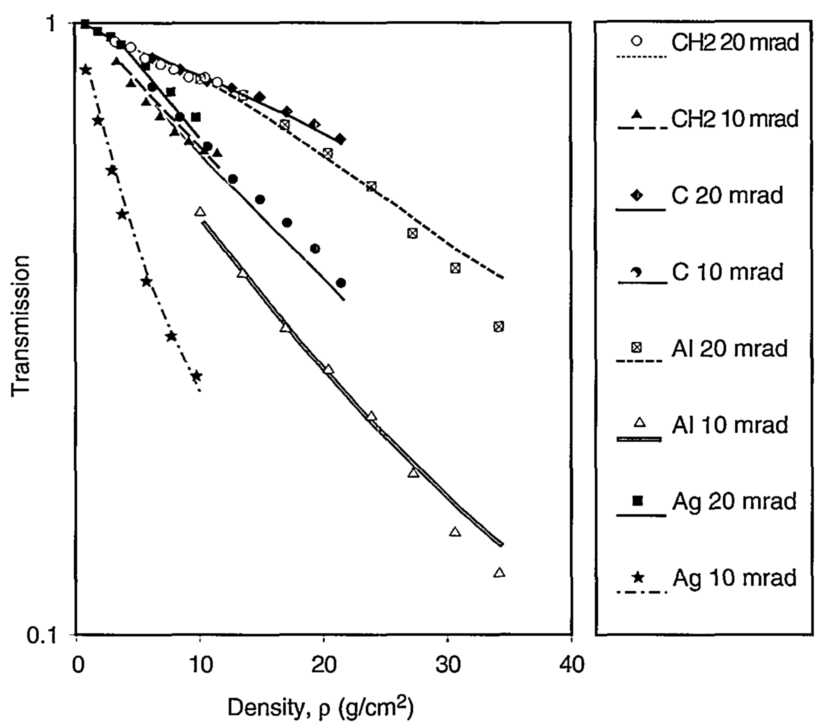
Advanced Dynamic Radiography with Protons

97618

Chris Morris

We are developing multiframe flash radiography using proton beams at the Los Alamos Neutron Science Center (LANSCE). In previous work we successfully demonstrated the use of proton transmission as a radiographic tool, which has broad applications in physics and is especially useful in clarifying the status and potential explosive impact of the nuclear weapons stockpile in the absence of testing. Our objectives for this year were to design, construct, and commission a three-lens proton radiography system in beam line C at LANSCE, which is currently being used in experiments supporting the Science-Based Stockpile Stewardship program.

The three-lens proton radiography system has a series of capabilities, many of which we have demonstrated on static test objects (results shown in the figure). These include tailoring and measuring the beam with the first lens in an upstream location and providing, with two additional lenses, two downstream image locations for material identification supplemented by more-conventional radiography. With this configuration, we achieved significant quantitative measurements of density changes in exploding systems. A set of dynamic experiments with a seven-frame camera system is under way.



Step-wedge (sample) data for four objects taken through the line C system: methylene (CH2), carbon (C), aluminum (Al), and silver (Ag). Transmission is the ratio of flux with the object in place to that with the object out. The difference in transmission between the large and small collimators, or imaging lenses (20 mrad and 10 mrad), can clearly be seen to be dependent on atomic mass (objects are listed in ascending order of atomic mass). This is the first demonstration of material identification in proton radiography in which two lenses are used simultaneously.

Publications

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Quantum Technologies

97615

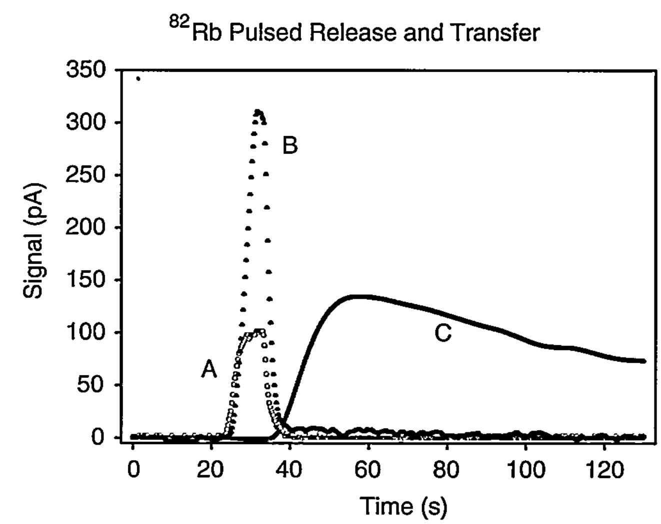
David Vieira

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 optical and magnetic 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.

During the past year we have developed a master-oscillator power amplifier for high-power generation of light at 397 nm from a frequency-doubled, 794-nm diode-laser system, and we will use this system to Doppler-cool and image calcium ions in an ion trap. (See the first figure for an image of Ca^+ ions cooled to rest.) We have demonstrated ultra-high-efficiency, interaction-free measurements and imaging. We have also developed a novel high-brightness source of entangled two-photon states and used this source to demonstrate quantum-mechanical nonlocality with unprecedented precision. We have investigated the mechanism for ion heating in trapped-ion quantum computers and developed new concepts for quantum computation in such systems.

In our atom trapping work, we have had great success in demonstrating the trapping of six million atoms of radioactive rubidium-82 in a magneto-optical trap (MOT) coupled to a mass separator. This represents a hundred-fold improvement in the number of trapped atoms over all previous radioactive atom trapping work. Using this new capability, we have measured the rubidium-82 atomic hyperfine structure of the D1 transition for the first time and remeasured the D2 transition with higher precision. In the area of ultrasensitive detection, we have pushed our detection sensitivity down to the 2000-atom level. Using a double MOT system, we have recently transferred atoms from one MOT to a second MOT with an efficiency of ~50% (see the second figure). A time-orbiting-potential magnetic trap has been constructed, and we are currently preparing to load this trap with polarized rubidium-82 atoms for a high-precision beta-asymmetry experiment.

Image of a string of four Ca^+ ions cooled to a crystallized string (20 μm spacing).



Pulsed release, trapping, and transfer of rubidium-82 in a double magneto-optical (MOT) system. Trace A is the optical pyrometer reading of the catcher foil temperature into which rubidium-82 is implanted. The foil is inductively heated to a temperature of ~750°C for ~10 s. Trace B is the lock-in fluorescence trapping signal from the first MOT. At the ~35-s mark, the first MOT is switched off, and the atoms are "pushed" with a second laser beam over to the second MOT, where they are retrapped. Trace C shows the trapping signal from the second MOT.

Publications

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Zhao, X., et al., "Developing Optical Traps for Ultra-Sensitive Analysis," *SPIE Proc.* **3270**, 70 (1998).

Multidisciplinary Science-Based Bioremediation

96620

James Brainard

Although bioremediation can provide cost-effective solutions to many of the DOE's and the nation's environmental cleanup problems, present bioremediation practices cannot easily destroy some contaminants, notably halogenated hydrocarbons. Many microorganisms contain halocarbon-degrading enzymes, but these enzymes are relatively poor catalysts: they have poor affinity for the contaminant, they have slow turnover rates even when the contaminant is bound, they are often inhibited by products of the reaction, and they sometimes generate toxic intermediates. The goal of this project is to develop the tools and understanding required to engineer and apply enzymes with enhanced properties for bioremediation of contaminants.

This year we solved the crystal structure of a *Rhodococcus* dehalogenase, one member of a previously structurally uncharacterized class of dehalogenating enzymes, gaining insights into its structural similarities and differences with respect to the archetypal and well-characterized dehalogenase from *Xanthobacter autotrophicus.* We developed a continuous spectrophotometric assay for dehalogenase activity, characterized the steady-state kinetics of a related *Rhodococcus* dehalogenase, performed site-specific mutations that were based on sequence homology and on functional comparisons in order to eliminate chloride product inhibition, and obtained vibration spectra of a novel type of reductive dehalogenase from *Dehalospirillum multivorans*.

We also demonstrated that the unusual reactivities of *Amphitrite-ornata* dehaloperoxidase and *Notomastus-lobatus* chloroperoxidase do not arise from a histidine imidazolate proximal heme iron ligand.

Finally, we developed a method, based on terminal restriction fragment analysis (RFA) of polymerase chain reaction-amplified DNA, for assessing the heterogeneity of microbial community populations in environmental samples. This method assesses microbial populations at the phylogenetic scale of families of microbes, between 16S clone library sequence analysis, which assesses at the species scale, and fatty acid methyl ester analysis, which assesses at the domain scale.

Publications

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Integrated Structural Biology Resource

97610

Angel Garcia

The purpose of this project is to develop our capabilities in x-ray structure determination, nuclear magnetic resonance (NMR), small-angle scattering, optical spectroscopy, and theory. We envision an Integrated Structural Biology Resource that is designed to target the structural biology of proteins and their biological functions. The initial focus of this resource has been the structure and function of DNA damage detection and repair systems. This focus has evolved to also include research on hyperthermophile proteins that can serve as model systems for studying human proteins, the structure-function relationship, and protein stability under extreme conditions of temperature, pressure, and salt concentration.

In the past year we made advances in the structure determination of hyperthermophillic proteins by using x-ray crystallography. This work led to the development of a structural protemics project, where we developed techniques for protein production, labeling, and purification. We used small-angle scattering to obtain global structural information pertinent to nuclease activity from FEN-1 D181A mutant. Our data indicate that FEN-1 and its mutant have a radius of gyration of 26 Å, and that the effect of magnesium alone on the scattering is insignificant. We constructed a 34-base DNA chain that forms a 5'-flap complex with FEN-1. The radius of gyration of this complex is 34 Å. A time course change in the scattering profiles arising from magnesium activation of the FEN-1/DNA complex is consistent with the protein completely releasing the DNA substrate after cleavage. We also used small-angle neutron and x-ray scattering with contrast variation to study the structure of DNA targeting component (Ku) of the DNA-dependent protein kinase and its complex with DNA. The scattering data are consistent with the idea that Ku treads onto the duplex DNA via a channel that can completely bury approximately 24 base pairs. The determination of the shape of the FEN-1/DNA and Ku/DNA complex are of great importance, because no other high-resolution structures have been determined by either x-ray or NMR. Other significant accomplishments were obtained in theory, NMR, DNA and protein labeling, and optical spectroscopy.

Publications

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Landscapes and Dynamics of Proteins

96612

Hans Frauenfelder

Our project goal was to advance our understanding of complex systems—proteins, in particular. A quantitative understanding of complex systems is important for fields such as new materials, improved medicines, and fundamental physics. Our approach is based on a close interaction between theory, experiment, and computation and involves biology, chemistry, and physics.

The last step in binding small molecules, such as CO, to myoglobin occurs inside the protein at the heme iron. The step is characterized by a distributed activation enthalpy and an abnormally small pre-exponential factor. These properties have been known for a long time, but they could not be explained quantitatively. In collaboration with the Laboratory's Theoretical Chemistry and Molecular Physics Group, we have solved the problem by using state-of-the art computation. In collaboration with the Biophysics Group, we constructed a new model for myoglobin that for the first time connects structure, energy landscape, dynamics, and function. Our studies with the Bioscience and Biotechnology Group provide insight into the fast-folding and beta-sheet formation in proteins. Finally, together with the Condensed Matter and Statistical Physics groups, we explored the charge ordering in a very different complex system, a transition metal oxide.

All of our work was performed with both Los Alamos and external collaborators. As a part of strengthening interactions outside the Laboratory, in 1998 we organized three international conferences and one workshop, and we presented more than ten colloquia.

Publications

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Next-Generation Biological Toxin Sensors

97617

Basil Swanson

Our overarching objective is to develop new biosensors for bacterial protein toxins in which signal transduction and amplification are triggered by protein-ligand recognition. Our approach is to use glycolipid cell membrane receptors that are optically tagged and imbedded in a fluid phospholipid bilayer membrane that mimics the cell membrane surface.

We have developed three optical transduction schemes in which close proximity of the tagged glycolipid receptors resulting from multivalent binding of glycolipids by cholera gives rise to fluorescence energy quenching and resonant energy transfer. Proximity-based resonant energy transfer provides an exciting approach because the protein-receptor binding event results in a two-color change that is highly specific. This triggered optical biosensor has been demonstrated for cholera using flow cytometry and is now being adapted to several other protein toxins. We have achieved the same sensitivity as the best laboratory-based immunoassay technique for the detection of cholera with the advantages of being faster, simpler (one step), more robust, and adaptable to multielement sensor arrays based on integrated optics and optical waveguides; our technique also requires no additional reagents. Other research highlights include measuring and modeling multivalent protein-receptor recognition for cholera, studying the mechanism of hybrid bilayer formation, and synthesis of functionalized monolayer films containing glycolipids for affinity-based chromotography.

Publications

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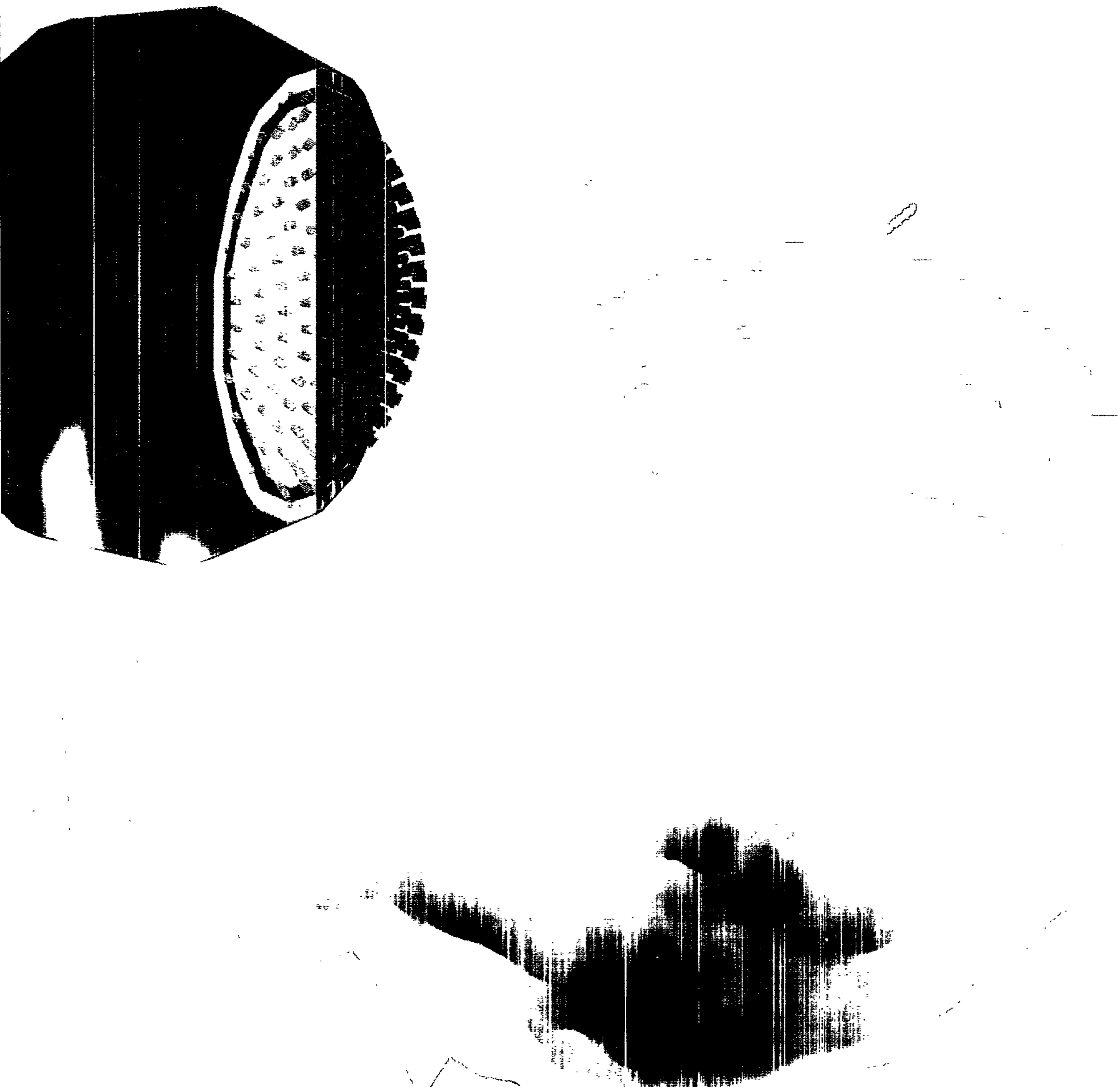

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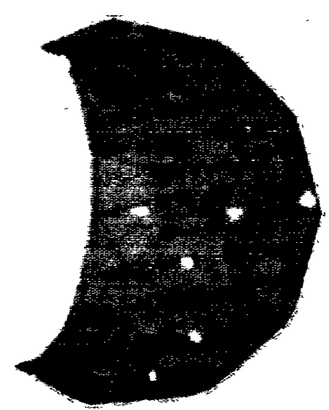
PROGRAM DEVELOPMENT PROJECTS



Materials Science

Alternate TRUPACT Foams

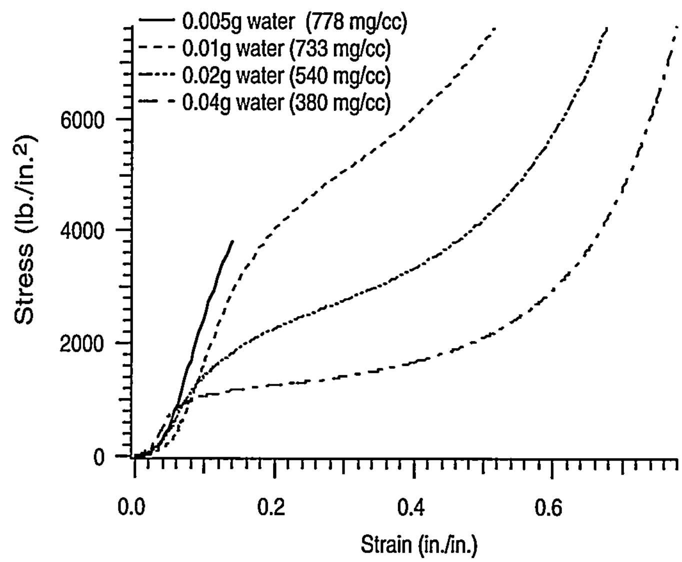
98514

Warren Steckle

Our goal is to enhance the safety of shipments of transuranic (TRU) waste to the Waste Isolation Pilot Plant (WIPP) from across the country, scheduled to begin shortly. TRU waste consists of clothing, tools, rags, and other disposable items contaminated with trace amounts of radioactive elements such as plutonium. The TRU waste package transporter (TRUPACT) containers for transporting waste to WIPP are specially designed to transport this waste and have been certified by both the Nuclear Regulatory Commission and the Department of Transportation.

Safe transport of the containers requires thermal stability and flame resistance, which are provided by polyurethane foams; standards for the strength of these foams are established by the TRUPACT-II Safety Analysis Report (SAR). Foams can be improved by stabilization—through the addition of antimony oxide or of halogen- or phosphate-containing polyols—and by maintenance of the foams' mechanical properties, a function of foam density. The accompanying figure shows how mechanical properties can be tailored by changing the densities of the foam. The foam with the lowest density meets the required strength outlined in the SAR, and the stiffer foams exceed these requirements.

To stabilize the foams, we used halogenated siloxane polyols synthesized in-house, as well as commercially available polyols. To control density, we altered the amount of blowing agent during processing; water was used as the agent as an alternative to environmentally harmful chloroflurocarbons. Densities of the resulting foams ranged from 7.5 lb/ft^3 to 50 lb/ft^3 (the SAR requires a nominal 8.25 lb/ft^3 polyurethane foam). The mechanical properties exhibited by these foams also met or exceeded those established in the TRUPACT-II SAR.



Stress/strain curves for a series of polyurethane foams made by varying the amount of blowing agent used. The numbers in parentheses are the densities of the foams.

Plutonium Aging: Investigation of Changes in Weapon Alloys as a Function of Time

98535

Barbara Martinez

We are using a novel new technique to study aging in plutonium-239. We blend gallium-stabilized δ-phase plutonium-239 with 5% plutonium-238 to accelerate the aging process at the rate of 15 years to one. We use a suite of sensitive, fundamental measurement techniques to characterize the doped material at periodic intervals in order to track subtle changes as aging progresses (see the accompanying table). The tests will be performed jointly with Lawrence Livermore National Laboratory (LLNL). Samples will be stored under carefully controlled conditions to simulate realistic stockpile storage environments, accounting for self-heating and thermally sensitive aging effects. Understanding how aging manifests itself in the enriched material will allow us to look for similar behavior in the stockpile.

This year we developed a program plan and obtained DOE approval to create the enriched material. We completed an aging chamber for use with a dry-bath incubator, completed long-term tests of temperature stability, and conducted heat sink tests to control sample temperature, which showed that the simple finned design works well. We determined the need for glove boxes, conducted facility reviews to upgrade them, and completed outfitting the LLNL glove box.

In addition, we constructed and tested a dilatometer based on linear variable differential transducers and began testing a laser dilatometry system. We began testing the stability of our density balance and performed experiments to characterize the effect of anticipated self-heating of our samples on the immersion density measurement. We dissolved, precipitated, and calcined plutonium-238 oxide and completed direct oxide reduction resulting in a button of plutonium-238 metal.

Test Program for Accelerated Aging of Plutonium[a]

Test	Technique	Information
Analytical chemistry	Atomic absorption, emission spectroscopy, radiochemistry	Impurity content, alloying elements, ingrowth of decay products
Density	Archimedes immersion	Swelling, phase composition
Dilatometry	Laser triangulation, linear variable displacement transducers	Swelling, phase stability, and transformation temperatures
Helium effusion	Sieverts pressure-volume-temperature	Helium diffusivity
Mechanical	Tensile and compression testing	Yield strength, ultimate tensile strength, strain hardening
	Kolsky bar, 40-mm gas gun	Equation of state, shock-loading behavior
Microscopy	Light microscopy	Grain size, phase content, morphology of inclusions
	Transmission electron microscopy	Bubbles, dislocation structure
	Oriented image microscopy	Texture
Positron annihilation	Gamma radiation spectroscopy	Types of atomic defects, location of helium atoms
X-ray diffraction	Diffractometry	Interatomic lattice spacing, texture, phase composition
	Low temperature	Self-annealing effects

[a]Activities are divided between the laboratories so as to make the best use of resources and capabilities available at each location.

Nanosized Aluminum Powders from the Exploding-Wire Process

98563

Joel Katz

The exploding-wire process is a viable method for manufacturing nanosized aluminum powders. There have been persistent reports that the aluminum powders produced by this method are unusually reactive, even when compared with other fine powders of similar size. Commercial quantities of "raw" powders manufactured by the exploding-wire process are currently available for testing from the Argonide Corporation. As part of our research project, we used such powders to investigate claims of their enhanced reactivity, to characterize the powders, and to perform a preliminary investigation of particle-separation techniques.

By using several techniques, such as differential thermal analysis, differential scanning calorimetry, x-ray diffraction, laser-light scattering, and high-resolution transmission electron micrography (HRTEM), we characterized an Argonide powder manufactured by exploding aluminum wire (ALEX powder). While the average particle size was about 150 nm, particles as large as 1000 nm were occasionally observed. The powder was also highly agglomerated; the mean particle size measured by light-scattering techniques was over 1200 nm because of agglomerates. Using our technique for particle deagglomeration/separation, we reduced the mean particle diameter and the standard deviation of the particle size distribution to the "basic" particle size observed by HRTEM. This technique also enabled us to separate a particular size from the general particle-size distribution.

Our research disproved claims of enhanced reactivity of aluminum powders produced with the exploding-wire process. Data from the thermal analysis of an ALEX powder shows a sharp exotherm between 579°C and 594°C during both heat-up and cool-down. Similar analysis of a coarser, commercially available micron-sized powder reveals a much broader exotherm near the same temperature range. Critical examination of x-ray data from an ALEX powder reveals that aluminum oxides, nitrides, and oxynitrides have formed during thermal analysis in spite of efforts to eliminate trace impurities from the environment during the analysis. Because we found no evidence of aluminum hydroxides, we do not think they are responsible for the observed exotherm. Similarly, we did not observe any anomalous behavior due to metastable effects, high defect density, or spontaneous sintering.

Publications

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Concentrating Low-Level Tritiated Water

98509

Robert Dye

We tested the concept for a tritiated water "enrichment" capability, removing tritium from tritiated water at low concentrations (near that of drinking water standard maximums), on waste streams from the Laboratory's liquid radioactive waste treatment facility. Our tests are based on the exchange of tritium on polymeric materials. Both the exchange rate and the hydrogen bonding strength vary between HTO, T_2O, and H_2O. With these differences in mind, we investigated materials that retain the tritiated water. The key to success was to optimize the hydrogen bonding strength between the water and the material in such a way that the material would retain the molecule if tritium were present and release the molecule if it consisted of hydrogen.

Most hydrogen-containing materials tend to retain the tritiated molecule because the hydrogen bonding strength is slightly higher. This tendency can be optimized by adjusting the material's hydrogen bonding strength. This effect alone is not enough to produce a tritium enrichment system. However, if a hydrogen exchange rate is also optimized within the same material, we will have a viable enrichment process.

We investigated a series of polymers with different bonding groups: –OH, –SH, and –NH. At this point, the data suggest that, indeed, the different polymers have different degrees of tritium retention. Although these scoping studies are necessary to establish feasibility, a much more intensive program is required before the concept can be used in the field.

Experimental and Computational Investigation of Ultrafine-Scale Materials with Strength Levels Close to the Theoretical Strength

98542

J. Embury

We are investigating fine-scale structures with strength levels close to their theoretical strengths, using both experimental methods and molecular dynamics simulations. The computer simulations and experimental studies are closely linked because they each deal with similar-scale structures which are embedded layers or fibers 5 nm in diameter.

This year both experimental and computational activities have concentrated on a copper-silver system. This includes multiple-layer copper-silver, cold-drawn copper-silver, and microwires fabricated by melting alloys in glass and drawing them into filaments at high temperatures by a method know as the Taylor wire technique. We are currently exploring new materials using this technique and exploring possible engineering applications of high-strength microwires.

We undertook molecular dynamics simulation studies of copper-silver composites. Both the computational and experimental results demonstrated that the dislocations are generated at the interfaces between copper and silver during the deformation. Using atomic-scale imaging and neutron defraction, we detected significant internal stresses in both copper and silver after deformation. We also observed distortion of the face-centered-cubic structure and related it to internal stresses. These internal stresses and the role of the interfaces are the features that appear to dominate plasticity in ultrafine-scale structures.

Publications

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Han, K., et al., "Internal Stresses in Wires for High Field Magnets," *Proc. Int. Conf. Electron Micros.* **3**, 359 (1998).

Han, K., et al., "Microstructure Aspects of Cu-Ag Produced by the Taylor Wire Method," *Acta Mater.* **46**, 13 (1998).

New Membrane Solutions for Hydrogen Isotope Issues

97502

Robert Dye

We are developing specialized nonporous membranes to improve our ability to separate tritium, an istotope of hydrogen, from hydrogen and another of its heavy isotopes, deuterium. More-effective separation and processing methods for tritium will help to maintain the nation's supply of this essential, limited-lifetime component of the nuclear weapons stockpile. Currently, tritium is not produced in the US; once production resumes, we will also need novel approaches for safely capturing and collecting it. These capabilities are vital to the DOE Defense Programs mission and can lead to improved safe cleanup of mixed waste. In this project we have conducted lifetime studies at room temperature and investigated isotope effects of permeation, using enhanced membranes that induce flows 20 times higher than those through standard palladium membranes.

We built on our earlier success of doubling the hydrogen flow rates through our membrane by constructing a tritium system for measuring tritium permeation through the metal membrane. However, tritium availability was too limited to conduct experiments. Other experiments in this effort were targeted to address specific issues related to hydrogen and hydrogen isotopes. We were also successful in our goal of program development: the initial monetary investment in this research yielded a return from external sources of over 4 to 1.

Development of a System for Endoscopic Imaging and Spectroscopy of Pit Interiors

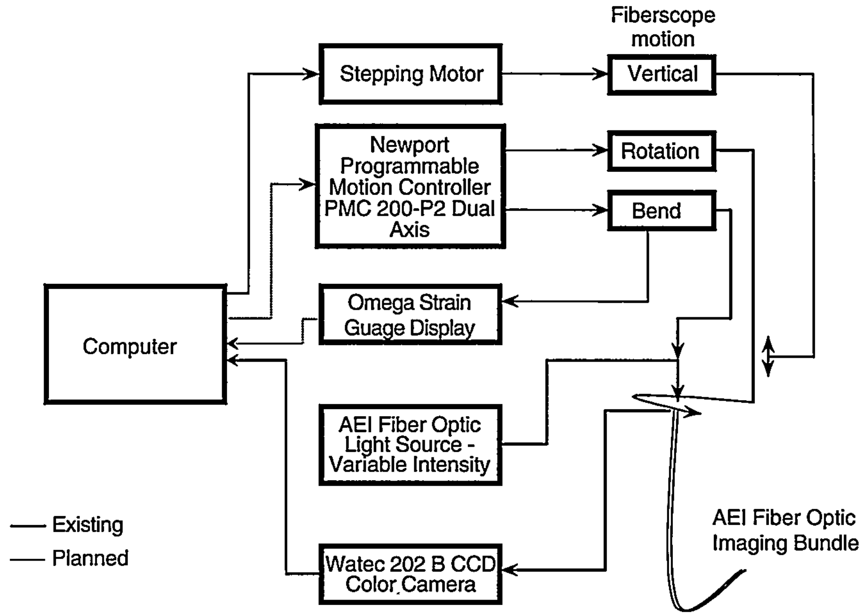
97514

D. Kirk Veirs

Each year, to ensure the safety and reliability of the nuclear weapons in the stockpile, some weapons are pulled at random for examination. Part of the examination includes checking the plutonium pit to see if any issues related to aging might require its replacement. The objective of this work is to develop and demonstrate a method to interrogate the inner surface of an intact pit using a fiber-optic probe inserted into the fill tube of the pit. We have fabricated a fiber-optic probe that contains an illumination bundle and an imaging bundle and is capable of bending 180° across the inside of the pit to access all of its interior regions. The fiber is coupled with a mechanical stage that precisely controls the rotational angle, vertical position, and the overall bending of the fiber. A color camera is used to obtain images, and a computer controls the motion of the probe and stores the images (see first figure).

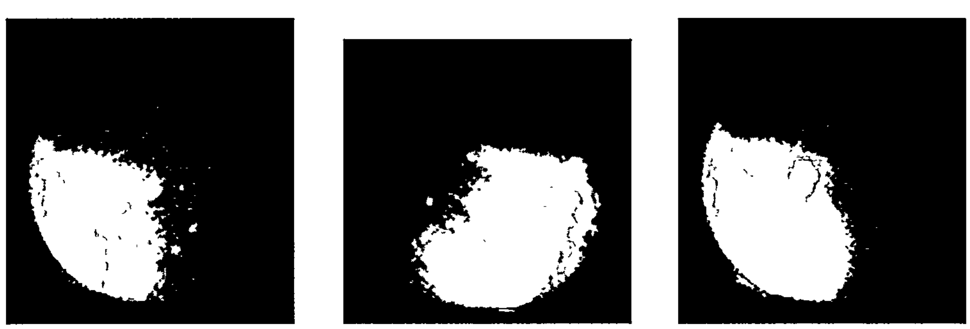
We have observed the weld line and machining grooves on the inner surface of a pit surrogate using this apparatus (see second figure series). The weld line was imaged in 5° segments over a total span of 300°. As you can see in the figures, anomalous craters were also observed in this segment of the weld. Our efforts thus far have demonstrated the capability of imaging more than 90% of the interior surface of a pit surrogate.

We have also investigated spectroscopically characterizing plutonium surfaces using fiber optics. In particular, we have demonstrated laser-induced breakdown spectroscopy of plutonium metal and quantitative determination of minor constituents using fiber optics. Our efforts to integrate the imaging and spectroscopic capabilities into a single fiber-optic probe should be aided by a report we received from Rensselear Polytechnic Institute that describes the damage mechanisms to fiber optics from high-peak-power pulsed lasers.



Schematic of the imaging system showing the motion controllers and their coupling to the fiber-optic probe. The motion controllers can position the probe tip precisely in three-dimensional space. Images are then transmitted to and archived on the computer.

Three images of the inner surface of a welded sphere. The reflection of the light source creates a large bright area in all three images. The weld line, bright spots due to small craters or bumps, and striations from the machining are seen in the first two images, and a very large crater situated just below the weld line is seen in the third image.



Application of High-Temperature Superconductors to Underground Communications

97517

David Reagor

Although the need to communicate with mobile personnel or equipment arises regularly in underground work areas, underground communications is still one of the most intractable problems found in mining and geophysics. Conventional radio signals do not propagate underground, but it is well known in the geophysics community that ultralow-frequency (ULF) signals can cover significant distances. However, the conventional semiconductor-based receivers for ULF signals are either too large or too noisy. We are studying the potential of high-temperature superconducting quantum interference devices (SQUIDs) to advance underground communications by replacing the conventional ULF receivers. During our research, we have also realized the potential of geophysical instruments based on SQUID receivers.

Our goals are to continue developing SQUID device structures, to develop control and interface electronics for SQUIDs, to develop a transmission antenna, to design and test preliminary orientation-independent receiver technology, to engineer cryogenic packaging, and to field-test a simple receiver.

We obtained our first results from a new process for making small, high-temperature superconducting junctions. Making smaller junctions is a key technical goal en route to building higher-performance SQUIDs. Last year we also designed and built a set of control electronics that was specific to the underground communications problem and fabricated a lightweight transmitter for testing underground communications systems. We designed a Dewar flask using mixed cryogens. Our work provided the technology needed to build the first portable underground radio that supports two-way voice communication through hundreds of meters of solid rock. We received a 1998 R&D 100 Award for this work.

Publications

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Mechanical Properties of Cellular Materials

98538

Johndale Solem

While a great deal of empirical information is available on elastomeric foams and while computer algorithms do a respectable job of simulating their mechanical response, the complexity of these cellular materials has generally prevented deep theoretical understanding at an analytic level, particularly in the nonlinear regime. Curiously, the disilicate open-cell foam of particular interest enjoys a stress-strain relationship that is an almost perfect fit to a hyperbola. Such an algebraically simple behavior strongly suggests an intrinsic symmetry that may lead to deeper theoretical understanding. Our aim was to undertake a coordinated experimental and theoretical investigation of (1) the feasibility of producing thin elastomeric foams either through forming or in situ foaming and (2) the response of the materials under stress to ascertain how the simple hyperbolic behavior is produced.

On the basis of an *ab initio* calculation, we have shown that the nonlinear region of the stress-strain relation for elastomeric foams should be well approximated by a hyperbola. This approximation is supported by data available on the behavior of some polyethylene, polyurethane, and silicone foams. The success of our calculation can be partially explained by Kachanov's method of superposition as it would be applied to find the interactions within an ensemble of spherical holes. This method hinges on finding the perturbation to mean stress at a cavity due to each of its cavity neighbors. Since the mean stress is uniform and unaffected by the presence of a hole, the superposition method would lead to the conclusion that the interaction has relatively little effect. In a similar vein, the competing effects of stress shielding and stress amplification balance each other—at least in the cases investigated.

Publications

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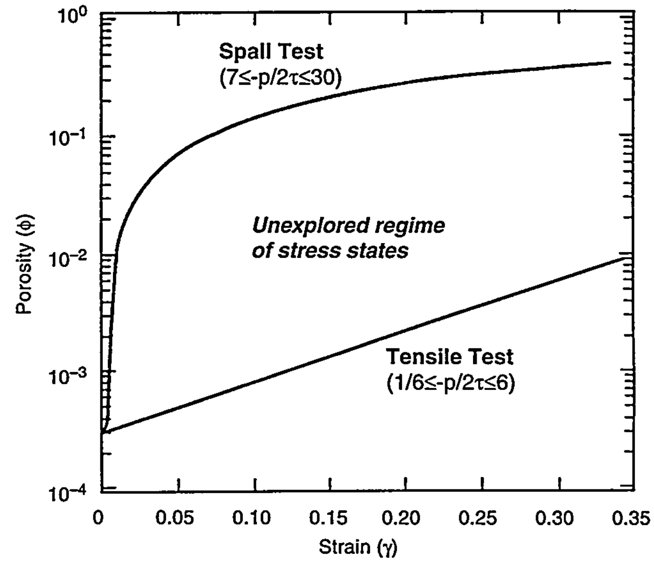
Ductile Fracture in Dynamic Tensile Regime

98539

Anna Zurek

Our research project is extending the current expertise and methodology in dynamic compression behavior of materials to the unexplored regimes of ductile dynamic fracture in high-rate tension (see accompanying figure). More specifically, we will be exploring fracture in an intermediate regime of strain rates (500–9000 s^{-1}) and stress states ($1/3 \le -p/2\tau \le 5$). Ductile fracture involves void nucleation (typically at material heterogeneities), void coalescence, and growth. The influence of strain rate on ductile fracture is important to several of the Laboratory's technical divisions (Materials Science and Technology, Theoretical, Dynamic Experimentation, and Applied Theoretical Physics Divisions, for example), and its experimental aspects serve to verify codes that calculate damage accumulation, crashworthiness, and spallation. New experimental input and model development can improve the utility of these codes for a wide range of materials.

Our project leads to the fabrication of a new experimental apparatus to explore fracture in a previously unexplored regime of intermediate strain rates (500–9000 s^{-1}) and range of stress states ($1/3 \le -p/2\tau \le 5$). This new experimental capability will provide data for yield surfaces and fracture limits of the investigated material systems. To date, we have acquired, fabricated, or purchased all the hardware we need to build the Tensile Hopkinson Bar apparatus. The assembly of the components is under way.



The figure shows the porosity change with respect to the strain in two cases of stress state. The upper line represents the porosity achieved in a dynamic fracture spall test with stress-state ranges of $7 \le -p/2\tau \le 30$. The lower line represents the porosity change in a quasi-static tensile test with a stress range of $1/3 \le -p/2\tau \le 5$. These data are characteristic for the tests commonly performed to characterize fracture behavior in materials. We are developing the techniques to understand and obtain data to fill the in the "unexplored regime of stress state" that exists between the two tests.

Measurement of Helium-3 Formation in Palladium

98532

Dale Tuggle

Our objectives for this project were to fabricate single-crystal palladium hydride samples; to measure the location of the hydrogen and helium atoms in the palladium hydride lattice using ion-beam channeling spectroscopy; and to measure, using resonant ultrasound spectroscopy, the elastic constants of palladium tritide and the changes in these elastic constants that occur as tritium decays to form helium-3. Understanding tritium chemistry is important in maintaining a safe, secure US defense arsenal.

We successfully fabricated single-crystal palladium samples and developed techniques for cutting, polishing, and aligning these single-crystal samples so that the 110 or 111 cube faces were exposed. Ion-beam channeling experiments confirmed the location of the hydrogen atoms at the octahedral sites in the palladium lattice. We started initial experiments to inject helium-3 into the lattice and watch it diffuse into the lattice. We measured the resonant ultrsound spectra of single-crystal palladium deuteride and tritide samples at room temperature, and the samples lost tritium at a slow rate that obscured changes caused by the retention of helium-3 in the lattice. In future work the measurements will be conducted at –70°C to minimize the tritium loss from the sample. The tritide measurements were the first ever conducted, and we are submitting our results for publication.

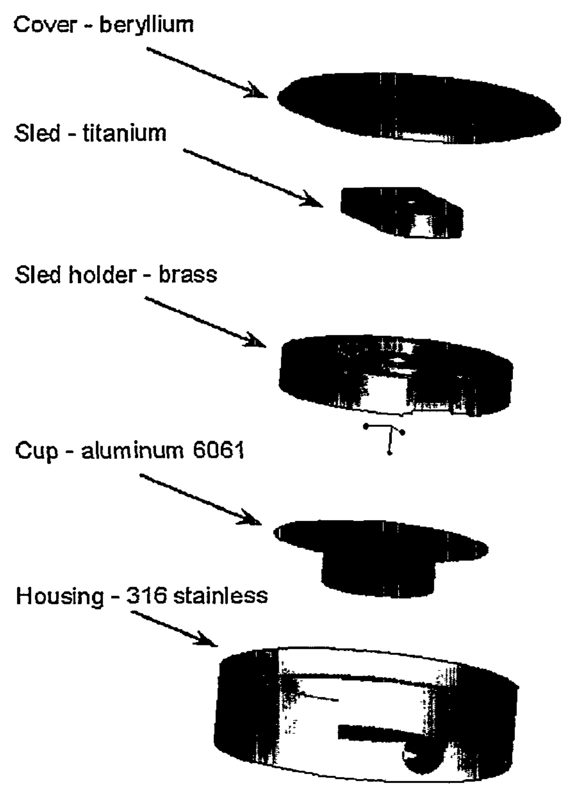
Simulating Materials Defects by Integrating Nondestructive Techniques with Engineering Models

98533

Thomas N. Claytor and Deborah A. Summa

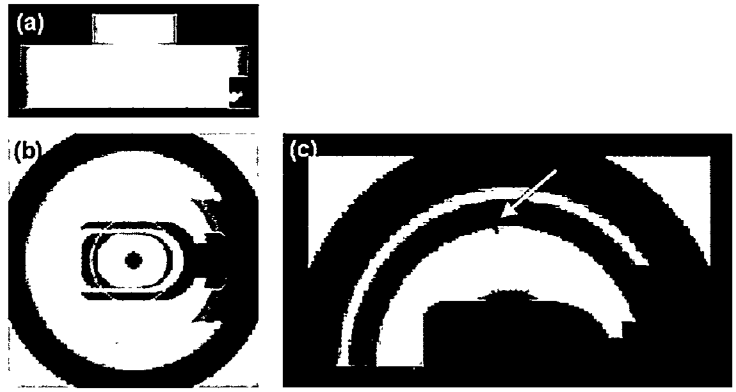
The numerical simulation of radiographs, computed tomography, and ultrasonic scans enhances surveillance reliability and efficiency by providing realistic quantitative answers to nontrivial, nondestructive evaluations of complex components. Simulation is not intended to replace experimental data but rather to guide efforts to obtain the best, most complete data set and interpretation possible using limited resources. Simulations have the added advantage of being able to be used in either a predictive (forward) mode or in an analytic or comparative (backward) mode. With a forward simulation, effects of varying geometry, orientation, material properties, etc., can be readily quantified. The information obtained is utilized to optimize inspection parameters and improve interpretation of complex examinations. With a backward simulation, the computational response is compared with experimental results. The required fundamental parameters necessary to produce the observed data set can be extracted, thus providing insight on material aging effects. Once early signs of aging are identified, appropriate measures can be taken to prevent or control associated degradation processes more effectively. Simulations can also be used to generate probability-of-detection maps that can be used for determining the likelihood that a defect of a certain size will be seen in any given section of the part and ultimately in designing critical sections for inspectability.

Our project focuses on integrating new radiographic simulation codes into the weapons inspection program. A comprehensive x-ray simulation code, XRSim, is being developed at the Iowa State University Center for Nondestructive Evaluation under a consortium agreement that includes the DOE. We are experimentally verifying source models, film models, and other aspects of the code, calibrating it to existing radiography equipment and working with developers to make appropriate modifications for weapons applications. We have upgraded the x-ray absorption coefficient database to include materials of interest in weapons systems (e.g., various fissile elements, alloys, polymers, and high explosives), and we have created an extensive interchangeable parts library that has been used to simulate unclassified components and assemblies (see accompanying figures). Models of classified components have also been secured for future analysis. An XRSim User's Guide and a comprehensive report detailing technical aspects of the project and examples of applications are expected to be available shortly.



Simulated radiographs for the solid detonator shown in the next figure. (a) CAD model of entire assembly. (b) Simulated radiograph with exposure parameters optimized to show detail inside assembly. (c) Simulated radiograph of the stainless steel housing and copper holder with exposure parameters optimized to reveal a small crack at the interface (note that this crack *would not* be seen using the typical inspection protocol). XRSim is helpful for custom tailoring exposure parameters (source voltage, current, exposure time, film, part orientation, etc.) to individual inspections, as well as for determining minimum detectable flaw size in critical components.

Exploded view of an unclassified model of a solid detonator. A CAD (computer-aided design) model of each individual subcomponent is imported into XRSim, where the user can then assign an appropriate material to each part and reassemble the entire component. Flaws that mimic voids, inclusions, cracks, and misalignments can be incorporated at any desired location. Part geometry, materials, and flaw characteristics can be easily changed to allow rapid evaluation of expected radiographic indications, or lack thereof, due to differing specimen configurations.



Materials Compatibility and Migration in Polymer Systems

98536

Carl Maggiore

The purposes of this project are to study the effects of materials migration by direct measurement of the diffusion and convective migration processes in complex polymeric materials and to develop appropriate predictive models. This study and modeling are necessary to completely understand the long-term stability of polymers in complex systems and to predict lifetimes and possible failure modes. Our central approach involves measuring transport properties of various volatile components in polymer/plasticizer systems and using those measurements to predict the overall transport behavior of combinations of composite materials derived from those polymeric systems.

This year we demonstrated the use of isotopically tagged probe molecules to measure in situ the diffusion of water in estane. We fabricated a special environmental cell with a thin window to enable real-time measurements under realistic conditions simulating actual operating parameters. Depending on the migrating species to be followed, the depth resolution needed, and the sensitivity required, the stable isotopic tag can be deuterium, carbon-13, nitrogen-15, or oxygen-18. We measured the depth profiles quantitatively using ion-beam methods available at the Los Alamos Ion Beam Materials Laboratory. The low-energy nuclear reactions that are particularly useful for these measurements are the positive Q-value reactions such as $d(^{3}He,\alpha)p$, $^{13}C(d,p)^{14}C$, $^{15}N(d,\alpha)^{13}C$, and $^{18}O(p,\alpha)^{15}N$.

We adopted the Williams-Landau-Ferry model as a general expression for diffusion of a volatile in a polymer. This model contains both thermal-activation and free-volume change effects to account for the changes in polymeric structure with temperature and physical properties as embodied in the glass transition temperature.

As a specific case, we deduced from data in the literature a full initial parameter set for water migration in polyurethane. We then used the parameters in an ideal one-dimensional, constant-temperature, constant-boundary concentration test problem for which an analytical solution is known. The code worked properly. In addition, the test case also showed that time steps on the order of 10 min are permissible, which is the order of the time required to collect data for one measurement in the ion beam.

Diamond and Diamond-Like Materials as Hydrogen Isotope Barriers

97511

Larry R. Foreman

Our research objective is to evaluate the application of dense polycrystalline diamond as a barrier layer to hydrogen permeation for use on implosion-target shells and tritium-processing equipment. Little is reported in the literature on the permeability, solubility, or diffusion of hydrogen in diamond, but measured values of hydrogen concentration are on the order of $10^{19}/cm^3$. This value suggests a low solubility of hydrogen in diamond that, combined with its dense structure, makes it a good candidate material for a barrier.

Permeability measurements of hydrogen through polycrystalline diamond require free-standing thin films. To make them, we have deposited films on silicon wafers and removed the wafer by chemical etch. Although the films were brittle and stressed, a sufficient amount of material remained intact for testing. These material samples were mounted in a high-vacuum system supported on a porous stainless-steel support, and a fixed pinhole leak was used to calibrate a mass spectrometer for hydrogen flux. However, using this method, our attempts to measure the flux at room temperature through 2-μm-thick diamond films failed because the permeability was below the detection limits of this system.

We now have a high-pressure and high-temperature system that has been constructed at the Los Alamos Tritium System Test Assembly to measure the permeability of hydrogen and its isotopes through stainless steel and a barrier material. Our next effort will be to use this test assembly to examine diamond coatings. We will also be examining a number of nitride and intermetallic systems, since reports indicate that 1,000- to 100,000-fold decreases in the permeability of hydrogen can be obtained with nitride and aluminide barrier coatings.

Optimized Charge-Transfer Materials for Protection of Sensors from Laser Weaponry

98505

Duncan McBranch

Presently there are no available materials that can protect ballistic missile, tank, and automated aircraft sensors, as well as soldiers' eyes, from pulsed-laser radiation intended to disarm or disable battlefield function over a broad and agile frequency range. Our main goal was to develop and test a prototype of the next generation of nonlinear optical molecules to provide passive protection (optical limiting). We designed our new systems to have large excited-state extinction coefficients in the visible spectrum, with long lifetimes as a result of intramolecular, photo-induced charge transfer. We have successfully synthesized a new molecular dyad consisting of a metal-phthalocyanine that is directly and covalently linked to a fullerene acceptor species. Our optical-limiting measurements demonstrated that the dyad showed enhanced performance relative to either pure compound, and our assignment of the enhancement as caused by an intra-dyad electron-transfer reaction was confirmed using femstosecond spectral transient-absorption studies.

Publications

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Structure and Dynamics in Quasi-Ordered Systems

98540

Juergen Eckert

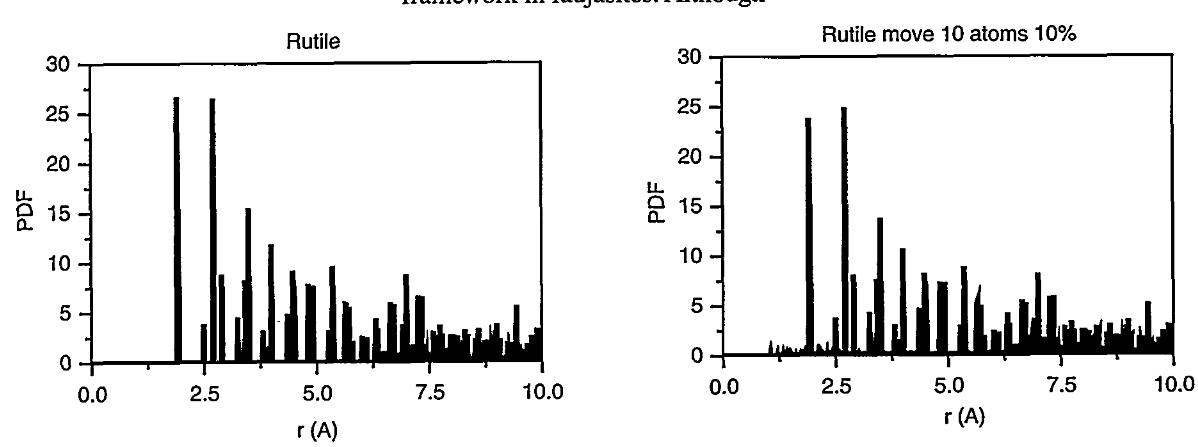
We are using neutron diffraction to locate metal ions randomly distributed inside microporous catalytic materials, pair distribution function (PDF) analysis to examine the local structure of these materials, and inelastic neutron scattering (INS) to study interactions between reactant and product molecules with the ions and the microporous matrix. These techniques are invaluable in elucidating the structure and function of these classes of catalysts. While we are examining specific systems of interest in catalysis, development of neutron-scattering capabilities as described will also allow us to create the capability to tackle structural issues in environmental systems (e.g., adsorbates on minerals). This year we had two main objectives: (1) to carry out neutron-scattering experiments in zeolite systems and (2) to develop computer codes for PDF analysis and inelastic neutron scattering experiments.

We have performed several experiments to determine the effectiveness of PDF methods for the study of local structure in catalytic materials. Very high quality data were collected at the ISIS facility (Rutherford Appleton Laboratory, UK) on six-atom iridium and rhodium clusters in faujasite at low temperatures. Unfortunately, these clusters were too dilute to show unambiguous structure in the PDF. We expect that this problem can be solved with the neutron flux increase to be provided by the new US neutron scattering facility, the Spallation Neutron Source, that will be built at Oak Ridge National Laboratory. We have designed a second experiment using a differential PDF with normal and deuterated chloroform to determine the location of absorbed chloroform molecules relative to the framework in faujasites. Although data analysis is still in progress, this type of experiment appears to be promising for a variety of such disordered adsorption complexes.

We also carried out experiments and calculations using INS to analyze the sorption of organic molecules inside zeolites. In addition, we have developed computer codes to carry out PDF analysis of partially ordered systems. Our main result from the calculations is to learn that experimental detection of displaced atoms is only possible if the displaced atoms are displaced by large amounts or if the fraction of displaced atoms is very large. The accompanying figure shows that for a 10% displacement of about 8% of the atoms in the unit cell of rutile, the difference between the PDF of the perfect and perturbed crystals is too small to be detectable.

Publications

Henson, N.J., et al., "Studies of Ethane and Ethene Sorption in Na-Y by Inelastic Neutron Scattering (INS) and Computation" (Twelfth International Zeolite Conference, Baltimore MD, July 5–10, 1998).



Comparison of pair distribution function (PDF) analyses for a rutile crystal (left panel) and a crystal in which 8% of the atoms are displaced by 10% from the lattice positions (right panel) to show that the difference between the perfect and perturbed crystals is too small to be detectable.

Carbon Management: CO_2 Sequestration

98513

Klaus Lackner

Fossil fuels supply more than 80% of the world's energy. Unfortunately, burning them produces CO_2, a greenhouse gas that is responsible for global warming. For fossil fuels to remain a viable energy resource, ways must be found to remove or recyle this harmful by-product. We investigated three promising CO_2 sequestration technologies: (1) increasing the carbon inventory held in biomass, (2) directly recycling CO_2 by conversion to useful fuels, and (3) fixing CO_2 in mineral form.

In our biomass studies, we have discovered that biomass growth (i.e., CO_2 uptake) can be increased significantly at a given nitrogen supply in two ways: by providing a natural signal metabolite and by genetically engineering overexpression of a key enzyme in the plants' nitrogen metabolism. We are now preparing patent disclosures for these processes, and several agencies have expressed interest in funding further R&D work related to them.

In terms of recycling CO_2, we investigated the potential for using sunlight to produce power or fuels such as methanol from CO_2. Our work on solar absorption was motivated by the recently discovered radical red shift in the absorption spectrum of CO_2 in the solar region. With the onset of an absorption avalanche, significant quantities of solar radiation can be converted into chemical energy. We demonstrated high-temperature photoabsorption in CO_2, which is expected to result in dissociation into CO and oxygen. A process based on this effect could produce electrical power or could be used to produce synthesis gas from which useful fuels such as methanol could be manufactured.

We also explored a permanent disposal option based on converting CO_2 to mineral carbonates such as magnesium or calcium carbonate. We developed process flow diagrams for forming the carbonates and conducted preliminary experiments to explore kinetic issues. The conversion scheme for producing magnesium carbonate is attractive since the overall chemical reaction is thermodynamically favored, the carbonate is stable and environmentally benign, and the mining operations for obtaining magnesium silicate will localize environmental damage. However, some global kinetic (and thus cost) issues—in particular, how to speed up the reaction kinetics—require further study. We also investigated a coal-to-power conversion process that uses calcia to extract CO_2. Calcia or CaO will bind CO_2, which in this way is removed from the flue gas stream. The resulting calcium carbonate is subsequently calcined to release the CO_2. This process appears to be an attractive route to "capturing" CO_2 in a relatively inexpensive manner.

Finally, we researched several cross-cutting technologies related to separating carbon dioxide at power plants. One separation scheme involves the use of clathrate-hydrate structures to capture the CO_2 (these structures could also be used to remove sulfur). Upon dissociation, the gases are released and available for sequestration or recyling. The use of high-temperature ceramic metal oxide membranes was also explored, with the goal of removing CO_2 from high-temperature process gas streams.

Salt Recycle in Support of Molten Salt Oxidation of ^{238}Pu-Contaminated Combustible Waste

98511

Kevin Ramsey

To enhance the recovery of plutonium-238 from contaminated combustible wastes, so that the plutonium and the wastes can be safely disposed, we sought to improve the process by which the waste is rendered noncombustible. The process is molten-salt-oxidation (MSO), which can be improved by recycling portions of the salts it produces. MSO produces a spent salt containing sodium carbonate, sodium chloride, and ash (i.e., oxides of plutonium, uranium, iron, aluminum, etc.); a molten alkali salt (e.g., sodium carbonate) serves as a catalyst for the conversion of hydrogenous material (i.e., organic solids and liquids) and oxygen to water and carbon dioxide.

Our initial objective was to optimize the recycle efficiency for Na_2CO_3, and we used CeO_2 as a surrogate for PuO_2. We conducted tests using evaporative crystallization as the separation process and found that approximately 70% of the initial weight of Na_2CO_3 can be recovered from a salt containing 33 wt % NaCl and 67 wt % Na_2CO_3. To take the process further, in order to develop a separation process for plutonium recovery, we pursued an understanding of phase formation and chemical speciation during the MSO process by preparing and analyzing surrogate spent salts. This was done by heating Na_2CO_3, NaCl, and CeO_2 at 900°C in air for four hours. When we conducted x-ray diffraction analysis of surrogate salts containing Na_2CO_3 with 7 to 19 wt % NaCl and 18 to 21 wt % CeO_2, we found that the ash consists primarily of CeO_2, thus confirming the absence of cerium chloride formation and the stability of CeO_2 during MSO processing.

Uranium Chemistry Collaborations

98530

Dan Knobeloch

This joint project with the Oak Ridge Y-12 Enriched Uranium Operations Facility involved the development and implementation of a chemical-processing technology modernization strategy. Specifically, our objective was to identify improvements that have been made in the selection and configuration of hardware or in chemical-processing techniques that could increase safety, productivity, and reliability while reducing capital expenditure and operating costs.

We selected two uranium chemical operations to study further: (1) laser-induced reduction to generate uranium metal from oxides and (2) ion-exchange technology for liquid-waste-stream polishing. These candidate processes were selected because they increased safety and productivity, even though assessment data indicated they also had higher operating costs.

Our selection of laser-induced reduction technology was based on several chemical processing improvements that included a chemical reaction mechanism that initiates at room temperature, a more uniform reaction rate to minimize elevated temperature spikes, and the minimization of waste by eliminating the need for a salt-based solvent system.

We found that ion exchange enhances productivity because it involves fewer process steps for liquid feed treatment and minimization of wastes. Safety enhancement was a lesser consideration: large volumes of organic liquids used in the existing solvent extraction system could be replaced with ion exchange resins that are stable for long periods of time and will tolerate variations in process parameters, such as pH and temperature.

We conducted experiments to evaluate whether the two chemical processes could be adjusted for full-scale production operations at the Y-12 facility. The experimental plan for the laser-induced reduction process involved studying the chemical-reaction parameters, including the stoichiometry, of calcium metal and uranium tetrafluoride so that we could provide a chemical mixture that uses precise quantities of chemicals in order to minimize waste. We also designed a magnesium oxide reaction-containment crucible and assessed the purity of the metal buttons that resulted from the chemical reaction.

To test the ion-exchange technology, we conducted ion-exchange experiments on a dilute solution of uranyl nitrate to determine resin-bed capacity, binding rates, and an extraction solution to remove the pure uranium from the resin.

Methane Conversion to Fuels and Chemicals: Opportunities and Approaches

98559

Mark Paffett

The conversion of alkanes to olefins, in particular ethane to ethylene, is a very important process in the chemical industry and is normally accomplished with relatively inefficient steam cracking of ethane. Converting ethane to ethylene by partial oxidation in short-contact-time reactors has been identified as a very promising new technology to replace the more conventional steam reforming of ethane. This project seeks to understand the fundamental limitations of this technology and actively addresses other novel chemical processes that may be part of the Global Climate Change Initiative and other future program development opportunities. The goal of this program development project is to seek out and develop new DOE and industrially sponsored programs.

This year we have identified an industrial partner that we have successfully teamed with in a larger DOE Office of Industrial Technology (OIT) project addressing engineering limitations and fundamental science issues that are being overcome to bring this partial oxidation technology to market. At Los Alamos we are using two-dimensional modeling to derive a new, more complete, kinetic mechanism (with corresponding rate parameters) that couples homogeneous gas phase chemical processes to heterogeneous surface chemistry. Our modeling work is being done in conjunction with a Los Alamos experimental program (funded under the OIT project) to verify the predictions. So far, the agreement between our predictions and the experimental results is excellent. We have also teamed with Sandia National Laboratories (SNL)/California and SNL/New Mexico to provide additional experimental evidence required for full implementation of this predictive modeling capability. Such mechanistic work is expected to provide broader applicability and understanding so that reliable simulations can be achieved for a wider range of experimental or industrial configurations beyond just methane conversion.

Publications

Zerkle, D., et al., "Two-Dimensional Modeling of the Partial Oxidation of Ethane to Ethylene," in *Proceedings of the International Combustion Symposium, Boulder, CO, August 2–7, 1998* (Combustion Institute, Pittsburgh, PA, in press).

Developing Assessment Strategies for Biotic Actinide Remediation Processes

98546

James Brainard

The DOE's Natural and Accelerated Bioremediation Research (NABIR) Program is principally focused on research that is applicable to the remediation of radionuclides and toxic metals in the subsurface of the earth. Bioremediation has traditionally been applied to organic contaminants that biological systems can degrade and ultimately mineralize. Clearly this strategy cannot be applied to the remediation of metal contaminants since no such degradation pathways exist. However, biologically mediated changes in the speciation of metals can greatly affect their transport and toxicity. We and other researchers have proposed the manipulation of biological systems in order to either stabilize or mobilize radionuclides for the purposes of subsurface remediation, but the ability to understand, control, and ultimately prove bioremediation of radionuclides and toxic metals depends on the ability to distinguish biotic transformations (those induced by living organisms) from those which came about by abiotic mechanisms. Currently, there is no way to determine which process was responsible. The objective of our project is to devise, develop, and test new strategies to establish the existence of biotic transformation mechanisms in actinide-contaminated subsurfaces.

During the past year we have worked with NABIR Program managers, science team leaders, and principal investigators, as well as with other investigators at Los Alamos, to identify the most crucial assessment needs in the NABIR Program and the best scientific strategies for meeting those needs. With other Laboratory investigators, we selected and developed the most promising of these strategies. We wrote and submitted four scientific proposals for the NABIR science call this summer, including one for an in situ hybrid biological-chemical sensor that will monitor speciation changes in metal contaminants, and another for assessing the role of fermentative microorganisms in mediating metal speciation changes under varying environmental conditions. Although all four of the proposals received high marks for scientific content, unfortunately none were approved for funding in this round of competition. We expect, however, that one or more of these proposals will be revised for the next NABIR call in this fiscal year.

Smart Film Optical Sensors for Chemical Warfare Agents

97534

Basil Swanson

We have developed species-selective thin films for chemical sensing based on waveguide Zeeman interferometry for rapid detection of chemical warfare (CW) agents. The species-selective films are based on guest-host inclusion complexes formed by bucket-shaped cyclodextrin molecules and the CW agent in question. We have prepared several different cyclodextrin molecules and incorporated them into thin films formed by covalent attachment to functionalized self-assembled monolayers on surface acoustic wave (SAW) and optical waveguide transducers. We have approached the problem of finding more sensitive multilayer films in several ways, and the most successful has been to attach the cyclodextrin molecules to hydrophobic polymers, which are then covalently attached to functionalized SAMs on the transducer surfaces.

We have measured the sensitivity and time response of SAW and optical transducers using both monolayer and polymer films. The sensitivity to dimethylmethoxyphosphonate, a sarin simulant, is in the range 10 to 100 ppb, and the sensitivity to mustard simulants is in the range of >100 ppb. We demonstrated waveguide Zeeman interferometry transduction using these thin films and showed this approach to be as sensitive as SAW devices. We expect to improve on the sensitivity of the waveguide Zeeman approach, based on optimization of the optical waveguides. We have designed and fabricated optimized wavegides, and they are now being evaluated.

Publications

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Grace, K.M., et al., "Real-Time Chemical Detection using Species Selective Films and Waveguide Zeeman Interferometry" (to be published in *SPIE*).

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Yang, X., et al., "Molecular Host Sol-Gel Thin Films for Surface Acoustic Wave Chemical Sensors," *Sens. Actuators, Chemical* **45**, 79 (1997).

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Mathematics and Computational Science

Moment Equations for Two-Phase Flow in Random Porous Media

97528

Dongxiao Zhang

We are improving the mathematical methods for analyzing the movement of underground water and petroleum resources, methods which have broad potential for better understanding the mechanisms of uncertainty in general. Our goal is to develop partial differential equations for the moments of two-phase (oil-water) flows in heterogeneous porous media, as an alternative to the commonly used Monte Carlo simulations. Workable moment equations would have some important benefits over Monte Carlo. 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; therefore these equations could be solved on a relatively coarse grid. Third, the moment equations would be available in analytic form, even though they would be solved numerically in applications.

This year we developed a theory and a computer model for transient single-phase flow in heterogeneous porous media. The model has been partially validated by Monte Carlo simulations. We have also developed and solved moment equations for two-phase displacement in random porous media. Good agreement is found between the moment equation results and Monte Carlo simulations.

Publications

Harter, T., and D. Zhang, "Water Flow and Solute Spreading in Heterogeneous Soils with Spatially Variable Water Content" (to be published in *Water Resour. Res*).

Xin, J., and D. Zhang, "Stochastic Analysis of Biodegradation Fronts in Heterogeneous Media," *Adv. Water Resour.* **22**, 103 (1998).

Zhang, D., "Nonstationary Stochastic Analysis of Transient Unsaturated Flow in Randomly Heterogeneous Media" (to be published in *Water Resour. Res.*).

Zhang, D., "Numerical Solution to Statistical Moment Equations of Groundwater Flow in Nonstationary, Bounded Heterogeneous Media," *Water Resour. Res.* **34**, 529 (1998).

Zhang, D., "Quantification of Uncertainty for Fluid Flow in Heterogeneous Petroleum Reservoirs" (to be published in *Physica D*).

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Design of an Indexing Scheme for Knowledge Management at Los Alamos National Laboratory

98566

Isabel Sandoval

Because of a ban on testing and consolidation of data archives across the DOE complex, legacy information is becoming a critical link between simulation, verification, and experimentation. It is estimated that the Los Alamos holdings include over 42 million records in various file formats and media types (e.g., paper, film, aperture cards, microfiche, and radiographs), including records that have been transferred from Mound, Pinellas, and Rocky Flats plants.

The project objective is to explore development of a state-of-the-art indexing scheme that might dramatically improve management and retrieval of information and also examine applications and interfaces necessary for possible implementation. Such a scheme would have significant impact on the nuclear weapons program and also have wide application in other information arenas.

In the first year of this exploratory effort, it is essential that we gather data from several involved organizations in order to understand the nature of their data and processes and to help us focus the thinking of our subject matter experts toward possible methodology development and implementations.

This year we developed a document called "Indexing Analysis Questions" and conducted interviews regarding weapons-related information and drawings with weapons organizations, research libraries, and facilities at Los Alamos and Sandia National Laboratories. Approximately 30 people participated in the interviews; 14 projects were represented; and 246 metadata fields were identified. We identified different needs and focus areas (e.g., beryllium, gas transfer, engineering, and design).

An area of concern is the need to know, a convention that has kept data separate and classification at the lowest level possible. Other concerns are that field definitions and coding conventions are inconsistent or nonexistent, documentation is limited, and numerous electronic information sources exist.

Our current focus is on aperture card media and drawings, of which there are over 1 million. The aperture card collection is heterogeneous in composition, with information located in inconsistent structures. Our research showed that the indexing process from aperture cards throughout the industry is accomplished manually, which is unacceptable practice for such voluminous material. Our research found only one commercial resource that could potentially address our needs in this area.

Responding to the Helsinki Accords and Proposed START III Reductions

98553

Darryl Butt

This project is divided into three separate but integrated areas: US weapons complex modeling, Russian weapons complex modeling, and weapons dismantlement transparency (WDT). Although the models and data generated under this project are useful for a variety of applications, the focus of our research was on producing tools and data that would be useful to the probable, upcoming START III treaty processes.

Initially, the aim of the US modeling effort was to construct a model that would track weapons, plutonium, and highly enriched uranium so that the transparency team could run various scenarios dealing with the impact of transparency on the US nuclear weapons complex. This milestone was met with the model called START III, which was expanded into a multilayer model called T^2.

Inherently, the Russian weapons complex modeling was comparatively lacking in data, but through our research we were able to acquire and document considerable data into various models, many of which took the form of maps and complementary materials flow sheets. These two parallel efforts were integrated through our WDT effort, which involved using these data to write a draft protocol statement for WDT under a START III regime.

Publications

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Signal Integrity Verification

98555

David Cartwright

Traditional design approaches assume that the time delay across electronic devices is mainly determined by transistor rather than electrical-connection (interconnect) characteristics. However, in future generations of smaller devices, 80% or more of the time delay across an electronic device will be determined by the interconnects. Therefore, traditional approaches to designing electronic devices will not accurately predict either time delay or signal integrity (crosstalk).

Two types of three-dimensional (3-D) design tools will have to be developed: (1) scalable, physically based, engineering computer-aided design (ECAD) tools that can extract capacitance and inductance from general, 3-D shapes of metals/dielectrics and (2) tools that would predict the crosstalk between real 3-D interconnect elements. Because future generations will require "clock speeds" in excess of 500 MHz, the lack of these 3-D design tools is now viewed as a serious obstacle to future capability.

Aimed at eliminating this obstacle, our research project is composed of two parts: calibration of existing Motorola electromagnetic design tools with the finite-difference time-domain (FDTD) code created at Los Alamos and development and implementation of a new code for extracting capacitance and inductance from complex, 3-D interconnect structures.

This year we ran three different reference interconnect geometries that validated the Los Alamos FDTD code as accurate for the "slow-wave" and "skin-depth" limiting cases for which commercial design tools exist. A new geometric case, provided by Motorola to test the effect of partially conducting substrates, was run and shown to be closer to the latter limiting case. Motorola started fabrication of a more-advanced test case whose performance we will predict with the FDTD code.

We also implemented into software a new series of algorithms based on solutions to Poisson's equation for extracting capacitance and inductance matrices from 3-D multiple-material interconnect structures of general shape. The code was tested against the spherical capacitor (analytical solution), the infinite cube capacitor (numerically exact solution), and a single flat plate (numerically exact solution). We have generalized the code to allow treatment of multiple dielectrics for which there is currently no design tool available, and Motorola will fabricate test structures against which this code will be validated.

A Hierarchical Simulation R&D Test Bed for Test and Evaluation

97535

Stephen Upton

The project objective is to develop an integrated collection of computer simulations and methodologies that can be applied to the analysis of war-fighting systems. Analysts use simulations to test and evaluate war-fighting problems; for example, simulations can be used to understand survivability scenarios of a fighter aircraft. Our goal is to provide an expanded capability beyond what analysts currently possess by combining an existing war-fighting simulation architecture with available statistical methodologies, existing physics-based codes, and current hardware.

This year the following accomplishments moved us closer to our goal of developing an integrated simulation and analysis hierarchy. First, we developed the theoretical statistical methodology to address simulation input uncertainty with a small number of variables. We then applied the developed statistical methodology to a simple engagement simulation; specifically, we simulated an engagement between a fighter aircraft and a surface-to-air missile defense system. Finally, we assessed the application of the statistical methodology to the engagement simulation; the assessment revealed a significant utility for this methodology, particularly in the area of simulation testing. 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.

Publications

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Statistics of Rare Events

98550

Len Margolin

In this one-year project, we studied the subject of rare events (sometimes termed "large deviation theory") in two areas: mathematical models of traffic flow and turbulence.

Traffic theory has been receiving much attention recently, and a number of approaches have been suggested, including fluid mechanics, cellular automata, particle hopping, and ballistic motion. We introduced a simple model to study one-lane rural traffic. This model incorporates passing and clustering and can be analyzed using a modified Boltzmann equation. We have solved analytically for various steady-state properties such as the flux and car/cluster velocity distribution. Our results indicate that a single dimensionless parameter determines the nature of the steady state. Analogous with the Reynolds number in turbulence, this number allows us to write the equations in a dimensionless form. This number is simply a dimensionless measure of the collision frequency. For small collision numbers, the flow is uninterrupted, while for large collision numbers, large clusters occur. In this strong interaction case, the flux is suppressed algebraically. Meanwhile, the velocity distribution develops a boundary layer structure, which means that cars faster than a threshold velocity are strongly affected by the presence of slower cars. The statistics of the slowest cars characterize the nature of the steady-state velocity distribution.

Some of the predictions of the theory were also confirmed by direct observations. Furthermore, by introducing the Maxwell collision integral (which approximates the collision rate), we obtained relaxation properties of the velocity distribution and the size distribution. The relaxation of the car and cluster velocity distributions toward steady state is characterized by a wide range of velocity-dependent relaxation scales. These relaxation time scales decrease with the velocity, with the smallest scale corresponding to the decay of the overall density. Again, extreme statistics of the initial velocity distribution characterize the nature of the relaxation toward the steady state. The steady-state cluster-size distribution follows an unusual scaling form in the sense that it is characterized by the square of the average cluster size, rather than by the average itself. This distribution is algebraic except at its very large tail, where it is exponential.

The Navier-Stokes Equations (NSE) are widely believed to provide a satisfactory mathematical description of turbulent flows. However, in the context of numerical simulation, it is not possible to resolve the full range of important physical scales except in the most idealized cases. The practical alternative is large eddy simulation (LES), in which the effects of the unresolved scales on the resolved scales are modeled. A particular model that has been recently proposed is based on the Camassa-Holm Equations (CHE). This model, termed an "alpha model" because of the presence of a new parameter, alpha, has been shown to reproduce the mean statistics of turbulent flow in channels and pipes (as validated by experimental data). In this project, we have studied the variability of the simulated flows, using a pseudospectral code to perform direct numerical simulations of both the NSE and the CHE. We then compared these results to LES simulations, using the same code at lower resolution. The variability of the flow can be estimated in terms of the normalized ratios of higher moments to lower moments of the flow velocity. We have performed a suite of simulations for different values of the parameter alpha and are currently analyzing the data. First indications are that the alpha equations reduce the variability of the flow.

Publications

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E. Ben-Naim, P.L. Krapivsky, "Maxwell Model Traffic Flows" (submitted to *Phys. Rev. E*).

Simulation of Thin-Film Formation

97513

Robert B. Walker

In our research, we are seeking to develop algorithms and software for a fully three-dimensional integrated modeling, simulation, and design capability for predicting the behavior of materials. This capability includes being able to predict how the deposition depth and texture of thin films of various materials will vary according to their position on the original material surfaces being studied.

We are collaborating with Motorola, Inc., to develop reliable simulation software capable of predicting these behaviors in deposited thin films of metals and metal oxides. We have integrated a variety of techniques and disciplines, using the Laboratory's high-performance computing expertise to draw from large-scale molecular dynamics and Monte Carlo simulations, continuum modeling, and gas-phase and surface chemistry. To establish the validity of these computer simulation tools, we compare code predictions with the results of well-designed experiments.

Our ability to predict the deposition thickness of films relies, in part, on being able to predict in what way the probability of an impacting atom sticking to a surface depends on the collision energy and the angle of the atom when it hits the material's surface. We have recently studied these processes in great detail.

Our preliminary simulations have led to the development of an improved potential energy function to describe the interaction between copper atoms. The improvements in the potential function concentrate on interactions at higher maximum energies. With this new potential, we have re-examined the sputtering physics that occurs when copper atoms collide at high energies with copper surfaces. New results for this copper-atom/copper-surface system show some unexpectedly high sticking coefficients for events where incidence atoms strike the surface at low angles and again when they strike at high angles. For intermediate impact energies of the atom, this angle-dependent sticking coefficient does not (as expected) go to zero for small (grazing) angles, but instead goes through a minimum and then increases again. The sticking coefficient for the sputtering of platinum atoms on a platinum surface also behaves in a similar fashion.

In contrast, simulations using rough metal surfaces do not show this upturn in the sticking coefficient at grazing-incidence angles. This observation is consistent with our intuition—an atom skimming along a smooth flat surface has difficulty scattering off into the vacuum, but on a rough surface, an atom can more easily scatter off a step or other protrusion. Our calculations show that the effect of surface roughness on the sticking coefficient will be the greatest at grazing-incidence angles and intermediate (10–50 eV) energies.

Our developing capability to predict the texture (microstructure) of deposited films must be validated against experiments. Some initial promising results have been obtained from x-ray microscope experiments performed on two of Motorola's metal samples (an alloy made of copper and aluminum and another alloy made of copper, aluminum, and silicon). Diffraction patterns were generated and texture values were successfully determined (±4% disorientation for the AlCu alloy and ±6% for the AlSiCu alloy).

Publications

Boehm, R., et al., "Theoretical Studies of H_2 Desorption from Boron-Doped Surfaces" (submitted to *Surf. Sci.*).

Coronell, D., et al., "Molecular Dynamics-Based Ion-Surface Interaction Models for Ionized Physical Vapor Deposition Feature Scale Simulations" (to be published in *Appl. Phys. Lett.*).

Hanson, D., et al., "An Interatomic Potential for Reactive Ion Etching of Si by Cl Ions" (to be published in *J. Chem. Phys.*).

Hanson, D., et al., "Reactive ion Etching of Si by Cl, Cl_2, and Ar Ions: Molecular Dynamics Simulations with Comparisons to Experiments" (submitted to *J. Vac. Sci. Technol., A*).

Kress, J., et al., "Molecular Dynamics Simulations of Cu and Ar Ion Sputtering of Cu(111) Surfaces" (submitted to *J. Vac. Sci. Technol., A*).

The Effects of Operator Splitting on the Predictability of Numerical Models

98551

William Rider

Our research focuses on identifying and alleviating the effects of operator splitting in the integration of complex systems. Operator splitting is commonly used to simplify the simulation of complex systems. Despite its ubiquitous use, its impacts on the solutions are largely uncharacterized.

This year we have identified various effects of operator splitting. Chief among them is the generic loss of accuracy associated with the application of operator splitting to nonlinear equations. Furthermore, operator splitting near critical points produces significant anomalous effects. It can result in pathological structures in the simulation of mixing layers. The nonphysical perturbations result in reduced accuracy and unrecoverable lack of resolution. We have identified other effects in nonequilibrium radiation transport, where splitting can produce intrinsically incorrect solutions.

We have also identified various methods to remove operator splitting. Chief among these are Newton-Krylov methods for the efficient, nonlinearly consistent integration of systems of equations. We have developed unsplit methods for hyperbolic equations and tested them on mixing phenomena. In addition, we have developed new equations and methods for coupling hydrodynamics and radiation.

Publications

Knoll, D.A., et al., "An Efficient Nonlinear Solution Method for Nonequilibrium Radiation Diffusion" (to be published in *J. Quant. Spectros. Radiat. Transfer*).

Lowrie, R.B., and J.E. Morel, "The Coupling of Radiation and Hydrodynamics" (submitted to *Astrophys. J.*).

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

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," *J. Comput. Phys.* **141**, 112 (1998).

Rider, W.J., et al., "A Multigrid Newton-Krylov Method for Multimaterial Equilibrium Radiation Diffusion" (submitted to *J. Comput. Phys.*).

A Composition Environment for the Virtual Simulation Test Bed

98508

W. Greg Lacey

We have developed a new tool for rapid development of simulations: a composition environment, containing reusable model components, in which an analyst can compose simulations or composite components to model and simulate scenarios. The objective of the project is to enable users to create new simulations and to provide simulation-based analyses more quickly and with greater confidence in the product than is currently possible. The analyst or subject-matter expert is freed from building new simulations, and from rebuilding existing models, and can therefore focus on understanding the problem at hand and examining simulation results and alternative scenarios.

This one-year project generated not only the composition environment but also an initial set of player models for one ongoing program, biological defense, and several other program-development efforts, e.g., Anti-Personnel Land Mine Alternatives, targeting of hardened and deeply buried targets, and Crisis/Consequence Management Information System. The test bed software, highly instrumental in obtaining funding for the programs, features a fully distributed, machine-independent architecture upon which completely portable components can be developed, warehoused, and combined to respond to practically any simulation demand. Our results have been prepared in report form and are being submitted for publication, and we are receiving a continuing stream of requests for new uses for the test bed.

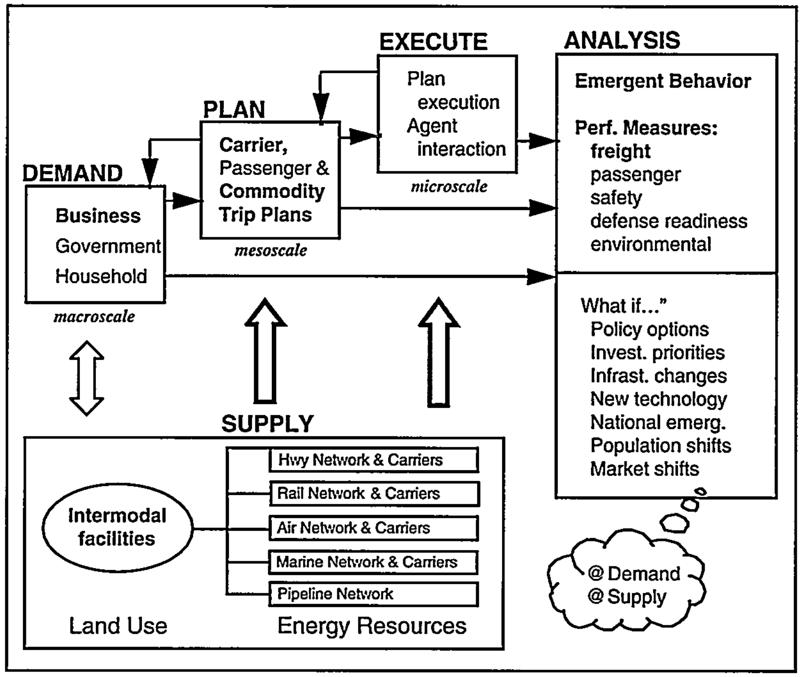
National Transportation System Analysis Capability

98556

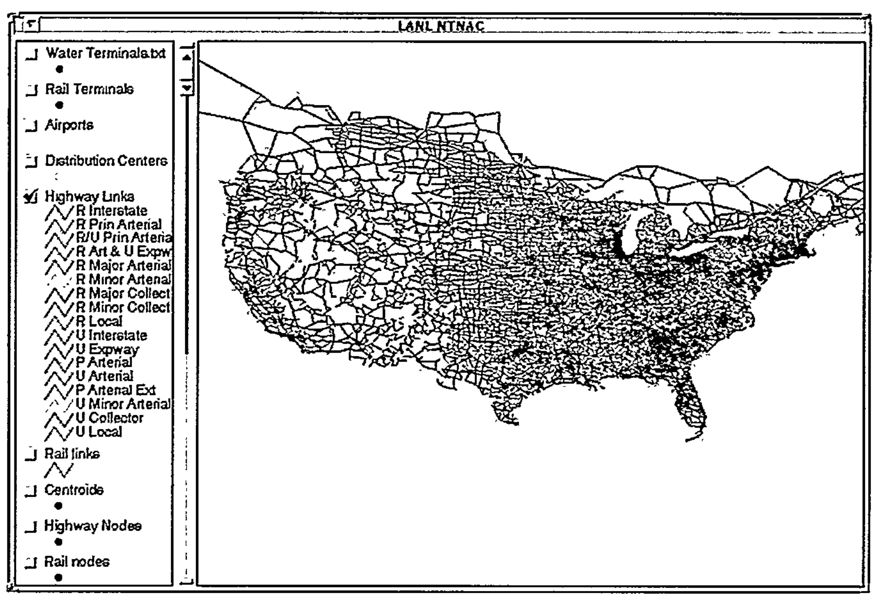
Douglas Anson

The Department of Transportation (DOT) and Department of Defense (DoD) need a National Transportation Network Analysis Capability (NTNAC) that simulates the movement of individual carriers and loads over the national transportation system. The goal of this project is to demonstrate the feasibility, in the form of a NTNAC prototype, of building a simulation-based model that integrates the national highway, rail, air, water, and pipeline systems and subsystems.

This year we achieved three main objectives. First, we designed the overall framework of the NTNAC based upon an analytical decomposition of a large-scale transportation system first developed in the Laboratory's Transportation Analysis and Simulation System (TRANSIMS) model of regional transportation systems (see first figure). The framework partitions the problem according to functional, temporal, and spatial dynamical characteristics in the form of supply, demand, plan, execute, and analysis modules. Second, we obtained large, independently developed data sets and integrated and modified them to meet the requirements of the supply and demand modules. Data was drawn from public and private sources such as the DOT, the Bureau of the Census, Oak Ridge National Laboratory, the American Trucking Associations, and Yellow Freight, Inc. Third, we developed and integrated network and carrier initialization routines (supply module); county-to-county, carrier-sized commodity-shipment algorithms (demand module); and an agent-based simulation of the highway (see second figure) and rail networks (plan module). These three modules are the core of our NTNAC prototype.



The analytical decomposition of a large-scale transportation system used in the National Transportation Network Analysis Capability (NTNAC). The prototype addresses those portions in bold type.



The national highway network consists of over 95,000 links, 67,000 nodes, and 3 million nonlocal trucks.

Development of an Integrated Global Energy Model

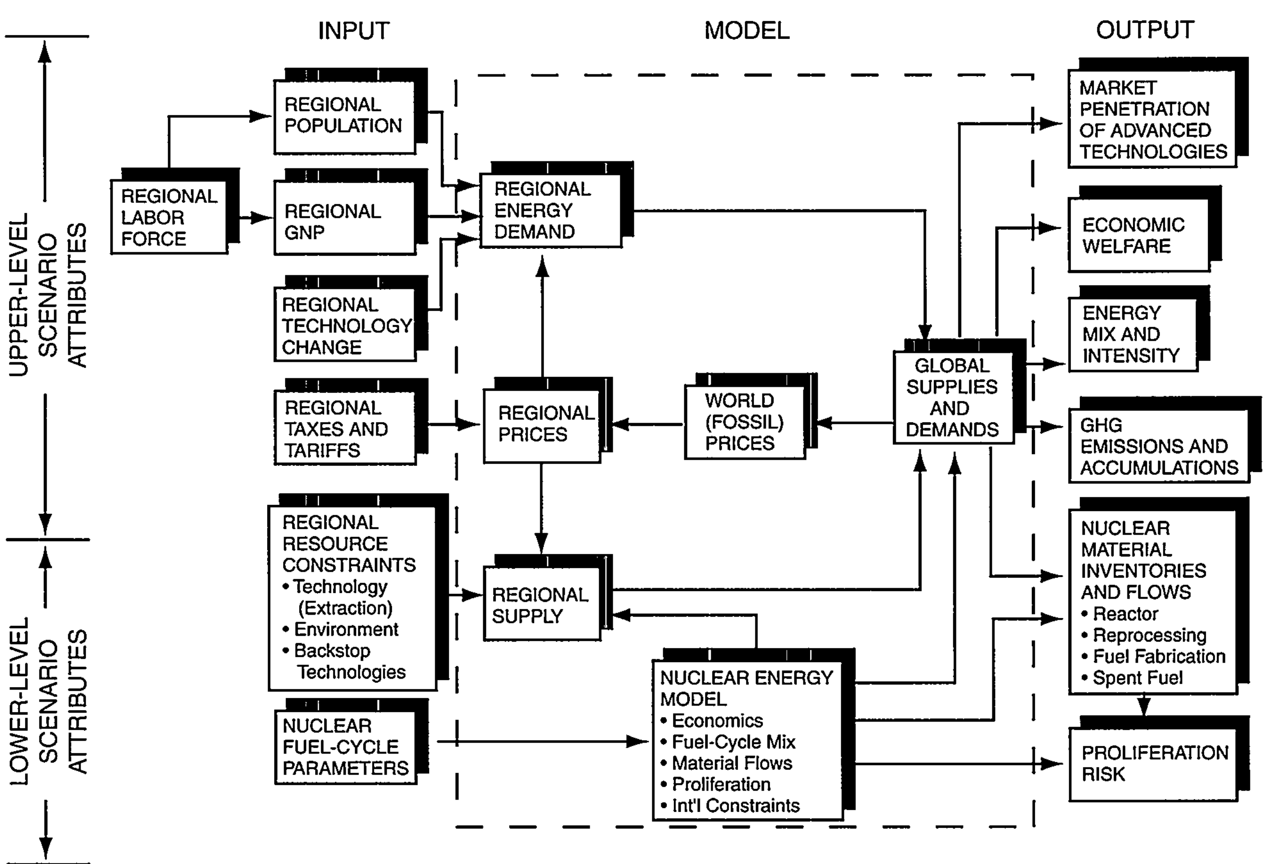
98534

Robert Krakowski

The primary goal of this project is to develop a global energy-economics-environmental (E3) analysis tool and to better understand long-term nuclear energy and nuclear materials futures. We have taken an existing multiregional E3 model and adopted, modified, and used it to develop a range of long-term scenarios that might transpire under a range of energy demand-side and supply-side forces. The scope of this project is best illustrated by the input/output variables that form the essential elements of the modified Edmonds-Reilly-Barns (ERB) macroeconomic model used in these studies (see first figure). Of particular interest to these studies are the E3 tradeoffs related to supply-side economic drivers, security issues, and environmental concerns.

Viewed through the "lens" of these interconnected E3 impacts, the second figure illustrates the avoided increase in average global surface temperature as the market share of nuclear (and other noncarbon) energy sources is increased by making fossil energy more expensive through the imposition of a carbon tax at the indicated rates. The benefit of reduced global warming is accompanied by increased costs related to reduced productivity (reduced gross domestic products brought about by increased energy prices) and elevated risks associated with nuclear proliferation (increased inventories of commercial plutonium). The resulting benefit-cost tradeoffs illuminated by the E3 model depend sensitively on the carbon-tax structure implemented and the nuclear fuel cycle adopted for the particular scenarios being considered.

Studies of these kinds point to directions wherein the benefits of nuclear technologies can be optimally derived while reducing risks associated with both fossil and nuclear energy over a long run where both populations and demands for overall well-being are increasing. Furthermore, results obtained to date illuminate research that can lead to increased modeling realism through endogenizing the coupling between energy resource depletion, prices, and technology innovations, and between productivity, economic growth, per-capita well being, and population growth.



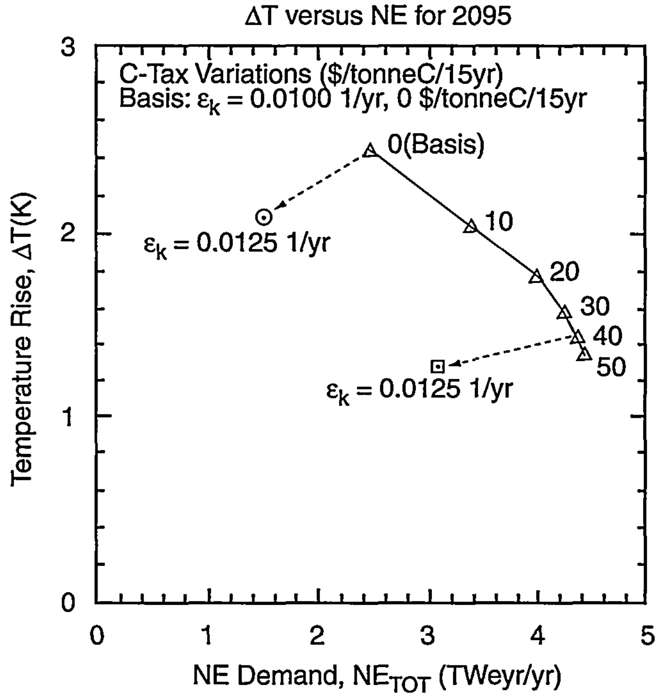
Input-output layout of Edmonds-Reilly-Barns model, as modified and adapted to the needs of our present study.

Publications

Krakowski, R.A., "Re-Engineering Fission: Reactors for Safe, Globally Sustainable, Proliferation-Resistant, and Cost-Effective Nuclear Power" (to be published in *Aspen Climate Change Institute Workshop on Innovative Energy Systems and CO_2 Stabilization*).

Krakowski, R.A., "The Role of Nuclear Energy in Mitigating Greenhouse Warming," in *Global Foundation International Conference on Environment and Nuclear Energy,* B.N. Kursunoglu, Ed. (Plenum Press, New York, New York, 1999).

Krakowski, R.A., et al., "Nuclear Fission for Safe, Globally Sustainable, Proliferation-Resistant, and Cost-Effective Energy," in *Global Foundation International Conference on Environment and Nuclear Energy,* B.N. Kursunoglu, Ed. (Plenum Press, New York, New York, 1999).



Sample results showing impact of carbon taxes imposed at the indicated rates on reductions in global warming and increased nuclear-energy capacity in the year 2100; also shown is the impact of increased rate of end-user energy efficiency, e_k.

Numerical Methods for Stochastic Partial Differential Equations

98549

David Sharp

We are investigating various issues regarding the derivation and numerical solution of nonlinear, stochastic, partial differential equations (SPDEs). While convergence properties of stochastic ordinary differential equations (ODEs) are by now well understood, the situation for SPDEs is still somewhat unclear, especially when the stochastic forcing terms are spatio-temporal and nonlinear (multiplicative noise). The first problem we tackled was the control of errors in one-dimensional SPDEs satisfying a fluctuation-dissipation relation: errors arise from the finite time step as well as from the spatial discretization. By writing the functional Fokker-Planck equation, which corresponds to the particular SPDE at hand, we showed how to isolate errors arising from the finite time step without actually needing to solve the Fokker-Planck equation. Our methodology was tested by comparing the analytic predictions to high-resolution simulations of a Landau-Ginzburg theory, with excellent results. Our error control techniques were applied to a study of statistical mechanics.

We have developed a new second-order algorithm for multiplicative noise, which required only a single random-sequence generation per time step. By comparison with analytic solutions, this algorithm has been proved for the case of stochastic ODEs where it is clearly superior to the standard Heun algorithm. Moreover, it is easily extendable to the case of SPDEs. Finally, based on a series of lectures given by two of us at the Center for Nonlinear Studies at Los Alamos, we worked on a review article on the physics and mathematics of a large class of SPDEs with the aim of making the mathematical literature accessible to chemists, physicists, and engineers who use these equations in their applications.

Publications

Glimm, J., and D. Sharp, "Stochastic methods for the Prediction of Complex Multiscale Phenoemena" (to be published in *Quart. J. Appl. Math*).

Habib, S., et al., "Statistical Mechanics of Double Sinh-Gordon Kinks" (to be published in *Physica D*).

Khare, A., et al., "Exact Thermodynamics of the Double Sinh-Gordon Theory in 1+1-Dimensions," *Phys. Rev. Lett.* **79**, 3797 (1997).

Mineev-Weinstein, M., "Selection of the Saffman-Taylor Finger Width in the Absence of Surface Tension: An Exact Result," *Phys. Rev. Lett.* **80**, 2113 (1998).

Rapid Prototyping of DoD Simulation Entities

98506

Randy Michelsen

The objective of this project was to extend a prototype software framework developed in previous DoD work for others. A software framework provides a set of general mechanisms designed to assist in the construction of a specific class of applications and a computational environment within which to apply them. A framework also specifies the operational protocol or architecture in which simulation components are designed to interact. Our goal was to provide a more comprehensive, flexible software framework for simulation construction.

Our Integrated Virtual Environment for Simulation (IVES) is a component-based framework that allows the construction of simulations and simulation elements using a compositional paradigm, or "plug and play" design. In this context, a component is a well-delineated, relatively independent, and replaceable part of a software system that performs a specific function. The IVES framework provides software components representing such common simulation elements as events, event-stepped execution controllers, and a variety of other typical infrastructure elements, such as graphical user interfaces and databases. Advantages of the component-based approach include increased software reuse, enhanced ability for validating or verifying the software, and reduced development costs in the delivery of focused, tailored simulations specific to analysis requirements.

We have successfully developed a framework for developing composition-based simulations and demonstrated its applicability to problems ranging from DoD war fighting to manufacturing processes. The resulting software is actually a second-generation product derived from the earlier work that now incorporates unique technical features and capabilities. The framework presently supports "nested" composition of software components to any depth. Compositions may be controlled through the definition and use of constraints (e.g., "only two subcomponents allowed" or "maximum weight is *x*"). These constraints are implemented as software objects, either chosen from a predefined set or created by a developer, that define restrictions to be applied at the time of constructing a simulation or simulation element. The complete set of defined elements, including the constraint mechanism, visualization components, and other framework components, is readily usable and extensible.

Publications

Holland, J.V., et al., "Composing Simulations Using Persistent Software Components" (submitted to *Advanced Simulation Technologies Conference, ASTC '99).*

Generative Analysis for Future DoD Concepts

98503

Stephen Upton

The objective of this project is to integrate elements of the Integrated Virtual Environment for Simulation (IVES), a composable simulation environment derived from earlier JointSim work. Our approach is "generative analysis," which investigates issues through a robust directed search over a large region of a problem space. This method is unlike the traditional comparative-analysis approach, which tends to limit the search over a small region of a problem space.

Our overall goal for this year was to conduct a proof-of-concept demonstration with a few simple entities. We focused on developing the requirements for allowing actors (individual simulation components that can interact with other such components) to evolve based on input goals and objectives, actor capabilities, and the synthetic environment of the proposed system. Additionally, applying the methodology involved the exploration of future Department of Defense (DoD) concepts. We developed limited behavioral representations of actors at the individual/platform level and used them to populate a medium-scale war-fighting simulation. The demonstration indicated that more work is needed but that the overall generative-analysis concept appears feasible.

End-to-End Radiographic Systems Simulation

98537

Allen Mathews

Our goal is to improve radiograph simulation and analysis capabilities by developing a validated end-to-end radiographic model that can be applied to both x-rays and protons. The specific objectives are to link hydrodynamic, transport, and magnetohydrodynamic (MHD) simulation software for purposes of modeling the radiographic process and radiographic systems and to develop optimization and analysis algorithms to validate physical models and optimize systems. These efforts will provide the means to make the best use of data from both existing and new radiographic facilities, and they will help determine performance requirements and facility design for the next generation of radiographic facilities by assessing capability to meet the needs of science-based stockpile stewardship.

Last year we rewrote the MERLIN Particle-In-Cell MHD code to run on computer platforms developed by Silicon Graphics, Inc., and on platforms developed at the Laboratory as part of the Accelerated Strategic Computing Initiative. We verified the code's accuracy using a test suite and began links to the Monte Carlo Neutron and Photon transport code, called MCNP. We installed the new Integrated Tiger Series 3.0 cross-section library in MCNP and validated the calculated Bremsstrahlung angular distribution and cross sections, using published results.

We developed new models for proton cross sections that are applicable to proton radiography. In addition, we developed a new method for inverse reconstruction of radiographs, using a regularized least squares method and direct matrix inversion, and applied the method to x-ray and proton radiographs using oblique views.

We developed a method for reconstructing three-dimensional (3-D) objects by using arbitrary azimuth and elevation angles and applied the method to synthetic radiographs of 3-D objects.

We began the development of a "designer-assisted analysis" tool for analyzing radiographs and other diagnostic data by connecting them directly to hydrocode calculations. Thus, the effects of changes in design simulations can be validated by simultaneous comparison with a variety of experimental measurements. We also began investigating the use of "wavelets" for image analysis and modeling on multiple scales. These techniques promise to be more flexible and robust than traditional Fourier methods.

Atomic, Molecular, Optical, and Plasma Physics, Fluids, and Beams

Magnetized-Target Fusion: Experiments and Modeling

96388

Richard Siemon

The magnetized-target fusion (MTF) approach begins with fusion fuel in a magnetized and preheated target plasma and then heats the fuel to thermonuclear temperatures by compression with a hypervelocity liner implosion. The purpose of this project was to generate MTF target plasmas experimentally and to simulate the formation process with magnetohydrodynamic (MHD) codes. Also, an assessment of MTF's potential relevance to Science-Based Stockpile Stewardship (SBSS) was needed because MTF, if successful, would be a valuable experimental source of intense neutrons.

We used the Colt capacitor-bank facility at Los Alamos, which drives a current of 1–2 MA in 2.5 μs, to form target plasmas. The initial mass for the plasma was provided either by a polyethylene thread along the *z*-axis or by a uniform static fill of hydrogen gas. Operation with frozen deuterium fibers was not possible with the available vacuum system. We studied plasma dynamics using B-dot probes, fast framing photography, filtered silicon photodiodes (analyzed with previously developed computational tools), a HeCd-laser interferometer, a time-resolved visible monochromator, and a gated, optical, multichannel visible spectrometer. Results suggest that hot plasma cools in 1–3 μs, whereas cool plasma (with energy less than 10 eV) persists for about 25 μs. The results we achieved were qualitatively similar to simulations that suggested polyethylene fibers would not show a late-time Kadomtsev-stable profile. We established that existing MHD tools will need some improvement to simulate implosions including both plasma and liner dynamics. Promising applications of MTF neutron sources to SBSS were identified.

Based on the results from this project, the MTF team prepared a proof-of-principle proposal and submitted it to DOE's Office of Fusion Energy Sciences. Participants from the University of Washington, Westinghouse, the Air Force Research Laboratory, Lawrence Livermore National Laboratory, and General Atomics also contributed to the proposal. DOE's review of the proposal was favorable.

Publications

Siemon, R.E., et al., "Why Magnetized Target Fusion Offers Low-Cost Development Path for Fusion Energy" (to be published in *Comments Plasma Phys. Controlled Fusion*).

Remote Atmospheric Measurements and Modeling (RAMM) Project

98567

John Schultz

Schemes that have been proposed for Doppler lidar wind sensing include coherent detection and two approaches using Fabry-Perot interferometers. We have developed a concept for an unequal-path two-beam interferometer, in a Mach-Zehnder configuration, that will have the required sensitivity in a simpler and more stable instrument. In addition, calibration will be much simpler than for the other devices. Our objectives in this study were to demonstrate that (1) the device would have the sensitivity to measure velocity to an rms (root mean square) accuracy of 1 m/s and (2) the required measurement accuracy could be obtained with as few as 100 photon counts.

A breadboard interferometer was constructed with a path difference of 1.6 m between the two beams. The fringe shift is given by $f = 2(v/c)(OPD/\lambda)$, where f is the fringe shift, (v/c) is the velocity divided by the velocity of light, and OPD/λ is the ratio of optical path difference to wavelength. This results in a shift of one fringe for a velocity of 60 m/s, showing that the required accuracy can be obtained.

In a separate interferometer, variable-intensity fringes were formed on a multichannel photomultiplier, and the accuracy of measuring the fringe position was determined. The result showed the expected variation of accuracy as a function of photon count, 1/50 fringe being obtained with fewer than 100 counts. The experiment thus met both of our objectives.

Engineering Science

Physics-Based Damage Predictions for Simulating Testing and Evaluation (T&E) Experiments

97515

Frank Addessio

The primary objective of this research project is to develop an accurate and efficient numerical modeling technique that is capable of simulating fully coupled fluid/solid interactions. Presently, attention is being focused on predicting structural damage that results from hydrodynamic ram and related physical phenomena. The modeling capability that is sought requires the development of a computational technique that can calculate the dynamic interaction of multiple fields of interpenetrating media, as well as physics-based material models that describe the complex dynamic deformations and motions of the solids, liquids, and gases involved.

This year we have developed a fully three-dimensional computational technique for solving fully coupled fluid/solid interaction problems. Both homogeneous and micromechanics-based, heterogeneous solid-material models have been incorporated into the code for describing the large-deformation, high-strain-rate behavior and failure of both elastoplastic and viscoplastic solids. We have included arbitrary equations of state for describing the density- and temperature-dependent pressures that develop in both the fluids and the solids. These state equations allow for arbitrary fluid velocities ranging from fully incompressible to hypersonic regimes.

We also have incorporated classic plate and shell theory into the code in an effort to mitigate length scale resolution issues and are considering including a higher-order shell theory to better represent the thin, composite structural components often found in fluid-containment systems. In addition, we have identified several sample calculations as potential code and model validation problems and have already used the results of two separate simulations to assist in the design of several fluid/solid interaction experiments.

Radio Frequency Weapons Technology Assessment for Aircraft Defense

98507

Michael Fazio

Traditionally, radio frequency (RF) and microwave weapons have used a modulated electron beam. However, it is difficult to produce electron beam-based systems that are compact, rugged, and inexpensive. We have been investigating electromagnetic weapon systems that are based on solitons. Solitons (or solitary waves) can be generated in nonlinear ceramics. Arrays of solitons can be generated at frequencies (a few gigahertz) and powers (up to a gigawatt) necessary for electromagnetic weapons. These compact, all-solid-state, instant-start systems can be used to protect aircraft from infrared or RF missiles or to attack surface-to-air-missile sites.

This past year we focused on two areas. The first area is solid-state, high-voltage switches for driving soliton-producing transmission lines. These switches must switch tens of thousands of volts (~30 kV) at currents of tens of thousands of amps. We have been developing a set of thyristor-based switches that are instant start and very compact, that switch the necessary voltage and current, and that run at repetition rates of up to 1 MHz.

Our second area of focus was soliton-generating materials, where we have been developing barium-strontium titanates as well as antimony-sulfur iodide. These materials have a very high initial dielectric constant (~15,000), high nonlinearity, high relaxation frequency, low loss tangent, and high dielectric strength. In addition to the materials research, we have been developing the test equipment needed to characterize these materials.

Thermal Energy Transport with MIC

98564

Lawrence Hull

Materials called metastable intermolecular composites (MICs) are highly energetic and can be "tuned" by varying their physical parameters from behavior similar to ordinary thermite to very fast reacting or "superthermite" behavior. Certain applications require that energy in the MIC be deposited onto a target of interest. Our objective is to obtain information on the energy deposition mechanism and associated expected thermal effectiveness in an example configuration.

The configuration we examined consisted of molytrioxide/aluminum MIC burned in a roman-candle-like device and directed upward at a flat steel target plate. The tube was completely filled by 25.3 g of MIC at the pour density of 0.2 g/cm^3 and placed so that the mouth of the tube was separated from the plate by 127 mm. Analysis of the temperature history of the steel plate shows that about 40% of the energy originally contained in the MIC was deposited in the plate. Further, the peak temperature increase that we recorded was above 1200°C, and the duration was on the order of 2 s. For comparison, when the MIC was replaced by an energy-equivalent amount of gunpowder, the maximum temperature increase was below 50°C.

This work demonstrates that the mechanism of thermal transport using MIC is mass deposition on the target through condensation and freezing. The thermal deposition mechanism for the gunpowder baseline is primarily gas convection because the gunpowder reaction products are predominately gaseous at room temperature. Thus, MIC can deliver a highly effective thermal pulse to a target.

Technology Development for Cold-Neutron Spallation Science

98552

Geoffrey Greene

The purpose of this project is to identify and develop novel technologies that enhance and exploit the unique capabilities of pulsed (spallation) cold-neutron sources. The use of spallation neutron sources can be enhanced by increasing the "useful" intensity of the neutron pulse. This can be done in two ways. The first approach is to make the incident proton pulse more intense. The second approach is to make the target/moderator system more efficient or more appropriately matched to the experiment. DOE/ Defense Programs is currently supporting a major upgrade to the Los Alamos Neutron Science Center (LANSCE) spallation source using the first approach.

Our LDRD project has supported moderator studies to explore the second approach. The work involved the application of Monte Carlo codes to assess the performance of cryogenic neutron moderators. The project also supported the development of novel technologies that exploit pulsed neutron sources. An essential feature of a pulsed source is the high phase-space density of the pulse. We have developed a new method to allow the high intensity of the short pulse to be transformed into a very narrow range of neutron momenta for neutron reflectometery. The technique involves the rapid (20-Hz), extremely well controlled (less than 1 arc-minute accuracy) oscillation of the reflectometry sample in synchrony with the spallation target. A detailed engineering study of the technique has been carried out. The final design (including fabrication drawings) of an apparatus to demonstrate the method has been completed and will be tested in FY99.

A Miniature Flux-Gate Magnetometer System for Mapping and Identification of Ferrous Bodies

97536

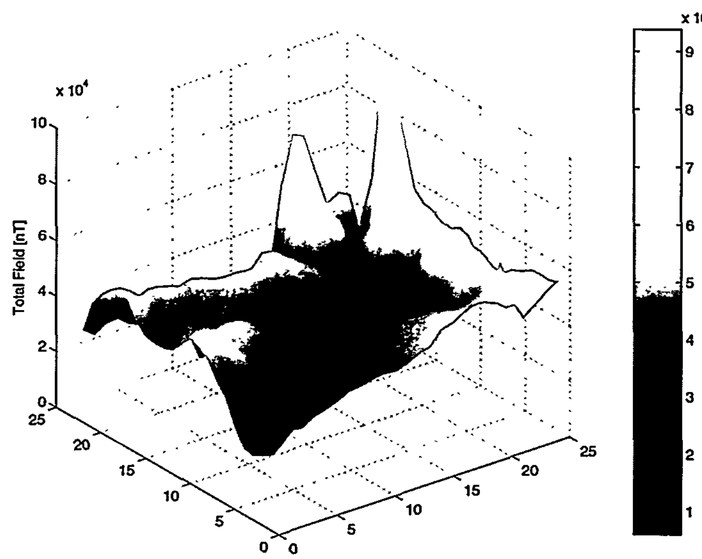
R. Clayton Smith

Conventional munitions test ranges in the US collectively contain thousands of pieces of unexploded ordnance (UXO) that render the ranges unsafe for human occupation. The land affected by UXO amounts to millions of acres. In order to reclaim this land, UXO must be identified and located for removal. Currently there is no single instrument capable of identifying and locating UXO. Our objective was to design a prototype magnetometer instrument that is low power, lightweight, and capable of being mounted on an autonomous platform that can enter a range containing UXO and gather magnetic information without the risk of human casualty.

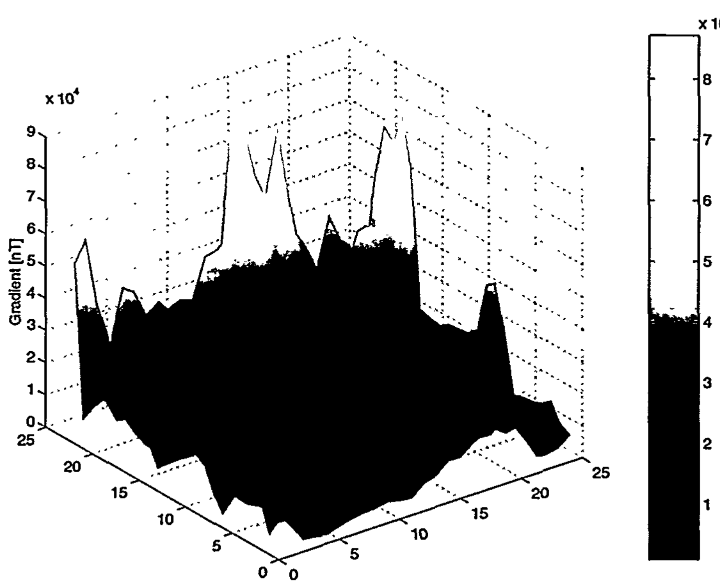
We developed numerical models for simple shapes of ferrous metal objects placed on or below the earth's surface. Application of these models to various UXO configurations helped us define the sensitivity requirements for the instrument. We have designed and built an instrument to measure ~20- to 400-nT anomalies using the total field and gradiometer measurement techniques. The instrument is a three-axis vector sensor with a three-axis total field sensor at its origin, and three magnetometers on axis with the origin magnetometers for gradiometric measurements.

We have tested the instrument in an area that is known to have local magnetic fields that vary greatly in a large space. The first figure shows a magnetic image taken with just the total field magnetometer. The second figure shows the gradiometric data for the same area. In the first figure, there is a relatively large flat area where small anomalies are overwhelmed by the magnitude of the earth's field (~40,000 nT) and cannot be seen. In the plot of the gradiometric data, the field of the earth has been effectively canceled and many features can now be distinguished in the area that was relatively flat in the total field image.

We have clearly demonstrated that it is possible to use a small, lightweight, low-power, fluxgate magnetometer system to take magnetic images of areas containing ferrous bodies. This magnetometer has an integrated geolocation capability, making it useful to navigate a robotic platform or to map the locations of magnetic anomalies created by ferrous metal debris from unexploded ordnance.



Total field data.



Gradiometer data.

Demonstration of Hydrothermal Plutonium-Combustible Waste Treatment Process

98510

Laura Worl

The goal of our project was to demonstrate a treatment based on hydrothermal processing for stabilizing transuranic combustible materials. Hydrothermal oxidation offers the ability to destroy radioactive combustible wastes to CO_2 and H_2O and is a favorable alternative to incineration or costly storage options.

We conducted a series of experiments with actinide-contaminated organic material. The accompanying table lists the types of actinide-contaminated organics that were processed and the destruction efficiencies. Organics included plutonium- and americium-contaminated analytical solutions, bromobenze, trichloroacetic acid, tributylphosphate mixtures, and ion exchange resins. The organic mixtures, processed at 540°C and 46.2 MPa, were reacted with 30 wt % H_2O_2 at an organic flow rate of 0.5 ml/min. The total organic carbon concentrations at the inlet of the pressure vessel were between 1,800 and 25,000 mg/L, depending on the organic. In the reactor, the organic was oxidized to levels below 20 mg/L and in most cases below 5 mg/L, resulting in >99.9% organic destruction efficiencies. (See process unit schematic and treatment flow sheet in accompanying figures.)

We also found that most of the actinide species entering the reactor are transformed to insoluble species. The solid plutonium component is easily removed from the effluents by simple filtration and is attributed to a plutonium (IV) oxide or plutonium (VI) carbonate solid because of the very low solubility of product in the solutions. A titanium liner in the reactor and heat exchanger provides corrosion resistance for the oxidation of chlorinated organics. The treatment of solid material (plastic bottles, rags) is accomplished either by pyrolysis to liquefy the organics, or particle-size reduction and the addition of a viscosity-enhancing agent to generate a pumpable mixture.

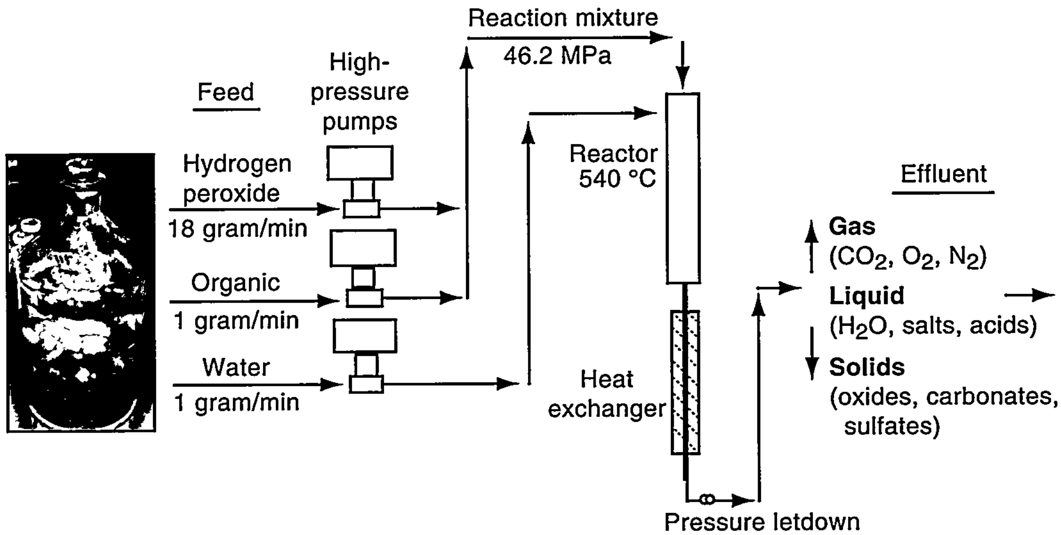
Publications

Worl, L.A., et al., "Hydrothermal Processing of Radioactive Combustible Waste," in *Mixed Waste Management I, 1998* (Spring Meeting of the American Institute of Chemical Engineers, New Orleans, LA, March 9, 1998).

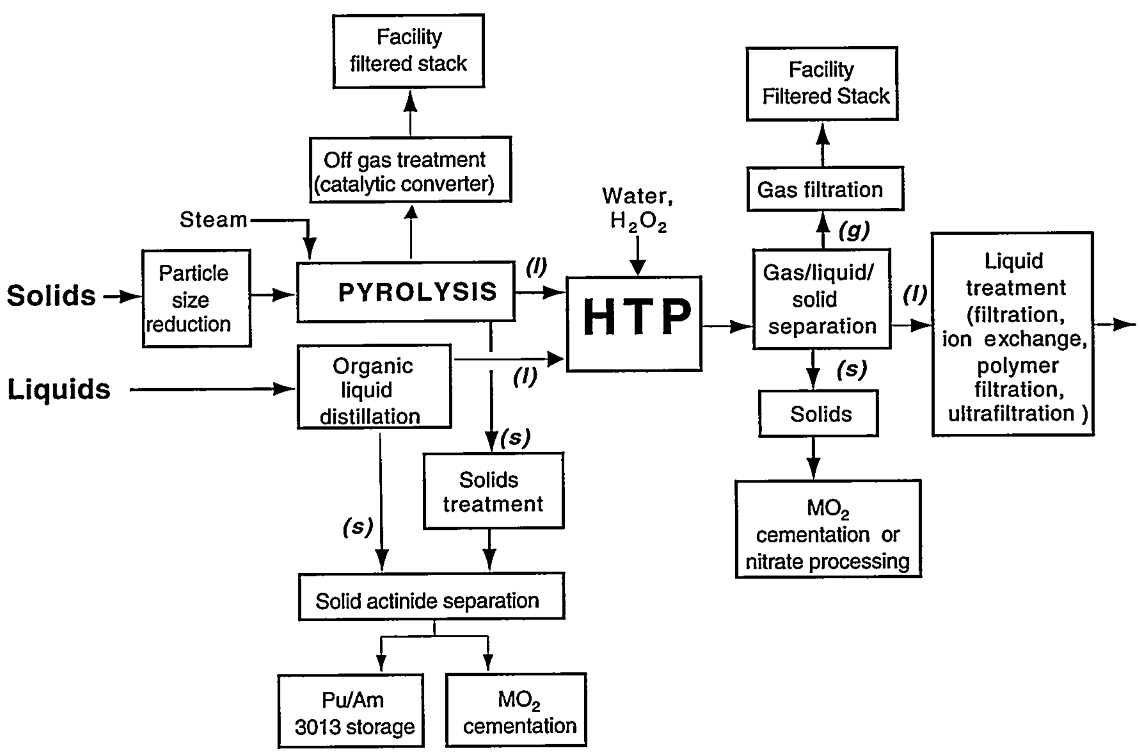
Hydrothermal Processing of Actinide Combustible Material

Waste Type	Vol (ml)	Organic Composition	Actinide	Effluent pH	TOC Results (mg/L) Influent	TOC Results (mg/L) Effluent[b]	Organic Destruction
Vacuum pump oil	20	Petroleum distillate; $(CH_2)_n$	Plutonium (Pu)	4.0	21400	<2	99.99%
Analytical solution	310	Ultima Gold-AB DIPN[c]	Pu (IV) nitrate or $AmCl_3$	4.0 - 2.5	22500	5	99.99%
Solvent extraction solution	200	70% dodecane 10% decanol 20% TBP[d]	Pu (IV) nitrate or $AmCl_3$	2.5	16100	6.6	99.95%
Dense liquid	100	Bromobenzene	$AmCl_3$	2.0	18000	7	99.96%
Cl heteroatoms	100	1.5 M trichloroacetic acid	Pu	1.0	1800	<1	99.94%
Pyrolysis liquids	100	Partially pyrolyzed polypropylene rags	Pu	4.0	18000	4.5	99.98%
Water soluble polymer	50	Methylpolyvinyl-pyridine $(C_7H_3N)_9(NO_3)$	Pu	2.5	5100	4	99.92%
Reillex HPQ ion exchange resin	20	$(C_8H_{10}N)^+n$ 36:62:2 ratio of resin/water/CMC	Pu	1.0	23000	6	99.97%

[a]Reaction conditions of 46.2 MPa, 60 s, 550°C
[b]Average results from multiple tests
[c]Diisopropyl naphthalene
[d]Tributyl phosphate



Schematic of the laboratory-scale process unit. Organic material is pressurized and mixed with pressurized hydrogen peroxide (30 wt %). The reaction mixture is fed into a high-temperature, high-pressure reactor and allowed to react for ~1 minute. At the end of the reactor, the mixture is cooled in a heat exchanger and then depressurized. In the reactor, the organic components of the wastes are oxidized to carbon dioxide by reacting with the supercritical water and oxidant. Heteroatoms such as chlorine, sulfur, and phosphorus are oxidized and converted to acids or salts depending on the pH of the solution. At temperatures above 500°C, reactions are rapid, and conversions of greater than 99% can be achieved in seconds. The reactor is fitted with a titanium liner to protect the pressure vessel from corrosion.



Flow sheet for treatment of plutonium-combustible materials. Items that we investigated in this project include pyrolysis and solids treatment, hydrothermal processing (HTP) and solids separation, and liquid treatment. Solids are liquefied by pyrolysis, then pumped through HTP. The ash from the pyrolysis unit contains most of the inorganics (iron, plutonium, and silica). The organic liquids are pumped through HTP, and any solids that form are removed by filtration.

Detection of Underground Structures, Tunnels, and Objects

96507

Joseph Mack

We are pursuing hard and deeply buried target defeat (HDBTD) at Los Alamos through unified collaboration with the other two DOE nuclear weapons laboratories—Lawrence Livermore National Laboratory and Sandia National Laboratories at Albuquerque. One of the objectives of this collaboration is the final documentation of the deep-target electromagnetic computations and tracking code, DETECT. Other objectives are the evaluation of resonant, microstrip patch antennas as a novel means for detecting metal or nonmetal buried objects such as land mines and the development of related electromagnetic technologies for underground communications.

The focus of our HDBTD project is to address the total systems needs for defeating the target: detection, characterization and monitoring, and neutralization. Sensing buried conductors from near-surface to beyond 30-m depths is possible from the earth's surface. Evidence suggests that electromagnetic surveys might be performed by using distant antennas or selected airborne platforms. Establishing depth and resolution limits for these methods remains.

We are documenting our wave-propagation assessment computer code, which can now model a variety of transmitter configurations, receivers, and long compact targets. We have completed the final report on resonant-antenna land mine detection, and the instrument is undergoing testing. We developed a data-processing algorithm that enables detection with reduced height-above-soil sensitivity. In addition, we completed further successful tests of our underground communications system.

Environmental Technology Analysis Using Complex System Simulation Techniques

97507

R. Wayne Hardie

We are investigating and applying a fundamentally new approach to environmental management policy analysis and assessment. In particular, using advanced simulation science techniques for analyzing complex systems, we are developing an environmental technology evaluation tool for use by environmental management policymakers. The model is being designed to evaluate issues such as how to spend the budget to achieve the best set of prevention/remediation options over the next decade, given the various constraints and considering new treatment and mitigation technologies. This model will specifically combine existing processing and transport models for the Los Alamos Plutonium Facility at Technical Area 55 (TA-55). Then a packaging module will be added. The resulting model will be reviewed for application to other DOE sites.

This year we combined the two primary modules (the model of Los Alamos TA-55 and the model of Waste Isolation Pilot Plant [WIPP] transport and storage) into one application—Envirosim. Now we can model the simulated waste being generated by TA-55 operations and being fed to storage, packaging, and transport simulation entities. The three simulation scenarios we ran demonstrated the usefulness of Envirosim as a policy analysis tool for use in planning WIPP operations. Since April, extensive changes to Envirosim allow the application to compile and run on most systems that support a C++ compiler that is American National Standards Institute compliant.

We also implemented a graphical user interface using Interactive Data Language (IDL), which allows the analyst to easily view simulation results. Although IDL is not necessarily the graphics interface that will be selected for the production version of Envirosim, it does provide some powerful data manipulation capabilities, and it runs on a variety of platforms.

Targetry Development for the Production of Research Radioisotopes

97523

Eugene Peterson

The primary objective of this project was to define the targetry requirements for a new 100-MeV proton irradiation facility at the Los Alamos Neutron Science Center (LANSCE) to produce medical radioisotopes. We have been evaluating various target concepts to produce the most probable isotopes and designing a target system and shielding to implement the best concept.

We made significant progress this year, including the creation of a conceptual design for a credible target assembly and shielding system for the irradiation facility. Our design was reviewed by a panel of experts brought to Los Alamos from around North America. We also visited TRIUMF (Tri-University Meson Facility) in Vancouver, British Columbia, and NAC (National Accelerator Centre) in Cape Town, South Africa, to view operating irradiation and targetry facilities with characteristics similar to those envisioned for the LANSCE facility. We presented the experts at these facilities with the conceptual design and asked them to review its viability.

After making detailed shield transmission and activation calculations demonstrating the viability of the conceptual design, we used the results to obtain congressional funding for designing and constructing the new facility, beginning in November of 1998.

Instrumentation and Diagnostics

Covert Facilities Detection

98519

William Priedhorsky

Our research project explores new approaches to covert facility detection, which is essential to proliferation defeat. The project is composed of three elements: the conceptual development of a hybrid system for remote sensing of effluents, application of information from Los Alamos' FORTE satellite to synthetic aperture radar (SAR), and development of systems analysis tools to optimize detection and characterization of underground facilities.

This year we developed a strawman concept of a satellite hybrid system, determined its critical tracking and pointing requirements, and studied its utility for detecting effluents from chemical weapons production facilities. This concept formed the basis of a well-received proposal to the Tri-Service Space Prioritization Board of the Department of Defense. An airborne version of the system has become the top-priority, new initiative for the Nonproliferation and International Security (NIS) R&D Program.

We also obtained and analyzed data from the FORTE satellite, which showed that, after transit through the ionosphere, impulse signals could be reconstructed with nanosecond resolution. This type of resolution is a requisite for low-frequency, SAR image formation, which can be applied to foliage penetration for detection of hidden facilities.

As most of the covert facilities will be located underground, we developed a novel method to assess the usefulness of existing and anticipated sensors against essential elements of information for underground facilities. We expect this effort to continue as part of the Tri-Lab Hard and Deeply Buried Target Defeat Program.

Real-Time Personal Neutron Dosimeter

98528

William Casson

It is desirable to develop a real-time, electronic, neutron personal dosimeter for monitoring neutron doses to laboratory personnel. The technology and associated benefits have been demonstrated with the current generation of gamma and beta electronic dosimeters throughout the nuclear industry. The development of neutron systems has been hampered by a lack of demand and the technological challenges associated with neutron dose measurements. Los Alamos National Laboratory is unusual in the industry in that the neutron dose represents as much as 75% of the total dose equivalent reported annually to the DOE. The primary objectives of this project are to development a small, sensitive neutron detector that can fill the energy gap from thermal to approximately 100 keV and another detector to extend beyond 100 keV.

We have modified a thermal detector by designing and incorporating a moderator/filter over the detector. This has resulted in a dose equivalent response up to the 50- to 100-keV range (the detector response remains flat in this energy region, but the dose conversion factors increase dramatically). Work conducted on a high-energy detector with an initial 2-MeV threshold resulted in extending the range down in energy an unknown amount but still much higher than 100 keV (probably just below 1 MeV). Proof-of-concept has been established with the current configuration, which is awaiting prototype development and monoenergetic energy validation.

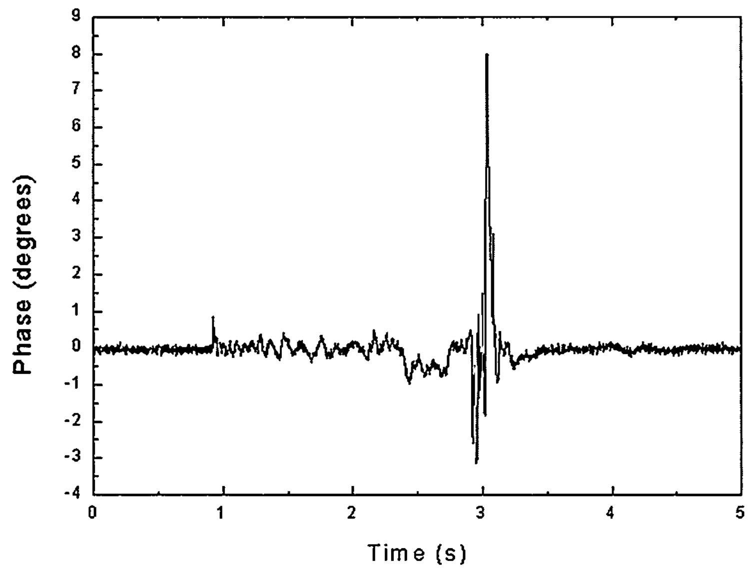
The Ultrasonic Detection of Alpha-Emitting Surface Contamination

98512

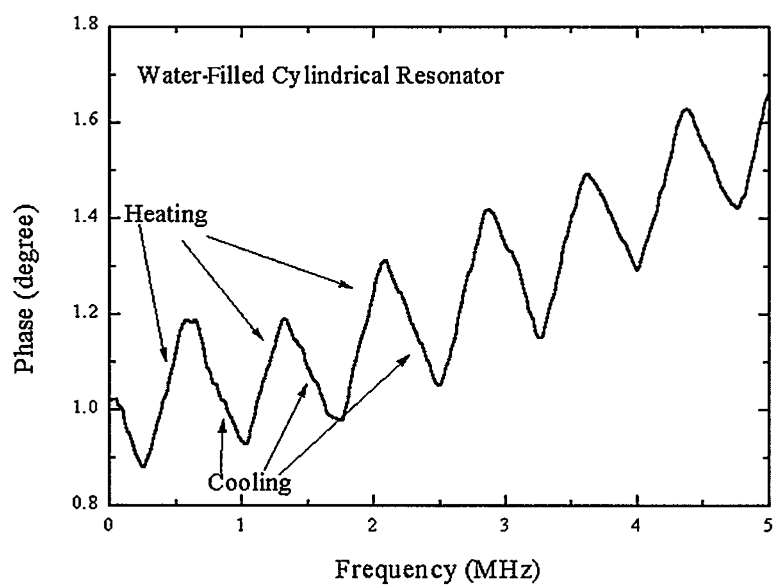
Dipen Sinha

We sought to determine whether ultrasonics can detect the presence of alpha particles in depositions on glove-box walls from the opposite side of the wall. The technique is based on the high sensitivity of a metal plate when it is driven into its thickness-mode resonance. The Q-factor of the resonance, as high as 1,000, amplifies all disturbances on the plate surface or in the interior. In the case of alpha emissions, if the alpha particles enter a small distance into the plate and thermalize, the effect may be observable from the temporal variation in the resonance characteristics of the plate.

We carried out tests in water-filled cylindrical resonators and thickness-mode plate resonators and found that the proposed technique is sensitive enough to detect even a single grain of salt (approximately 10 μ in diameter), as well as 10-mK changes in temperature (see accompanying figures). However, the measurement with thin, disk-shaped, electrodeposited americium and plutonium sources did not show any discernible results. One possible reason was that the Q-factor of the alpha-source disk was only 35. Another reason was that the disintegration rate of the source (5,000–30,000 disintegrations per minute) was too high to allow detection of individual impact: the effect was smeared out because of the finite mass of the metal disk. The technique we developed is highly sensitive and merits further exploration with better-designed experiments.



Effect of dropping a single grain of salt inside a water-filled cylindrical resonator. The spike near 1 s occurs when the grain first impacts the water surface. The large disturbance near 3 s occurs when the grain reaches the central region of the cylinder where the electrodes are.



Light modulation causes alternate heating and cooling of water in a cylindrical resonator. The upward drift is caused by the cumulative heating of the water.

High-Throughput Flow Analysis for Pathogen Identification

98526

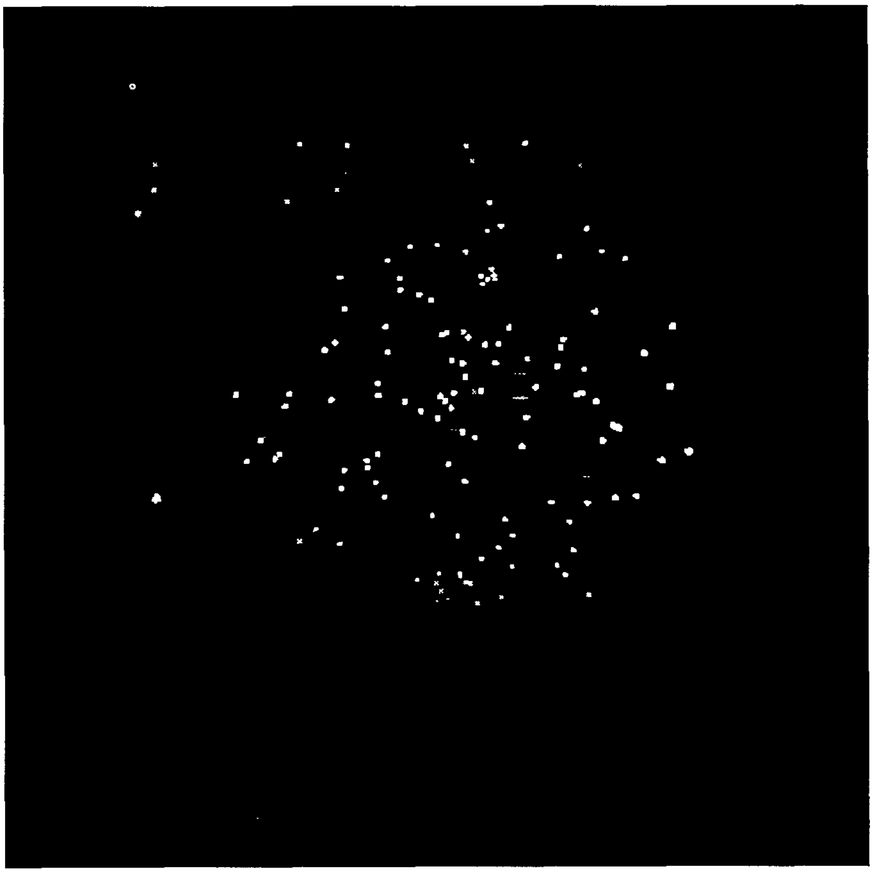
Richard Keller

We have developed a flow cytometry approach to DNA fragment sizing that is approximately one thousand times faster and one million times more sensitive than conventional gel electrophoresis. DNA fragments are stained with a fluorescent intercalating dye and passed individually through an ultrasensitive flow cytometer constructed at Los Alamos. The intensity of the fluorescence from each fragment is a measure of the fragment length. A histogram of the fragment lengths is a DNA fingerprint.

This technology is being used to characterize clones of phage artificial chromosomes and bacteria artificial chromosomes, for the construction of DNA libraries and for bacteria fingerprinting in the identification of species and strain of potentially hazardous bacteria. We received a 1997 R&D 100 Award for this invention. For the applications described above, it is desirable to increase the sample throughput. We used the LDRD award to develop and characterize an innovative approach to increasing sample throughput.

In our present approach to DNA fragment sizing, samples are introduced into a 250-µm, square-bore sample cell from a 40-µm-i.d. capillary. The capillary is surrounded by a sheath stream to focus the analyte stream to a diameter of ~10 µm. A laser beam, focused to a diameter of ~20 µm, crosses the sample stream ~50 µm downstream from the capillary outlet. Fluorescence from individual, fluorescently stained DNA fragments is collected with a high numerical aperture microscope objective and focused onto a silicon avalanche photodiode. Fragments are processed serially at a rate of ~100 fragments/s.

We are exploring the possibility of increasing the throughput of DNA fragment sizing by introducing fragments into the entire cross section of the 250-µm, square-bore sample cell crossed by a sheet of laser light. The sample cell is viewed end-on, and fluorescence images are recorded on a charge-coupled-device (CCD) camera. The intensity of each spot on the CCD readout is proportional to a fragment size. Hundreds of fragments are in the laser beam at the same time (see the accompanying figure). In a proof-of-principle experiment, fluorescence from approximately 100 microspheres was recorded in 30 ms, an increase in throughput over serial analysis of ~13.



Simultaneous imaging of hundreds of fluorescent microspheres in flow.

Development of a Real-Time Beryllium Air Monitor Utilizing Microwave-Induced Plasma Spectroscopy

98531

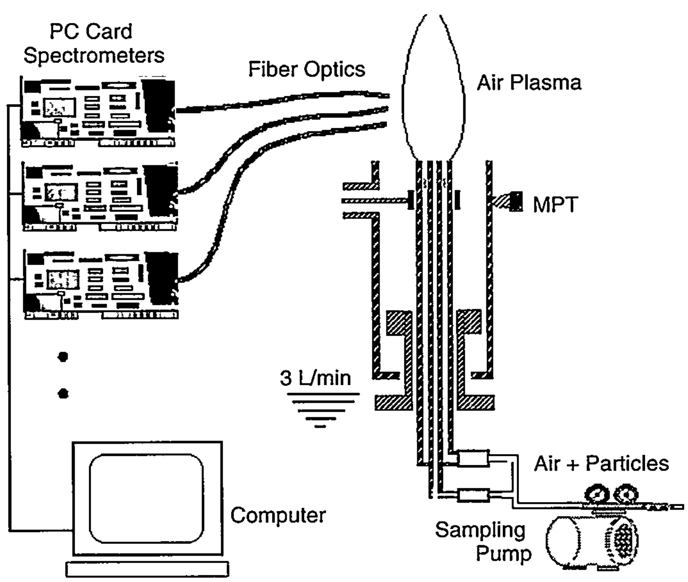
Stephen Abeln

Because assessment of beryllium exposure is important for site workers and the current techniques for assessment have significant restrictions, there is an urgent need to develop a high-sensitivity beryllium-air-particle monitor that is inexpensive and can be used in varying situations. Our goal is to develop an on-line, real-time beryllium monitor, and an associated microwave plasma source to induce it, that can be used in beryllium technology facilities and other beryllium-affected sites to monitor human exposure.

This year we built a bench-top instrument in our laboratory. To build this instrument, we designed and machined a microwave plasma torch, set up a small computer plug-in spectrometer and a monochromater for optical-beam collection and detection, and built a high-efficiency nebulization and desolvation system for sampling and testing (see figure). Instrument characterization has been performed and the experimental parameters have been optimized. We can detect beryllium signals in a 10-ppb sample solution with the small spectrometer, compared to a detection limit of about 1 ppb with our currently used bench-top instrument. This solution detection limit is equal to approximately 0.1 $\mu g/m^3$ in air. We are making further efforts to improve the instrument performance and reduce the instrument cost.

Publication

Duan, Y., et al., "A Field Portable Spectrometer for Environmental Hazardous Monitoring and On-Line, Real-Time Analysis" (Federation of Analytical Chemistry and Spectroscopy Society Conference, Austin, TX, October 11–15, 1998).



Schematic for the layout of the microwave-induced plasmas instrument that shows the computer, small (PC card) spectrometers, fiber optics, air plasma, microwave plasma torch (MPT), and sampling pump.

Noninvasive Characterization of High Explosives: Dielectric Relaxation Analysis of PBX

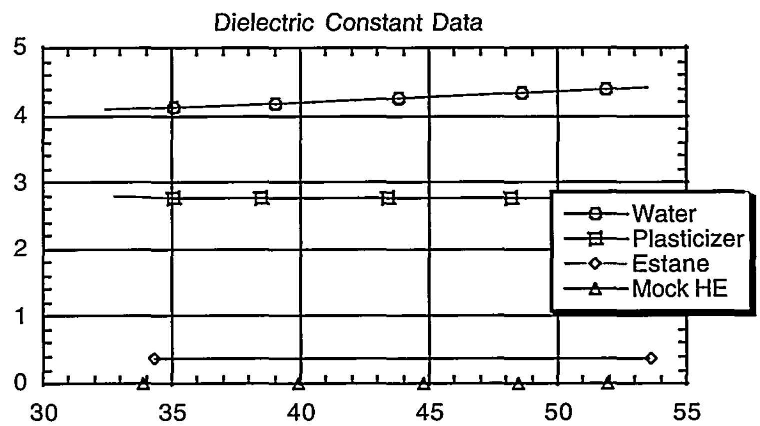
97526

Mark Smith

Our objective is to evaluate the potential of dielectric relaxation spectroscopy for noninvasive determination of the state of high-explosive (HE) material. This ability would allow for a material and/or component evaluation with little or no disassembly of stockpile weapons. The main challenge lies in the sensitivity of the technique to the high-explosive components. Of particular interest is the status of the binder, composed of 50% polyester urethane polymer and 50% plasticizer. This binder only accounts for 5% of the total high-explosive composition.

Specific events targeted by the dielectric analysis include molecular weight change of the polyester urethane, plasticizer content, moisture adsorption, and the remaining stabilizer concentration. Our main efforts have been to increase the sensitivity of the technique by increasing the frequency span of measurement, improving contact between the dielectric probe and material surface, adjusting the surface area of the probe to minimize loss, and minimizing dielectric signal interference resulting from the presence of material defects.

The accompanying figure demonstrates the best response attainable for water, the plasticizer, the polyester urethane (Estane), and a sugar mock of high explosive. While the individual responses are well separated, with the highest signal for water (as expected), the Estane and mock explosive are very similar. The sensitivity of the combination of material is rapidly decreased as a result of the inclusion of solids (sugar in the mock sample, HMX [high melting explosive] in actual systems). Since this behavior was observed over the entire measurable frequency range, the utility of dielectric relaxation spectroscopy for characterizing high explosives is not feasible without extensive developments in the technology.



Dielectric constant measured as a function of temperature at 200 kHz. Mock high explosive (sugar used as the surrogate HMX), the polyester urethane binder component (Estane), plasticizer, and water response are included. The response of the liquid components, plasticizer and water, is much higher than for the polymer and mock material.

A Precision Thickness Probe for On-Machine Gauging Using Laser Ultrasound

98529

Thomas C. Hale

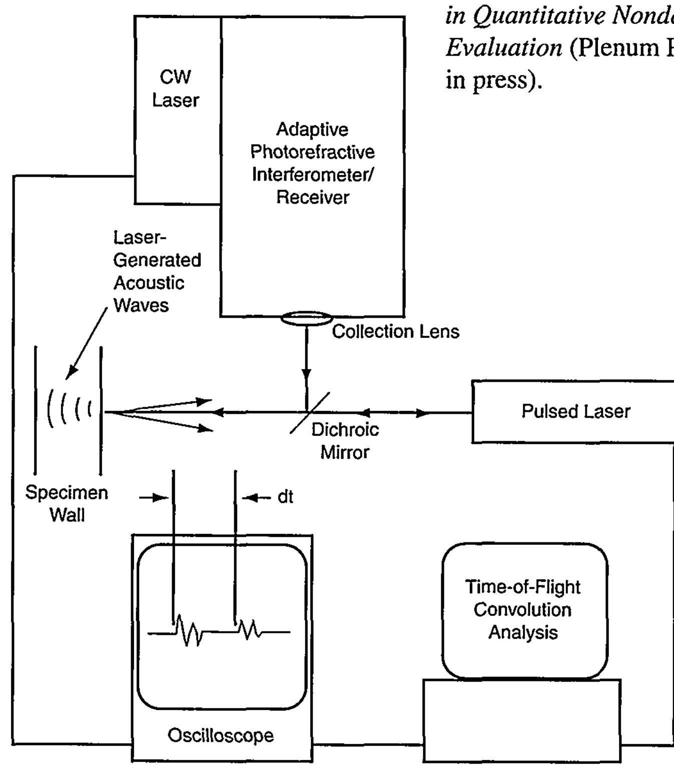
The objective of this project is to investigate the use of noncontact laser ultrasonic methods to measure wall thickness in metallic components. This year we successfully demonstrated the capability of this technology to measure laser-generated acoustic-pulse time-of-flight down to 1-ns resolution (standard deviation between successive measurements). Assuming uniform density and temperature, this resolution translates to a material thickness resolution of less than 2.2 µm (or 0.0001 in.) in plutonium.

The system uses a "ping and listen" sonar-like approach that uses two lasers: one pulsed and one continuous wave (CW). The pulsed laser is used to generate an acoustic impulse at the front surface which propagates into the material. Reaching an interface such as the back surface or a defect, the acoustic impulse is reflected back toward the front surface. The CW laser beam, also incident on the front surface, has its phase perturbed by the arrival of the reflected acoustic impulse. This phase perturbation is converted into a voltage signal by the adaptive photorefractive receiver/ interferometer for easy analysis by an oscilloscope. The time between generating and receiving the acoustic information or time-of-flight is a direct measure of material thickness.

Although several improvements to the system have been identified, the initial adjustment would involve increasing the optical-reception power while reducing the damaging optical pulsed energy. At present, we have to "ping" the specimen very rigorously (with the pulsed laser) because we are not able to "listen" (with the CW laser) well enough (see the first figure). Adjusting the equipment would bring the system operation from the ablative regime to the thermoelastic regime, where all nondestructive testing applications are usually implemented. This improvement will be implemented next year on a CRADA project called laser ultrasonics for process monitoring and on-line material characterization. We will be partnering with Lasson Technologies, Inc., of Culver City, California.

We have upgraded the optical receiver and the pulsed laser using capital equipment funding. We are also seeking to upgrade the CW laser from 0.15 to 5.0 W, which should have a significant impact on the implementation of this technology on real weapons components.

The optical receiver is a transduction device based on optical interference in photorefractive GaAs crystals that converts phase perturbations in the coherent CW laser beam into photo-electromotive force (emf) currents. These currents are then analyzed to yield information concerning the acoustic behavior of the specimen. The second figure shows examples of contact and noncontact detection of ultrasonic acoustic impulse reflections in an Inconel plate.

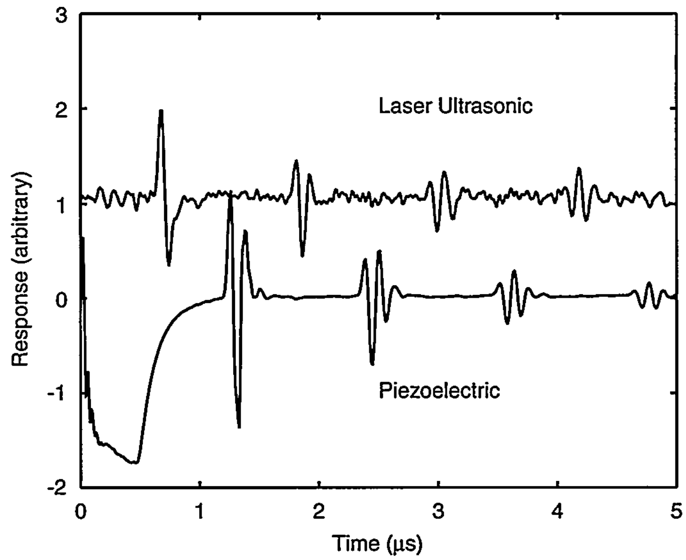


Experimental setup for high-resolution thickness measurement employing laser ultrasonics with photo-emf detection. The adaptive photo-emf interferometer converts acousto-optic phase perturbations into intensity perturbations that are then received and analyzed by the oscilloscope.

Publications

Hale, T., and M. Klein, "High Resolution Time-of-Flight Measurements Employing Laser Ultrasonics with Photo-EMF Detection," D.O. Thompson and D.E. Chimenti, Eds., in *Review of Progress in Quantitative Nondestructive Evaluation* (Plenum Press, New York, in press).

Detection of ultrasonic acoustic impulse reflections through a 3/16-in. Inconel plate. The top trace shows the ultrasonic wave detected by the noncontact photo-emf-based interferometer, and the bottom trace shows the wave detected by the contact piezoelectrical transducer (PZT). The PZT-detected acoustic wave is 180 degrees out of phase with the optically-detected wave because the optical probe is on the opposite side of the plate from the PZT. Both instruments detect the acoustic impulse reflections, which are a measure of material thickness.



Novel Instrumentation for Real-Time Measurement of Drug Concentrations in Tissue

98561

Irving Bigio

The primary goal of this project is to demonstrate the technology and develop instrumentation for noninvasive, real-time in vivo measurement of the concentrations of various drugs in tissue, using a fiber-optic detection method whose probe geometry can be amenable to easy access to different organ sites in test animals.

An extension of the elastic-scattering spectroscopy techniques, previously developed for the detection of cancer, is applied to the quantitative measurement of absorbing compounds in tissue. Such instrumentation can be valuable to the pharmaceutical industry, permitting acquisition of otherwise difficult-to-obtain data on tissue drug concentrations in real laboratory research circumstances, especially for generating data on site-specific pharmacokinetics. The technology could shortcut major portions of the animal-testing process in drug development by permitting continuous time-dependent measurements on various organ sites of a single animal.

This year we have made measurements of the tissue concentrations of two chemotherapy agents in vivo on an animal tumor model. The method is based on elastic-scattering spectroscopy with a fiber-optic-probe spectroscopic system. The results of the work include measurements of the time course of the drug concentrations in the tumors, as well as a comparison of the optical measurements to high-performance liquid chromatography (HPLC) analysis of the drug concentrations at the time of sacrifice. System sensitivity proved sufficient for the stated goals. We found that the optical measurements correlate linearly with HPLC measurements, but further research will be required to improve absolute quantitation.

Thanks to these preliminary results, our funding proposal to the National Institutes of Health has received a good score, and we have recently received notification of forthcoming funding for continuation of this research.

Publications

Mourant, Judith R., et al., "Noninvasive Measurement of Chemotherapy Drug Concentrations in Tissue: Preliminary Demonstrations of In Vivo Measurements" (submitted to *Phys. Med. Biol.*).

Geoscience, Space Science, and Astrophysics

Geoscience Methods Applied to the Development of High-Performance Concrete

98554

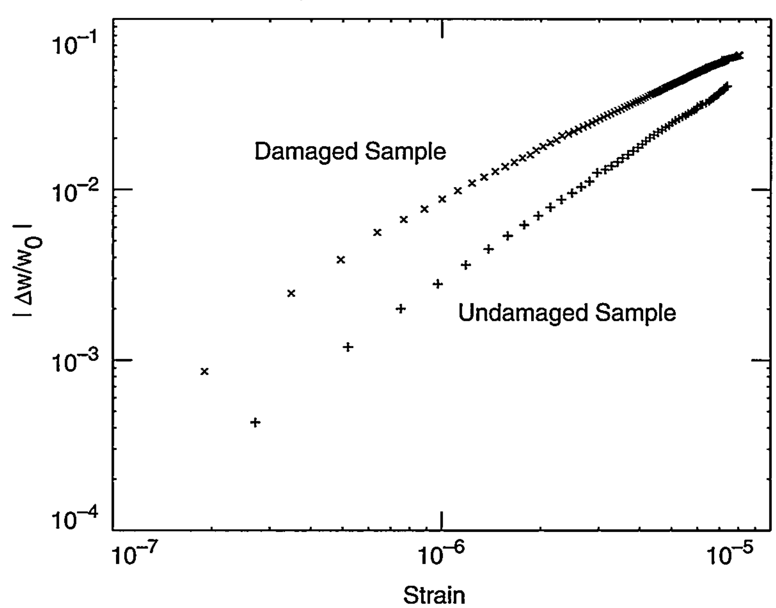
James Carey

The strength and durability of concrete are controlled by structural and chemical factors that correspond closely to geochemical and geophysical properties used in the analysis of rocks. The purpose of this project is to build upon existing geoanalysis and geomodeling capabilities to predict the lifetime performance of concrete structures as a function of mix design and interactions with environmental media. This predictive model will have applications in DOE for infrastructure design, waste repository design, waste disposal, nuclear reactor design, and oil, gas, and geothermal drilling technologies.

During this fiscal year we focused on a common degradation problem in concrete: alkali-silica reaction (ASR). ASR is a geochemical reaction between aggregate and the alkali hydroxide that concentrates in the fluids migrating within the pores and fractures in concrete. We conducted research in three areas. In studies of the geochemical mechanisms of ASR, we investigated the conditions leading to condensation of the swelling gel produced by ASR. Our research suggests a more complex mechanism than previously described in which a reduction in pH must occur. We also employed nonlinear geophysical methods, including resonant ultrasound spectroscopy, to analyze ASR in situ. The studies show that fracture properties of concrete with ASR are distinct (see accompanying figure) and may provide a basis for nondestructive evaluation of concrete damage. In the third area of research, we investigated a possible microbial role in the generation of ASR. We used carbon and oxygen isotope methods and biological stains to identify potential signatures of biological activity in concrete.

Publications

Guthrie, G.D. Jr., and J.W. Carey, "A Simple Environmentally Friendly and Chemically Specific Method for the Identification and Evaluation of Alkali-Silica Reaction," *Cem. Concr. J.* 27, 1407 (1997).



The frequency shift of ASR-damaged concrete versus intact concrete as a function of the induced strain. The difference in slope and position of the curves demonstrates the potential of nonlinear resonant ultrasound spectroscopy for nondestructive evaluation.

Space Sciences

98515

David McComas

We are promoting scientific excellence in space science and exploration at Los Alamos. To do this, we focused on four broad areas that have the greatest potential for success and synergism with current Laboratory capabilities: space physics and astrophysics, planetary exploration, space power/propulsion and materials, and biological sciences. We made substantial progress this year in four areas highlighted below.

The most outstanding accomplishment was in evaluation of laser-induced breakdown spectroscopy (LIBS) for use in potential planetary exploration. This LDRD experiment was accommodated as a component in the larger effort on remote analyses by mass spectrometry and emission spectroscopy. The LIBS technique was successfully tested on a Mars simulant soil in a simulated Mars atmosphere, and it was verified that the technique is indeed highly applicable to extraterrestrial exploratory applications. This work was so successful that NASA awarded a follow-on, 3-year, $1.1M grant to develop a LIBS instrument for potential Mars rover use. A similar LDRD-funded evaluation of laser-induced mass spectrometry (LIMS) is in progress but is not as far advanced, although we have successfully constructed a system with two vacuum chambers connected by a long ion conduit. Final evaluation of the LIMS technique as a planetary exploration tool will follow.

In space physics and astrophysics, we translated the measurement requirements for both generic solar wind and magnetospheric mission instruments into specific instrument geometries and pixel resolutions, allowing for rapid, detailed responses in many space-science applications. We also developed, using special, thin-window, solid-state detectors, a conceptual design for an advanced miniature plasma spectrometer (AMPS) instrument that can measure ion energy, charge, and mass. The design should enable identification of hydrogen, helium, and oxygen for magnetospheric plasmas and identification of these three, plus possibly silicon and iron, in the solar wind.

In space power/propulsion and materials, we completed the initial development of a gas-core nuclear rocket design code, which couples hydrodynamics, neutron transport, and radiation transport in three dimensions (3D); uses an unstructured grid; and is written in fully portable Fortran 90 for parallel and massively parallel platforms. To ensure that the hydrodynamic algorithms are accurate, we compared the code with a nozzle experiment and a cold-flow scaled gas-core experiment. The agreement with both experiments is very good, and we now can perform full-scale, 3D gas-core nuclear rocket design calculations. In another project, we developed detailed cost and schedule estimates for two types of Heatpipe Power Systems (HPS). These estimates provide a convincing case that an HPS could be built in less than 5 years for under $100M.

In biological sciences, a Mars radiation study revealed that the radiation doses to which astronauts may be subjected may be much higher (a factor of 2) than previously predicted. If left unshielded, the astronauts could be exposed to their total lifetime dose from the planet's surface during a currently envisioned 3-year mission. The result of this study could have a tremendous impact on NASA mission planning.

Publications

Berte, M., and B. Capell, "Balance of Plant Options for the Heatpipe Bimodal System," in *Space Technology and Application International Forum—1998,* M.S. El-Genk, Ed. (Springer-Verlag, New York, 1998), p. 1200.

Bish, D.L., et al., "CHEMIN: A Miniaturized CCD-Based Instrument for Simultaneous X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) Analysis," *47th Annual Denver X-Ray Conference, Book of Abstracts* (International Centre for Diffraction Data, Newtown Square, PA, 1998), p. 55.

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Coupling of Nonlinear Acoustics during Material Fatigue and Heterogeneous Chemical Interactions: Exploration and Development of Cross-Disciplinary Research Collaborations

98541

Paul Johnson

Basic geoscience research at Los Alamos has strong capabilities, particularly in the areas of materials and chemical and computational sciences. This project couples investigation of nonlinear acoustics behavior in geologic materials with detailed spectroscopic investigation of chemical interactions that take place within the cracks and pores of geologic material. We are using spectroscopic tools, as well as information on metal speciation and structural environment, as probes to characterize the detailed interactions that occur during nonlinear acoustic stimulation, while simultaneously investigating chemical behavior in dynamic rather than static systems.

Our work this year focused on study of the mechanism of nonlinear response in materials. We reviewed the literature and discovered that the adsorbed fluids in geomaterials and other damaged materials, such as cracked plastics and metals, may be the primary cause of nonlinear response. Applied force microscopy experiments illustrate that, when adsorbed fluids are present (as they always are under ambient conditions), mesoscale hysteretic effects take place at grain contacts and within cracks. The hysteretic effects are due to the strong Van Der Waals forces associated with the adsorbed fluids.

The implications of this discovery are enormous, both scientifically and economically. We may have finally discovered the nonlinear mechanism in rocks. Furthermore, this same mechanism may explain why damaged materials exhibit the same behavior.

Our studies, which were aimed at nondestructive evaluation, may have inadvertently received a boost from our work on the nonlinear response in materials. If the response mechanism is known, we can base our prediction of the nonlinear response of damaged material on a physical description, something we do not presently have.

Toward an Umbilical Drilling System—Developing a Seismic Technique for Measurement While Drilling

98557

Michael Fehler

A capability to take measurements during drilling that would provide information about rock formations ahead of the drill bit would be useful for steering the drill bit toward a target and for anticipating and planning future drilling operations. Numerous methods for seismic measurement while drilling have been proposed and tested. However, this technology is still at an early stage of development. The overall goal of this project is to extend the state of the art using the expertise at Los Alamos National Laboratory in small-hole seismic tool development and in seismic data analysis. Such development will be applicable to both DOE and civilian missions.

The first step is to develop a seismic-monitoring system to monitor the noise generated while drilling. Results from a micro-borehole project last year suggest that a hydraulic-hammer component of a rotary-percussion system may focus acoustic energy forward, a good effect for our approach. We are performing small-scale tests to characterize the acoustic signal from this type of borehole analysis to gain crucial information we need to design and build a seismometer package capable of providing the required measurements.

Earlier this year we built a system capable of measuring the seismic noise generated by drill bits. The system can be deployed in shallow boreholes located near drilling operations and can detect vibrations within the earth induced by drilling. We have collected several sets of data during shallow drilling operations, and we are currently analyzing the signals to determine their characteristics as part of our initial effort to design a seismic "measurement-while-drilling" system that will be deployed within the hole being drilled. We also hope to use our understanding of the seismic signals generated by drilling to develop signal-processing schemes for analyzing the signals to determine earth structure ahead of a drill bit.

Nuclear and Particle Physics

Determining Quark Masses

98544

Tanmoy Bhattacharya

The quark masses are fundamental parameters of the standard model of particle physics. They are essential ingredients in the phenomenological predictions of the model and thus necessary in uncovering possible new physics from the experimental data. An example of the predictions is the CP violation parameter ε'/ε (where CP stands for the symmetry under simultaneous charge-conjugation and parity transformations). The quarks, however, are not asymptotic states, so their masses cannot be measured directly in experiments. The three theoretical methods usually employed to calculate them are chiral perturbation theory, sum-rule analyses, and lattice quantum chromodynamics (QCD). The first of these can provide estimates of the ratios of masses, but not their absolute values. Therefore, our objective is to focus on the sum-rule analyses and lattice QCD approaches to determining the quark masses.

This year we studied the uncertainties in the existing sum-rule analyses. We showed that these arose from three sources: the techniques used to fix the normalization of the hadronic spectral function in the existing sum-rule calculations had large systematic errors; assumptions about the location of the onset of duality influenced the extracted quark masses significantly; and the perturbative errors had often been underestimated in the literature. A combination of these uncertainties could lower the previous estimates of quark masses by a factor of 2. In addition, we derived lower bounds on quark masses using the positivity of the spectral function and showed that they too suffer from uncertainty about the scale at which the theory becomes perturbative.

We also analyzed the world data on quark masses determined using lattice techniques. We showed that the major uncertainties came from three sources: the renormalization constants needed to match the lattice to the continuum theory, the extrapolation to the continuum limit, and the inclusion of dynamical quarks in the calculations. To better control the first two of these systematic uncertainties, we plan to calculate the quark masses using improved techniques. Toward this end, we have developed the codes required to determine the renormalization constants and carry out the analysis of quark masses using the SGI Origin 2000 computers (built by Silicon Graphics) at the Advanced Computing Laboratory.

Publications

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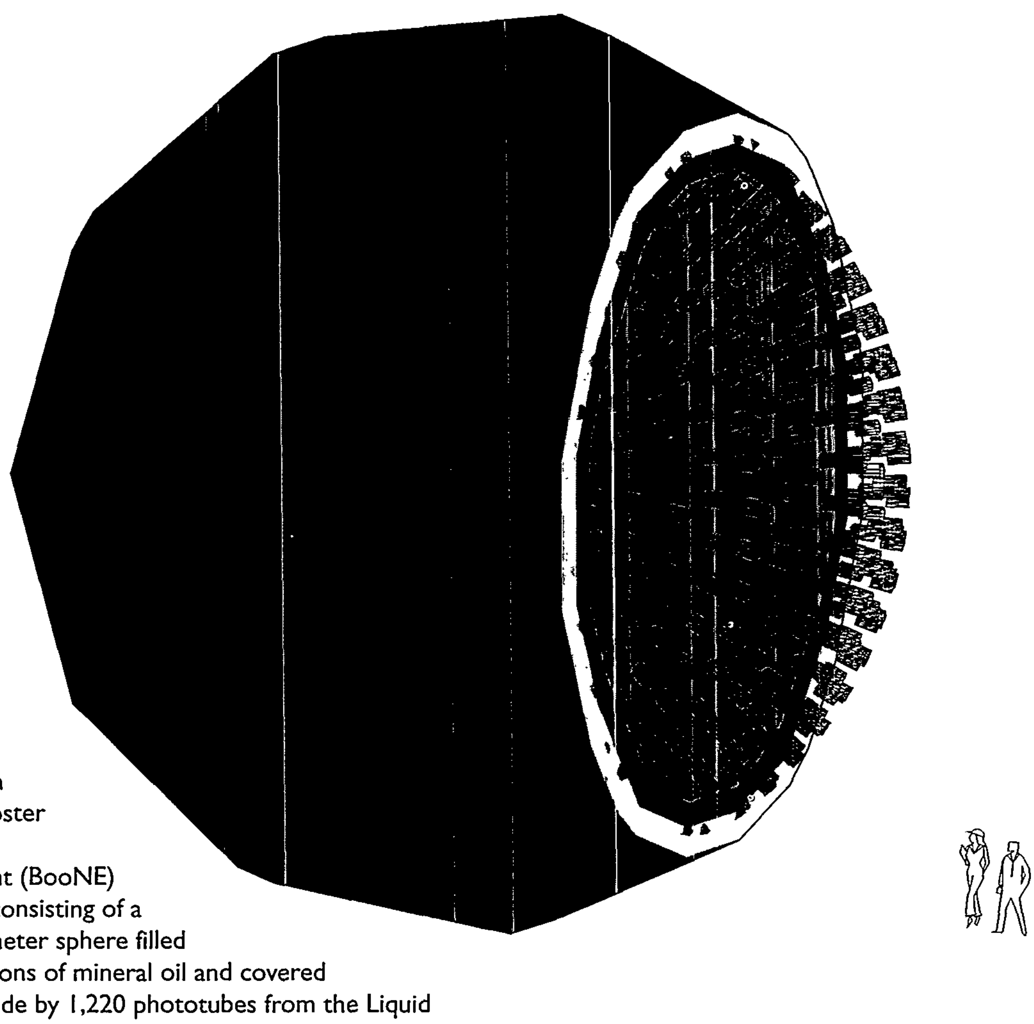
Exploratory Engineering Studies and Detector Development for the BooNE Experiment at Fermilab

98543

William Louis

The Booster Neutrino Experiment (BooNE) at Fermilab is designed to be a definitive test of the evidence for neutrino oscillations obtained by the Liquid Scintillator Neutrino Detector (LSND) experiment at Los Alamos. The BooNE detector will consist of a 12-m-diameter sphere filled with 770 tons of mineral oil, covered on the inside by 1,220 phototubes from the LSND experiment. The detector will be located 500 m downstream of a new neutrino beamline that is fed by the 8-GeV proton booster at Fermilab. Our objective was to perform exploratory engineering studies and development of the detector.

We first carried out extensive Monte Carlo simulations of the beam and detector and determined that a spherical geometry was best for maximizing the fiducial volume of the detector and for reconstructing neutrino events. We also demonstrated that a 10% photocathode coverage was sufficient for event reconstruction and for separating electron events (the oscillation signature) from background muon and neutral-pion events. Next, we made measurements of the Cerenkov and scintillation light produced by electrons in pure mineral oil and showed that this light was adequate for the particle identification. Finally, as shown in the accompanying figure, we performed a detailed design of the excavation and construction of the spherical tank. Based on this exploratory design work, we submitted a full proposal to Fermilab in January 1998 and obtained stage-one approval in May 1998.



The design of the Booster Neutrino Experiment (BooNE) detector, consisting of a 12-m-diameter sphere filled with 770 tons of mineral oil and covered on the inside by 1,220 phototubes from the Liquid Scintillator Neutrino Detector experiment.

Development of an Isotope Separator for Studies of Radioactive Species

97530

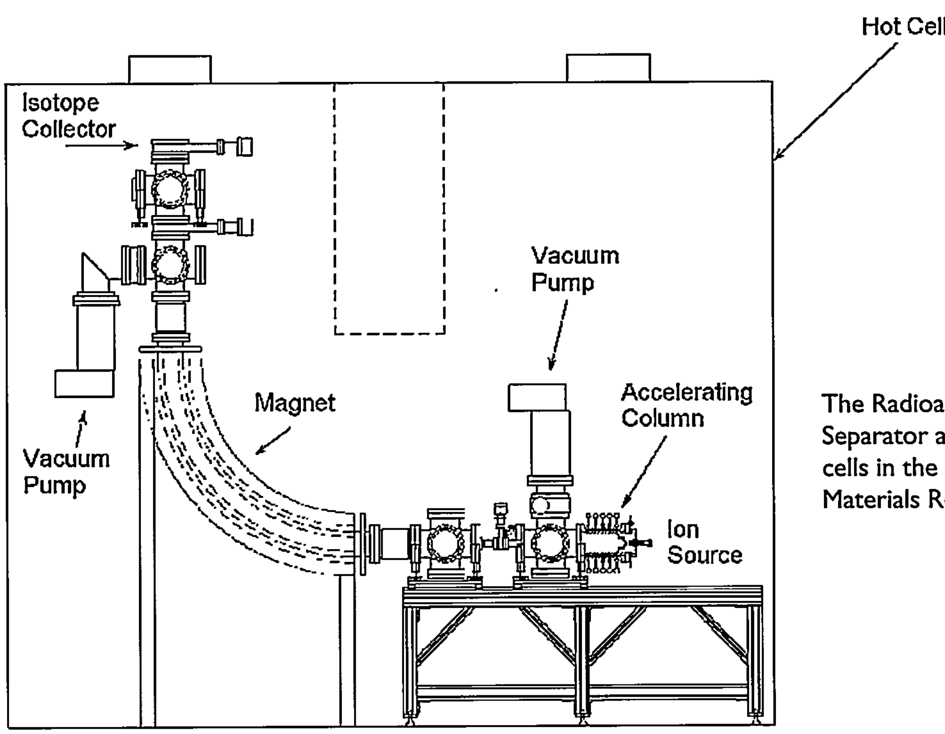
Jerry Wilhelmy

The objective of this project is to produce a Radioactive Sample Isotope Separator (RSIS) that will have the dual purpose of supporting Science-Based Stockpile Stewardship efforts and the Medical Isotope Production Program. The goal is to use the RSIS capability to obtain material for neutron-induced reaction studies on radioactive targets for stockpile stewardship, basic research, and medical applications.

The isotopes of interest for capture-reaction studies to support interpretation of weapons diagnostics include: thulium-168, -170, -171; lutetium-173, -174m,g; gold-195; and iridium-192. Also, neutron-induced, proton-out reactions on zirconium-88 and yttrium-88 are of interest. The target requirements for irradiation at the Weapons Neutron Research Facility/Los Alamos Neutron Science Center are on the order of 10^{18} atoms. A dedicated radioactive isotope separator can also be used to produce targets of interest to DOE's Energy Research programs, including the study of reactions of interest for astrophysics and nuclear structure. Smaller amounts of material might be of interest to DOE's Basic Energy Sciences researchers for implantation studies.

For environmental and safety reasons, it is important to have a dedicated device that can be located in a very controlled environment. The radioactive species will be obtained from nuclear reactor irradiations and from spallation reactions on beam-stop irradiated targets used in medical isotope production. Chemical processing in hot-cell facilities will select the desired elements, but since the production process can make a wide spectrum of products, it will be necessary to perform isotopic separation to obtain the high-purity specific isotopes. This can be done with the high-current RSIS (see accompanying figure) that is confined in a dedicated hot cell in the Chemistry and Materials Research (CMR) Building at Los Alamos.

We achieved substantial milestones this past year. (1) Construction of RSIS was completed; (2) an improved thermal ionization source was fabricated and tested; (3) control and safety systems were designed, constructed, and tested; (4) stable beams of strontium were successfully extracted for commissioning tests; (5) the design goal of 50% collection efficiency for strontium was demonstrated; (6) an Unresolved Safety Question Determination for installation and operation of RSIS in the CMR Building was completed and approved; and (7) the RSIS was dismantled from its test site and installed in the CMR Building. Full operation of RSIS with radioactive species separation is expected next year.



The Radioactive Species Isotope Separator as deployed in hot cells in the Chemistry and Materials Research Building.

Quantum Transport of Nonlinear Coherent Structures

98545

Salman Habib

In recent years significant progress has occurred in nonequilibrium quantum field theory. Interest has grown considerably in the study of dissipative effects in specifically quantum mechanical systems, driven largely by the impressive progress in the fabrication and control of sensitive microdevices. As a result of recent advances in high-performance computing, quantitative attacks are now possible on a host of outstanding, but until recently quite intractable, problems in quantum transport theories under nonequilibrium conditions. In this project, we are using recently developed techniques in nonequilibrium quantum field theory to study the quantum transport of nonlinear coherent structures such as kinks, domain walls, and vortices.

This year we investigated transport properties of nonlinear coherent structures in several systems. The first area of research was one-dimensional field theories: (1) We investigated the statistical mechanics of kinks in a new theory based on a quasi-exactly-soluble quantum mechanical model. This research provided many important analytical results that will be very useful for benchmarking of numerical simulations. (2) We carried out a study of kink nucleation and dynamics in the Landau-Ginzburg model. To describe this system, we introduced a new model based on a dynamical classification of kink types. A powerful consequence of this model is that it provides a complete physical explanation for the kink statistical mechanics in this system. (3) We studied self-consistent kink solutions for sine-Gordon and Landau-Ginzburg theories by including fluctuations around the mean-field solutions. These studies are helpful in understanding the phonon corrections to kink-kink interactions in these models.

In addition, we initiated studies of vortex dynamics in two-dimensional theories based on Langevin simulations on parallel supercomputers.

Publications

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Developing the Groundwork for a Protein Structure Initiative

98547

Thomas Terwilliger

The human and microbial genome initiatives are proving to be spectacular successes. Inspired by these accomplishments, discussions are now widespread about how best to take advantage of the sequences from the genome projects in order to understand, modify, and even engineer proteins, nucleic acids, and cellular function. There is now a groundswell of interest in a major effort to determine structures of entire "basis sets" of protein structures that could be used for modeling or predicting structures of any protein molecule.

Our purpose is to carry out experiments that will be needed to address the feasibility of such a project. In particular, we are using existing classifications of protein structures to (1) determine the relationship between protein sequence similarity and structural similarity, (2) estimate the number of protein structures required to form a basis set that covers essentially all natural protein structures at sequence similarities of 20% and 50% identity, and (3) assess the feasibility of a production approach to structure determination by applying such an approach to a model protein.

During the past year we began assessing the feasibility of a production approach to structure determination by applying such an approach to a model organism. We chose a thermophile organism (*Pyrobaculum aerophilum*), purified six proteins from the protein, and crystallized and determined structures of two of them. Our feasibility study showed that it is indeed possible to determine structures of proteins rapidly by using a high-throughput strategy. This means that a large-scale protein-structure-determination effort is indeed feasible and timely.

Publications

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Biological Smoke Alarm

98562

Gary Salzman

Our goal was to develop a continuously operating, autonomous sensor to warn of a terrorist or military bioaerosol attack. The sensor, a single-particle aerosol flow cytometer, could in principle discriminate between background aerosol particles and biological aerosol particles (bioaerosols) because of a specific effect that occurs when UV light makes contact with living microorganisms: the light excites fluorescence in the reduced nicotinamide adenine dinucleotide (NADH) found in such organisms. Field tests at Dugway Proving Ground in Utah have demonstrated the principles for the cytometer by achieving single-particle NADH excitation and fluorescence detection with a large, 350-nm pulsed-laser system. Our approach is to find a smaller, lower-cost, continuous-wave light source.

During the year we investigated the use of arrays of 371-nm light-emitting diodes (LEDs) as light sources for the cytometer. The two LEDs we had acquired produced 0.7 mW each in a 15-nm band around 371 nm. Although we then designed an array of eight LEDs, production quantities of the LEDs did not become available until October 1998 after the project funding ended. In the absence of LEDs, we also modified a miniature flow-cytometer platform to accommodate a titanium-sapphire laser, which was purchased with LDRD capital equipment funds. The laser output was specified as 3.5 mW at 376 nm. The laser's initial problems were corrected, and it began operating in October 1998.

Advanced Spectroscopic Diagnostics for Breast Cancer
97505

Irving Bigio

The goals of this project were to further develop the technology that would permit proof of principle and advance the state of the art for a novel diagnostic technique for breast cancer. The first application of elastic-scattering spectroscopy uses an optical-fiber needle probe to provide real-time in vivo "biopsy" diagnosis (to replace surgical lumpectomies that are performed for diagnosis of breast lesions). The second application is for use during open surgery. The elastic-scattering spectroscopy could instantly assist the surgeon in identifying tumor margins.

Following the experience of probe designs executed during 1998, we designed new optical needle probes, as well as probes designed specifically for use during open surgery. We also took measurements in tissue "phantoms" to determine the optical geometry of the zone examined by these new probe designs. Working with medical collaborators, we made new optical measurements on freshly excised breast tissues (subsequent to mastectomies), and these measurements were compared with histopathology results from the same tissue sites. These comparisons provided information about which spectral signatures correlate best with the state of the tissue. The period of support for this project this year was four months (April through August 1998). Toward the end of this period, we received funding from an external granting agency to continue this research project.

Nuclear, Biological, and Chemical (NBC) Threat Reduction
98523

John Phillips

This project focused on three research areas: a phage cocktail for biological threat agent identification, point sensors for chemical and biological agents, and neutron and gamma-ray detection. Here we report on our third research goal of developing simple, sensitive, colorimetric methods for measuring ionizing radiation. Historically, chemical radiation dosimeters have suffered from low sensitivity and instability. New autocatalytic reactions allow a radiation event to initiate a chain that produces 10^7–10^{10} chemical reactions, enough amplification to allow the reaction products to be visualized. We have developed a laboratory prototype of a visible, colorimetric threshold dosimeter that can be set from 17 to 2000 mR. We have also designed prototypes for field testing and have begun testing them in the laboratory.

Publications

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Application of Ring Probe Instrumentation in Large-Scale DNA Sequencing
98560

Joe Gatewood

The DNA containing the genetic code for human beings is approximately three billion base pairs in length and contains all the genes responsible for human development, life, and reproduction. The full potential of DNA sequence information will only be realized when we have the capability of rapidly sequencing individual genomes. Ultimately, DNA from millions of individuals will be sequenced to identify disease genes, locate sites of DNA damage, and provide an absolute basis for identification. The routine application of total genomic sequencing will require a "quantum" leap in sequencing instrumentation. In a separate project, we are developing the foundation for a new, innovative sequencing technology that has the potential of reducing the cost and completion time of sequencing an entire human genome by orders of magnitude.

In this project we are working with industrial partners in the production of small pores for use in our DNA sequencing technology. We are also working with industry in the refinement of nanolithography capabilities for etching the sensor patterns required. This past year we established proof of principle for our approach, an accomplishment that could have a major impact on large-scale DNA sequencing. We have filed a patent disclosure and a patent application is in progress.

Molecular Recognition and Inactivation of Bacterial Toxins

98504

Bruce Lehnert

Biomolecules are capable of recognizing and binding to intracellular targets with exquisite selectivity. The ability of biomolecules to perform such diverse and specific molecular interactions is based on the large number of potential three-dimensional structures they can adopt. In order to exploit these properties, we have initiated an effort directed toward engineering biomolecules to form highly specific molecular interactions with target molecules of interest. This effort will yield unique, low-molecular-weight ligands, which will provide significant therapeutic advantage for abating the actions of bacterial toxins in vivo. Also, because the ligands will have high affinity and selectivity for specific target molecules, we additionally envision that they will serve as the recognition units in molecular sensors and biosensors, including point-sensors for remotely detecting chemical and biological agents. The approach we have chosen to accomplish this is a technique called in vitro evolution of RNA molecules. RNA molecules are particularly well-suited for in vitro evolution combinatorial methodologies because they provide a template for their own duplication.

In the last year we have created a library of starting molecules containing a high degree of molecular diversity (1×10^{14} random nucleic acid molecules). This initial library was then used to select for individual nucleic acid molecules that are capable of binding a recombinant version of *Bacillus anthracis* protective antigen (PA). We are currently characterizing the library of RNA molecules selected against PA and are continuing to use this library to select for RNA molecules capable of binding to other proteins.

A Computational Study of Blunt-Body Trauma to the Head

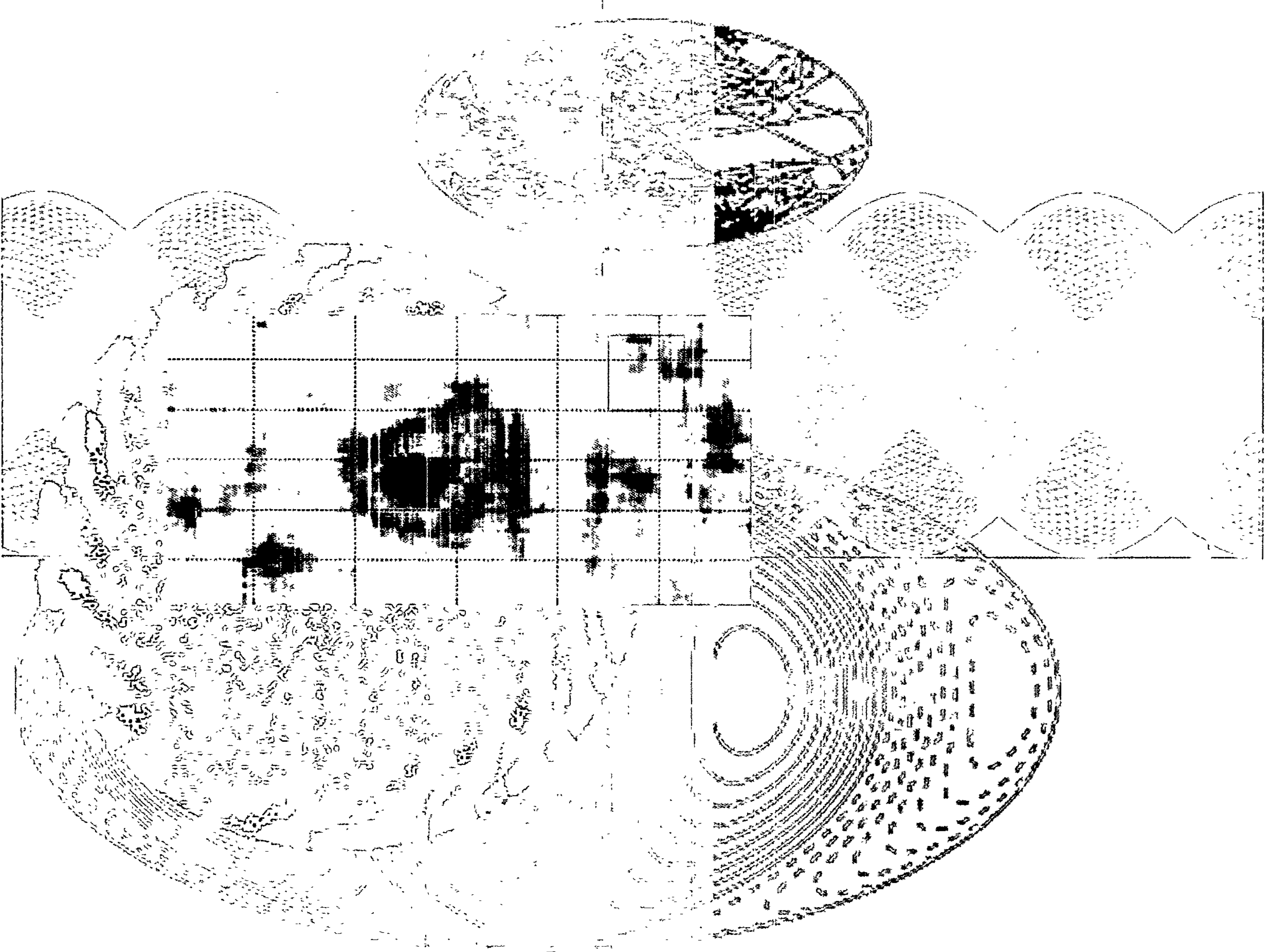
98565

William Wray

In this project we are using a detailed finite element model of the human head to study the mechanics of blunt-body trauma to the head.

In particular, we have used the model to simulate a series of impacts to the head from various angles. We analyzed certain engineering parameters, such as tensile and shear stresses, pressure, deformation, velocity, and acceleration. The extremes (maximum or minimum) of those parameters at any given time can be located within the brain. Furthermore, it is possible to connect the locations of extremes of those parameters at different times to generate so-called four-dimensional paths. Those paths will enable us to tell when and where certain parameters reach their extremes. By comparing these results with clinical findings of head injuries, we can help researchers and medical professionals make fair predictions about which parameters are most likely responsible for particular types of head injuries.

We based the model on the Visible Human Dataset provided by the National Library of Medicine. The dataset has approximately 60,000 nodes (computational points) and provides an average spatial resolution of 4 mm. The model includes the outer layer of soft tissue, the skull, and the brain. The skull has three layers, including the outer and inner tables and diploe. The brain is composed of the cerebrum, cerebellum, and brainstem. The cerebrum is subdivided into two hemispheres by the falx, a tough, leatherlike dura partition, and the tentorium separates the cerebrum and cerebellum. Each of these components is modeled individually and may be assigned its own constitutive relations. We use variable mesh densities on the resulting three-dimensional geometry to achieve an optimal balance between the level of detail and the cost of executing transient dynamic analyses on high-performance computers.



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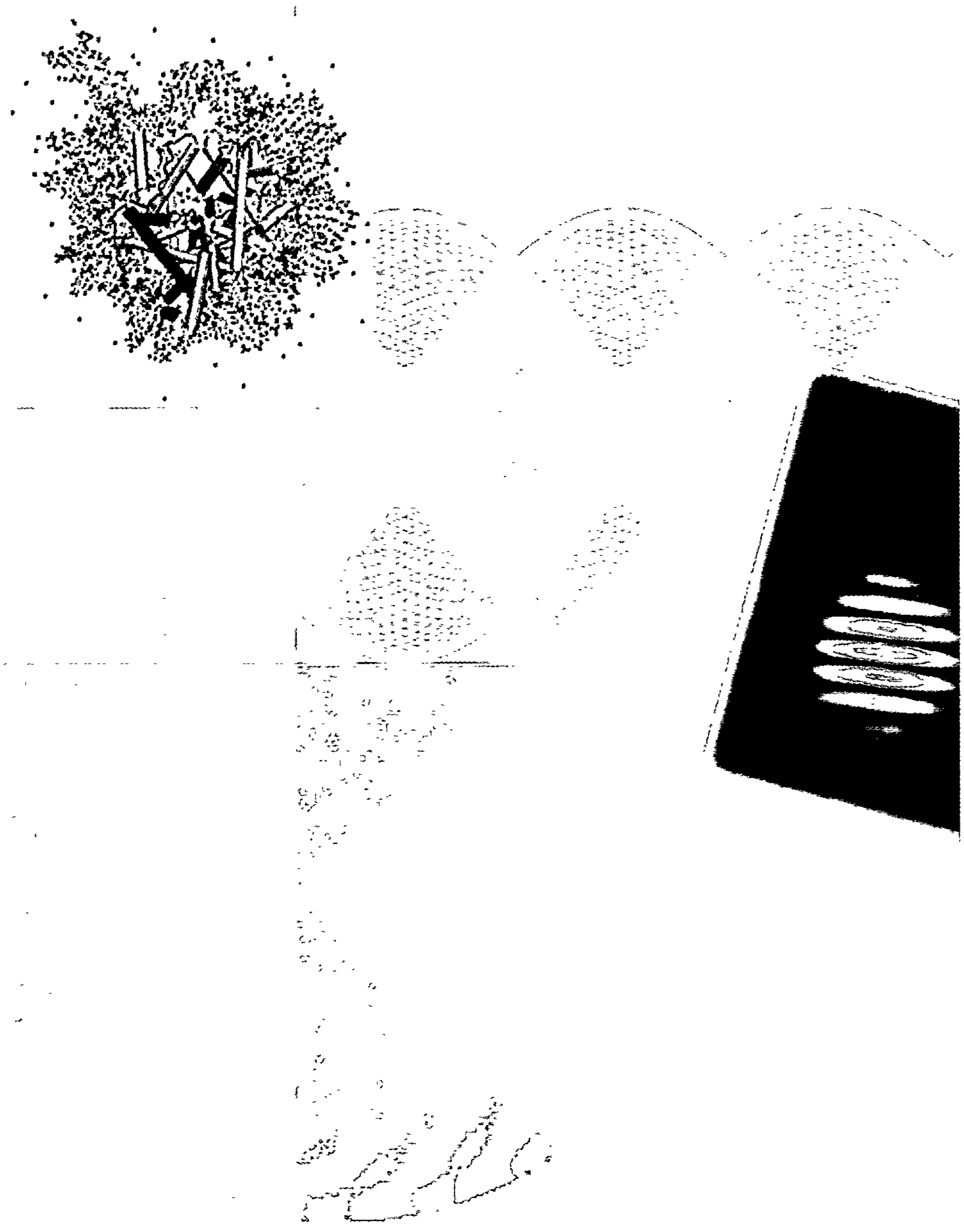
Individual Projects



Materials Science

Interfacial Charge Transport in Organic Electronic Materials: The Key to a Revolutionary New Electronics Technology

96236

Darryl Smith

The aim of this project 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 electro-optic 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 communications. Many design options are available for organic electronic materials and devices both at the molecular and mesoscopic scales. At present, the basic understanding of the properties of these materials and devices is insufficient to allow these design options to be used intelligently. Progress is currently limited by a lack of fundamental understanding. 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.

Two basic classes of questions can be asked about interfaces involving organic electronic materials: (1) the relative position of energy levels in the two constituent materials forming the interface and (2) the cross section for charged excitation transfer across the interface. We are investigating metal/organic interfaces and organic/organic heterojunctions. These are the two essential kinds of interfaces in organic electronic and opto-electronic devices. We are using a combination of experimental and theoretical methods.

This year we investigated the fundamental electronic excitation energies in the prototype conjugated-polymer MEH-PPV. Another of our accomplishments was a combined theoretical/experimental study of the energy relation between charged excitations in a conjugated polymer and the metal at a polymer/metal interface. We investigated charge accumulation at polymer/polymer interfaces in multilayer polymer light-emitting diodes using electro-absorption techniques. We also developed a theoretical model that explains injection currents at polymer/metal interfaces and settled a major puzzle in the field.

Publications

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New Vortex Phases in Irradiated High-Temperature Superconductors

98037

Lev Boulaevskii

We are investigating new structures of the vortex state and the vortex phase diagram in the magnetic field (B)-temperature (T) plane. These structures appear when anisotropic high-temperature superconductors (HTS) are irradiated with energetic heavy ions to produce nanometer-diameter amorphous columnar tracks. In the most anisotropic HTS compounds, vortex lines decompose into point-like "pancake" vortices that are weakly coupled between adjacent layers and that form a liquid of pancakes at high temperatures. Our goals are to establish the relation between the c-axis vortex structure and the Josephson plasma frequency and to obtain the phase diagram for layered HTS with columnar defects in the B-T plane. Such information is important for all HTS applications because vortex structure and dynamics control losses in the superconducting state.

Previously, we obtained resonance frequency as a function of B and T in pancake liquid plasma using the high-temperature expansion. We developed this approach further and derived a general relation connecting plasma frequency with the density correlation function of pancake liquid. We obtained c-axis magnetoresistance and current-voltage measurements in longitudinal magnetic field in single crystals of bismuth-2212 irradiated with heavy ions, which display distinct anomalies near fractional filling of the columnar defects created by the irradiation. The anomalous behavior extends over a limited range of B-T and in many crystals is manifested by a minimum in the resistivity as a function of B. Our measurements show directly that a reduced pancake and phase-slip mobility is associated with columnar defect filling in the vortex liquid state.

From our data, we estimate the average length of pieces of vortex lines confined inside columnar defects as a function of the filling factor. We find that the maximum length of approximately 15 interlayer pancake distances is reached near a filling factor of approximately 0.3. We have performed a microscopic calculation of the dielectric response function in highly anisotropic layered superconductors and obtained the frequency-dependent London penetration length and conductivity in the case of d-wave pairing for currents perpendicular to the layers. We use a BCS (Bardeen-Cooper-Schrieffer) model with coherent interlayer tunneling of electrons and take into account contributions from both superconducting electrons and quasiparticles. We show that quasiparticles change the low-temperature behavior of the penetration length in the intermediate frequency range, where the frequency is smaller than the superconducting order parameter but larger than the inverse quasiparticle scattering time.

Publications

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A Molecular Architectural Approach to Novel Electro-Optical Materials

96274

DeQuan Li

Our objective is to build thin-film materials for nonlinear optical applications by means of supramolecular systems. This year we synthesized novel barbituric acid and melamine derivatives that spontaneously self-assemble into a supramolecular ribbon according to their complementary hydrogen-bonding motif. This supramolecular ribbon stacks into a polar multilayer structure (see the accompanying figure) as verified by the sum-frequency generation ($\omega_1 + \omega_2$) and second harmonic generation (where $\omega_1 = \omega_2$). The second harmonic generation yields a nonlinear susceptibility (d_{33} = 3.2 pm/V) for the self-assembled films, and the sum-frequency generation shows a net polar orientation of the methyl groups in the multilayer structure along the surface normal. X-ray diffraction confirms the layered structure with a periodicity of 41 Å, which corresponds well to the width of the supramolecular ribbons (~40 Å).

Publications

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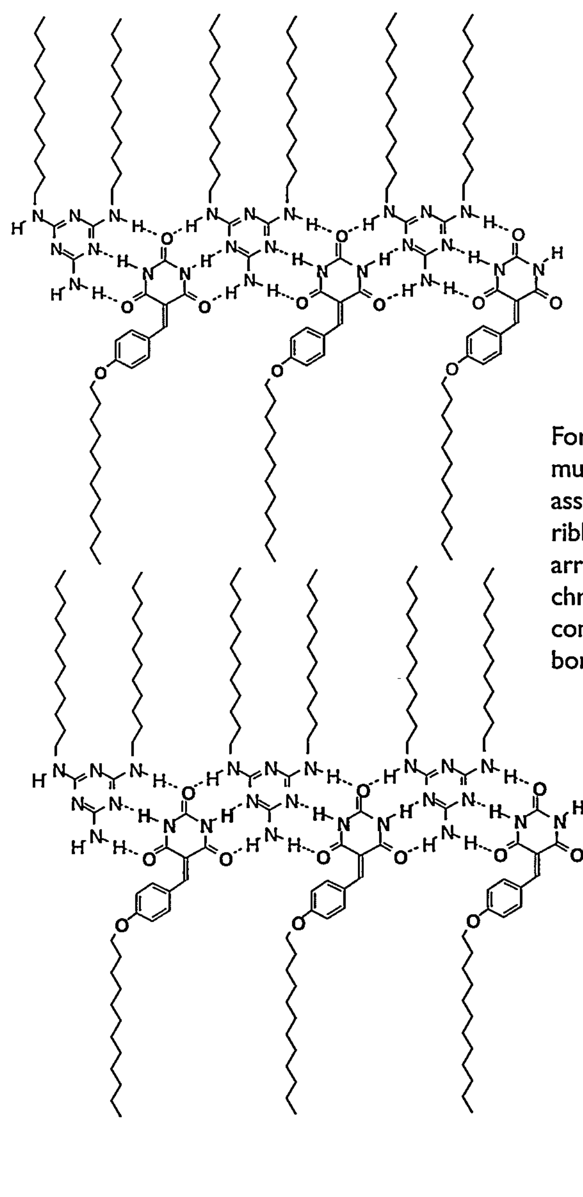

Formation of a polar multilayer system based on assembling supramolecular ribbons, which consist of an array of nonlinear optical chromophores with a complementary hydrogen-bonding motif.

Development of a Fundamental Understanding of Chemical Bonding and Electronic Structure in Spinel Compounds

96257

Kurt Sickafus

We are investigating the nature of chemical bonding, point defects, and electronic structure in compounds with the spinel crystal structure. This crystal structure is common to many ceramic compounds that are used as insulators, superconductors, and magnetoresistors. A better understanding of the spinel crystal structure is needed for more fully exploiting the widely varying physical properties of spinels.

This year we performed x-ray and neutron diffraction studies on nonstoichiometric (alumina-rich) magnesio-alumina spinel crystals ($MgO{\bullet}nAl_2O_3$) to determine the extent of cation disorder and the nature of the cation vacancy distribution; i.e., where charge-compensating cation vacancies reside in the spinel lattice. We used least-squares refinement procedures to determine from the diffraction data the average scattering "power" from the tetrahedral and octahedral cation lattice sites. Cation disorder was calculated assuming models in which the vacancies reside on tetrahedral sites, on octahedral sites, or on both kinds of sites. We found that no degree of cation disorder was consistent with the tetrahedral vacancy model and that vacancies in nonstoichiometric spinel most likely reside on octahedral sites. Cation disorder, specifically the degree of cation inversion, ranged from 15% to 26% in all samples, which is typical for synthetic spinel.

We performed shell-model total-energy calculations to assess point-defect formation and migration energies in magnesio-aluminate spinel. The magnesium Frenkel defect (interstitial vacancy pair) has the lowest formation energy of all the constituents of spinel (4.59 eV/defect), as well as the lowest migration energy (0.34 eV for the magnesium vacancy and 0.91 eV for the Mg^{2+} interstitial). The aluminum Frenkel defect has the highest formation energy (6.37 eV/defect) and the highest migration energy (1.87 eV for the aluminum vacancy and 2.83 eV for the Al^{3+} interstitial).

Publications

Sheldon, R.I., et al., "Cation Disorder and Vacancy Distribution in Non-stoichiometric Magnesium Aluminate Spinel" (submitted to *J. Am. Ceram. Soc.*).

Intrinsic Fine-Scale Structure in Complex Materials: Beyond Global Crystallographic Analysis

97016

Albert Migliori

Our picture of solids is largely based on the model of atoms occupying lattice positions corresponding to the global crystal structure. Structural probes on various scales have consistently demonstrated that this picture is not correct in many classes of complex materials, including perovskites, martensites, and "high-T_C" materials. Local deviations may persist for long periods of time compared to a phonon vibration. These "fine-scale" structure effects are present even in a material that is nominally free of the usual materials defects and microstructure (for example, grains). Recently studied cases demonstrate that fine-scale structure profoundly affects the mechanical (for example, elasticity), electronic, magnetic, and optical properties of many materials.

This project mounts a combined experimental and theoretical attack on this difficult problem, taking advantage of distinguishing Los Alamos capabilities, particularly pair-distribution function (PDF) analysis with pulsed neutrons, resonant ultrasound spectroscopy (RUS), nonlinear analysis, and many-body expertise in strongly correlated electronic materials.

This past year we analyzed and published elastic and magnetic effects of fine-scale structure development near phase transitions in colossal magneto-resistive manganites. We also systematically studied the evolution of fully (elastically) compatible martensitic microstructure in two dimensions; the local sensitivity of microstructure to compositional fluctuations and disorder; and the role of anisotropic, long-range elastic interactions with Ginzburg-Landau dynamics. We completed measurements with RUS and neutron PDF on martensitic nickel-aluminum single crystals, detecting a previously unobserved anomaly and producing results that provide insights into precursor effects in martensites. Finally, we developed new ideas about the stability of plutonium near phase transitions, and motivated the recent RUS measurements of gallium-stabilized plutonium.

Publications

Bussman-Holder, A., and A.R. Bishop, "Competing Length Scales in Anharmonic Lattices: Domains, Stripes and Discommensurations," *Phys. Rev. B* **56**, 5297 (1997).

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Dimitrov, D.A., et al., "Neutron and X-Ray Scattering Signatures of Rotational and Radial Local Distortions in Model Systems with Average Long-Range Order" (to be published in *Phys. Rev. B*).

Kerr, W.C., et al., "Role of Elastic Compatibility in Martensitic Texture Evolution" (to be published in *Phase Transitions*).

Louca, D., et al., "Local Jahn-Teller Distortion in $La_{(1-x)}Sr_xMnO_3$ Observed by Pulsed Neutron Diffraction," *Phys. Rev. B* **56**, 8475 (1997).

Roeder, H., and A.R. Bishop, "Theory of Colossal Magnetoresistance," *Current Opinion in Solid State and Materials Science* **2**, 244 (1997).

Saxena, A., et al., "Hierarchical Structures in Martensitic Materials," in *Fractal Analysis and Modeling of New Materials: New Directions* (World Scientific, Singapore, 1997).

Yu, Z.G., et al., "Charge Localization and Stripes in a Two-Dimensional, Three-Band Peierls-Hubbard Model," *Phys. Rev. B* **57**, 3241 (1998).

Unusual Metal Behavior in Taylor Microwires

98034

John Petrovic

The Taylor microwire technique of melting materials in glass tubes and drawing the composite to very small diameter wires is an elegantly simple method of combining both rapid solidification and deformation in an experimentally convenient manner. This approach can be employed to produce both ultrafine-scale and various nonequilibrium microstructures. The potential properties of these materials are of great scientific interest both because their strength can approach the theoretical strength of solids and because they offer a new combination of properties. A major advantage of this approach is that the ultrafine scale of these materials lends itself to detailed analysis using the techniques of analytical and high-resolution electron microscopy and to detailed atomistic simulations.

The goal of our research is to develop the Taylor microwire approach and employ it for fundamental studies of both ultrafine-scale and nonequilibrium structures in selected metal alloys. Our major objectives are to (1) explore fundamental scale aspects of the deformation and fracture of ultra-high-strength materials, (2) develop an understanding of the synthesis and thermodynamic stability of microwires, and (3) explore new and unique combinations of electrical, magnetic, and mechanical properties produced by the microwire approach.

We studied the microstructure and phase transformations of copper-16 at. % silver fabricated by the Taylor wire technique of drawing molten metals in a glass tube. As the diameter of the wire became smaller and the cooling rate increased, we refined the scale of the microstructure. When the wire diameter was less than 10 μm, we observed nanocrystals of supersaturated solid solution with a 10-nm grain size. When the diameter of the wire was larger than 20 μm, we found proeutectic copper and the copper + silver eutectic. In the region where the copper-silver eutectic was formed, a (111) twin-orientation relationship was found between copper and silver.

We obtained and installed an induction heating unit to heat and draw the Taylor wires. Then we assembled a Taylor wire laboratory to accommodate the research and develop experimental techniques for Taylor wire synthesis.

Publications

Han, K., et al., "Microstructural Aspects of Cu-Ag Produced by the Taylor Wire Method," *Acta Mater.* **46**, 4691 (1998).

Innovative Composites through Reinforcement Morphology Design—A Bone-Shaped Short-Fiber Composite

96232

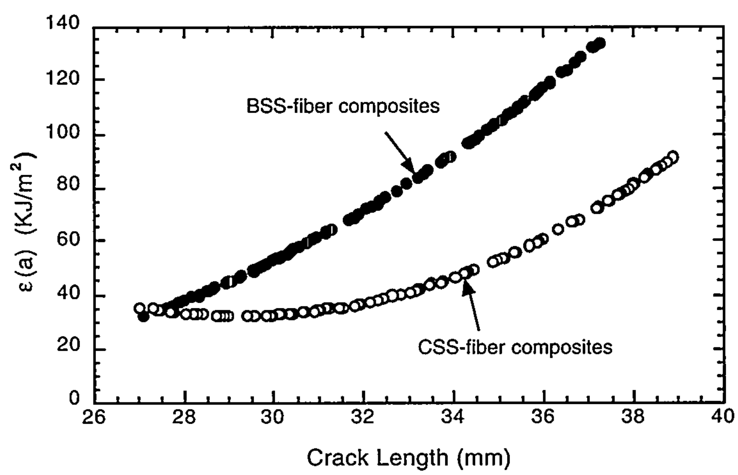
Yuntian Zhu

Our objective is to prove the innovative concept that fiber morphology, rather than interfacial strength, solves the problem of low strength and toughness in short-fiber composites. The problem is apparent in the paradoxical dilemma faced by the designers of conventional-shaped short (CSS) fiber composites: a weak interface results in low strength because of ineffective load transfer, and a strong interface results in low toughness because of stress concentration.

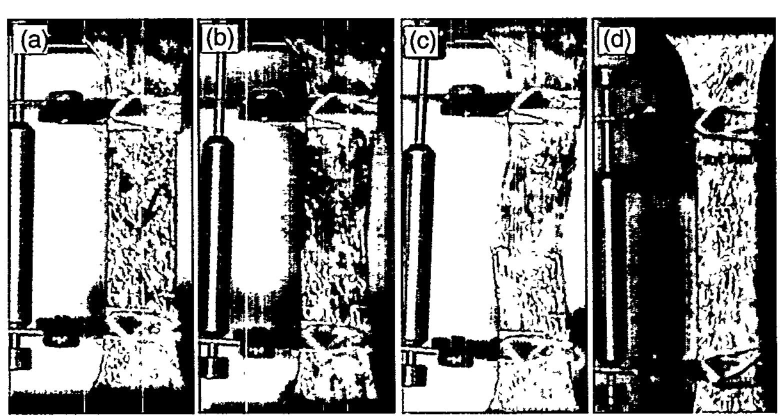
We have fabricated a model polyethylene composite that has a polyester matrix reinforced with bone-shaped short (BSS) fibers. Double-cantilever-beam tests revealed that with a fiber volume of only 5%, the BSS-fiber composites improved approximately 100% in resistance to crack propagation (fracture toughness) over CSS-fiber composites (see the first figure). Tensile tests showed that BSS-fiber composites did not fracture catastrophically as did CSS-fiber composites (see the second figure), attesting to the excellent capability of BSS-fibers in crack bridging.

In addition, we found that a BSS fiber is 150% more effective than a CSS fiber in improving composite strength. Single-fiber pull-out tests revealed an even higher reinforcing potential of BSS fibers. For example, pulling out a BSS fiber from the matrix requires eight times more force and 16 times more energy than pulling out a CSS fiber requires (see the third figure).

We developed a computational model to simulate crack propagation in both BSS- and CSS-fiber composites, accounting for stress concentrations, interface debonding, and fiber pull-out. The model predicts all major features observed during the double-cantilever-beam tests. It will be useful in optimizing BSS-fiber morphology and other material system parameters.



Supplied energy for a crack to propagate by a unit length, e(a), as a function of crack length for double-cantilever-beam tests of both BSS- and CSS-fiber composites with fiber lengths of 3 mm.



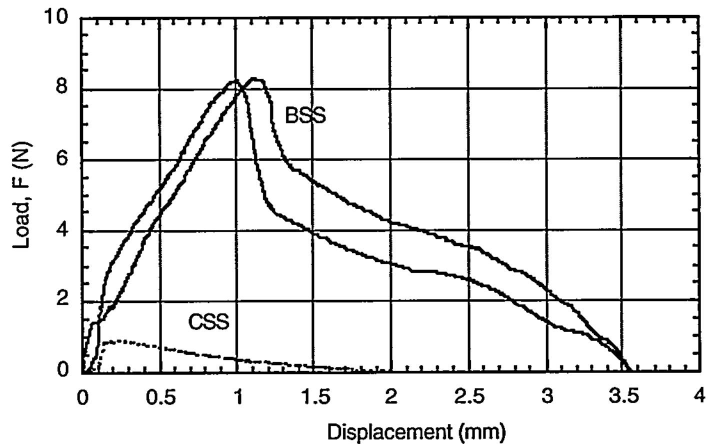
In situ pictures of tensile specimens during tensile testing: (a)–(c) reinforced with CSS fibers and (d) reinforced with BSS fibers. The engineering strain states are (a) 5%, (b) 13%, (c) 20%, and (d) 25%.

Publications

Zhu, Y.T., et al., "A Composite Reinforced with Bone-Shaped Short Fibers" (to be published in *Scr. Mater.*).

Zhu, Y.T., et al., "Influence of Reinforcement Morphology on the Mechanical Properties of Short-Fiber Composites," in *Processings of Metals and Advanced Materials: Modeling, Design and Properties,* B.Q. Li, Ed. (The Minerals, Metals, and Materials Society, Warrendale, PA, 1998), p. 251.

Zhu, Y.T., et al., "Mechanical Properties of Bone-Shaped-Short-Fiber Reinforced Composites" (submitted to *Acta Mater.*).



The load-displacement pull-out curves of both BSS and CSS fibers with an embedded length of 3.5 mm.

Enhanced Cemented Wasteforms for Low-Level Radwaste Immobilization

98045

James Rubin

We are conducting experiments on an innovative transformation concept using a traditional immobilization technique that may significantly reduce the volume of hazardous or radioactive waste requiring transport and long-term storage. The standard practice for the stabilization of radioactive salts and residues is to mix them with cements, which may include additives to enhance immobilization. Many of these wastes do not qualify for underground disposition, however, because they do not meet disposal requirements for free liquids, decay heat, head-space gas analysis, or leachability.

Our treatment method alters the bulk properties of a cemented wasteform by greatly accelerating the natural cement-aging reactions. This method produces a chemically stable form having reduced free liquids, as well as reduced porosity, permeability, and pH. These structural and chemical changes should allow for greater actinide loading, as well as for reduced mobility of the anions, cations, and radionuclides in aboveground and underground repositories. Simultaneously, the treatment process removes a majority of the hydrogenous material from the cement. Our treatment method allows for on-line process monitoring of leachates and can be transported into the field.

Publications

Rubin, J.B., and C.M.V. Taylor, "Partial Replacement of Portland Cement with Fly Ashes and Kiln Dusts Using Supercritical Carbon Dioxide Processing" (Holnam Process Engineers Meeting, Salt Lake City, UT, October 12–16, 1998).

Rubin, J.B., et al., "Enhancement of Stabilized Ash Cements by Supercriticial CO_2 Carbonation" (NATO Advanced Study Workshop, Turning a Problem into a Resource: Remediation and Waste Management at the Sillamäe Site, Estonia (Tallinn, Estonia, October 5–9, 1998).

Rubin, J.B., et al., "Extraction of Hydrogenous Material from Cemented Wasteforms by Supercritical Fluid Carbonation" (American Nuclear Society 1997 Winter Meeting, Albuquerque, NM, November 16–20, 1997).

Rubin, J.B., et al., "Radioactive Wastes Dispersed in Stabilized Ash Cements" (1997 International Ash Utilization Symposium, Lexington, KY, October 20–22, 1997).

Rubin, J.B., et al., "Supercritical CO_2 Carbonation of Standard and Modified Portland Cements" (8th International Symposium on Supercritical Fluid Chromatography, St. Louis, MO, July 12–15, 1998).

Optimum Design of Ultrahigh-Strength Nanolayer Composites

97042

Huijou Harriet Kung

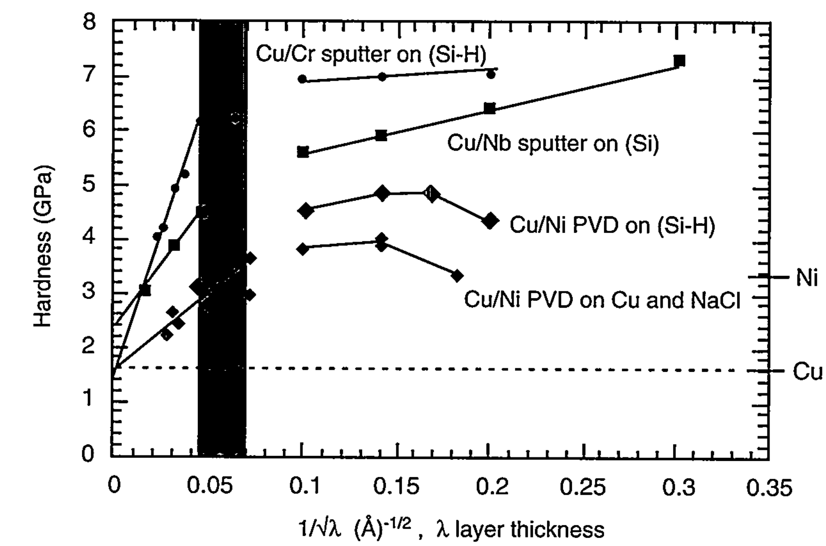
The objective of the project is to understand the relationship between macroscopic mechanical behavior and microscopic strengthening mechanisms in nanocrystalline metallic composites. Mechanisms of plastic deformation and fracture of nanocrystalline materials constrained by interfaces are not clear; when these effects are added to the length scale effect, thin films and multilayers exhibit very large residual stresses.

We have therefore focused our work on finding evidence for a change in the classic dislocation-based plastic behavior of metals as the scale of the structure decreases, and on evaluating and tailoring the residual stresses by ion implantation. Atomic-scale numerical simulations developed for the copper/nickel system are an invaluable tool in studying the local mechanisms responsible for deformation and the role of the interface.

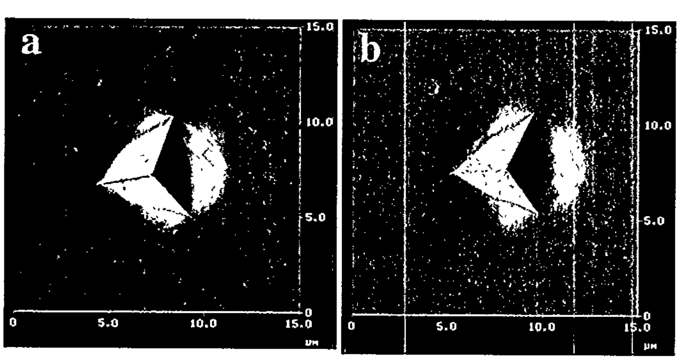
This year we have studied copper/nickel and copper/chromium multilayers. Hardness values were evaluated by nanoindentation techniques (see the first figure). Microstructure was characterized by using high-resolution transmission electron microscopy (TEM), atomic force microscopy (AFM), and x-ray diffraction techniques. We have designed a series of innovative experiments to deform the layers in a controlled manner. These experiments provided the first evidence of the absence of conventional dislocation activity in a layer less than 500 Å thick, despite the large plastic strain it achieved (7%) (see the second figure). Films on substrates also exhibited an unusual fracture behavior.

By using a substrate curvature technique and ion implantation, we have measured and tailored residual stresses in chromium thin films (see the third figure). This finding is of crucial importance for the development of nanolayer composite structures for wear coating applications.

In the theoretical part of our project, we have developed analytical models to produce scaling constitutive laws of the mechanical behavior of nanocrystalline structures. The models will be benchmarked by the experimental results obtained on copper/nickel and copper/chromium multilayers.



Hall-Petch plot of different copper/X (X: chromium, niobium, nickel) multilayers synthesized by physical vapor deposition (PVD) techniques, where λ is the layer thickness (1-μm-thick films). The change in the Hall-Petch slopes indicates a deviation from the conventional Hall-Petch behavior when the scale is reduced to below ~50 nm.



Height derivative AFM images showing the plastic deformation near indents on multilayers with (a) λ = 500 Å and (b) λ = 25 Å. For the 500-Å multilayers, a square rosette pattern made of the trace projection of the {111} planes ([100] surface]) is visible, whereas for λ = 25 Å, a similar upheaval is present but without any strong slip traces along {111} planes. These results provide the first direct evidence of the lack of conventional dislocation activity in ultrathin multilayers.

Publications

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Misra, A., et al., "Electrical Resistivity of Sputtered Cu/Cr Multilayered Thin Films" (to be published in *J. Appl. Phys.*).

Misra, A., et al., "Microstructures and Mechanical Properties of Sputtered Cu/Cr Multilayered Thin Films," *MRS Sym. Proc.* **505**, 583 (1998).

Misra, A., et al., "Residual Stresses and Ion Implantation Effects in Cr Thin Films" (to be published in *Nucl. Instrum. Methods Phys. Res. Sect. B*).

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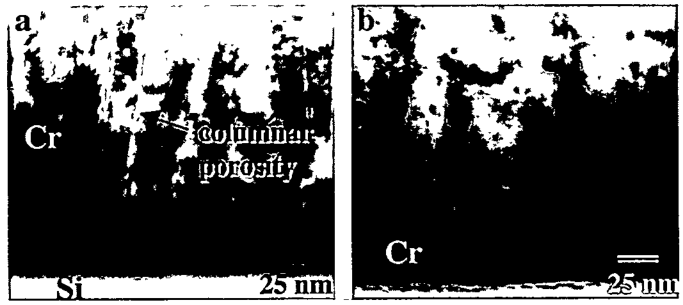


Cross-section TEM micrographs from (a) as-deposited and (b) ion-irradiated chromium film. The film in (a) has a large residual tensile stress and columnar porosity and has an elastic modulus lower than that of bulk. The film in (b) is under a large compressive stress and has density and modulus equal to that of bulk chromium.

Nastasi, M., and S. Fayeulle, "Stress-Related Interdiffusion in DC Sputtered TiN/B-C-N Multilayers" (to be published in *Appl. Phys. Lett.*).

Nastasi, M., et al., "The Influence of Stress on Ion Beam Mixing" (to be published in *Mater. Sci. Eng. A*).

Verdier, M., et al., "Comparison of Different Techniques to Study the Energetics Process of Recovery of Al2.5Mg Alloys" (to be published in *Philos. Mag. A*).

Verdier, M., et al., "Plastic Behavior of Cu/Ni Multilayer" (to be published in *MRS Sym. Proc.*).

Experimental Determination of Statistical Parameters for Improving a Micromechanical Model of Ductile Fracture

97001

Anna Zurek

The objective of this project is to develop and apply experimental techniques to determine quantitative microstructural descriptors of the ductile fracture process. The quantitative descriptors will further the development of a micromechanical model of ductile fracture.

In the methodology currently used for studying dynamic ductile fracture, a wide quantitative gap exists. Although a fine level of microstructural detail is built into numerical micromechanical models of the process kinetics, the overall experimentally quantifiable parameters (such as impact velocity, shock pressure, and spalled surface velocity) are macroscopic. Sample recovery techniques permit posttest microscopy. However, current microscopic techniques allow only 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. Our goal is a micromechanical model with far greater physical accuracy.

This year we concentrated on expanding the quantitative descriptors and appropriate theoretical constructs for the micromechanical model. We improved our image analysis procedures using commercial and custom-written software. We developed procedures to combine data obtained from optical micrographs and optical profilometry. The new descriptors developed in the past year include a multivariate description of void size with respect to position. Also, we performed several incipient-spall tests to provide input samples for the analysis.

Publications

Thissell, W.R., et al., "Quantitative Microstructural Damage Evolution in Copper under Spallation Conditions" (submitted to *Metall. Trans.*).

Thissell, W.R., et al.,"Quantitative Microstructural Damage Evolution in Incipiently Spalled 10100 OFHC Copper" (submitted to *Int. J. Plast.*).

Experimental and Theoretical Investigation of Fracture and Deformation of a Revolutionary High-Temperature Gamma-TiAl Alloy

97011

George Gray III and Zhe Jin

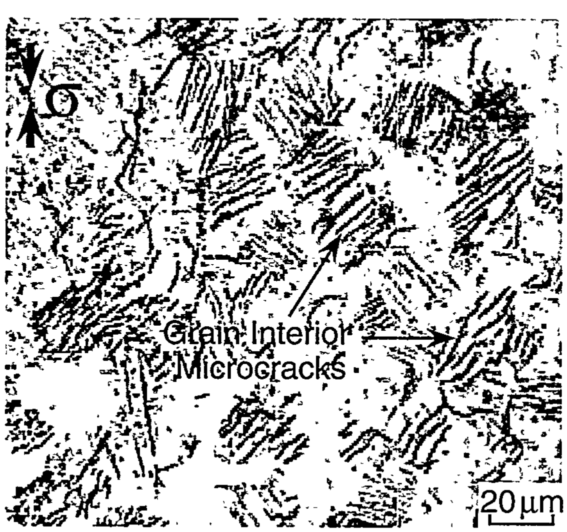
In developing new lightweight, high-strength materials for jet engines, we are investigating the deformation mechanisms that cause low ductility in gamma titanium aluminides (gamma-TiAl), which are alloys with great promise if their fracture characteristics under stress can be overcome. To provide a scientifically driven, physically determined basis for improving ductility, this year we systematically clarified the mechanisms of microcrack nucleation and propagation in both polysynthetically twinned (PST) TiAl crystals and polycrystalline gamma-TiAl alloys.

For the PST-TiAl crystals, we found that under compression, interlamellar microcracks form by a shear mode, and translamellar microcracks form by a tensile mode. We also found that grain interior microcracks in polycrystalline gamma-TiAl alloys (first figure) are formed by a shear displacement along particular crystallographic planes. The mechanical response of polycrystalline gamma-TiAl alloys, as shown in the second and third figures, was also systematically studied over a wide range of temperatures and strain rates. In tests of pure shear loading in particular crystallographic directions of PST-TiAl crystals, we found that both the yield stress and mechanical response of PST-TiAl crystals were different in different crystallographic directions.

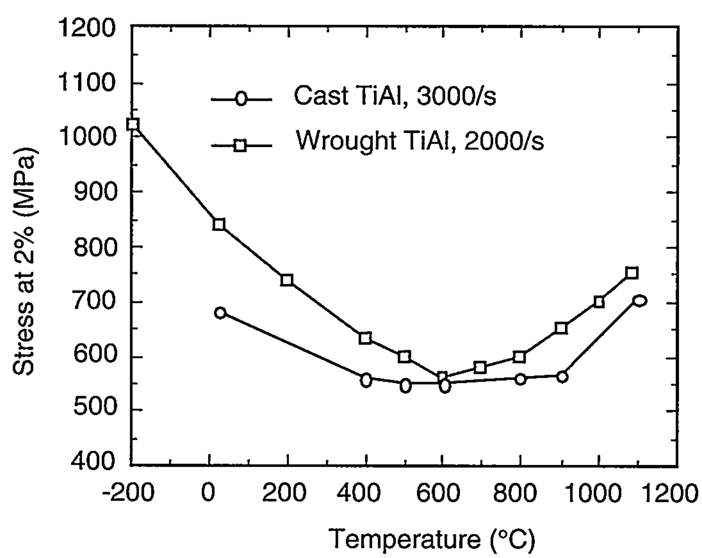
The effects of jogs (shifts in dislocation from one plane to another) and twin boundaries on the mobility of ordinary screw dislocations (spiral ramps of atomic planes) were also studied with a large-scale molecular dynamics code and an embedded-atom method (as seen in the fourth figure). An interstitial-produced jog, involving extra atoms, was found to drag a dislocation motion more significantly than a vacancy-produced jog. The blocking effect on dislocation motion by a twin boundary was found to depend on the orientation between the twin boundary and the dislocations.

Publications

Jin, Z., et al., "Deformation of a $45^\circ\langle\bar{3}21\rangle$ Oriented Polysynthetically Twinned (PST) TiAl Crystal at a High Strain Rate and a High Temperature" (to be published in *Philos. Mag. A*).



Typical microcracks observed in polycrystalline gamma-TiAl alloys under dynamic loading (2000/s) and temperatures below 25°C. The specimen was deformed to 6% true strain.



The change of yield stress as a function of temperature at high strain rate for cast and wrought gamma-TiAl alloys. The yield stress is represented by the 2% flow stress because the stress-strain curves at high strain rates do not accurately reflect true stress. The yield stress of gamma-TiAl alloys shows a significant anomalous temperature dependence at temperatures above 600°C under dynamic loading conditions. This is independent of the alloy grain size.

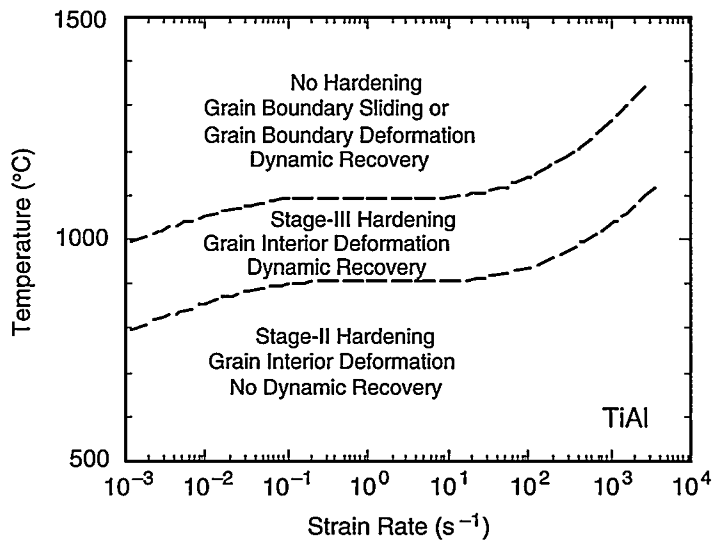
Jin, Z., et al., "Mechanical Behavior of a Fine-Grained Duplex Gamma-TiAl Alloy" (to be published in *Metall. Mater. Trans. A*).

Jin, Z., et al., "Mechanical Twinning in a 45°< $\bar{3}$21> Oriented Polysynthetically Twinned TiAl Crystal at a High Strain Rate and a High Temperature," *Philos. Mag. A* **78**, 239 (1998).

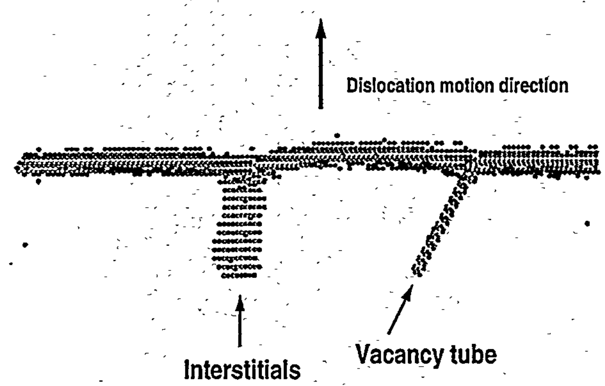
Jin, Z., et al., "Microcracking in Polysynthetically Twinned (PST) TiAl Crystals under Compression" (submitted to *Gamma Titanium Aluminides [Intl. Symp. Gamma TiAl, ISGTA '99]).*

Li, M., and S.J. Zhou, "Investigation of Jog Motion in Gamma-TiAl via Molecular Dynamics" (submitted to *Philos. Mag. Lett.*).

Vaidya, R.U., et al., "High Temperature Oxidation of Ti-48Al-2Nb-2Cr and Ti-25Al-10Nb-3V-1Mo," *Oxid. Met.* **50**, 215 (1998).



The summary of mechanical response of gamma-TiAl alloys at strain rates from 0.001/s to 3000/s. Three temperature regimes are observed in relation to the deformation, work hardening, and dynamic recovery exhibited by the alloys.



Snapshot of the motion of a jogged screw dislocation in a system containing ~1.1 million atoms. Atoms within a specific energy range—from –3.884 eV to –3.139 eV—are displayed to show the dislocation line. Under loading, the dislocation line bows out around the two jogs and creates interstitials and vacancies behind it.

Theoretical and Experimental Investigation on the Low-Temperature Properties of the $NbCr_2$ Laves Phase

96233

Dan J. Thoma

Laves phases are the most abundant yet least utilized intermetallic alloy. Historically, these phases have been avoided because of their perceived brittle nature; however, as a class of materials, Laves phases exhibit outstanding high-temperature properties. The successful use of these intermetallics in various applications is based on optimizing the deformability of the crystal structures. Three approaches have been used to achieve such optimization:
(1) alloying of the monolithic intermetallic, (2) incorporating ductile phases, and (3) tailoring of microstructures through processing pathways.

Significant advances have been made by applying these three defined approaches in the Nb-Cr-Ti system (see figure). Our efforts last year focused on strategies (2) and (3). As a result, we have developed materials with high room-temperature yield strength (1.6 GPa), compressive ductility (16%), and reasonable fracture toughness (>15 MPa–$m^{1/2}$) that still have high strength (1 GPa) at 1000°C. These properties exceed the strengths obtained in almost all metallic systems.

Publications

Chen, K., et al., "Microstructures and Mechanical Properties of Two-Phase Alloys Based on $NbCr_2$" in *High-Temperature Intermetallics (*Materials Research Society, Boston, MA, in press).

Chen, K., et al., "Processing and Properties of Dual Phase Alloys in the Nb-Cr-Ti System," in *Advanced Processing and Materials* (TMS, Warrendale, PA, 1998), p. 1431.

Chu, F., et al., "Phase Stability of C15 MV_2 (M=Zr, Ta, or Hf): An Electronic Structure Investigation," *Philos. Mag. B* **7741**, 121 (1998).

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Kotula, P.G., et al., "Defects in Nb-Cr-Ti C15 Laves Phase Alloys" (Microscopy and Microanalysis, Atlanta, GA, July 1998).

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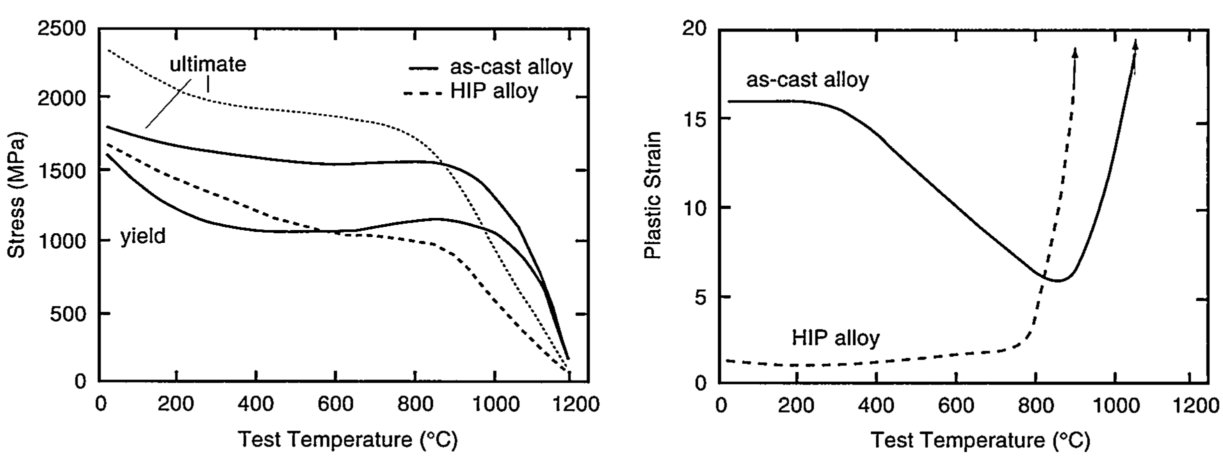
Thoma, D.J., et al., "Elastic and Mechanical Properties of C15 Laves Phase $Nb(Cr,V)_2$," *Mater. Sci. Eng.*, A **240**, 251 (1997).



On the left, the graph illustrates yield and ultimate stress, and on the right, we have shown plastic strain as a function of the compression test temperature of the as-cast (bold lines) and the HIP alloy (dashed lines) in Nb-36Cr-27Ti.

Unconventional Superconductivity and Violation of Time-Reversal Invariance

98036

Roman Movshovich

The goal of this project is to identify the origin of the low-temperature anomaly in the thermal conductivity of a nickel-doped Bi2212 high-temperature superconductor. We suggest that this anomaly (a very sharp drop in thermal conductivity at 200 mK) is a result of the second low-temperature phase transition from the first into the second superconducting state. To investigate whether this identification is appropriate, we performed several experiments.

Measuring the specific heat of several nickel-doped Bi2212 samples in magnetic fields up to 9 T, we found a broad anomaly centered about 1 K, which moves up in temperature with increasing magnetic field. At the low-temperature side of the anomaly, specific heat is linear in temperature. These features are analogous to those of canonical spin-glass systems. The freezing of the impurity spins is one ingredient of the theory of interaction of these spins with a d-wave superconducting order parameter, developed by Alexander Balatsky, a coinvestigator of this project.

We measured the low-temperature thermal conductivity of an insulating analog system to identify the origin of the anomaly, substituting yttrium for calcium in Bi2212 ($Bi_2Sr_2CaCu_2O_8$), which drives the compound from superconducting (Bi2212) into an electrically insulating ground state (Y-Bi2212). We measured the thermal conductivity of Y-Bi2212, which gives the upper bound of the lattice or phonon thermal conductivity. We determined this thermal conductivity to be less than 20% of the total thermal conductivity in nickel-doped Bi2212 at the temperature of the anomaly of 200 mK. Therefore, we can conclude that the observed anomaly is of electronic origin.

Publications

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

Bulk Ferromagnetic Metallic Glasses

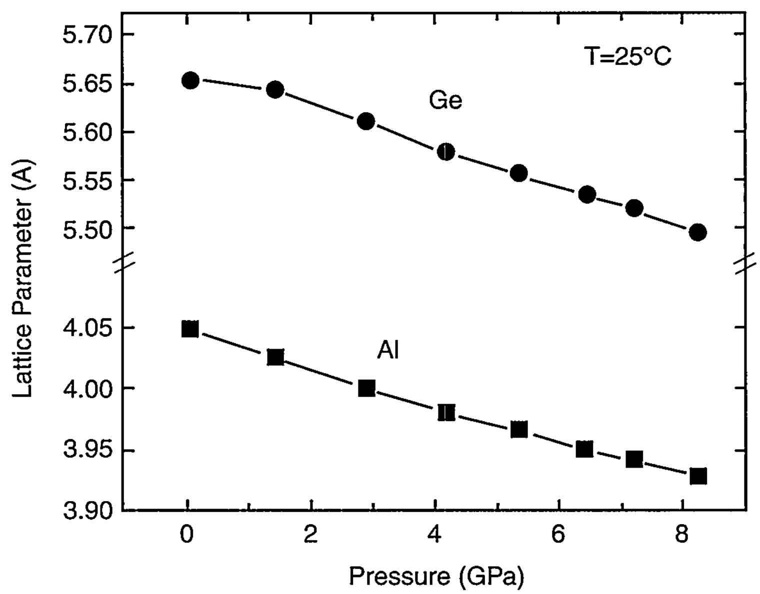
98035

Ricardo Schwarz

This project addresses the science needed to develop a new synthesis route for bulk ferromagnetic metallic glasses. The new techniques are based on pressure-induced solid-state reactions between two or more elemental crystalline phases.

During the review period we analyzed x-ray diffraction data collected during two experiments conducted at Brookhaven National Laboratory (BNL). We prepared fine two-phase mixtures of aluminum and germanium by (a) mechanically alloying mixtures of elemental powders and pressing these powders in a die and (b) rapidly solidifying molten alloys inside 1-mm ID quartz tubes. Reactions between the aluminum and germanium were investigated at BNL as a function of pressure and temperature by in situ, synchrotron x-ray diffraction at the six-axis multi-anvil press.

We found that a two-phase aluminum-germanium 30/70 mixture reacts under pressure and forms a metastable orthorhombic aluminum-germanium phase. At a pressure of 9.8 GPa, this alloy melts at approximately 380°C, which is 40°C lower than the eutectic melting temperature at zero pressure. On cooling and releasing the pressure, the alloy solidifies into a metastable state consisting of a majority amorphous aluminum-germanium phase and pure germanium (see figure).



Pressure dependence of the lattice parameters of aluminum and germanium measured on a fine, two-phase aluminum-germanium alloy by in situ x-ray diffraction. From these data, it follows that the elastic moduli of aluminum and germanium are 91.07 GPa and 94.12 GPa, respectively.

Chemistry

Recombination Kinetics: Correcting the Textbooks

97007

Russell Pack

Our overall objective is to change the way chemists treat recombination reactions. Most textbooks and literature assume that recombination reactions occur only via sequences of two-body collisions. We are showing by actual calculations on real systems that true three-body collisions also contribute significantly. Our work is paying off: this year an author has promised to include our studies in a textbook he is currently writing.

During the past year we have made great progress. We calculated more cross sections for the reaction Ne + Ne + H -> Ne_2 + H, thermally averaged these cross sections to get rate constants, and did kinetics studies. Our results show that, for the above reaction (and probably also for many other reactions), three-body collisions dominate in the kinetics at all pressures. As the accompanying figure shows, including only a sequential two-body mechanism gives a recombination rate coefficient that is a factor of 2.3 times too small at low pressures and orders of magnitude too small at high pressures.

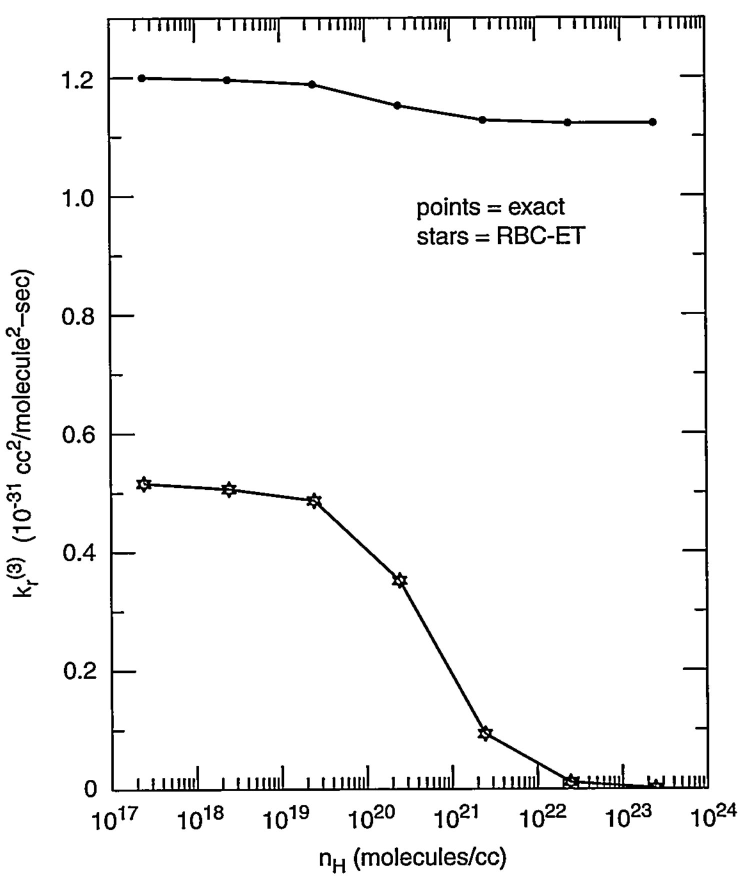
We completed much of the programming for exact calculations using arrangement-channel hyperspherical coordinates, and we are getting close to results. We also got computer programs that use symmetrized hyperspherical coordinates to work well at small-to-moderate hyperradii. We've parallelized these programs and are writing three papers on this work.

Publications

Kendrick, B.K., et al., "Comment on 'On the Longuet-Higgins Phase and Its Relation to the Electronic Adiabatic-Diabatic Transformation Angle' " (to be published in *J. Chem. Phys.*).

Pack, R.T., et al., "Three-Body Collision Contributions to Recombination and Collision-Induced Dissociation. I. Cross Sections," *J. Chem. Phys.* **109**, 6701 (1998).

Pack, R.T., et al., "Three-body Collision Contributions to Recombination and Collision-Induced Dissociation. II. Kinetics," *J. Chem. Phys.* **109**, 6714 (1998).



Recombination rate coefficients versus third-body number density. The points represent an exact solution of the master equations, while the stars represent the Roberts, Curtiss, and Bernstein Energy Transfer (i.e., the sequential two-body) mechanism.

Uses of Novel Selenium-Containing Chiral Derivatizing Agents

96068

Louis Silks

The development of new and effective pharmaceuticals critically depends on the synthesis of molecules that display a variety of homo- and hetero-carbon bonding in complex stereochemical arrays. The discovery of new chemical reactions that allow for the efficient stereoselective synthesis of target chiral molecules has a direct impact on the drug discovery process. We have uncovered a new chemical process that allows for the generation of two defined chiral centers in one chemical step by using our chiral selone reagents.

Reaction of an aldehyde with the titanium-based enolate of a propionyl selone yielded a product in which the two new chiral centers are in a *syn* relationship. 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% yield), and the stereoselectivity was greater than 98%. Using alpha-alkoxy groups on both reacting partners of this selone-mediated aldol reaction has also been successful. The relationship of the two new chiral 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 nuclear magnetic resonance (NMR) experiments indicated the *anti* relationship. In addition, reaction of alpha sulfide aldehydes gives rise to good yields of the thio-ether terminated aldol product (see first figure).

The most exciting feature of this work has been the additional apparent discovery of a C- H···Se through-space interaction. Single-crystal x-ray analysis of the aldol selone adduct with crotonyl aldehyde has indicated a H···Se distance of 2.63 Å (see second figure), and two-dimensional $^{1}H/^{77}Se$ NMR spectroscopy has clearly indicated a unique correlation between this proton and selenium.

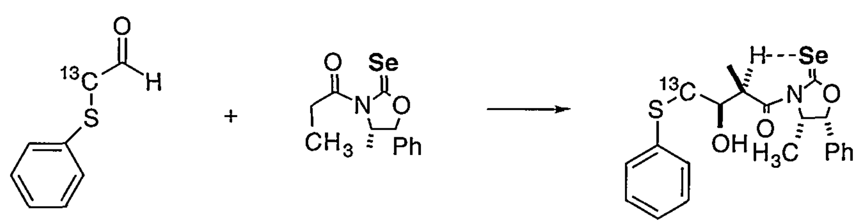
Publications

Salzmann, M., et al., "Labeling of Biomolecules with ^{77}Se: NMR Characterization of L,L-Selenocystine" (submitted to *J. Biomol. NMR*).

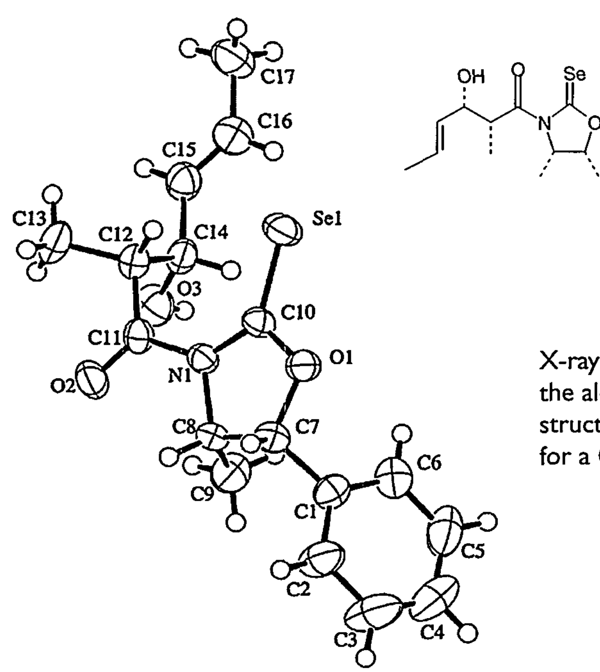
Silks, L.A., et al., "Synthesis and Applications of Chiral Selones," *Phosphorus, Sulfur, Silicon and Related Elements* **136–137**, 209 (1998).

Stocking, E.M., et al., "Synthesis of L-Selenocystine, L(Se-77)selenocystine and L-Tellurocystine," *J. Chem. Soc., Perkin Trans. 1*, **16**, 2443 (1997).

Wu, R., et al., "Recent Progress in the Synthesis and Application of Chiral Selones," in *Recent Research Developments in Organic and Bioorganic Chemistry* (Transworld Research Network, Trivandrum, India, in press).



Chiral-selone-promoted aldol reaction with $TiCl_4$.The product 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 the aldol product.This structure provides evidence for a C-H···Se interaction.

Solvation and Ionic Transport in Polymer Electrolyte Membranes

96054

Thomas Zawodzinski

We have developed the most extensive and realistic model of transport in ion-conducting polymer membranes to date. The first phase of this work combines theory and experiment to generate a qualitative framework of experimentally observed transport properties in hydrated, proton-conducting membranes. The framework not only provides a starting point for rationally designed polymer-membrane technologies for such applications as fuel cells and electrochemical reactors, but also could contribute to research in biological cell membranes for medical applications.

In developing the framework, we performed molecular quantum chemical calculations that probed minimum energy structures of membrane molecular fragments. Further, we identified key elements that influence solvation and transport, gaining an understanding of local structure and dynamics of polymer components. To compare membrane structures with different functionalities, we then used relatively simple calculations that focus on these key elements. These simpler computations were made on model compounds and water solvates. From the extensive calculations on clusters of various numbers of water molecules with small analog acid molecules, we produced a new quasi-chemical theory that describes the chemical thermodynamic properties of these clusters and is a useful link to experiments.

We are developing the first full theory of transport in these particular membranes that involves molecular-level detail using two parallel approaches. In one approach, the electronic-structure data define a periodic potential in the pores. This information is used to compute the friction tensor for interaction of a hydronium ion with the pore wall, giving a prediction of conductivity. In the second approach, which involves detailed Monte Carlo simulation of transport dynamics, the electronic-structure data define the influence of ion traps on protonic mobility. The latter approach can be extended to an arbitrary network of pore shapes and sizes.

Publications

Paddison, S.J., and T.A. Zawodzinski, "Molecular Modeling of the Pendant Chain in Nafion" (11th International Conference on Solid State Ionics, Honolulu, HI, November 16–21, 1997).

Paddison, S.J., et al., "Conformations of Perfluoroether Sulfonic Acid Side Chains for the Modeling of Nafion" (submitted to *J. Am. Chem. Soc*).

Paddison, S.J., et al., "High Frequency Dielectric Studies of Hydrated Nafion" (to be published in *J. Electroanal. Chem.*).

Paddison, S.J., et al., "Ion and Water Transport in a Nafion Membrane Pore: A Statistical Mechanical Model With Molecular Structure" (194th Electrochemical Society Meeting, Boston, MA, November 1–6, 1998).

Paddison, S.J., et al., "Theoretical Structures of Triflic Acid-Water Clusters and the Molecular Mechanism of Proton Dissociation" (194th Electrochemical Society Meeting, Boston, MA, November 1–6, 1998).

Zawodzinski, T.A., et al., "Mechanics of Transport in Proton-Conducting Membranes" (11th International Conference on Solid State Ionics, Honolulu, HI, November 16–21, 1997).

Zawodzinski, T.A., et al., "New Insight into Transport in Membranes for Polymer Electrolyte Fuel Cells" (193rd Electrochemical Society Meeting, San Diego, CA, May 3–8, 1998).

Reactivity of Metal Ions Bound to Water-Soluble Polymers

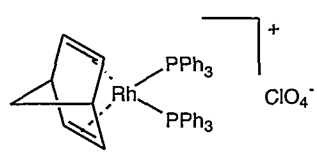
96088

Nancy Sauer

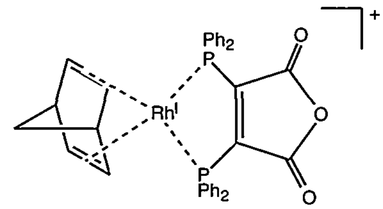
The objectives of this research were twofold: to complete the evaluation of polyamine-supported rhodium-phosphine catalysts for hydrogenation (see accompanying figure); and to synthesize, characterize, and evaluate supported soluble catalysts for carbon-bond-forming reactions. Our studies demonstrated that catalyst systems modified for tethering to polymeric supports were active hydrogenation catalysts with rates similar to known homogeneous catalysts (see accompanying table). However, tethering the catalysts to polyamines resulted in systems that had very low or no hydrogenation activity. It appears from 31-P and ICP data (which show that rhodium is still present in the system but not coordinated to phospine) that support amine binds the rhodium, inactivating it for hydrogenation.

We then began developing phase-separable surfactant systems for catalysis to determine whether a water-soluble transition-metal complex bearing surfactant-like phosphine ligands could promote micelle formation in a biphasic reaction system. In addition to increasing the rate of reaction by producing a large interfacial surface area, it was expected that the catalyst could readily be separated from the organic products by simple water/organic phase separation.

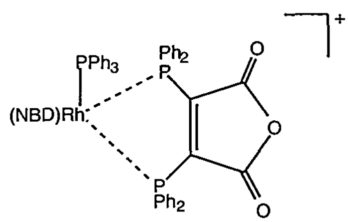
We synthesized a phosphine ligand with a surfactant-like structure and employed this ligand to prepare catalytically active complexes of palladium. Then we used the palladium complexes in Heck-type coupling reactions (for example, coupling of iodobenzene and ethyl acrylate to produce ethyl cinnamate) under vigorously stirred biphasic reaction conditions, and found that their performance was superior to that of a standard water-soluble palladium catalyst under analogous conditions.



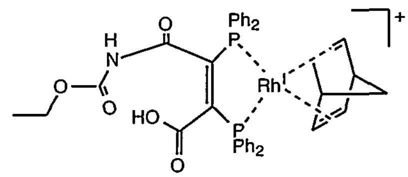
$[Rh(NBD)(PPh_3)_2]^+ ClO_4^-$



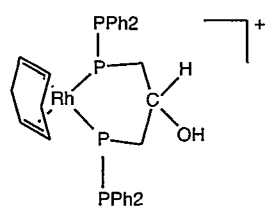
$[Rh(NBD)(PMA)]^+ ClO_4^-$



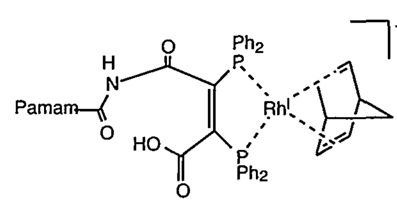
$[Rh(NBD)(PMA)(PPh_3)]^+ ClO_4^-$



$[Rh(NBD)(PMAET)]^+ClO_4^-$



$[Rh(NBD)(PPR)]^+ ClO_4^-$



$Rh(NBD)(PMA\text{-}Pamam)^+ClO_4^-$

Structures of rhodium hydrogenation catalysts.

Hydrogenation (k_1)and Isomeration (k_2) Rates for Hydrogenation Catalysts

Catalyst*	k_1 (s^{-1})	k_2(s^{-1})
$Rh(NBD)(PPh_3)_2$	16. 0	90.2
Rh(NBD)(PMA)	2.0	2.5
$Rh(NBD)(PMA)(PPh_3)$	0.5	0.9
Rh(NBD)(PMAET)	2.4	6.9
Rh(NBD)(PPR)	17.2	13.4
Rh(NBD)(PMA-Pamam)	very limited catalysis	

*All ClO_4^- salts. All rates × 10^6

Solvation Dynamics of Ion Pairs

98010

Antoinette Taylor

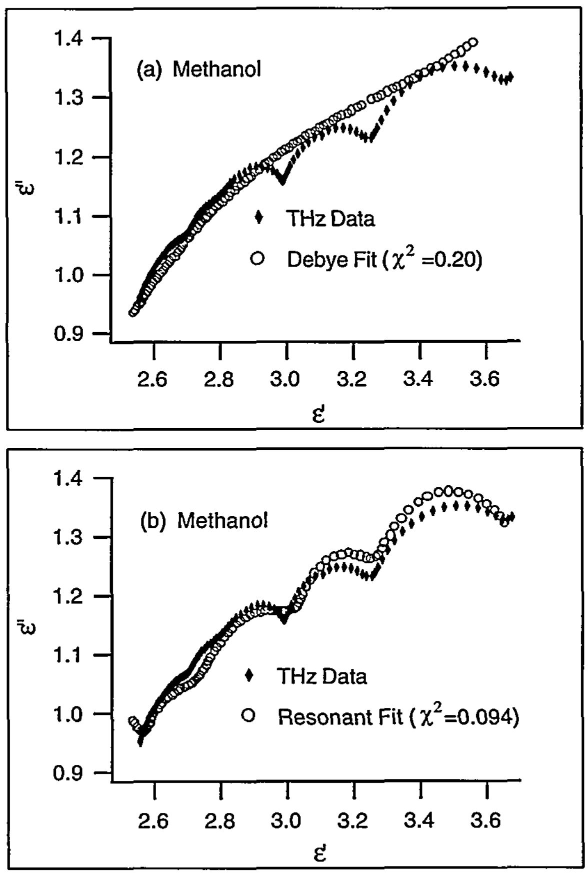
The dynamics of how a solvent responds to a solute still remains an unsolved but critically important scientific problem. The objective of this project is to probe the mechanisms and time scales involved in both the formation and dynamic behavior of ion pairs in solution. To accomplish this goal, we will further develop the technique of femtosecond-terahertz time-domain spectroscopy for the study of dynamic molecular interactions in liquids, closely coupling the spectroscopic results obtained to the molecular dynamics calculations. The project will proceed from background studies on pure liquid and some novel room temperature molten salts through studies on both dilute and concentrated solutions of salts in appropriate liquids.

This year we set up our femtosecond time-domain spectrometer and used it to measure the complex dielectric function of a variety of liquids in the terahertz frequency range. Previous studies interpreted such data in terms of a Debye-based relaxation model that describes the dielectric relaxation of collective, diffusive reorientational motions in a liquid. In contrast, we found for all liquids studied that Debye-based models are inadequate in their description of the dielectric function because significant resonant absorption occurs throughout this frequency range. Our measured dielectric functions can, however, be described by a model that combines resonant absorption with diffusive behavior, thus demonstrating for the first time that the nature of the dielectric function is changing from diffusive to oscillatory in character in the terahertz frequency range.

In the accompanying figure, we demonstrate this claim with Cole-Cole plots of the measured dielectric function for methanol. The best Debye fit (panel a) does not reveal the fine structure seen in the data, whereas a fit including resonant absorption (panel b) matches the data well. These low-frequency modes correspond to collective motions of molecules.

Publications

Asaki, M.L., et al., "The Nature of the Dielectric Response of Polar Liquids" (submitted to *Phys. Rev. Lett.*).



(a) Cole-Cole plot of the measured dielectric function for methanol, fitted with a three-term Debye relaxation model. (b) Cole-Cole plot of the same measured dielectric function for methanol fitted with our model, which combines Debye relaxation and resonant absorption. In both plots, ε′ and ε″ are the real and imaginary parts, respectively, of the dielectric function.

Classical Kinetic Mechanisms Describing Heterogeneous Ozone Depletion

97023

Bryan Henson

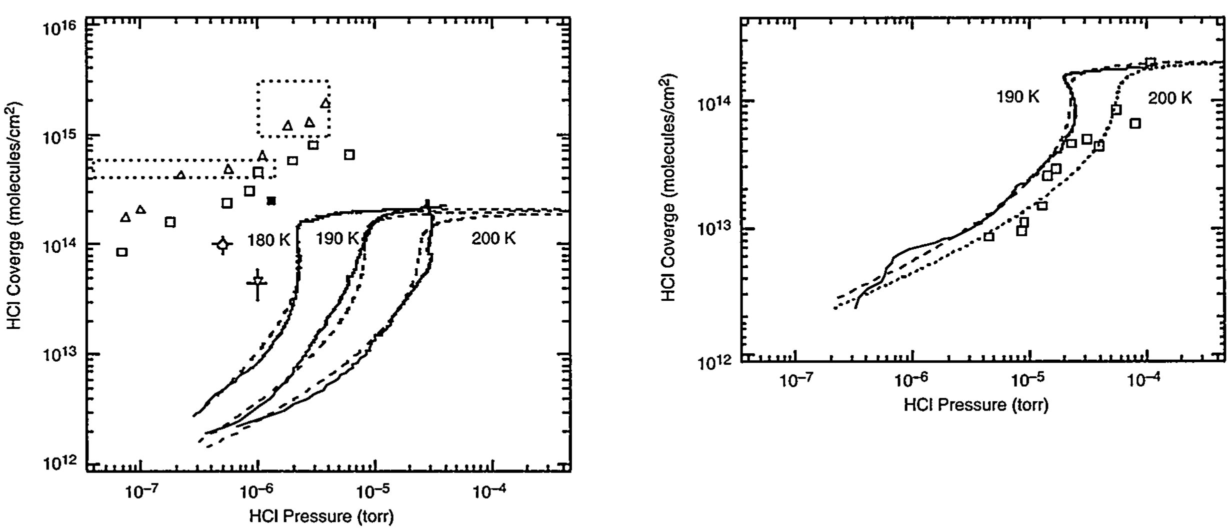
One of the most important trace gas species involved in stratospheric ozone depletion is HCl. We have continued experiments on HCl uptake on surrogate stratospheric aerosol surfaces coupled with detailed modeling in order to relate interfacial HCl phases to the well-known bulk thermodynamics of HCl/H_2O or $HCl/H_2O/HNO_3$ solutions. The coverage of HCl on two surfaces as a function of pressure and temperature is shown in the first figure. The data strongly indicate classic physical adsorption to form true two-dimensional phases. We therefore derive a first principles model of the pressure isotherms based on a Langmuir and Bragg-Williams theory that incorporates a dissociative equilibria to stabilize adsorption, shown by the equation below. In the equation, P is the HCl pressure; m is the HCl mass; k and h are Boltzman's and Plank's constants; θn is the vibrational temperature; ΔH_{sol} and ΔH_{sub} are adsorption energies for dissociated and undissociated species, respectively; ε is a lateral interaction energy; θ_T is the total coverage of HCl; θ_I is the coverage of dissociated hydrogen and chlorine; and $\theta = \theta_I + \theta_T$.

The agreement between the model and the data indicates that this simple theory captures the complex physics of this heterogeneous interaction. Furthermore, the calculated energies are directly comparable to measured values for the heat of solution and bulk hydrates, so that now modeling of the complex interface can be informed by over 100 years of classical solution and solid thermodynamics, as shown in the second figure.

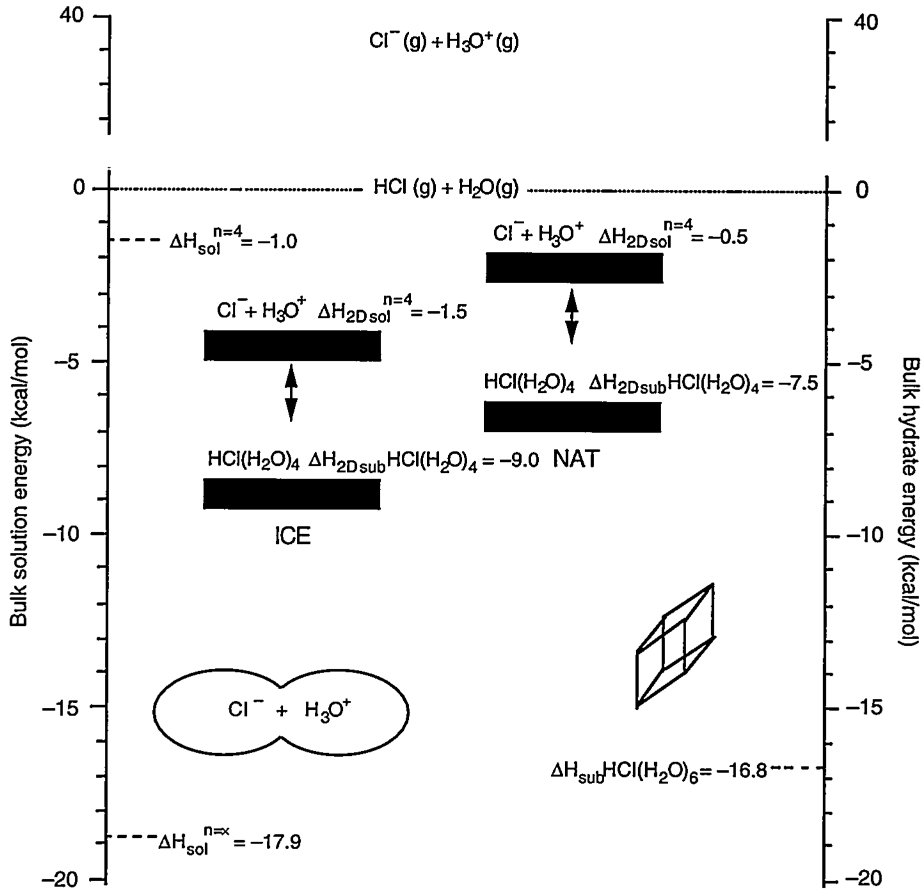
Publications

Henson, B.F., et al., "Eley-Rideal Mechanism of N_2O_5 And $ClONO_2$ Reactivity on Simulated PSC Surfaces," *Abstr. Pap. Am. Chem. Soc.* **214**, 130 (1997).

$$P = \left(2\pi mkT/h^3\right)^{3/2} kT/h\left[e^{-\theta v/2T}/\left(1-e^{-\theta v/T}\right)\right]^{-3f}(\theta/1-\theta)^f$$
$$\times\ \exp\left(-f\left\{\left(\theta_I/\theta_T\right)\Delta H_{sol}+\left[1-\left(\theta_I/\theta_T\right)\Delta H_{sub}\right]+f\theta\varepsilon\right\}/RT\right)$$



Uptake of HCl on ice as a function of pressure. The solid lines, with the temperatures labeled, represent pressure over the ice—measurements obtained by mass spectrometry. The dashed lines are calculations. The individual data points (shown as squares and triangles) are isolated measurements from other laboratories. In the left panel, the uptake as a function of pressure over ice is shown at 180 K, 190 K, and 200 K. In the right panel, the uptake as a function of pressure over nitric acid trihydrate ice is shown at 190 K.



Schematic energy diagram of HCl bulk solutions and hydrates and the interfacial analogs of these phases. The heats of solution and hydrate sublimation energy are plotted on each axis. The equilibria between liquid-like and solid-like phases are shown schematically for the HCl system on H_2O ice (ICE) and nitric acid trihydrate (NAT), with the adsorption energy for each phase labeled.

Characterization of Propane Monooxygenase: Initial Mechanistic Studies

97024

Pat Unkefer

The industrial use of chlorinated hydrocarbons has led to widespread contamination of soils and groundwater. A number of bacterial strains have been shown to degrade chlorinated compounds, including trichloroethylene (TCE). For example, methanotrophic bacteria degrade TCE by way of the enzyme methane monooxygenase (MMO), which converts TCE to epoxide. However, the effectiveness of methanotrophic bacteria in bioremediation schemes is limited because of the toxicity of TCE epoxide. We are investigating 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 by formation of the epoxide, PMO could provide an important alternative to MMO for bioremediation of TCE-contaminated sites.

This past year, we made significant progress toward our goal of further characterizing the propane monooxygenase enzyme. We determined that the PMO is actually a multiprotein complex and that this complex is composed of at least three, and perhaps as many as four, proteins. Two of these proteins are enzymatically active. One is a reductase, the other is an oxygenase. The third protein appears not to be catalytically active; we have hypothesized that it has a regulatory function. The three components have been separated and recombined with retention of enzymatic activity.

We also demonstrated that, contrary to previously published reports, this enzyme activity could be induced in cells grown on a rich nutrient source, if the rich nutrient source were replaced with propane as the carbon source. The demonstration allowed us to obtain increased amounts of this fragile enzyme. We are preparing a stockpile of the enzyme for next year's work, which will begin with the specific objective of characterizing the substrate specificity and products generated by PMO.

Unraveling Heterogeneous Surface Reaction Kinetics

98012

W. Patrick Ambrose

When a system of molecules is heterogeneous, a bulk measurement masks the behavior of molecules that are different from the mean. Unexpected heterogeneous behavior was uncovered recently even in nominally homogeneous systems when individual molecules are examined. We have built optical microscopes that we are using to investigate the optical properties of single molecules. In this project, we are observing and studying the nature of the interactions between single immobilized and single solvated molecules. The unique concept that sets our work apart from other surface reaction techniques is that interesting details will be uncovered that are normally masked by bulk averaging.

We accomplished five goals toward direct observation of the interactions between single pairs of solvated and immobilized molecules at an interface. First, we developed efficient methods for analyzing time-resolved, fluorescence lifetime data from single rhodamine-6G molecules on a silica surface. Second, we developed the capability to perform video-rate detection and imaging of the fluorescence from single molecules at air/glass and water/glass interfaces (with continuous imaging at 33 ms per picture). With our apparatus, we have successfully obtained fluorescence images of individual molecules of B-phycoerythrin binding from buffer solution to a fused silica surface using both slow-scan and video-rate imaging. We also explored three types of total internal reflection (TIR) excitation and found that prism TIR produces the best signal-to-background ratio, whereas through-objective TIR excitation produces the best total signal before photo-bleaching (see the accompanying figure). Finally, we have improved a sensitive proteinase assay that will be combined with observation of the interactions of molecules at an interface. These results were described in four invited seminars, and several interested companies have visited our laboratory to discuss possible collaborative investigations.

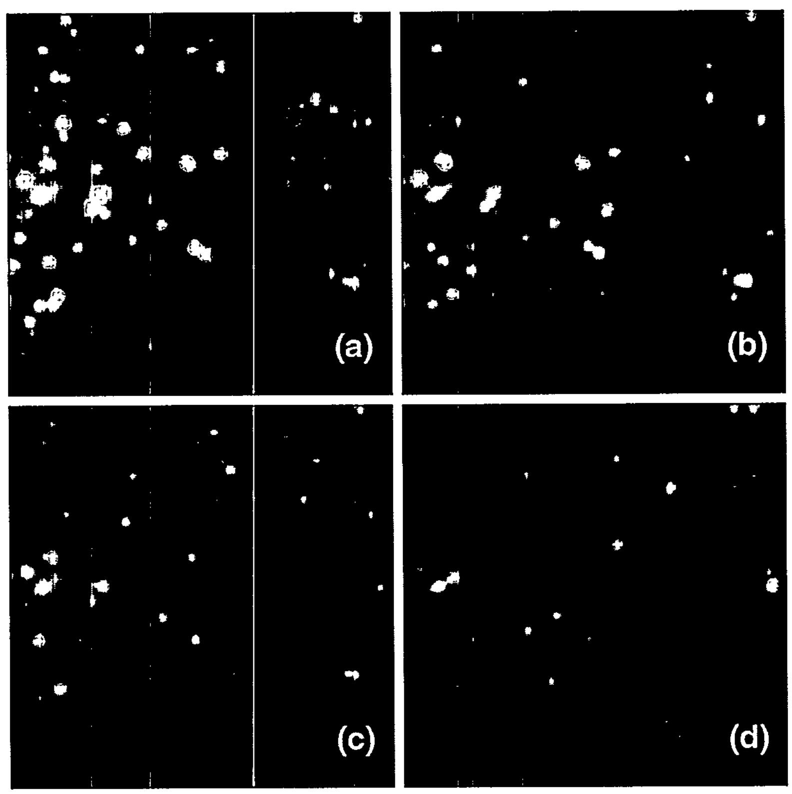
Publications

Ambrose, W.P., et al., "Detection System for Reaction-Rate Analysis in a Low-Volume Proteinase Assay," *Anal. Biochem.* **263**, 150 (1998).

Ambrose, W.P., et al., "Probing the Heterogeneous Fluorescence Lifetime Behavior of Single Rhodamine-6G Molecules on Fused Silica," *SPIE Proc. on Methods of Ultrasensitive Protection* **3270**, 190 (1998).

Ambrose, W.P., et al., "Single Molecule Detection with Total Internal Reflection: Signal to Background and Total Signal in Different Geometries" (submitted to *Bioimaging*).

Enderlein, J., et al., "Molecular Shot Noise, Burst Size Distribution, and Single Molecule Detection in Fluid Flow: Effects of Multiple Occupancy," *J. Phys. Chem. A* **102**, 6089 (1998).



A sequence of fluorescence images showing the locations and brightnesses of individual B-phycoerythrin (BPE) molecules adhering to a silica-water interface. Molecules of BPE diffuse through an aqueous solution, bind to a silica surface, are excited with laser light using total internal reflection at the silica/water interface, and are imaged with a sequence of 0.5-s integration times. The disappearance of bright spots from one picture to the next is the result of photo-bleaching. Photo-bleaching individual molecules provides a direct measurement of the total available signal per molecule.

Ultrafast Solid-State Electron Transfer in Donor-Acceptor Conducting Polymers

97025

Duncan McBranch

Electron transfer in conducting polymers is a promising new field of photochemistry. In these systems, 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 nine orders of magnitude. The goals of this project are to (a) apply 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); and (b) explore applications of these materials by studying methods to use the charge transfer state to drive photovoltaic and nonlinear optical effects.

This year we have finished our comprehensive studies of nonlinear interactions of excitons in conjugated polymers at high excitation densities, including the first observation of biexcitons. We have demonstrated universal spectroscopic features on femtosecond time scales resulting from photoexcitations (intrachain excitons and interchain excimers and charge-transfer states) in many different phenylene-based conjugated polymers. Our broad-band polarized transient absorption measurements have allowed the first direct measurement of charge-transfer states in oriented polymer films (in which polymer chains are aligned in a common direction). We have, for the first time, extended the range of femtosecond spectroscopy in the mid-infrared range to map the low-energy photo-induced optical transitions in these materials. We have further developed a new theory of the optical properties and energy structure of conducting polymers.

Publications

Kirova, N., et al., "Excitations and Optical Properties of Phenylene-Based Conjugated Polymers and Oligomers," *Opt. Mater.* **9**, 472 (1998).

Klimov, V., and D.W. McBranch, "Femtosecond High-Sensitivity, Chirp-Free Transient Absorption Spectroscopy," *Opt. Lett.* **23**, 277 (1998).

Klimov, V., et al., "Biexcitons in π-Conjugated Oligomers: Intensity-Dependent Femtosecond Transient Absorption Study," *Phys. Rev. B* **58**, 7654 (1998).

Klimov, V., et al., "Exciton and Biexciton Signatures in Femtosecond Transient Absorption of π-Conjugated Oligomers," *SPIE Proceedings, Optical Probes of Conjugated Polymers* **3145**, 58 (1997).

Klimov, V., 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).

Kraabel, B., et al., "Ultrafast Spectroscopic Holography in Conjugated Polymers" (to be published in *Synth. Met.*).

McBranch, D.W., et al., "Signatures of Excitons and Polaron Pairs in the Femtosecond Excited-State Absorption Spectra of Conjugated Polymers and Oligomers" (to be published in *Synth. Met.*).

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

Soluble Polymers for Enhancing Biocatalysis

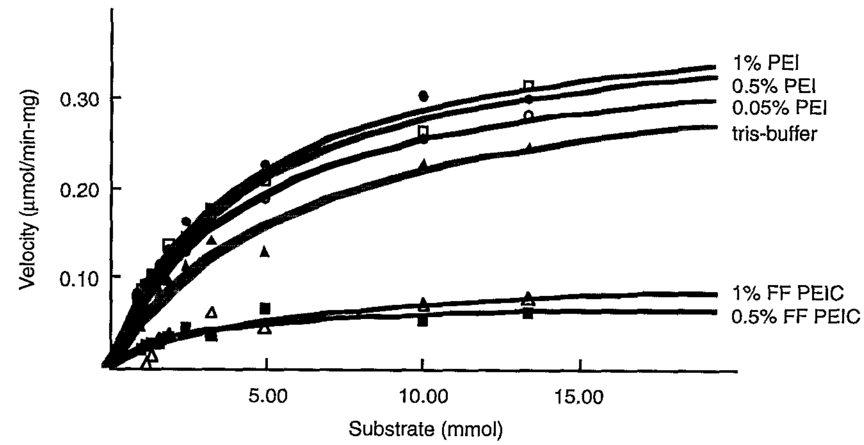
98013

Nancy Sauer

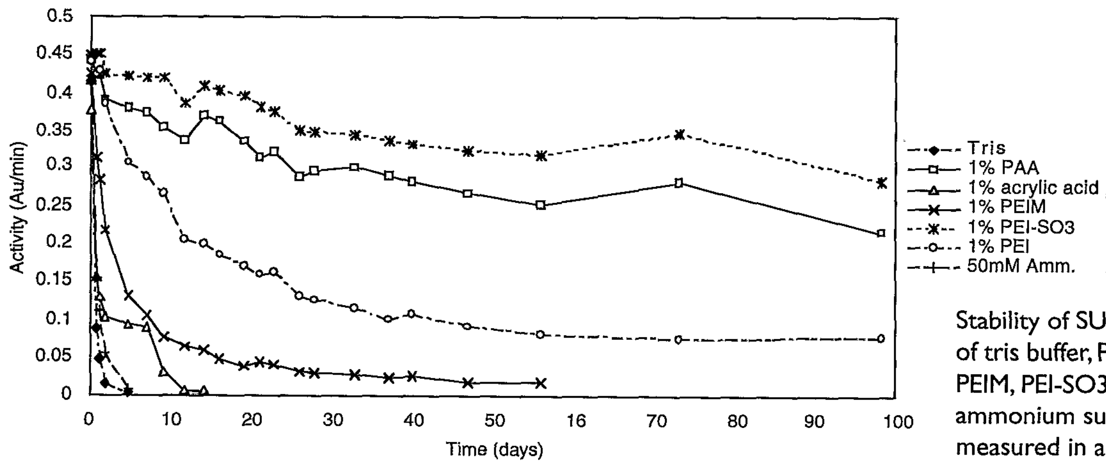
This research aims at increasing the utility of biocatalysts for chemical synthesis, sensors, waste treatment, and decontamination. To do this, we seek to understand the molecular interactions between soluble synthetic polymers and enzymes affecting biocatalysis. We are using organic and aqueous soluble polyamines of three structural types (linear, branched, and spherical) as backbones for enzyme stabilizing systems. Two enzymes have been selected for study: yeast alcohol dehydrogenase (ADH) and subtilisin (SUB).

We have evaluated the stability of ADH and SUB under a range of conditions in the presence of four water-soluble polymers: polyethylenimine (PEI), polyacrylic acid (PAA), permethylated polyethylenimine (PEIM), carboxylated polyethylenimine (PEIC), and sulfonated PEI (PEI-SO3). When we monitored enzyme activity over time for ADH, we found that both PEI and PEI-SO3 increased enzyme lifetime from 48 hours to 2,000 hours compared with untreated buffer solutions and that PEI provided some protection against enzyme denaturization at elevated temperatures. However, not all polymers tested enhanced ADH lifetime. As shown in the first figure, which shows the conversion velocity of the ethanol substrate as a function of PEI and PEIC concentrations, adding PEI to ADH increases reaction velocity, but adding PEIC decreases velocity. Initial enzyme activity (Kcat) for ADH in PEI-containing solutions increased three-fold over the enzyme in tris buffer solution alone. In addition to these experiments, we have determined the polymer-ADH ratio that imparts stabilization for ADH by using ultrafiltration and colorometric assays of enzyme activity. For ADH, polymer-to-enzyme mole ratios of 1:1 increased enzyme lifetime; however, stable polymer-enzyme complexes formed at polymer-to-enzyme ratios of 70:1.

The second figure shows the activity of subtilisin in the presence of polymeric and monomeric ionophores. Although we observed no elevation of initial activity of SUB relative to unstabilized solutions, a dramatic enhancement of enzyme lifetime was achieved with PEI-SO3 and PAA. For both SUB and ADH, several polymers inhibited substrate conversion and the lifetime of the enzyme. Clearly, stabilization is not simply the result of the polymers holding the enzymes together and not allowing denaturization. These studies have shown that electrostatic interactions strongly affect enzyme stabilization at varying temperatures and pH.



Effect of polyethylenimine concentration on ADH activity. Plot shows relative velocity vs. substrate (ethanol) concentration. PEI increases ADH activity, and PEIC inhibits it.



Stability of SUB in the presence of tris buffer, PAA, acrylic acid, PEIM, PEI-SO3, PEI, and ammonium sulfate. Activity is measured in absorbance units per minute (Au/min).

Heterogenization of Homogeneous Catalysts: The Effect of the Support

96082

William Earl

The goals of this project were to (1) develop novel methods of characterizing heterogeneous metal catalysts, (2) investigate the effects of unusual solvents (supercritical CO_2) on heterogeneous catalytic reactions, and (3) attempt to synthesize heterogeneous catalysts using the mesoporous MCM-41–type supports.

This year for our first goal, we completed the successful development of a novel method of using nuclear magnetic resonance (NMR) spectroscopy with paramagnetic metals to characterize the state of the metal in the heterogeneous catalyst. This method relies on the interaction between the paramagnetic electrons and the NMR nuclei of interest. The NMR spectrum of those nuclei close to the paramagnetic metal are broadened and shifted. The linewidth, shift, or disappearance of signal can be used to understand the nature of the bonding of the metal into the support structure.

To meet our second goal, we completed our studies of catalytic reactions in supercritical CO_2. A very small number of the reactions show novel selectivity, which we do not fully understand.

For our third goal, we were successful in adsorbing a known homogeneous catalyst in MCM-41 and retaining catalytic activity. This rhodium catalyst ([(R,R)-DuPhosRh(COD)]Otf) is an enantiomeric hydrogenation catalyst. It retains good activity and very high enantiomeric selectivity when supported on MCM-41. Under some conditions the metal can be leached out of the solid support, but in nonpolar solvents it retains its activity through several cycles. This supported catalyst has important implications in the synthesis of enantiomeric compounds for agricultural and pharmaceutical uses.

Publications

de Rege, F.M., et al., "Asymmetric Hydrogenation Using MCM-41 Immobilized Diphosphine Rhodium Cations" (submitted to *Chem. Commun.*).

Labouriau, A., et al., "Probing the Structure of Metal-Substituted Molecular Sieves by Solid-State NMR" (12th International Zeolite Conference, Baltimore, MD, July 5–10, 1998).

Pesiri, D., et al., "Palladium Cross Coupling Reactions in Supercritical Carbon Dioxide," *Chem. Commun.* 1397 (1998).

Pesiri, D., et al., "Selective Epoxidation in Dense Phase Carbon Dioxide," *Chem. Commun.* 1015 (1998).

Fundamental Processes in Polymer Light-Emitting Electrochemical Cells

98011

Darryl Smith

A light-emitting electrochemical cell (LEC) is a recently invented electroluminescent device in which coupled electronic and ionic transport in a luminescent polymer electrolyte is exploited to form an efficient light-emitting structure. These devices have potential fabrication, performance, and cost advantages that make them very attractive for lighting and display applications. The use of blended polymeric components, polymeric electrolytes, and dopable light-emitting polymers to form a mixed conducting plastic is a key new feature of LECs. With modest improvements in device performance, particularly faster response times, the advantages of LECs will lead to wide commercial application. We are conducting a multidisciplinary study of these issues to better understand and then improve the operation of LECs. We have three specific goals: (1) to develop an understanding of ionic/electronic transport in polymer blends and determine how these processes influence the operation of LECs, (2) to use measured material properties to develop and validate device models describing the steady-state and transient operation of the LECs, and (3) to use the basic understanding of the individual materials and composites to guide the synthesis of improved polymeric components for use in LECs.

We have investigated the structure of the electrochemical junction formed during the operation of the LECs using electrical ac impedance spectroscopy. Using small-signal impedance spectroscopy at frequencies high enough that the electrical response of the LEC is dominated by electrical and not ionic transport, we determined the capacitance is due to the electrical junction formed in the LEC. This electronic capacitance determines the effective separation of the electrons and holes forming the junction. The width of the electrical junction is extremely important. In operation, light is emitted when electrons and holes recombine radiatively in the undoped region of the polymer layer that separates the *n*- and *p*-doped regions. If this region is too thin, the electrons and holes will not recombine in it. Instead they will be injected into the doped regions where they will predominantly recombine nonradiatively. Learning to control the width of this junction is a key to improving the LEC quantum efficiency.

Publications

Campbell, I.H., et al., "Capacitance Measurements of Junction Formation and Structure in Polymer Light Emitting Electrochemical Cells," *Appl. Phys. Lett.* **72**, 2565 (1998).

Mathematics and Computational Science

Elliptic Solvers for Adaptive Mesh Refinement Grids

96310

Daniel Quinlan

We are developing 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. This library will directly benefit efforts in scientific computing, geoanalysis, fluid dynamics, mathematical modeling and analysis, radiation transport, and others. 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 (AMR) applications.

This year we focused on putting the software together, completing the final AMR++ library, writing tutorials and manuals, and building example applications. We implemented the Fast Adaptive Composite Grid method as the principal elliptic solver, presenting our results at the Overset Grid Conference and other more AMR-specific conferences. We made significant advances in optimizing the software's serial and parallel performance; several papers were published on the details of this work.

Publications

Bassetti, F., et al., "C++ Expression Templates Performance Issues in Scientific Computing" (Twelfth International Parallel Processing Symposium, Orlando, FL, March 30–April 3, 1998).

Bassetti, F., et al., "A Comparison of Performance-Enhancing Strategies for Parallel Numerical Object-Oriented Frameworks" (First International Scientific Computing in Object-Oriented Parallel Environments Conference, Marina del Rey, CA, December 8–11, 1997).

Bassetti, F., et al., "Loop Transformations for Performance and Message Latency Hiding in Parallel Object-Oriented Frameworks" (1998 International Conference on Parallel and Distributed Processing Techniques and Applications, Las Vegas, NV, July 13–16, 1998).

Bassetti, F., et al., "Optimizing Transformations of Stencil Operations for Parallel Object-Oriented Scientific Frameworks on Cache-Based Architectures" (Second International Scientific Computing in Object-Oriented Parallel Environments Conference, Santa Fe, NM, Dec 13–16, 1998).

Bassetti, F., et al., "OVERTURE: An Object-Oriented Framework for High Performance Scientific Computing" (High Performance Networking and Computing Conference, Orlando, FL, November 7–13, 1998).

Bassetti, F., et al., "Temporal Locality Optimizations for Stencil Operations within Parallel Object-Oriented Scientific Frameworks on Cache-Based Architectures" (Parallel and Distributed Computing Symposium, Las Vegas, NV, July 13–16, 1998).

Bassetti, F., et al., "Toward FORTRAN 77 Performance From Object-Oriented C++ Scientific Frameworks" (The High Performance Computing Conference, Boston, MA, April 5–9, 1998).

Brown, D., et al., "Overture: An Object-Oriented Framework for Solving Partial Differential Equations" (First International Scientific Computing in Object-Oriented Parallel Environments Conference, Marina del Rey, CA, December 8–11, 1997).

Quinlan, D., "AMR++: Object-Oriented Design for Adaptive Mesh Refinement" (The High Performance Computing Conference, Boston, MA, April 5–9, 1998).

Designing a Micromechanical Transistor

96305

Ronnie Mainieri

Micromechanical electronic systems, essentially chips with moving parts, are fabricated with the same cost-effective techniques used to manufacture electronic chips. These micromechanical chips can also contain electronic components. When mechanical parts are combined with electronic parts, chips can process signals mechanically at room temperature. Achieving designs comparable to those obtained with electronic components requires a mechanical device that can change its behavior in response to a small input—a mechanical transistor. This project developed some of the design tools for these complex-shaped resonant structures by using the geometrical ray technique. The project also explored nanometer-scale electronic structures that can work at room temperature.

During the project, we determined the limitations in computing the resonances for complex shapes and the ways in which they are altered by noise. We also studied the possibility of using the disorder in polysilicon to create a nanometer-scale electronic device that operates at room temperature, as well as the possibility of using the mechanical devices for quantum computing.

We discovered that computing resonances to a fixed accuracy requires an exponential amount of computer work for typical shapes. This is to be contrasted with the polynomial time that was expected. In computing the effect of noise on the resonances, we discovered a new resummation for Feynman diagrams. With this resummation, the number of terms to be summed grows geometrically, rather than factorially, with the order of the expansion.

Publications

Aronov, I.E., et al., "AC Transport and Collective Excitations in a Quantum Point-Contact, *Semicond. Sci.Technol.* **13**, 104 (1998).

Aronov, I.E., et al., "On Properties of Boundaries and Electron Conductivity in Mesoscopic Polycrystalline Silicon Films for Memory Devices," *Semicond. Sci. Technol.* **13**, 33 (1998).

Berman, G., et al., "Introduction to Quantum Computing" (submitted to *World Sci.*).

Berman, G., et al., "Molecular Dynamics Simulations of Grain Boundaries and Electron Conductivity in Mesoscopic Polycrystalline Silicon Films for Memory Devices" (submitted to *Semicond. Sci. Technol.*).

Cvitanovic, P., et al., "Trace Formula for Stochastic Evolution Operators: Weak Perturbation Theory" (submitted to *J. Stat. Phys.)*.

Mainieri, R., and J. Rehacek, "Projective Synchronization in Chaotic Systems" (submitted to *Phys. Rev. Lett.*).

Particles and Patterns in Cellular Automata

96287

Erica Jen

A cellular automation is a discrete dynamical system that consists of a lattice of sites whose values evolve through 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 patterns of electrical activity in simple neural networks—generated by systems consisting of a large number of simple, identical, locally connected components.

In the past two years we have obtained results in the mathematics of one-dimensional cellular automata that promise to provide for the first time a method for analyzing the relationship between an automaton's microscopic-level specifications through its rule table and certain macroscopic features—including pattern formation and computational capabilities—that characterize its evolution. The central point is that these automata are characterized by their generation of mesoscopic structures that can be viewed as particles and backgrounds. The global behavior of the automata may be described through a reformulation of the automation in terms of collision rules on this mesoscopic scale.

This past year we extended these results to develop a "rule-table mathematics" for cellular automata (i.e., a mathematics for functions specified in rule table, or look-up, form). This mathematical approach will help to address questions that are fundamental to the use of cellular automata in simulations, such as the effects of perturbations in rule tables or in initial conditions.

Efficient Multilevel Iterative Methods for Nonlinear PDEs

98029

Dana Knoll

Nonlinear systems of partial differential equations (PDEs) often contain multiple time and length scales. Krylov subspace methods are a dependable means of solving the linear systems associated with implicit time differencing, which is employed to handle the multiple time scales.

Multiple-length scales require fine spatial resolution, which results in large linear systems of equations, usually taxing linear iterative methods. This motivates us to use multigrid methods. In many applications there is a demand for increased dependability from the nonlinear iteration, coupled with a demand for increased efficiency and dependability from the linear iterative methods. Our goal is to develop a hybrid methodology that has Newton-like, nonlinear convergence properties, multigrid-like efficiency, and Krylov-like dependability. Our specific objective is to combine matrix-free Newton-Krylov methods with the multigrid method to develop a new, robust, multilevel methodology for solving systems of nonlinear PDEs. The results of this research could have wide-ranging impact in such areas as computational fluid dynamics, combustion/reactive flow modeling, molding and casting simulation, and magnetohydrodynamics.

We made significant progress during the past year and initiated several external collaborations. As preconditioners to the matrix-free Newton-Krylov method, we developed and studied a large variety of linearizations and multilevel iterative methods. We have considered such applications as radiation diffusion, the Fokker-Planck equation, and the Navier-Stokes equations. We have also developed a parallel, multilevel, iterative method for unstructured grids.

Publications

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Knoll, D.A., et al., "An Efficient Nonlinear Solution Method for Nonequilibrium Radiation Diffusion" (to be published in *J. Quant. Spectrosc. Radiat. Transfer*).

Knoll, D.A., et al., "A Multilevel Iterative Field Solver for Implicit, Kinetic, Plasma Simulation" (submitted to *J. Comput. Phys.*).

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Rider, W.J., and D.A. Knoll, "Multigrid Newton-Krylov Methods for Nonlinear Heat Conduction," *International Mathematics Association for Computing and Simulation* **179** (1998).

Rider, W.J., et al., "A Multigrid Newton-Krylov Method for Multimaterial Equilibrium Radiation Diffusion" (submitted to *J. Comput. Phys.*).

Scalable Run-Time System Research

98055

Suvas Vajracharya

Our project goal is to explore, design, and build SMARTS, a dependence-driven run-time system to support numerical computation over clusters of shared-memory multiprocessors. In contrast to conventional models of computations, where executions are driven by operators or functions, a dependence-driven system is an asynchronous model of computation where the availability of operands or data determine the flow of execution. A dependence-driven model can be implemented either in hardware, as in data-flow computers, or in software, as a coarser grain of blocks of loop iterations or as functions. Our specific objective is to build a coarse-grain data-flow system driven by data and dependencies at the granularity of C++ objects.

This year we have successfully accomplished the project objectives, and SMARTS is now running on the Origins machine built by Silicon Graphics, Inc. (SGI). We have designed an interface for compilers, users, or other frameworks for SMARTS. We have installed the POOMA scientific template library as part of SMARTS and run SMARTS on different architectures including SGI, a parallel cluster of Pentium II computers running on a symmetric multiprocessing (SMP) Linux operating system, a Sun Workstation running Solaris, and Digital Equipment Corporation workstations. Initial performance analysis shows favorable results.

An Agent-Based Model to Investigate the Spread of Influenza

98053

Robert Farber

Our objectives were to push the limits of epidemic modeling technology by developing massively parallel agent-based simulation software that would integrate and model epidemic data plus the associated software for a visualization system. (Agent-based software explicitly models the interactions of individuals within a population and from those interactions predicts the overall behavior for the population itself.)

Our modeling project had three stages: the first two have been completed, and we are now working on the third.

Stage 1: Develop the simulation software and verify its performance and functionality. We produced a flexible agent-based epidemic simulation software that runs on both inexpensive Linux computers and the supercomputers of the Laboratory's Accelerated Strategic Computing Initiatiave (ASCI). We also developed a Java-based global information system (GIS) epidemic viewer.

Stage 2: As part of the Laboratory's collaboration with the Centers for Disease Control and Prevention (CDC), implement a simple rate-of-infection model in the agent-based software and verify it against a published equation-based model. For verification, we implemented a simulation based on the equation-based model presented by Antoine Flahault et al. (see "A Mathematical Model for the European Spread of Influenza," *European Journal of Epidemiology* **10**, 471, 1994). This model calculated the rate of change between categories of individuals within the populations studied. Our simulation replicated Flahault's results, which were based on actual data and simulated the spread of influenza throughout the populations of nine cities interconnected by air travel.

Stage 3: In collaboration with the CDC, extend the agent-based software to more accurately represent CDC influenza data. This additional work is currently under way as part of a separate LDRD project (Epidemiology: Molecular Causes to Macroscopic Consequences).

Multigrid Homogenization of Heterogeneous Porous Media

98033

Joel Dendy

The numerical simulation of flow through heterogeneous porous media has become a vital tool in forecasting reservoir performance, analyzing groundwater supply, and predicting the subsurface flow of contaminants. Consequently, the computational efficiency and accuracy of these simulations are paramount. However, the parameters of the underlying mathematical models (for example, permeability and conductivity) typically exhibit severe variations over a range of significantly different length scales. Thus the numerical treatment of these problems relies on a homogenization or up-scaling procedure to define an approximate coarse-scale problem that adequately captures the influence of the fine-scale structure.

Inherent in such a procedure is a compromise between the competing objectives of computational efficiency and numerical accuracy. Although techniques that balance these conflicting demands exist for a few specific fine-scale structures, this is not the case in general. Moreover, two recent reviews have identified up-scaling as a crucial and open problem.

We are developing new, efficient, numerical multilevel methods for homogenization in models of flow through heterogeneous porous media. Our motivation derives from our observation that multiple-length scales are captured automatically by robust multilevel iterative solvers, such as the multigrid method that was developed at Los Alamos National Laboratory. In recent work on two-dimensional single-phase saturated flow for periodic heterogeneous media, we developed an algorithm based on the operator-induced variational coarsening of black box multigrid that offers a significant improvement in the compromise between accuracy and efficiency.

In our current work, we are extending this algorithm to general boundary conditions in two and three dimensions, which we expect to result in the first algorithm for accurate tensoral up-scaling of the permeability that does not require internal artificial boundary conditions. We are also investigating the extension of the algorithm to multiphase and unsaturated flows.

Publications

Moulton, J.D., et al., "The Black Box Multigrid Homogenization Algorithm," *J. Comp. Phys.* **142**, 80 (1998).

Extending the Theory of Resonant Perturbations to Partial Differential Equations, with Applications to Nonlinear Optics

98030

Roberto Camassa

The exponential growth of the Internet has created intense pressure for new, higher-capacity, fiber-optic communication lines. The most promising designs for the next generation of communication systems presently use soliton pulses as bit carriers. To increase the system capacity, it is necessary to use a regime with multiple soliton channels (operating at different wavelengths) in a single fiber. Recent experiments show that this approach, although feasible, is adversely affected by two types of resonance effects: resonant interactions between channels and resonances with periodic amplification. A novel transmission technique, which uses strong periodic variation of dispersion with distance, or dispersion management, has recently been introduced to cope with these effects. This project's main goal is to provide a theoretical understanding of resonance effects, thereby optimizing the performance of transmission lines based on dispersion management.

In the past year we provided a theoretical explanation of how dispersion management, by suppressing the primary resonance, can dramatically reduce the process of accumulation of four-wave mixing and therefore can stabilize a bit pattern. We also developed parallel codes for efficient simulation of multiple soliton channels and implemented diagnostic tools based on a novel hybrid numerical and analytical approach.

Publications

Burtsev, S., and I. Gabitov, "Four Wave Mixing in Fiber Links with Dispersion Management" (submitted to *Opt. Comm.*).

Burtsev, S., et al., "NRZ-to-Soliton Data Conversion Problem" (submitted to *J. Opt. Soc. Am. B*).

Burtsev, S., et al., "Numerical Algorithms for the Direct Spectral Transform with Applications to Nonlinear Schrodinger Type Systems" (to be published in *J. Comput. Phys.*).

Zoldi, S., et al., "Parallel Implementations of the Split-Step Fourier Method for Solving Nonlinear Schrodinger Systems" (submitted to *SIAM News*).

Quantum Information Capacity in the Presence of Noise

97057

Raymond Laflamme

Many researchers in the field of computing now believe that quantum computation (which operates at the atomic level rather than with electronic components) may be a likely outgrowth of the drive toward the miniaturization of circuits (Moore's law). However, before the advantages of quantum computers can be realized, much work remains to be done to design practicable ways to implement quantum error correction and fault-tolerant computation and to better understand the physical nature of actual noise processes that occur in practicable devices. It is important to stress that even in the first generation of quantum computers, error correction will play an important role. Just to factor the number 15 on a quantum computer will require roughly 1,000 gates (i.e., operations at the atomic level) with the present algorithms—a very difficult task to achieve without any form of error control.

We have recently discovered that quantum computation can be protected against errors by applying quantum mechanical techniques of error correction. By using hierarchical (concatenated) quantum codes, we can successfully perform arbitrarily long computations, providing 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 hold for specific (but realistic) types of noise.

Publications

Habib, S., et al., "Decoherence, Chaos and the Corresponding Principle," *Phys. Rev. Lett.* **80**, 4361 (1998).

Knill, E., et al., "Resilient Quantum Computation," *Science* **279**, 342 (1998).

Paz, J.P., and W.H. Zurek, "Continuous Quantum Error Corrections," *Proc. R. Soc. London, Ser. A* **454**, 355 (1998).

Zurek, W.H., "Decoherence, Einselections and the Existential Interpretation (the Rough Guide)," *Philosophical Transactions: Mathematical, Physical and Engineering Sciences* **356**, 1793 (1998).

Completely Parallel ILU Preconditioning for Solution of Linear Equations

97036

Wayne Joubert

We are developing completely parallel versions of incomplete Cholesky and incomplete lower-upper (ILU) preconditioners for solving sparse systems of linear equations. It is widely recognized that the most computationally expensive part of many modeling and simulation processes is the solution of the sparse linear equations that arise from the finite-difference or finite element discretizations. Unfortunately, the widely used and powerful incomplete Cholesky preconditioning and related techniques are sequential in nature, and thus they have not been effective on parallel computer platforms.

However, we have developed a new formulation of the ILU algorithm that solves this problem for an important class of linear systems of interest for large-scale modeling and simulation applications. The technique gives almost perfect speedup even on large numbers of processors without sacrificing the strong convergence properties of the global ILU and modified ILU (MILU) preconditioners. We are applying these parallelization techniques to a broad range of structured problems, performing basic research on the convergence properties and parallelization possibilities of ILU preconditioners, and exploring the possibility of using these techniques for the preconditioning of unstructured problems.

This year we have made an extensive comparative numerical study of parallelization strategies for ILU preconditioning techniques. To our knowledge this is the first study of its type performed, and the study has produced a number of surprising results—for example, the effectiveness of multicolor orderings for certain problems. We have completed and written up our numerical experiments with orderings for small problems and the associated theoretical results. Our work on parallel ILU strategies for structured problems has been completed, and the paper is currently in review.

Investigation into applying similar parallelization strategies to general unstructured problems has shown that parallelizing the unstructured case with the same approach would be considerably more difficult, for a number of reasons. Future research will focus on alternative approaches.

In addition, we have spent some time investigating hybrid ILU/multilevel methods and have developed a computer code to experiment with these methods. As a result, we have developed some theoretical insight into the behavior of these methods that will hopefully lead to improved algorithmic approaches.

Publications

Benzi, M., et al., "Numerical Experiments with Parallel Orderings for ILU Preconditioners" (submitted to *Electron. Trans. Numer. Anal.*).

Joubert, W., et al., "Fully Parallel Global M/ILU Preconditioning for 3-D Structured Problems" (to be published in *SIAM J. Sci. Comput.*).

3-D, Unstructured, Hexahedral-Mesh S_n Transport Methods

97035

Jim Morel

The objective of this project 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, but the successful use of hybrid finite element meshes will result in a unique and powerful capability for modeling complex, 3-D geometries.

This year we continued to verify the 3-D, hexahedral-mesh research transport code that we developed during the first year of this project by performing various benchmark calculations. To add time dependence to our code, we used a new linear-discontinuous, finite element, temporal-discretization technique. In addition, we generalized our two-level, diffusion-based, multigrid technique for accelerating basic source iterations to make it compatible with the new temporal-discretization technique.

We also generalized our algorithm to solve the Boltzmann-Fokker-Planck equation for charged-particle transport. This modification required the addition of a linear-discontinuous, finite element discretization for the continuous-slowing-down operator, together with modifications to our multigrid acceleration technique. Finally, we developed and implemented a parallel solution technique, based upon spatial domain decomposition, for solving our discrete, hexahedral-mesh transport equations on massively parallel computing platforms.

A Self-Consistent Multiscale Theory of Internal Wave, Mean-Flow Interactions in the Ocean

96286

Darryl Holm

To quantify uncertainty in ocean and climate modeling caused by turbulence, we formulated these models as Euler–Poincaré systems (that is, the Lagrangian analog of Lie-Poisson Hamiltonian systems) defined on semidirect-product Lie algebras. This is the natural mathematical structure of the nonlinearity in continuum dynamics. We first gave a derivation of the Euler–Poincaré equations for a parameter-dependent Lagrangian by using a variational principle of Lagrange d'Alembert type. Then we derived an abstract Kelvin-Noether circulation theorem for these equations and determined how to apply them to the analysis of continuum mechanics and fluid dynamics.

By using asymptotic expansions, two-timing, and averaging in Hamilton's principle for an ideal incompressible fluid, then introducing viscosity semiempirically as diffusion of an appropriate momentum, we used our Euler–Poincaré theory to derive a new closure model for three-dimensional (3-D) incompressible turbulence. The steady solutions of this model compared well with experimental data for mean fluid-velocity profiles in pipes and channels at high Reynolds numbers. We also used the resulting equations as the basis for a Large Eddy Simulation (LES) numerical model. The foregoing may be regarded as a new, one-point closure model for incompressible turbulence.

We extended this work by developing a new, second-moment closure model for 3-D incompressible turbulence. This model acts like an adaptive LES numerical model and gives a dynamical equation for how the diffusivity tensor for passive scalars responds to shear forcing. We used this approach in formulating a new, second-moment closure model for 3-D oceanic turbulence. The utility of this model for large-scale computations in ocean and climate modeling is now being tested.

Publications

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Cendra, H., et al., "The Maxwell-Vlasov Equations in Euler–Poincaré Form," *J. Math. Phys.* **39**, 3138 (1998).

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A Theoretical Description of Inhomogeneous Turbulence

97018

Leaf Turner

Most of the current fundamental theoretical calculations elucidating turbulence focus on the statistically homogeneous case, a case that is never realized in nature. All mean quantities of such models are spatially uninteresting, having vanishing spatial gradients. We are developing a state-of-the-art analytical and numerical model of inhomogeneous turbulence. We expect this work to have implications for improved phenomenological models for engineering applications that address drag forces on transportation vehicles, energy efficiency in automotive combustion, meteorological phenomena, forest fires, volcanic eruptions, and weapons physics.

During this past year we have used our fundamental theory of inhomogeneous fluid turbulence to study the spatial structures of macroscopic mean quantities, structures that are intrinsically interesting and significant for the dynamics of any natural (necessarily inhomogeneous) flows. We have numerically calculated triple-velocity correlations vs. pressure-velocity correlations from "first principles" in the context of the Eddy-Damped Quasi-Normal Markovian model (see first figure). We have also demonstrated theoretically and numerically that a mean fluid-flow structure can develop in a channel out of the interaction of the turbulent eddies even when no mean flow is initially present (see second and third figures). Finally, we have calculated two-point velocity correlations and have convincingly demonstrated spectacular agreement of direct numerical simulations with our formalism assuming the random-phase approximation (see fourth figure).

Publications

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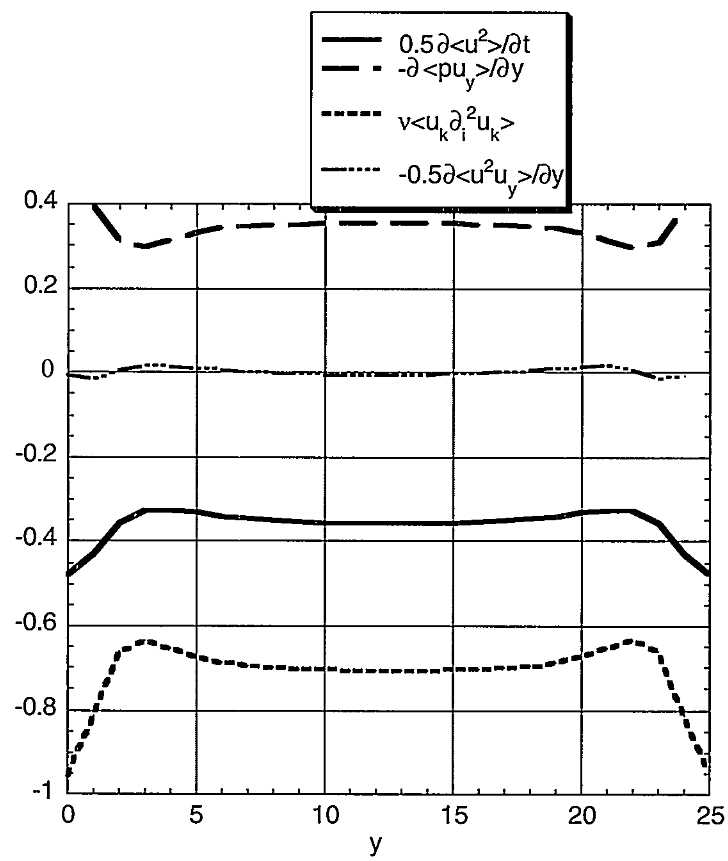
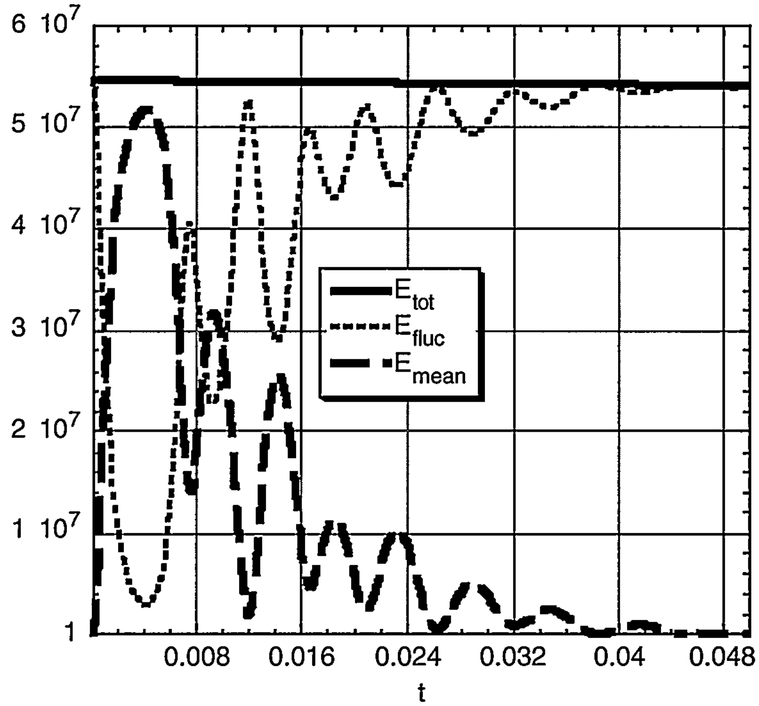
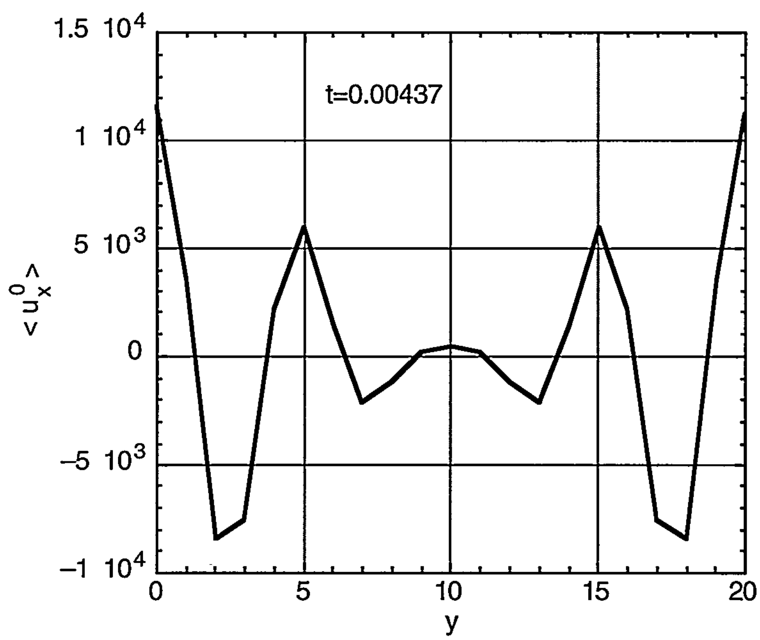


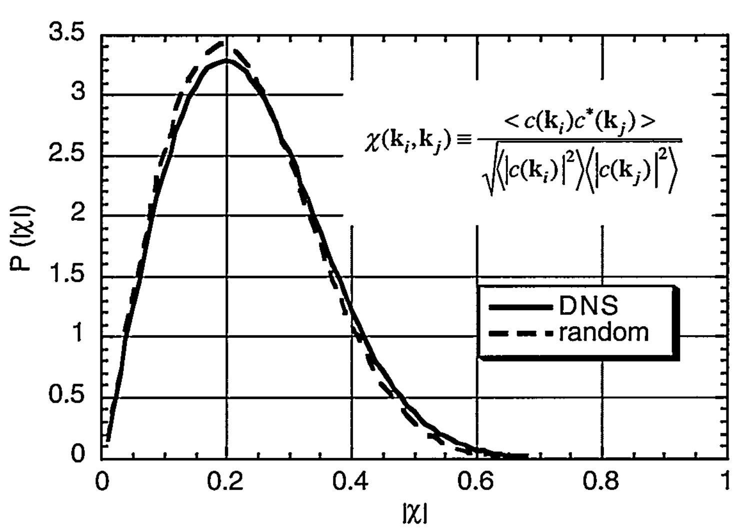
Shown are the negative gradients of both the pressure-velocity correlation and half of the triple-velocity correlation when the mass density is taken to be unity. When we add these gradients to the depicted local rate of energy density dissipation, we obtain the depicted local rate of change of kinetic energy density. The flow is bounded between two planar free-slip boundaries normal to the y-axis.



The mean flow kinetic energy, absent initially, undergoes remarkably large oscillations that damp out as a result of the nonlinear terms stirring away the initially non-mirror-symmetric components of the energy spectrum. Observe that the sloshing about of the mean flow energy comes about through a compensating sloshing of the kinetic energy of the fluctuations, conserving the total energy, aside from a gradual viscous dissipation of total energy.



A snapshot of a course-grid calculation of a mean-flow velocity profile. Notice that momentum is conserved because the integral of the profile across the channel is zero.



Comparison of two probability density functions of the correlation between two spectral coefficients: one calculated from a random phase distribution and the other from an ensemble of direct numerical simulations (DNS) of the Navier-Stokes equation of an incompressible fluid. The very slight discrepancy is due to the presence of a finite viscosity.

Diffusion in Porous Media and Stochastic Advection

97033

Shi-Yi Chen

We are combining numerical simulation and theoretical research to study diffusion by intermittent stochastic velocity fields. The principal application is to diffusion of substances in porous media. In the past year we have obtained significant results.

We computed the advection of a passive scalar field by a rapidly decorrelating, random velocity field with power-law scaling by simulations in a cyclic square at resolutions of 4096^2 and 8192^2 grid points. We measured structure functions of the scalar field and determined inertial-range scaling exponents; we then found the conditional mean of the scalar-field dissipation term and its moments. Our results compared well with theoretical predictions and with other recent simulations.

We determined the cylindrical vortex profile that yields maximal moment ratios, and we have used the moment ratios for cylindrical vortices to interpret differences in scale dependence of enstrophy and dissipation previously found in numerical simulations.

We developed a hierarchical model for the joint moments of the passive scalar dissipation and the velocity dissipation in fluid turbulence. This model predicts that the joint probability density function of the dissipations is a bivariate log-Poisson. We calculated the scaling exponents of structure functions of the passive scalar for this hierarchical model; our results show good agreement with the results of direct numerical simulations and experiments.

We studied the fractal properties of passive scalar isosurfaces in fluid turbulence using data from direct numerical simulation. For the two-dimensional case, the velocity field varies rapidly and has Gaussian statistics yielding a fractal dimension of 1.38. We have solved the three-dimensional Navier-Stokes equations and the advection-diffusion equation simultaneously. Our results yield constant fractal dimensions for any given isosurface, in agreement with theoretical predictions and previous experimental measurements. Our results support the idea of self-similarity of the passive scalar isosurfaces in fluid turbulence.

Using direct numerical simulations and large-eddy simulations of velocity and passive scalar in isotropic turbulence, we examined directly and quantitatively the refined similarity hypotheses as applied to passive scalar fields with Prandtl number of order one.

Publications

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Foias, C., et al., "A Connection Between the Camassa-Holm Equations and Turbulence Flows in Channels and Pipes" (submitted to *Phys. Fluids*).

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He, G., et al., "Hierarchy of Structure Functions for Passive Scalars Advected by Turbulent Flows," *Phys. Lett. A* **246**, 135 (1998).

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He, X., et al., "On Three-Dimensional Rayleigh-Taylor Instability" (submitted to *Phys. Fluids*).

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Unitary Symmetry, Discrete Mathematics, and Combinatorics

98032

William Chen

There is a wealth of underlying discrete mathematics that point to new directions in the application of symmetry methods to many-particle quantum systems. One of the most important of these directions is the primary role of MacMahon's Master Theorem, a foundational result in combinatorics, in deriving generating functions for the basic invariants of angular momentum theory. These invariants are the basic objects needed to unravel the spectra of molecules, atoms, nuclei, and particles. This confluence of physical theory and modern combinatorics points the way toward a comprehensive theory of such invariants. Unitary symmetry and discrete dynamical systems also involve discrete mathematics amenable to combinatorial methods. These subjects have been advanced by the discovery of fundamental results that bring group theory under the purview of combinatorics and of the relationship between the combinatorics of abstract words and the behavior of dynamical systems.

One of our major accomplishments during the past year includes proof that the MacMahon's Master Theorem and the concept of a double Pfaffian give completely the generating function for all recoupling coefficients in the quantum theory of angular momentum.

Another accomplishment is a combinatorial proof of a multiplication law for a class of polynomial functions defined over the elements of an arbitrary square matrix that gives group multiplication when the matrices form a group.

We also developed the proof that a certain set of ordered words on two letters alternates in their parity. This result identifies the intervals in parameter space in which bifurcations of maps of the interval occur.

Publications

Chen, W., and J.D. Louck, "The Combinatorics of a Class of Representation Functions" (to be published in *Adv. Math.*).

Chen, W., and J.D. Louck, "Enumeration of Cubic Graphs by Inclusion-Exclusion" (to be published in *J. Combin. Theory A*).

Chen, W., et al., "Angular Momentum and Umbral Calculus" (to be published in *Ann. Combin.*).

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Louck, J.D., and W. Chen, "Generating Functions for SU(2) Binary Recoupling Coefficients," in *Proceedings of the Fifth Conference on Symmetry and Structural Properties of Condensed Matter* (World Scientific, Singapore, in press).

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High-Performance Storage System (HPSS)

98046

R. Gary Lee

This project involves the High-Performance Storage System (HPSS) at Los Alamos and the specific effort to help develop a next-generation computational data storage system that will be able to store the extremely large volumes (petabytes) of data that are expected to be generated by the new ultracomputing platforms. Such a storage and retrieval system will need an ability to access this data at very high rates (in the tens to hundreds of megabytes per second).

Improvements in, and innovative approaches to, archival storage are needed to meet the future performance needs of users of ultracomputing systems. Toward this end, definitive performance analyses are needed to understand how the various elements of such advanced storage systems could be optimized.

Our specific objective is to better understand the potential impact of newer symmetric multiprocessor (SMP) architectures on storage systems. To meet this objective, we are investigating how HPSS would support a new 64-bit SMP architecture.

This year we carried out performance testing with HPSS. These tests, which are ongoing, have allowed us to improve storage system performance by 25% and to determine system bottlenecks where further improvements are needed. For example, we discovered that the transaction management software was limiting the performance on small file creates. We increased the segment size to reduce the number of transactions and improve performance somewhat. Further work with the transaction manager is needed to improve performance in this area.

Fiber-Optic Communications Using Solitons (FOCUS)

96326

Darryl Holm

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 Schrödinger equation, which 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 devices can be designed to actually exploit energy losses and pulse 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 analyzing individual devices to modeling and optimizing complete networks.

This year we continued our work on the mathematical models of transmission systems with periodically distributed components. We addressed three main issues: (1) modeling the fiber links with periodically distributed nonlinear semiconductor amplifiers, (2) developing the theory of optical signal transmission in the links with periodical dispersion maps, and (3) investigating the adiabatic reshaping of solitons with periodically distributed nonlinear elements—nonlinear amplifying-loop mirrors. We developed a mathematical model of pulse propagation in an optical-fiber link with in-line lumped semiconductor optical amplifiers. We described pulse propagation in the fiber by means of the nonlinear Schrödinger equation, and we used the Maxwell-Bloch type of equation to model the semiconductor amplifier. We derived new equations that describe optical transmission links with dispersion management. Currently, this dispersion map approach has become very popular and is promising for both upgrading the capacity of installed fiber networks as well as for creating new fiber links. We proposed regimes for an adiabatic reshaping technique that provide stable data transmission through optical-fiber links. These regimes exploit the negative slope characteristic of the nonlinear amplifying-loop mirror and, in addition to reshaping the solitons, can help suppress the accumulation of continuous radiation.

New Ways of Representing Functions

97034

George Zweig

We are investigating new sets of expansion functions that provide efficient representations of functions that have structure at many scales. Unlike the wavelets of continuous wavelet transforms, the new sets of expansion functions will have a longest scale and will not have the same shape at all scales. We will develop fast transform algorithms based on those functions by solving a partial differential equation for the transform coefficients. We expect significant applications to pattern recognition and noise removal. In particular, the primary objective is to find new useful ways of representing signals and images.

We have expressed medical magnetic resonance (MR) images in terms of their three-dimensional integer wavelet transform coefficients. This accomplishment allows us to create a novel lossless compression algorithm using zero-tree coding. We have extended the embedded zero-tree-coding algorithm to three dimensions and used context-based, adaptive arithmetic coding to improve its performance. The algorithm efficiently encodes image volumes by exploiting the dependencies in all three dimensions, while enabling lossy and lossless compression from the same bit stream. We obtained results on lossless compression of MR images and compared the results with those from other lossless compression algorithms. The progressive performance of the algorithm was also compared with the performance of other lossy progressive coding algorithms. For representative images, the algorithm produced an average decrease in compressed file sizes for MR images that was 25% greater than that achieved with the best available two-dimensional lossless compression techniques.

Publications

Bilgin, A., et al., "Loss Medical Image Compression Using Three-Dimensional Integer Wavelet Transforms" (submitted to *IEEE Trans. Med. Imaging*).

Invariant Discretization for Computational Gas Dynamics

98031

Mikhail Shashkov

The primary goal of this project is to create invariant discretizations that do not depend on any particular choice of coordinate system. Our objective is to formulate finite-difference schemes in terms of the coordinate-invariant characteristics of the grid, such as lengths of the edges, areas of the faces, volumes of the cells, and angles between edges. These new methods will more accurately preserve the symmetries and group properties of the underlying differential equations. We are applying these new methods to modeling problems where preservation of symmetry is very important; e.g., where small departures from spherical symmetry due to discrete errors can lead to unacceptably large errors in systems with large convergence ratios. Also, the uncertainty as to whether a nonsymmetric result is a result of numerical errors or of the physical design severely limits predictive capabilities and understanding of the dynamical behavior of such systems.

During the past year we developed two methods of reconstruction of the curvilinear grid for given logically rectangular distribution of points. Both methods are exact for straight lines and circles. The simplest method reconstructs the curve between two nodes as an arc of some local circle. This method is simple but produces curves with discontinuous tangents. We also constructed a method based on blending of local circles, which produces curves with smooth tangents. We constructed a new, conservative finite-difference method for Lagrangian gas dynamics in cylindrical coordinates that preserves plane, cylindrical, and spherical symmetries. This new method uses constructed curvilinear grids.

Publications

Margolin, L., and M. Shashkov, "Using a Curvilinear Grid to Construct Symmetry-Preserving Discretizations for Lagrangian Gas Dynamics" (to be published in *Comput. Phys.*).

High-Quality Finite-Difference Schemes for Partial Differential Equations and Discrete Vector and Tensor Analysis

96327

Mikhail Shashkov

Our primary goal 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 used discrete analogs of the invariant first-order operators—div, grad, and curl—on a logically rectangular grid to construct mimetic finite-difference methods for Maxwell's equations and equations of magnetic field diffusion. These new schemes are conservative, satisfy discrete analogs of divergence-free conditions exactly, and are free of spurious modes. We described how to apply our methodology to construct mimetic finite-difference methods for equations of solid mechanics, that is, how to discretize the gradient of a vector and the divergence of a tensor. We also described how to approximate boundary conditions for partial differential equations in the framework of our approach. In addition, we published a series of papers related to application of our methodology to discretization of the equations of Lagrangian gas dynamics.

Publications

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Caramana, E.J., et al., "The Construction of Compatible Hydrodynamics Algorithms Utilizing Conservation of Total Energy," *J. Comput. Phys.* **146**, 227 (1998).

Caramana, E.J., et al., "Formulation of Artificial Viscosity for Multi-Dimensional Shock Wave Problems," *J. Comput. Phys.* **144**, 70 (1998).

Hyman, J., and M. Shashkov, "Approximation of Boundary Conditions for Mimetic Finite-Difference Methods," *Int. J. Comput. Math. Appl.* **36**, 79 (1998).

Hyman, J., and M. Shashkov, "Mimetic Discretization for Maxwell's Equations and Equations of Magnetic Diffusion," in *Mathematical and Numerical Aspects of Wave Propagation, Proc. Int. Conf. Math. Numer. Aspects of Wave Propag., 4th*, J.A. DeSanto, Ed. (SIAM, Philadelphia, 1998) p. 561.

Hyman, J., and M. Shashkov, "Mimetic Discretization for Maxwell's Equations and Equations of Magnetic Diffusion" (submitted to *J. Comput. Phys.*).

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Shashkov, M., et al., "Local Reconstruction of a Vector Field from Its Components on the Faces of Grid Cells," *J. Comput. Phys.* **139**, 406 (1998).

Atomic, Molecular, Optical, and Plasma Physics, Fluids, and Beams

Nonlinear Atom Optics

96006

Peter Milonni

We are exploring aspects of nonlinear atom optics and condensates, particularly atomic matter wave analogs of nonlinear optical effects such as wave mixing, solitary wave formation, and spatial pattern formation. The recent observations of Bose-Einstein condensation in trapped cold-atom systems have led us to focus, especially during the past year, on atom-wave aspects of these condensates.

We have carried out calculations of the two-photon absorption spectrum that was used in the first experimental demonstration of condensation of atomic hydrogen by researchers at the Massachusetts Institute of Technology. In calculations thus far, we have dealt with only one spatial dimension, for simplicity; we are currently extending the calculations to three dimensions. We obtain a spectral width that appears to be consistent with the preliminary experimental reports. It is not clear yet what the detailed shape of the experimental spectra is, as the reports thus far are preliminary and have low resolution.

We are considering the properties of two-phase condensates produced when an applied magnetic field produces a Feshbach resonance. The interactions that create a quasi-bound molecule in the binary-atom Feshbach resonance create a second molecular condensate in the many-body Bose-Einstein condensate. The coexisting condensates interact by coherently exchanging pairs of atoms. The contribution to the many-body energy of this novel type of intercondensate tunneling depends on the relative phase of the atomic and molecular condensates, and reaches its minimum if the phases of the condensates differ by the constant p. The population dynamics of the double condensate reveal that the system has two distinct stationary states: the above-mentioned state with condensates of opposite sign and a state with condensates of the same sign. The particular state that the experimental system finds itself in depends on its history. If the condensate was brought near resonance by adiabatically decreasing the detuning, starting far above resonance, the condensates will have the same sign. If, on the other hand, the detuning was increased from below resonance, the atomic and molecular condensates can be of opposite sign. Interestingly, although the same sign mixture maximizes the energy, the corresponding system can nevertheless be stable. We also are investigating the implications of the molecular loss processes and the analogies with nonlinear optical processes.

Publications

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Fearn, H., et al., "Comments on 'The Ewald-Oseen Extinction Theorem,' " *Optics and Photonics News,* 4 (1998).

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Optical Wave Packets (Optical Bullets): A New Diffraction-Free Form of Light Travel

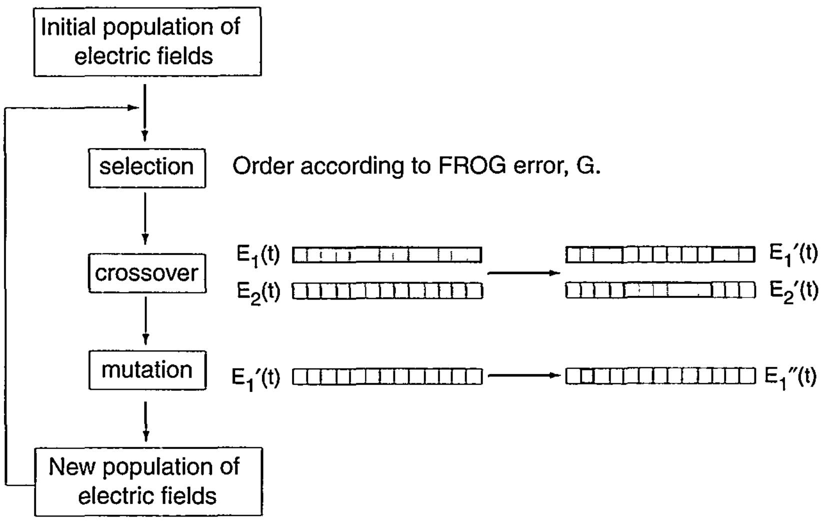
96003

David Funk

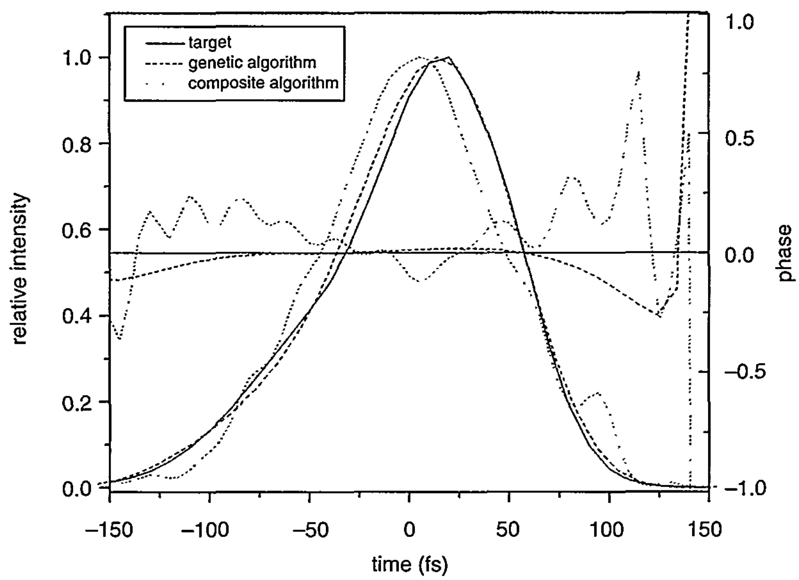
We are investigating "optical bullets," that is, the spontaneous formation and propagation of self-confined packets of light in air. At sufficiently high intensities, short (100-fs) laser pulses will not only self-focus but will propagate as fine filaments until the intensity of the pulse falls below a measurable critical value. The intensities are great enough that the pulse could leave a channel of ionized gas in its wake, giving these packets value as lightning discharge aids.

In addition, the packets may function as light sources for extremely long range light detection and ranging (LIDAR). Normal LIDAR is limited by diffraction, resulting in only a fraction of the light reaching long range with sufficient intensity to allow the measurement of returned signals. These pulses overcome this problem by concentrating the light at distances beyond what the diffraction limit allows. We are analyzing the phase changes and structure of these pulses above, near, and below the critical value to resolve the origin of this phenomenon and to determine the usefulness of optical bullets as tools.

We 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. This year we developed a new method to enhance the phase retrieval process using frequency-resolved optical gating (FROG) based on genetic algorithms (see accompanying figures).



A schematic of a single generation in the genetic algorithm, illustrating the selection, crossover, and mutation operators.



Target intensity and phase (solid lines), intensity and phase retrieved from the genetic algorithm (dashed lines), and the intensity and phase from the composite (Femtosoft) algorithm (dotted lines) in the second harmonic generation FROG case. The genetic algorithm returned an error of only 0.00175, and the composite algorithm returned an error of 0.00256. A generalized projection algorithm (result not shown) returned an error of 0.00270.

Publications

Nicholson, J.W., et al., "Evolving FROGS: Phase Retrieval from Frequency-Resolved Optical Gating Measurement Using Genetic Algorithms" (to be published in *Opt. Lett.*).

Determination of Optical-Field Ionization Dynamics in Plasmas through Direct Measurement of the Optical Phase Change

96367

Antoinette Taylor

The detailed dynamics of an atom in a strong laser field is rich in both interesting physics and potential applications. Ionization-based high-harmonic generation enables efficient and tunable sources of short-wave-length femtosecond radiation and may yield the first attosecond pulses. 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.

This year we performed the first quantitative measurements of high-field ionization rates of noble gases and diatomic molecules using the technique of multipulse interferometric frequency-resolved optical gating (MI-FROG). As MI-FROG recovers, to all orders, the phase difference between the pumped and unpumped probe pulses, it enables the determination of sub-pulse-width time-resolved phase and frequency shifts impressed upon the probe by the ionization front, independent of the probe structure. The measured ionization rates for noble gases, shown in the first figure, agree with those that were calculated by using a one-dimensional plasma-fluid model and Ammosov rates. In the case of homonuclear diatomic gases H_2 and N_2 (the second figure), we find similar close agreement with the model using published ionization potentials. However, in the case of O_2, we find a large discrepancy, as shown in the second figure, with tunneling theory overpredicting the ionization rate.

Publications

Nicholson, J.W., et al., "Evolving FROGs: Phase Retrieval from Frequency-Resolved Optical Gating Measurements using Genetic Algorithms" (submitted to *Opt. Lett.*).

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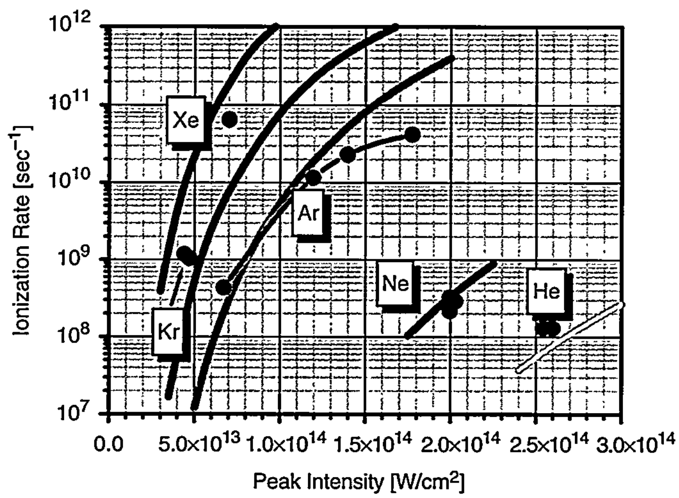
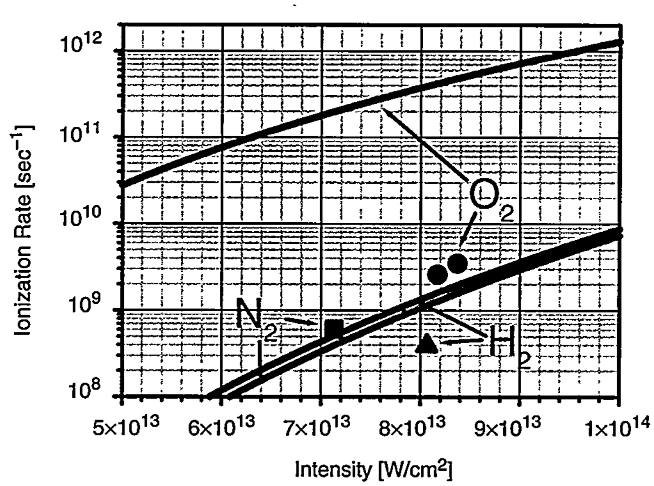


Measured and modeled ionization rates for noble gases.



Measured and modeled ionization rates for homonuclear diatomic molecules H_2, N_2, and O_2.

Artificial Atoms Probed by Femtosecond Pulses: Evolution of Optical Properties during the Bulk-Atomic Transformation

98001

Victor Klimov

The goal of this project is to study the evolution of electronic and optical properties of materials during the transformation from atomic to bulk-crystalline regimes. As model systems for these studies, we use nanoclusters with sizes from ~200 to ~10 Å generated by methods of colloidal chemistry. We concentrate on the following problems: (1) the evolution of the energy spectra during the bulk-atomic transformation, (2) influence of spatial confinement on the structure and intensities of optical transitions, (3) confinement-induced changes in mechanisms for light emission and optical nonlinearity, (4) effects of spatial confinement and size quantization on energy relaxation, (5) the role of surface/interface states in relaxation processes, and (6) the evolution of many-particle interactions during the transformation from bulk to atomic-like regimes.

Our studies of energy relaxation in CdSe nanoclusters with sizes ranging from 12 to 40 Å indicate that, in contrast to the predictions of "phonon bottleneck theories," the energy relaxation in nanoclusters is extremely fast (subpicosecond time scale), which is likely due to confinement-enhanced Auger-type energy transfer. Our comparative studies of mechanisms for optical nonlinearities and carrier relaxation in silicon nanoclusters and bulk silicon indicate that carrier relaxation in silicon nanoclusters is dominated by ultrafast surface trapping, leading to depopulation of quantized states on the 1- to 2-ps time scale. In both bulk silicon and silicon nanoclusters, optical nonlinearity is a result of excited-state absorption induced by intraband transitions. Application of femtosecond transient absorption in the visible and near-IR spectral ranges allowed us to separate electron and hole relaxation paths and to map the structure of inter- and intraband optical transitions in CdSe and CdS nanocrystals.

Publications

Klimov, V., "Femtosecond Broadband Spectroscopy of Inter- and Intra-band Transitions in Semiconductor Quantum Dots," in *Semiconductor Science and Technology '98*, Vol. 10, No. 11 (Institute for Advanced Physics Studies, La Jolla, CA, 1998), p. 117.

Klimov, V., "Linear and Nonlinear Optical Spectroscopy of Semiconductor Nanocrystals," in *Handbook of Nanostructure Materials and Nanotechnology*, H. Nalwa, Ed. (Academic Press, San Diego, CA, in press).

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Stochastic Resonance in Arrays with Tunable Nonlinearity and Coupling

98003

Alp Findikoglu

Nonlinear dielectrics, such as $SrTiO_3$, present unique opportunities to develop practical electrically tunable devices and also to explore novel scientific and technological concepts that exploit strong nonlinearities. Stochastic resonance (SR), a phenomenon in which random noise enhances a nonlinear system's response to a deterministic signal, presents one such opportunity. In particular, SR has the potential to improve the output signal-to-noise ratio of a signal-processing device in the presence of noise.

To study the potential of SR, we are designing compact, nonlinear, coplanar waveguide devices. We have begun by investigating the effect of noise on the nonlinear response using two microwave signals (at 700 MHz and at 2 GHz), direct-current bias, and noise (from 1 to 550 MHz) as the input and looking at the mixed microwave output, which is generated by the nonlinearity at the sum and difference of these two microwave signal frequencies (see first figure). The mixed microwave output increases by about 6 dB with the addition of noise (see second figure). However, we have not yet demonstrated signal-to-noise ratio increase unambiguously because it requires separation of external and internal noises in the noise floor.

Our theoretical modeling of the coplanar waveguide system uses a set of coupled partial differential equations in a nonlinear medium with two boundaries. The nonlinearity depends on the applied bias and is used to determine the input-output curve of the system, which allows us to calculate the conditions for the appearance of SR. Our setup will enable us to test fundamental theories about the relationship between nonlinearity and noise in a spatially extended nonequilibrium system.

Publications

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Findikoglu, A.T., et al., "New Potential Applications of Nonlinear Dielectrics: Microwave Solitons and Stochastic Resonance," *Integr. Ferroelectr.* **22**, 259 (1998).

Jansons, K., and G. Lythe, "Stochastic Calculus: Application to Dynamic Bifurcations and Threshold Crossings," *J. Stat. Phys.* **90**, 227 (1998).

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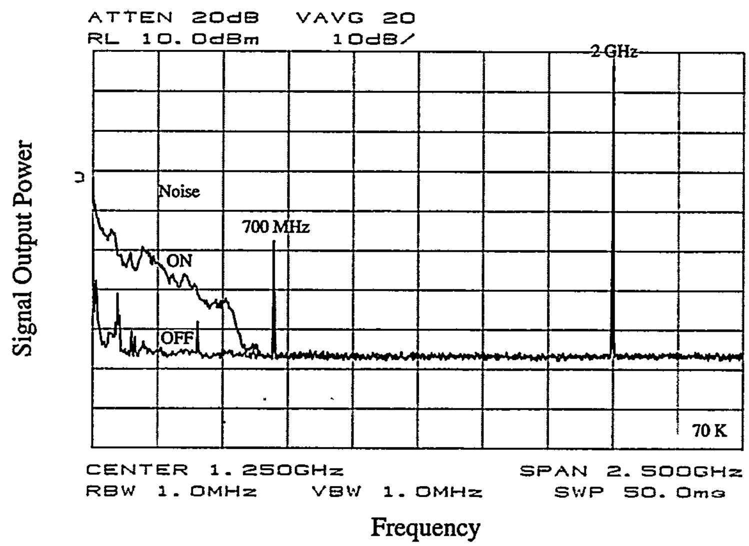
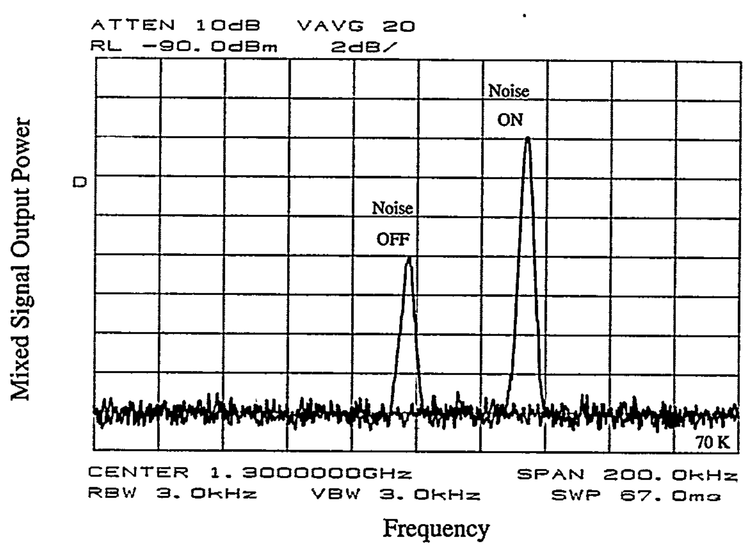


Output signal spectra with noise on and off.



Mixed microwave output with noise on and off.

Self-Amplified Spontaneous-Emission–Related Science Studies

98054

Richard Sheffield

The use of light sources has grown substantially. For example, present x-ray light sources are extensively used in the study of protein crystals and in x-ray absorption spectroscopy. A fourth-generation light source enhances, by 1 order of magnitude or more, some or all of the performance parameters of the presently used third-generation sources. Los Alamos is part of a collaboration proposing to build the Linac Coherent Light Source at the Stanford Linear Accelerator, a light source that is based on a self-amplified spontaneous-emission (SASE), free-electron laser (FEL). As part of that collaboration, we participated in the planning of the visible SASE (VISA). At this point, we have completed the initial design for the VISA and are well into its fabrication. We are also responsible for developing a plan for experimental work that is to be done at Brookhaven National Laboratory.

We also performed an experiment on an FEL at Los Alamos that demonstrated the fundamental physics underpinning the SASE. In FELs, an electron beam interacts with light, and the light builds up in power. One result of this interaction is the development of a periodic modulation of the electron beam, called microbunching. This microbunching, which occurs at the wavelength of the FEL light, forms the core of the FEL gain process. The microbunched charge produces coherent radiation that adds to the light field, giving exponential gain. We have demonstrated, for the first time, two critical aspects of the microbunching-induced coherent transition radiation (CTR)—the narrowing of the angular spectrum and the formation of line structure in the wavelength spectrum. Our observations have verified the microbunching-induced CTR predicted by theoretical analysis of the SASE theory.

Publications

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Plasma-Wakefield Accelerator

97010

Bruce Carlsten

Future advanced accelerators for high-energy linear colliders and short-wavelength free-electron lasers will require higher-gradient methods of acceleration than those currently available, which are limited to about 100 MV/m. High-density plasma excited by lasers has been shown to support gradients in excess of several tens of GV/m; however, it is not easy to excite such large gradients in plasmas with the required phase and amplitude control for accelerator applications. In this project, our goal is to demonstrate gradients of 1 GV/m in an electron-driven plasma-wakefield accelerator (PWA). The advantage of this type PWA over a laser-driven PWA is that the plasma-oscillation phase is easily controlled. In addition, we wish to demonstrate the high transformer ratios and other second-order effects required to scale the PWA to a practical device. Previous measurements on the Subpicosecond Accelerator (SPA) at Los Alamos showed plasma gradients of several hundred MV/m, but the resolution of the measurements was limited by instabilities in the SPA drive laser.

Our major efforts this year centered on upgrading the drive laser and constructing a differential interferometer that could measure transient plasma densities of $10^{16}/cm^3$. We made good progress on the interferometer, and it should be running in early next year. The drive laser work required more effort than originally thought, but we have now completed it. The compression of the electron bunch used to drive the plasma oscillations was better modeled in the upgraded drive laser, and we expect better control of the plasma excitation.

Quantum Computation and Nuclear Magnetic Resonance

98004

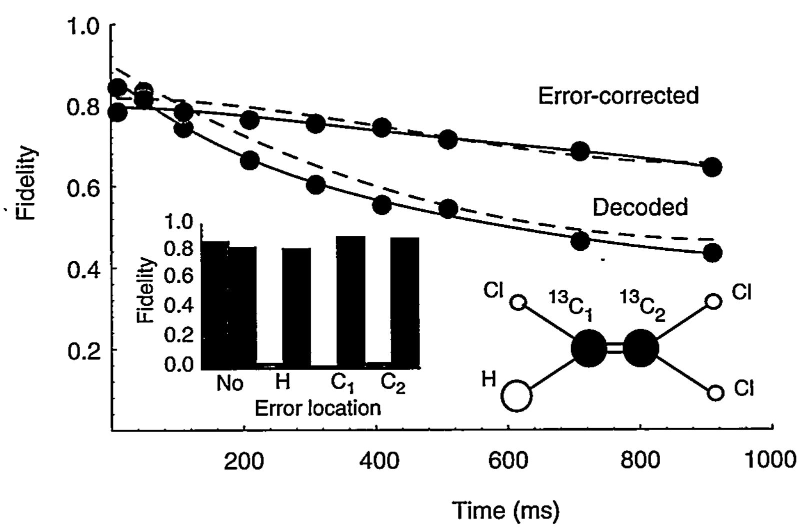
Raymond Laflamme

Recently, quantum computation has undergone a dramatic evolution—moving from an academic curiosity to a field that may revolutionize computer science, impact national security, and hold economic promise. A key to this evolution has been the discovery of quantum-error-correcting codes and the implementation of individual quantum gates (i.e., computer operations that follow the rules of quantum mechanics). These developments are important milestones on the way to replacing the electronic components of today's computers with much more powerful ones that operate at the atomic level. Based on quantum bits that can have any value between zero and one, quantum computers promise enormous increases in both how much and how quickly information can be manipulated and stored.

Our project aims at advancing quantum computation by (1) understanding quantum information capacity in the presence of noise and studying its implications for quantum logic circuits through computer simulations involving only a few quantum bits, and (2) using nuclear magnetic resonance (NMR) and our simulation results to implement quantum computations with small molecules (i.e., ones involving 2–10 nuclear spins). Our work is focused on creating Greenberger-Horne-Zeilinger (GHZ) states of matter, studying multiparticle entanglement, and demonstrating quantum error correction and quantum Fourier transforms.

In our first year we have created a GHZ state, which is an entangled state of three quantum bits. Our alanine molecule is shown as an inset in the accompanying figure. This is the first time that such a state of matter has been made in the laboratory. Its development was an essential precursor to implementing quantum error correction.

We have also achieved the first experimental implementations of quantum error correction and confirmed its expected impact on stabilizing the information stored in our alanine molecule (see accompanying figure). Our experiment carried out the 3-bit code for phase errors using liquid-state NMR.



Demonstration of the effect of quantum error correction in our alanine molecule (shown at lower right). The curves trace the amount of information preserved with error correction (black) and without error correction (gray); in both cases, the solid curve is experimental data, and the dashed is theoretical prediction. The ideal curve would be a horizontal line. Time, shown here in milliseconds, is proportional to the amount of error introduced. The histogram (left inset) shows errors introduced on specific hydrogen and carbon atoms and indicates their disastrous effects when left uncorrected (gray columns). For more details on this demonstration, see Cory et al.

Publications

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Laflamme, R., et al., "NMR GHZ," *Philos. Trans.: Math., Phys. Eng. Sci.* **356**, 1941 (1998).

Transient Quantum Mechanical Processes

96008

Lee Collins

Our principal objective centers on the development of sophisticated computational techniques for solving the time-dependent Schrödinger equation that governs the evolution of quantum mechanical systems. We have perfected two complementary methods, discrete-variable representation and a real-space-product formula, that show great promise for solving these complicated temporal problems. Both approaches readily yield to massively parallel architectures.

We have applied these methods to the interaction of laser light with molecules, intending not only to investigate the basic mechanisms but also to devise schemes for actually tuning the laser in order to control the outcome of microscopic processes. Lasers now exist that produce pulses short enough to probe a molecular process many times within its characteristic period, allowing the actual observation of an evolving quantum mechanical system.

This year we studied the potassium dimer as an example and found (1) agreement with the findings of a pump-probe experiment conducted at the Laboratory's Materials Science and Technology Division, (2) a sensitivity to the basic molecular properties, and (3) changes in the intermediate-state populations as a function of laser frequency. The latter effect represents a simple control prescription.

In addition, we employed elaborate quantum chemistry programs to improve the accuracy of basic input such as bound-bound and bound-free coupling moments. These time-dependent techniques have a far-ranging degree of applicability. We have now extended them to solve problems in Bose-Einstein condensates, where we are investigating the state of these dilute, trapped quantum systems at very low temperatures.

Publications

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Highly Dissociated Hydrogen and Nitrogen Plasmas for Ion Implantation

98050

Michel Tuszewski

There is a significant need in plasma ion implantation for high-density, low-pressure plasma sources that can yield a single ion species, which permits ion implantation at a well-defined depth. Atomic ions, such as H^+ or N^+, are desirable because the ion energy required for a given implantation depth is lower by comparison with the ion energy required for molecular species, such as H_2^+, H_3^+, or H_2^+. Hydrogen implants into silicon are a promising new technique for bonding thin silicon layers to insulator material (for example, silicon dioxide). Nitrogen implants into metals are known to improve surface hardness, a desirable benefit for many applications in which metal fatigue occurs. The objective of our project is to develop high-power, radio-frequency pulsed and continuous plasma sources that are suitable for hydrogen and nitrogen ion implantation.

This past year we developed pulsed and continuous radio-frequency hydrogen discharges that generate highly dissociated plasmas suitable for hydrogen implantation into silicon. The discharges were generated inductively inside a 0.2-m radius hemispherical quartz dome. The continuous discharges used 0.46 MHz radio frequency, while the pulsed discharges used 0.1 to 0.2 MHz radio frequency.

We generated pulsed hydrogen plasmas with 0.2 Pa gas pressure and 14 kW peak power. Plasma densities of 1.4×10^{17} m^{-3} were measured with Langmuir probes and with microwave interferometry 0.15 m downstream from the dome. We generated steady-state discharges with 0.1 to 1 Pa gas pressure and 0.5 to 1.5 kW power and used a compact ion mass spectrometer to measure the ion species. Plasmas with dominant (>90%) H_2^+ fractions were obtained at the lowest gas pressures and highest powers. Such discharges are suitable for high-dose hydrogen implantation into silicon, enabling us to bond thin silicon layers to silicon dioxide.

Laser-Plasma Interactions in Diffraction-Limited Beams

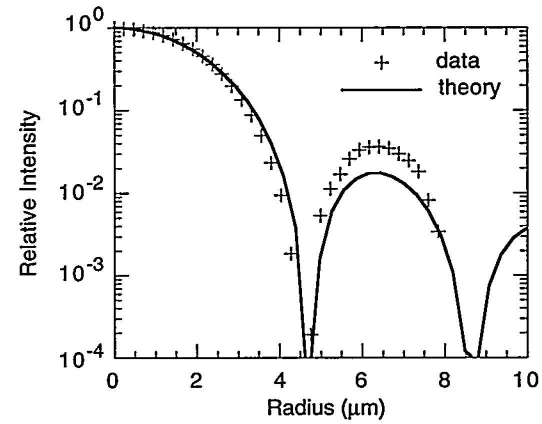
98002

David Montgomery

Laser-plasma interaction experiments using realistic lasers are often difficult to interpret, even for well-defined plasma conditions, because the laser-beam intensity and phase structure are often complicated. Parametric instabilities such as stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), and self-focusing can occur to varying degrees within individual "hot-spot" structures in such laser beams, and we don't know how instabilities produced in an ensemble of hot spots interact with each other. This uncertainty complicates understanding the onset and nonlinear behavior of these instabilities. These issues are still present in interpreting experiments using random-phase-plate-smoothed laser beams. Experiments studying the interaction between the plasma and a single hot spot, such as from a diffraction-limited laser beam, might increase our understanding of these instabilities.

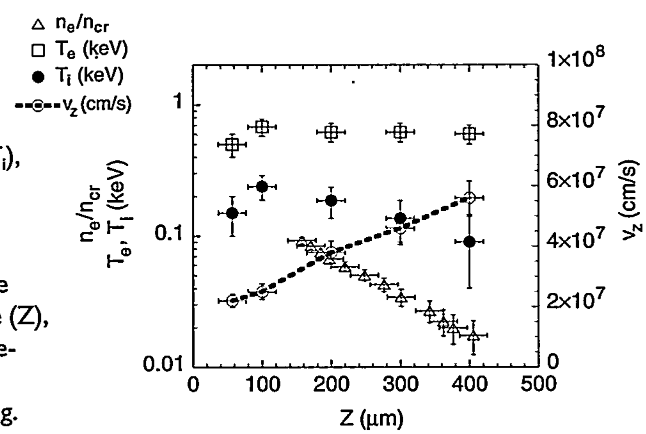
We have begun a series of experiments using the Los Alamos Trident laser facility to study SRS, SBS, and self-focusing in a diffraction-limited (single-hot-spot) laser beam. A separate heater beam is used to create a quasi-homogeneous, large-scale hot plasma with an electron temperature T_e of ~0.5 keV and a gradient scale length L of ~1 mm. We use imaging Thomson scattering from ion-acoustic and electron plasma waves to measure time-resolved spatial profiles of the electron density, electron and ion temperatures, and flow speed. A diffraction-limited beam interacts with this well-characterized plasma, and the plasma density and acoustic damping are systematically varied.

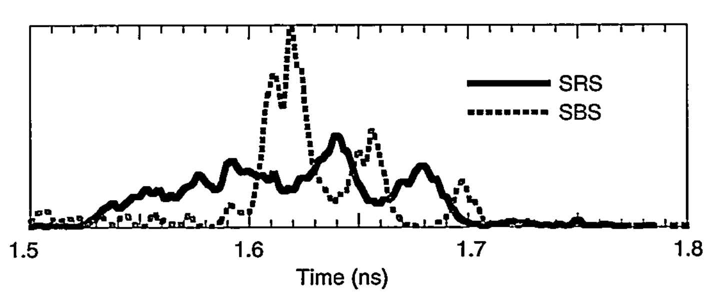
We have achieved diffraction-limited performance on one beam of the Trident laser facility. The far-field intensity (see first figure) and the near-field phase front were characterized for the interaction laser. A second, high-energy beam generated the plasma, and we measured time-resolved spatial profiles of the plasma's density, electron and ion temperatures, and flow speed using imaging Thomson scattering (see second figure). Initial experimental results show evidence of beam breakup by filamentation and beam steering in these high-Mach-number transverse-flow plasmas. We also observe SRS and SBS to be anticorrelated in time when both instabilities are present (see third figure). These experimental results will be used to "benchmark" state-of-the-art numerical models for laser-plasma interactions, which are currently being developed as part of this project.



Far-field intensity profile of the diffraction-limited beam, overlaid with a theoretical curve for an Airy profile.

Measured spatial profiles of electron temperature (T_e), ion temperature (T_i), electron density (n_e/n_{cr}), and flow speed (v_z) as a function of distance from target surface (Z), obtained using time-resolved imaging Thomson scattering.





Time dependence of reflectivity from SRS and SBS show competition between the instabilities when both are present. The *x*-axis shows the time in nanoseconds, and the *y*-axis the relative amplitude.

Publications

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Novel Plasma Sources for Restricted Geometries

98044

Carter Munson

The major objective of this one-year project was to extend research in plasma source ion implantation (PSII) and enhance our understanding of and capabilities in implanting the interior surfaces of selected targets. PSII is a technique that uses the physics of the transient plasma sheath formed around a pulse-based target to implant ions from the plasma into the target surface, producing improvements in the surface hardness, wear, or chemical characteristics. A collaborative program in PSII development involving the Laboratory, the University of Wisconsin, and several industrial partners was recognized in 1997 with an R&D 100 Award.

In this project, we focused specifically on the physics of plasma generation relevant to PSII (and related plasma-based surface modification) in severely restricted geometries (i.e., relatively small volumes that are nearly enclosed by solid, possibly electrically conducting material). Plasma-generating mechanisms and sources operative in this geometry type have a number of potential applications for surface modification within the weapons complex, the Department of Defense, and industry.

The bulk of the research dealt with characterizing and understanding the interactions in the plasma, plasma sheath, and target surface that occur in a relatively small, confined, plasma volume nearly surrounded by a large, complex PSII target. In this case, the target consists of a large assembly of 1008 surrogate automotive pistons that are mounted in four roughly parallel panels, with separation of approximately 25 cm between the panels. The total assembly has an effective surface area of ~16 m^2. Through numerical simulations and experimental measurements, we have obtained a much greater understanding of the interplay between the background plasma, target surface, secondary electrons (generated by ion impact on the target), and plasma sheath expansion. The fundamental physics of the sheath expansion away from the target is well understood and is demonstrated in the first figure, which compares the collision times predicted by several theoretical and numerical models with experimental measurements of the "overlap" or collision of sheaths expanding away from opposing target surfaces. The second figure demonstrates the impact of one plasma generation mechanism for this geometry, which is similar to the well-known hollow-cathode effect. The third figure demonstrates the presence of a beam-plasma instability that significantly increases the plasma density as well as the fluctuation levels in the plasma. Both experimental measurements and simulations indicate that the hollow-cathode operational regime produces undesirable implantation characteristics, whereas the beam-plasma instability regime is compatible with the desired target surface modification.

Publications

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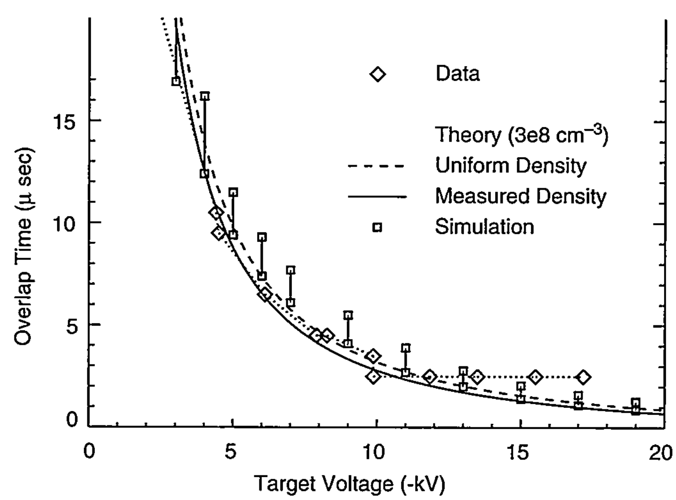
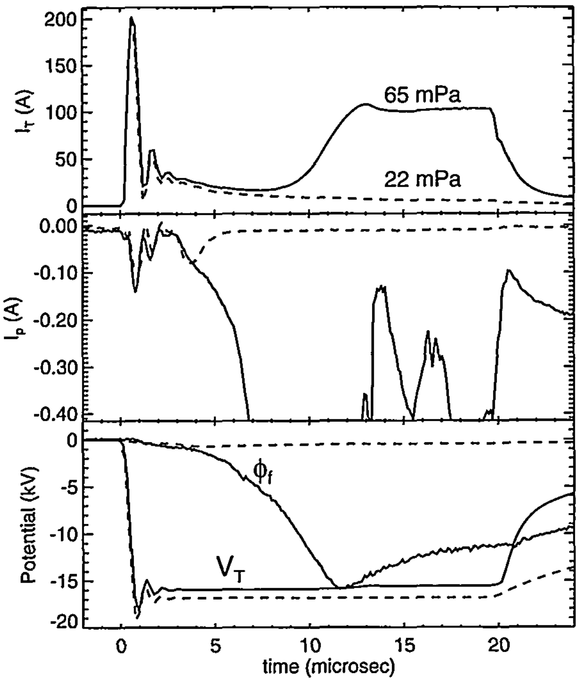
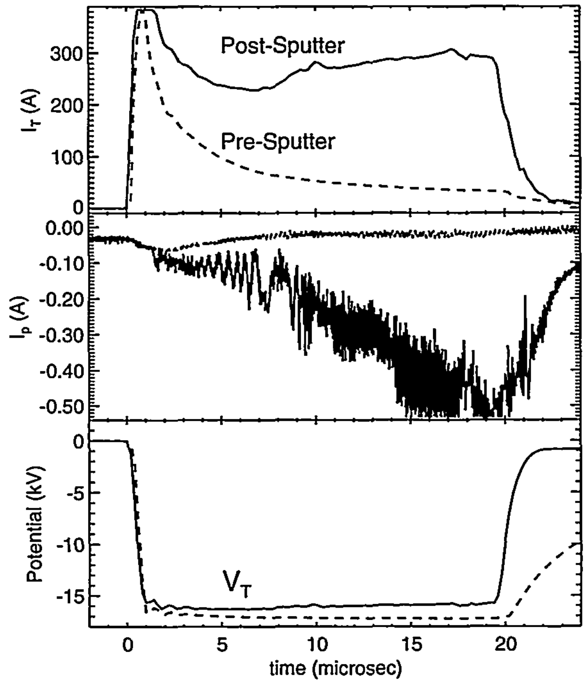


Comparison of predicted sheath overlap times from analytic theory, experimental measurements, and results of the numerical Particle-In-Cell (PIC) code. Experimental data is indicated by the diamonds, uniform density theory prediction by the dashed line, an analytic treatment accounting for the experimentally measured density profile by the solid line, and PIC simulation results by squares.





Experimental data showing the presence of a beam-plasma instability enhancement of the plasma density. The top graph gives the pre-sputter target current history (dashed line) and the post-sputter target current (solid line). The middle graph shows the Langmuir probe current measured in the target assembly inner space (dashed line representing the pre-sputter conditions), and the bottom graph shows the target voltage history (again the dashed line represents the pre-sputter conditions). In the post-sputter conditions, sufficient secondary electrons are present to trigger a beam-plasma instability. This is indicated by both the increased total target current and probe current, as well as the greatly enhanced high-frequency fluctuation level seen on the Langmuir probe current.

Experimental data showing the impact of the "hollow-cathode instability," which appears at 65-mPa, but not at 22-mPa gas-fill pressure. The top graph shows the temporal history of target current for 65 mPa (solid line) and for 22 mPa (dashed line). The middle graph gives the Langmuir probe current (again with the 65 mPa pressure represented by the solid line), and the bottom graph shows the target voltage (solid line for 65 mPa and dashed line for 22 mPa), as well as the floating potential of the probe (φ_f). The gradual target current rise starting at ~9 µs is due to the enhanced ionization of the background gas after the potential in the target inner space has dropped significantly and is approaching the target potential.

Laser Cooling of Solids

97015

Timothy Gosnell

We are developing materials and techniques for laser cooling of solids. Using anti-Stokes fluorescence in Yb^{3+}-doped fluorozirconate glass, we demonstrated the cooling of a condensed phase for the first time. Our primary objective is to demonstrate laser cooling to temperatures as low as possible. Additional objectives are developing thermodynamic models of the cooling process, understanding the fundamental limits of the cooling process, and developing new materials for optimizing the cooling effect.

This year we established a new world record for a laser-cooled solid: 65 K below room temperature. An additional discovery was the effect of upconversion in producing parasitic heating effects, an issue that will affect the cooling efficiency at high pump powers. Important modifications we added to the existing laser-cooling apparatus were a new sample mount that reduced thermal loads from the environment and double-pass pumping of the sample.

Publications

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Geometric Phase, Spatial Resonance, and Control in Spatially Extended Nonlinear Systems

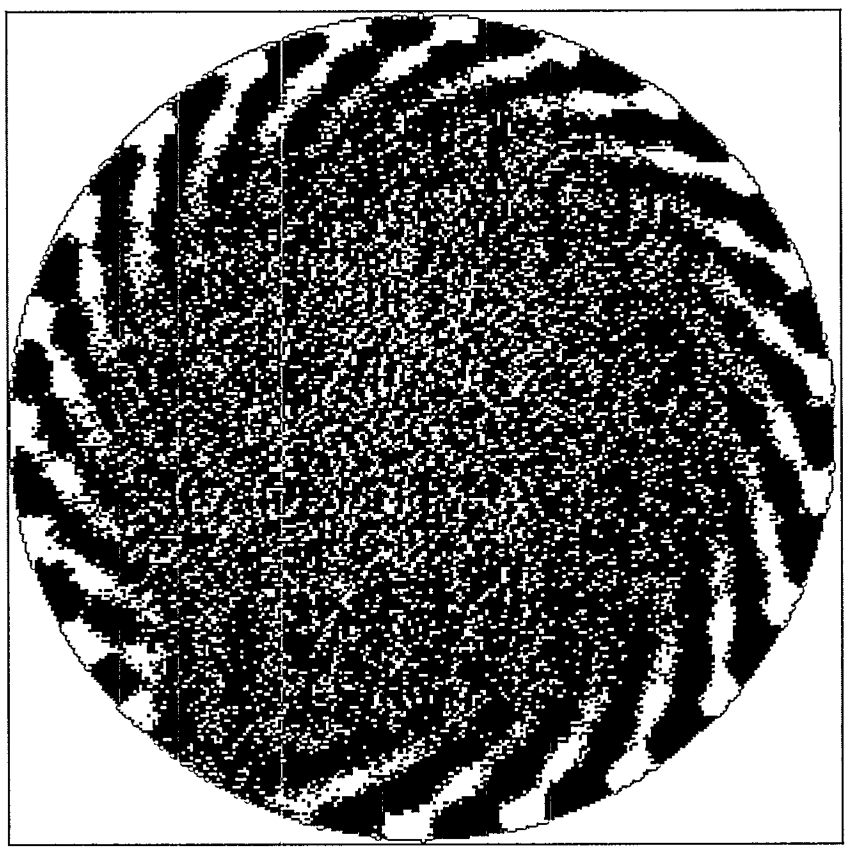
97002

Robert Ecke

Thermal convection with rotation provides a unique system for the study of novel phenomena in nonlinear dissipative systems. In particular, a one-dimensional, nonlinear traveling wave (see the accompanying figure) that we discovered is well described by the complex Ginzburg-Landau 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.

We used this wave to study several experiments ranging from fundamental to the more applied in nature. The first is an experimental search for the geometrical phase in dissipative systems, which was previously shown to apply to Hamiltonian systems, both quantum mechanical (Berry Phase) and classical (Hannay Angle). The second experiment is a model for topographic forcing of wave motion that can arise in geophysical systems. Finally, the 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.

This year we completed our analysis of experimental data on the Eckhaus-Benjamin-Feir instability and on phase diffusion of wave-number distortions. We improved our numerical calculations of linear stability for the traveling-wave mode and compared our results with those from experiments. We implemented side-wall heaters, measured pulse propagation, studied the influence of breaking the rotation symmetry of the system, and investigated the mechanism of wave-number selection in traveling waves.



Gray-scale coded image of nonlinear traveling waves in rotating convection, where azimuthal symmetry is broken by suppressing convection over a small distance along the side wall. The lack of periodic boundary conditions leads to a unique selection of a pattern wave number.

Publications

Choi, Wooyoung, et al., "Traveling Waves in Rotating Rayleigh-Benard Convection" (submitted to *J. Fluid Mech.*).

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Strongly Coupled Dusty Plasmas

97003

Dan Winske

This project examines the consequences of strongly coupled effects in dusty plasmas. This year we have conducted experiments in our newly constructed dusty plasma chamber. Using laser scattering diagnostics, we demonstrated that a strongly coupled dusty plasma is formed when particulates of roughly uniform size are injected into the discharge (with the strong coupling parameter equal to the ratio of potential to thermal energy >1000). Our experiments demonstrated that different configurations of dust grains can be produced, e.g., three-dimensional (3-D) crystals, 2-D arrays, and 1-D strings.

We have shown that strong coupling effects between dust grains are significantly modified when a dust grain is shielded by a nearby grain that is located upstream of it in flowing plasma. Such plasma flows occur in the experiments near the electrodes where the dust grains are located. We have modeled the plasma shielding in terms of a dipole interaction. Using this dipole model, we have carried out 3-D molecular dynamics simulations to show that different crystalline states are possible, depending on the strength of the dipole. An example of one calculation showing crystal formation is presented in the accompanying figure. In addition, we have shown how low-frequency, dust-acoustic waves excited in the crystal are modified by the dipole interaction.

During the final year of this project, we will conduct experiments to verify these results and to refine the parameters used in the model.

Publications

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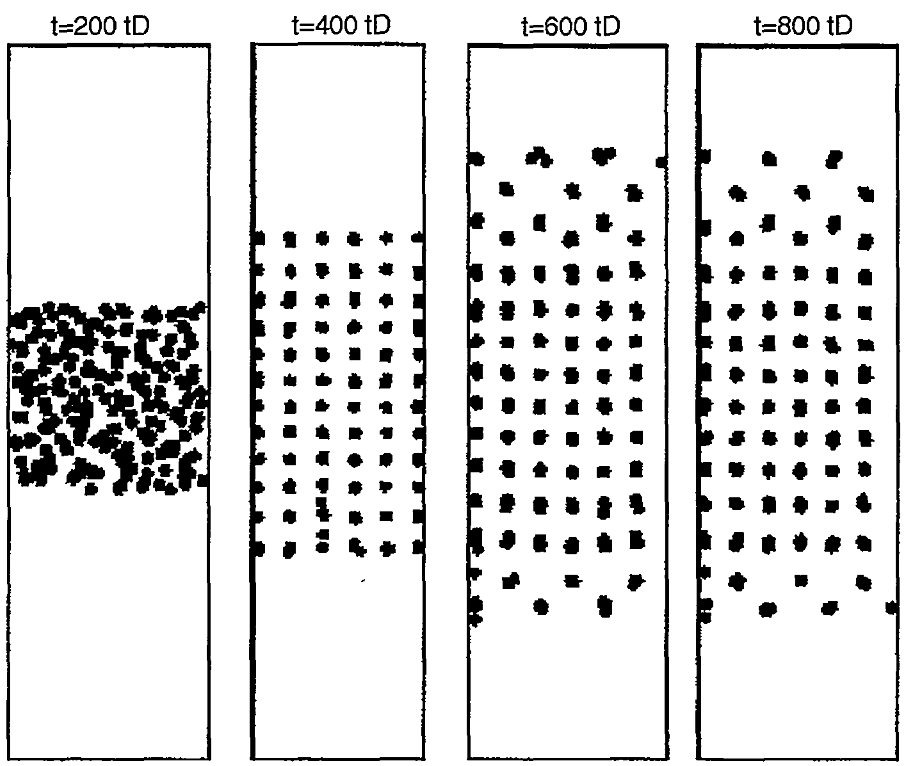


Results of a 3-D molecular dynamics simulation of dusty plasma showing the formation of a regular lattice array ("dusty plasma crystal"). With time (proceeding from left to right), the initially disordered dust grains form into a crystal that expands in the vertical direction. The outer layers are less well ordered.

Equation of State of Dense Plasmas

96391

John Benage

Our project involves the measurement of the equation of state (EOS) of dense, strongly coupled plasmas. 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.

In our technique we use a standard shock-driven EOS measurement, except that the shock is produced in a plasma jet by a laser. We are using state-of-the-art x-ray radiography to measure the shock velocity and the density of the shocked plasma. If we are successful, these results will be the first to address the EOS of materials at dense plasma conditions.

This year we planned to complete a proof-of-principle measurement of the EOS of a dense aluminum plasma, an objective that we were unable to achieve because of problems with the experimental facilities. However, we made significant progress toward the production of our dense plasma targets and with the laser x-ray backlighters needed for these experiments.

In addition, we completed construction of the laser being used to drive shocks in the plasmas. The experimental technique has shown enough promise that it will be used in preparing for experiments in support of the Atlas pulsed-power facility.

Publications

Workman, J., et al., "One-Dimensional X-Ray Microscope for Shock Measurements in High-Density Aluminum Plasmas" (to be published in *Rev. Sci. Instrum.*).

High-Intensity Laser-Matter Interaction Physics

96385

James Cobble

The interaction of a high-intensity laser beam with a preformed plasma is fundamental to research in high-energy-density physics and of special interest for investigation of the fast-ignitor concept of laser-induced fusion. In this context, high intensity implies irradiances exceeding 10^{18} W/cm^2. The laser electric field is hundreds of volts per angstrom; electrons, oscillating at speeds greater than 85% the speed of light, are stripped away from atomic nuclei. This research examines the question of how or whether the laser beam penetrates into a target plasma. Does the beam break up as it enters the low-density ionized atmosphere of the target or does it channel through to interact with the bulk of the plasma density?

We have used the Trident laser picosecond capability to demonstrate an unusual effect. Diffraction-limited focusing of a green laser beam has been achieved on a plasma target of density $>2.5 \times 10^{20}$ cm^{-3} with an f/1.85 off-axis parabola. The beam intensity, at 2×10^{19} W/cm^2, exceeds the threshold for ponderomotive electron cavitation in the plasma. In tunneling through the plasma, the laser is refracted such that its f-number increases to nearly f/8. This beam collimation is monitored by allowing the laser to pass through its best focus with the transmitted light scattered from a diffuser plate several inches behind the target. As the power is reduced to below the threshold (by lowering the plasma density), the beam focusing becomes less obvious. This experiment suggests favorable beam confinement for the alternate laser fusion concept known as the fast ignitor.

Publications

Cobble, J.A., et al., "Wavelength Scaling of High-Intensity Illumination of an Exploded Foil," *Phys. Plasmas* **5**, 4005 (1998).

Pulse Shaping in Explosive Pulsed Power Systems

98016

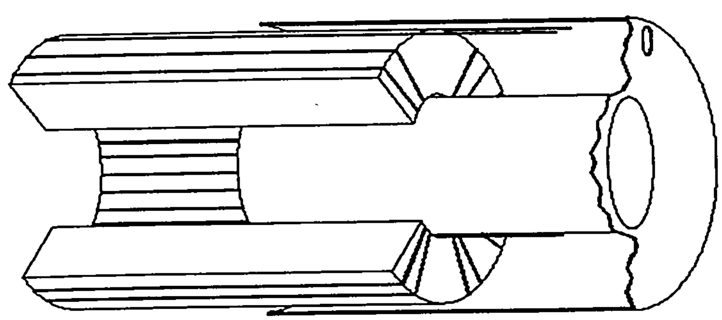
Michael Fazio

Explosive pulsed power systems use the chemical energy stored in explosives and convert it directly into fast, high-energy, electrical pulses. Because of the very high energy density of explosives (5 MJ/g), very compact power supplies can be built for driving compact electromagnetic weapon systems. However, the impedance, rise time, and pulse widths available from explosive pulsed power systems are not capable of directly driving microwave tubes. This project addresses modification of the pulse shape and impedance from explosive pulsed power systems so that compact, high-power, electromagnetic weapons can be developed. This type of weapon is becoming increasingly important for the modern battlefield for such applications as aircraft self-protection and suppression of enemy air defenses.

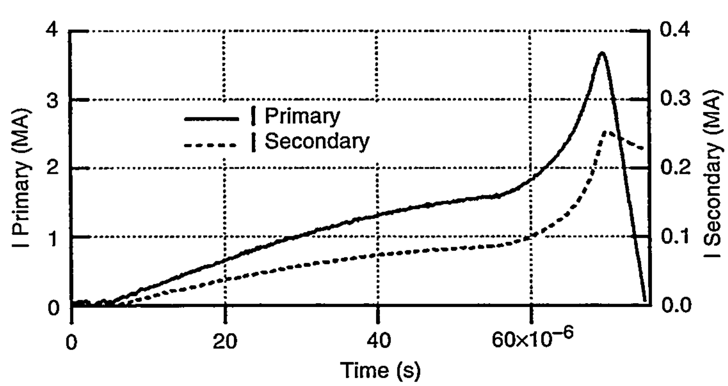
This past year we concentrated on the design and testing of high-coupling, high-voltage air-core pulse transformers for impedance matching (first figure). We designed a transformer capable of outputs of up to 500 kV with a coupling of greater than 0.9 and tested it on an explosive pulsed power system. In addition, we developed a transformer that can be repetitively driven by a large capacitor bank for optimization experiments. These transformers are designed to take up to 8 MA in the primary without the magnetic pressure destroying them. They currently have demonstrated outputs up to 300 kV at currents of up to 350 kA (second figure).

Publications

Fortgang, C.M., et al., "Efficient Air-Core Transformers for Explosive Pulsed Power Applications" (MegaGauss IX Conference, Tallahassee, FL, October 1998).



A cut-away schematic of a high-efficiency, air-core pulse transformer.



Output waveform from a transformer coupled to an explosive pulsed power system.

Virtual Bandwidth via Stochastic Polyspectra

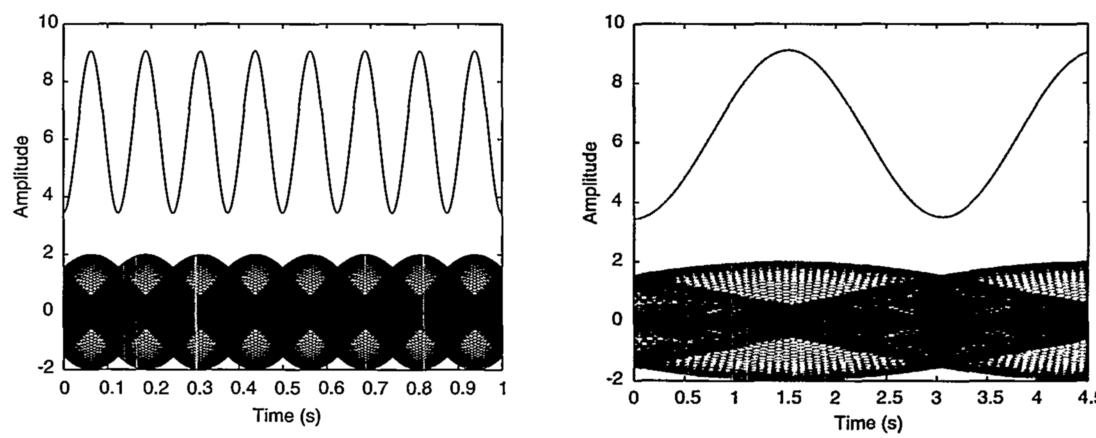
97028

Murray Wolinsky

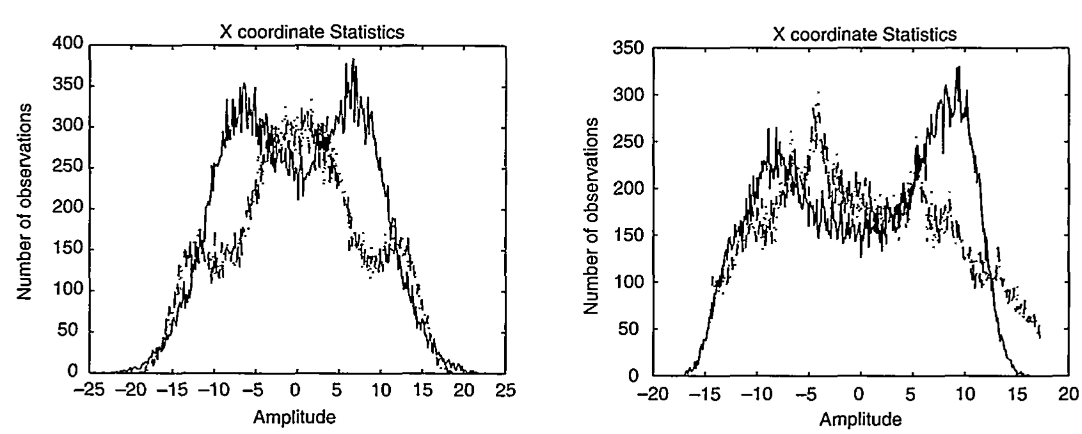
Our present research aims at a better understanding of the use of noise as a probe of signal properties. One continuing line of work investigates the feasibility of using polyspectra to provide noise-resistant communications. A second line of investigation involves the effects of noise on the predictability of dynamical systems.

Two major results were obtained this year. The first result is a definitive resolution of the controversy regarding the detectability of aliasing. We have shown that in the class of stationary signal processes, undersampling can indeed be detected (see the first figure), in spite of the argument based on the Shannon reconstruction theorem. The key insight is that reconstructions of undersampled stationary signals are not themselves stationary. In new work, we have extended the ability to detect undersampling to classes of signals that have no underlying notion of stochasticity. That is, we can—under very weak conditions—detect undersampling in single waveforms (see the second figure).

In the second major result, we have shown that high-frequency information can be used to identify the moments in time near which a dynamical system becomes sensitive to the effects of noise or other perturbation. Our methods allow one



Reconstructed, undersampled stationary signals are cyclostationary. Each figure shows sample ensemble from the reconstructed signal process (depicting nonstationarity of the signal envelope). The sixth moment of the reconstructed process is superimposed (upper curve). In the left plot, sampling is done commensurately with the signal period; in the right plot, the sampling is incommensurate with the signal period. Each original signal is generated by randomly shifting a periodic waveform.



Detection of aliasing in single waveforms (no underlying process assumed). The dotted and dashed plots provide sample statistics at two shifts of the sample comb and are nearly superimposed, showing sampling stationarity. Next a signal is reconstructed based on the data from the "dotted" samples and sampled at points corresponding to the "dashed" samples. The statistics of these samples are shown with a solid plot. Provided the original signal has stationary sample statistics, loss of this stationarity in the reconstructed signal implies undersampling. Results for data from the Lorenz system are shown on the left. Corresponding results for the Rossler system are shown on the right.

to identify these critical points in time from the time series alone, without knowledge of the dynamical system. If certain additional information is available, then the high-frequency behavior of the time series near the critical points allows one to determine whether the perturbation has fundamentally changed the dynamics. This result has important potential applications to the validation of simulations and to control of dynamical systems.

Publications

Luce, B.P., and D.E. Sigeti, "A Power Spectral Test for the Breakdown of Deterministic Dynamics by Low-Frequency Noise" (submitted to *Physica D*).

Using Metallic Glasses in Ceramic-Metal Joints

98014

Rajendra Vaidya

Our objective is to establish the feasibility of using metallic-glass material in ceramic-metal brazing. Metallic glass is relatively inexpensive, it melts uniformly, and it can be bent into complex shapes without kinking, making it advantageous over conventional materials used in brazing. Metallic glass also has a good high-temperature strength.

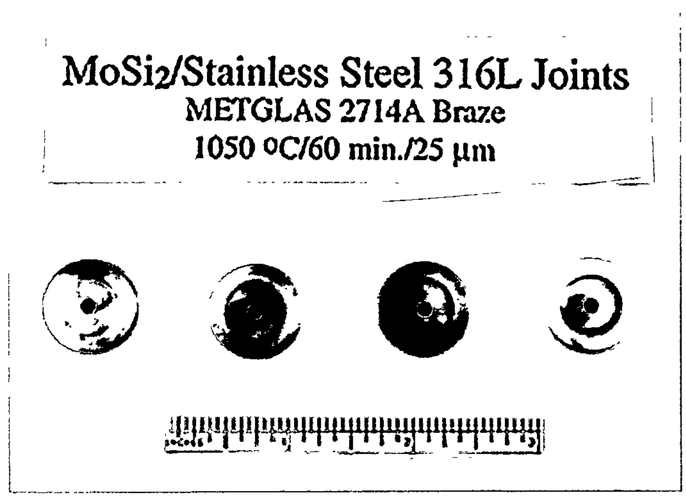
The experiments we conducted focused on optimizing the conditions required for making successful ceramic-metal joints. We conducted synergistic microstructural, mechanical, and residual stress evaluations on metallic-glass brazed joints and combined this research with computer modeling. The joint systems we chose were molybdenum-disilicide stainless steel and alumina stainless steel, both of which have been found to have many applications in ceramic-metal brazed joints.

We successfully produced joints between alumina stainless steel and molybdenum disilicide (see accompanying figure). We were also successful in defining the optimum temperature and time for the brazing cycle. A metallographic examination of the sectioned joints indicated excellent wetting between the metallic-glass material and the individual components of the joints, which is required for good joint strength.

Finally, we also established a "reactive" brazing system by diffusing the metallic-glass material into components. The finite element modeling we conducted on these joints established residual stresses in the joint system. We used this information to optimize the dimensions of the individual components of the joints.

Publications

Vaidya, R.U., et al., "Measurement of Bulk Residual Stresses in Molybdenum Disilicide/Stainless Steel Joints Using Neutron Scattering," *Acta Mater.* **46**, 2047 (1998).



Molybdenum-disilicide stainless steel joints produced using metallic glass. The molybdenum-disilicide tube is inside the joint.

The Plasma Fluidized Bed

97026

Robert Currier

Our goal is to develop novel fluidized-bed technologies centered on the use of cold plasmas. The basic concept is to form a nonequilibrium plasma by coupling radio-frequency or microwave energy into the fluidizing gas. Depending on conditions, the plasma may contain high concentrations of reactive free-radical species, all at modest bulk temperatures. We intend to use these species to initiate or promote desired chemistry. Examples include use of free radicals in extractive metallurgy, plasma-assisted thin-film deposition, and surface modification of solids. Our studies focus on electromagnetic coupling in fluidized beds, associated chemical kinetics, and plasma/solid-surface interactions.

We continued addressing three major topical areas: fluidized-bed dynamics, diagnostic probes, and electromagnetic design considerations. This year we began to screen various chemistries in dense bubbling beds (with input of microwave energy). Initial x-ray diffraction results from the dense beds suggest that atomic species generated in the discharge can convert solid oxides to the base metal. We also designed and fabricated a scaled-up circulating fluid-bed system this year. It can operate in the fast fluidized regime. Work is now under way in this scaled-up system to determine the optimal conditions for effective electromagnetic coupling as a function of solids size, dielectric constant, and density. We also designed experiments to measure pressure fluctuations in dense plasma fluidized beds as a means of indirectly probing the underlying dynamics. These efforts (experiment and analysis) will continue into next year.

Our diagnostics effort has led to construction of a fluidized-bed unit with a microwave applicator and sampling port for mass spectral sampling. This design permits analysis of gas composition (both with and without the glow discharge) as a function of key system parameters. This year we began using this system for off-gas analysis, product distribution, and estimating global reaction rates for plasma-assisted chemistry. We are also developing and testing a nonintrusive capacitance method for determining solids density within the beds and have installed an automated data acquisition system on our large circulating system.

Electromagnetic design has focused primarily on issues related to the ultimate scale-up of the laboratory reactors. This effort includes examination of applicators that would permit high-pressure (near atmospheric) operations. We also initiated laser light-scattering studies aimed at probing the dynamics of fine particles (e.g., resulting from particle attrition) in the plasma. This experimental system is just becoming operational and will be used over the next year.

Material Processing for Self-Assembling Machine Systems

96109

Klaus Lackner

Our objective has been to develop the material extraction and processing aspects of a new technology that is based on self-assembling or self-reproducing machine systems. We believe machine systems that are initially programmed to build more and more of themselves and then move on to build a specific type of "factory" could someday overcome resource limitations and control the deleterious side effects of human activities on the environment. Three examples we have considered are (1) a solar energy field, (2) a means to desalinate seawater for irrigation, and (3) a way to remove excess carbon dioxide from the atmosphere (greenhouse gases). Machine systems that build themselves promise an increase in industrial productivity as dramatic as that of the industrial revolution.

Such systems would need to procure all the raw materials they require from their surroundings. They would have to extract all the important chemical elements from common dirt. The extraction process would be limited to the use of common elements because any large-scale process that relies on rare elements would demand too much raw material. Although this process is completely different from conventional metallurgical extraction, the methods may interest conventional industry because they do not involve expensive and sometimes hazardous materials. Our design uses a thermodynamically viable extraction process, and we have experimentally demonstrated most of the steps that differ from common practice.

Through last year's effort, we have shown that if we are operating in a vacuum, we can use substantially lower temperatures than were originally estimated. We developed and built a small, disposable vacuum furnace system, and we succeeded in volatilizing, and thereby separating, silica from a mineral melt at about 1500°C rather than at the 1800°C to 2000°C required at atmospheric pressure. The elements we are extracting come from average crustal rock. We plan to publish our results concerning this important step toward a self-replicating machine system.

The Compliance Method for Measuring Residual Stress

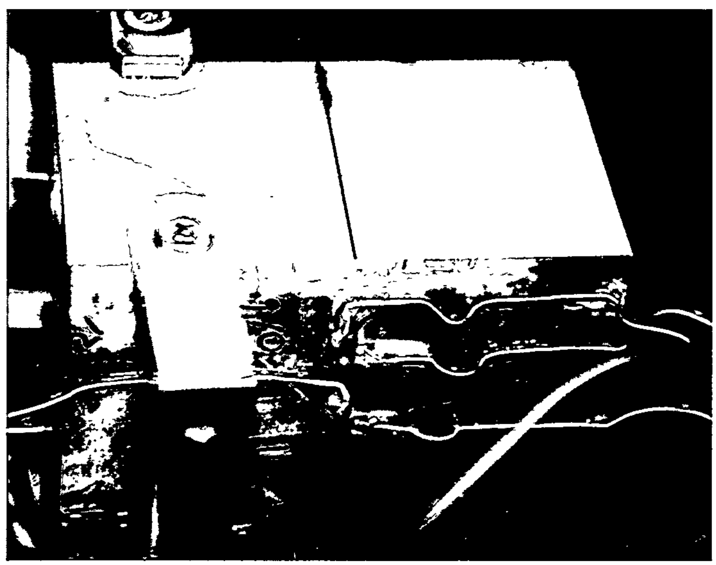
97017

Michael Prime

The purpose of this project is to develop the compliance method for measuring residual stress by successive extension of a slot. Our goal is to turn this fledgling method into a proven and versatile technique that can fill the gaps in current residual-stress measurement technology. In addition, the extended measurement capabilities resulting from our project will greatly aid the development and verification of predictive models.

This year we measured residual stresses in an aerospace aluminum alloy (see the figure). Our measurements took a few hours, whereas measurements using the conventional technique take several days; our results also gave better spatial resolution of the stresses. Residual stresses in these alloys cause major distortion of machined aircraft structural parts, and the required rework costs millions of dollars. Quicker and more accurate measurement of these stresses will allow manufacturers to more efficiently evaluate stress control measures.

We completed several other efforts this year. First, we extended the compliance method to a new capability—measuring both the residual stress and the resulting stress intensity factor in compact tension specimens. These specimens are routinely used to determine crucial material properties, and unmeasured residual stresses can lead to critical errors. Next, after a detailed study, we were able to identify the data reduction methods that are the least sensitive to experimental errors. Finally, we completed a feasibility test on an exciting new technique—measuring surface displacements after slot cutting instead of strains during the cutting. Such measurements are simpler, quicker, and could lead to improved results. We will study this technique further in the upcoming year.



Aluminum specimen after the compliance method measurement of residual stress. The slot made by wire electric-discharge machining is visible on top of the specimen. Strain gauges are visible on the front.

Publications

Prime M.B., "Measuring Residual Stress and the Resulting Stress Intensity Factor in Compact Tension Specimens" (to be published in *Fatigue Fract. Eng. Mater. Struct.*).

Prime, M.B., "Residual Stress Measurement by Successive Extension of a Slot: The Crack Compliance Method" (to be published in *Appl. Mech. Rev.*).

Rangaswamy, P., et al., "Comparison of Residual Strains Measured by X-ray and Neutron Diffraction in a Titanium (Ti-6Al-4V) Matrix Composite" (to be published in *Mater. Sci. Eng.*).

Tritium Recovery and Isotope Separation Using Electrochemical Methods

97027

Richard Willms

Techniques for the recovery of tritium gas and removal of tritium from hydrocarbon gases are crucial for maintaining the US nuclear stockpile. The recovery of tritium from the amassed mixed waste in the nuclear weapons complex is also of great national importance and therefore germane to environmental stewardship as well.

Our principal objective is to demonstrate a new tritium separation technology using ceramic, electrochemical ion pumps based on solid-oxide proton conductors. The ceramic materials are typically perovskite in structure and exhibit high conductivities for hydrogen isotope motion. Candidate materials are typically based on $(Sr,Ba)(Zr,Ce)O_3$ ceramics doped with lanthanide ions to enhance their ionic conductivity.

In our preliminary studies, we were able to determine the conductivity of hydrogen, deuterium, and tritium in $SrZr_{0.9}Yb_{0.1}O_{2.95}$. In general, the ionic conductivity in these perovskites is known to follow an Arrhenius-type equation where $sT = A\exp(-E/kT)$. We have determined the following values for the constants A and E:

	A (K/W·cm)	E (eV)
Hydrogen	8135	0.56
Deuterium	4917	0.58
Tritium	3325	0.605

The tritiated materials were stable over the two-day test period despite the high levels of beta radiation of 2% tritium gas. We demonstrated the electrolytic decomposition of water and deuterium oxide using these perovskite materials. The membranes separated hydrogen isotopes from carrier gases with 100% selectivity.

Publications

Mukundan, R., et al., "Tritium Conductivity and Isotope Effect in Proton Conducting Perovskites" (submitted to *J. Electrochem. Soc.*).

Development and Engineering of the Ion-Cut and Low-Temperature Direct-Bonding Process

98015

Michael Nastasi

We are developing an innovative integrated-circuit (IC) engineering technology that will enable effective and economical silicon-on-insulator (SOI) materials technology. SOI materials technology is critical to the production of high-performance radiation-hardened ICs, ICs that can operate at low power, and three-dimensional IC chips that are 250 to 500 times denser than conventional chips.

Our approach uses recent technological breakthroughs in plasma processing, ion-beam-stimulated cleaving of single-crystal silicon, and plasma-stimulated, low-temperature direct bonding of silicon to SiO_2. We plan to develop a plasma source ion implantation (PSII) chamber for implanting hydrogen in single-crystal silicon. Implanted hydrogen will stimulate the cleavage of a thin layer of silicon, which will be bonded to an insulating substrate. The bonding of the thin silicon layer and substrate will rely on the plasma activation of the bonding surfaces and a low-temperature annealing step.

We have designed and constructed a PSII system for implanting H^+ ions into 4-in.-diam silicon wafers. The PSII chamber is 70 by 70 by 60 cm with a spiral copper flat coil (five square turns 56 by 56 cm) inductive plasma source. The plasma power supply is under development. Hydrogen plasmas with densities in the range of 10^{11} to 10^{12} cm^{-3} have been produced in the pressure range of 1 to 5 mtorr. The pulse voltage modulator for ion implantation is under construction.

We have performed preliminary experiments to study systematically the effects of the hydrogen ion implant distribution, dose, silicon-doping level, and the presence of additional doping on the silicon interlayer cleavage mechanism. These experiments have been carried out with a traditional beam-line implanter. Surface cracking experiments have shown that the crack depth is controlled by the radiation damage, which forms during the hydrogen ion implantation, rather than the hydrogen implantation distribution. We are determining the influence of silicon doping and defect charge state on the cleavage process.

Quantum Cryptography for Secure Communications to Low-Earth-Orbit Satellites

96114

Richard Hughes

Cryptanalysis techniques and algorithms are advancing rapidly. By the start of the twenty-first century, they will necessitate new encryption technologies to ensure the security of communications to satellites. The aim of our project is the development of quantum cryptography to provide absolutely secure encryption of communications to low-earth-orbiting satellites. We are developing and demonstrating the cryptographic technology so that it can be feasibly incorporated into new satellites.

This year we designed, constructed, and tested a quantum cryptography system that creates and transmits (using single-photon transmissions) cryptographic random numbers at night between sending and receiving instruments separated by a 1-km outdoors optical path. The system is based on the propagation and detection of nonorthogonal polarization states of single photons in free space at a wavelength of 772 nm, for which the atmosphere has a very low attenuation.

We have now demonstrated point-to-point key distribution over 0.5 km in full daylight, and we are planning to extend the transmission range to more than 2 km in a new system that incorporates active control to compensate for turbulence-induced beam wander (see the accompanying figure). In addition, we are developing plans for a surface-to-satellite test experiment.

Publications

Buttler, W.T., et al., "Free-Space Quantum Key Distribution," *Phys. Rev. A* **57**, 2379 (1998).

Buttler, W.T., et al., "Free-Space Quantum Key Distribution at Night," *Proc. SPIE* **3385**, 14 (1998).

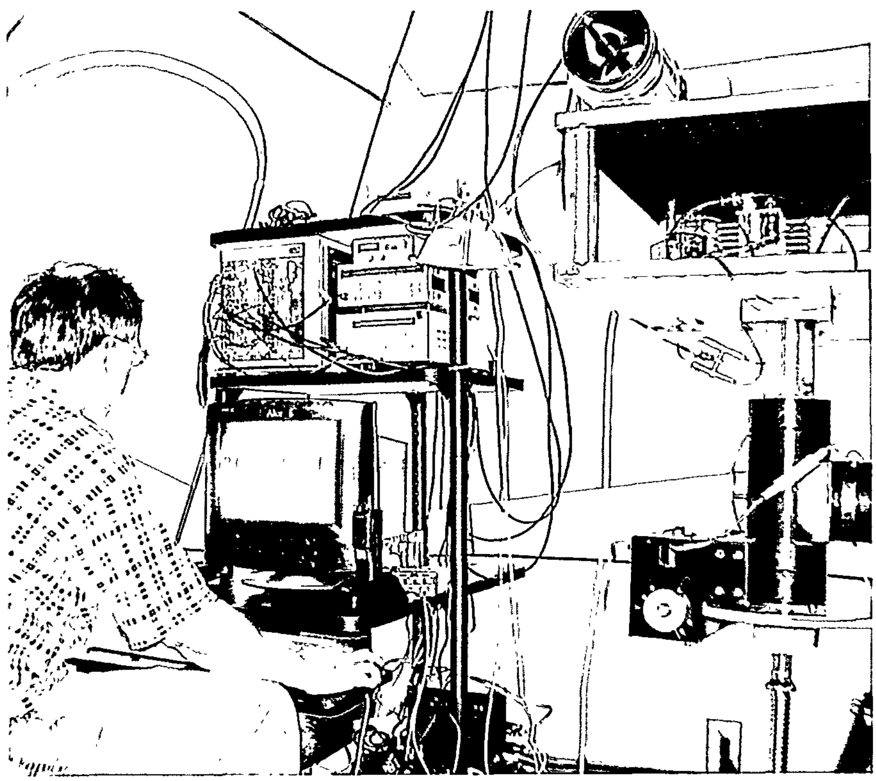
Buttler, W.T., et al., "Practical Free-Space Quantum Key Distribution over 1 km," *Phys. Rev. Lett.* **81**, 3283 (1998).

Hughes, R.J., "Secure Communications Using Quantum Cryptography," *OE Reports* **164**, 12 (1997).

Hughes, R.J., "Secure Communications Using Quantum Cryptography," *Opt. Process. Comput.* **8**, 1 (1997).

Hughes, R.J., et al., "Free-Space Quantum Cryptography" (Quantum Communications and Measurement Conference, Evanston, IL, August 1998).

Hughes, R.J., et al., "Practical Free-Space Quantum Key Distribution," in *Proceedings of the First NASA Quantum Computation and Quantum Communications Conference* (Springer-Verlag, New York, in press).



The free-space quantum cryptography transmitter for line-of-sight optical communications.

Acoustic-Network Refrigerators

98017

William Ward

We are developing a new class of refrigerators and heat pumps, which we call acoustic-network refrigerators. The acoustic-network refrigerator is a descendant of the thermoacoustic refrigerator developed at Los Alamos during the past 15 years. The acoustic-network refrigerator shares the thermoacoustic refrigerator's simplicity, but it uses an acoustic transmission-line network to cause the phasing between pressure and velocity to become that of the classic Stirling cycle.

This year we have confirmed the viability of the acoustic-network refrigerator concept. Based on numerical optimization and preliminary experiments, we have designed and started fabrication of a well-instrumented laboratory model refrigerator. The device is pressurized with helium to achieve a cooling power of several hundred watts. We have obtained a high-efficiency linear electric motor to drive the refrigerator. We isolated parasitic mean flow in the regenerator as the primary source of inefficiency, and we have developed and are optimizing methods to suppress it. In the course of the analyses, a number of improvements have been made to the thermoacoustic design code, DeltaE, to better model this specific configuration.

Instrumentation and Diagnostics

Novel Signal Processing with Nonlinear Transmission Lines

97031

David Reagor

Nonlinear dielectrics, such as $SrTiO_3$ (STO), offer uniquely strong and tunable nonlinearities. These properties make them attractive not only for immediate implementation in high-performance devices such as frequency-agile microwave filters, but also for the exploration of new technologies in the area of signal processing. Our objective is to develop time domain signal-processing concepts using integrated, nonlinear, dielectric, coplanar-waveguide devices that demonstrate general functionalities valuable to many systems.

We first developed measurement systems, photolithography masks, and device fabrication processes. The theoretical work focused on perturbation theory and simulations relevant to the type of transmission lines used in the experiments. In the past year we have studied pulse shaping in coplanar-waveguide delay lines made from superconducting $YBa_2Cu_3O_7$ electrodes on nonlinear dielectric STO substrates. The delay lines exhibited bias- and temperature-dependent nonlinearity and dispersion. At low temperatures and under dc bias, the interplay of dispersion and nonlinearity led to a variety of pulse shapes, some of which were preserved for long travel distances. The accompanying figure shows how pulse shape changes with pulse amplitude and dc bias at 20 K. The three sets of traces represent the same pseudo-Gaussian input pulse with three different attenuation levels. The top traces illustrate the effect in the large signal limit. At present we are investigating generation and manipulation of fast-risetime pulses and microwave solitons using our integrated coplanar-waveguide devices.

Publications

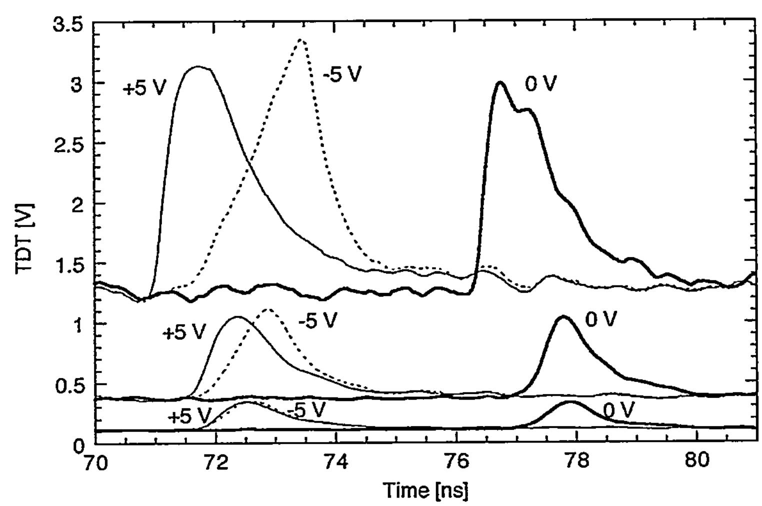
Bishop, A., and A. Sanchez, "Collective Coordinates, Lengthscale Competition and Complex Behavior in Soliton-Bearing Equations" (submitted to *SIAM Review*).

Cai, D., et al., "Resonance in the Collision of Two Discrete Intrinsic Localized Excitations," *Phys. Rev. E* **56**, 7246 (1997).

Kivshar, Y., et al., "Multiple States of Intrinsic Localized Modes," *Phys. Rev. B* **58**, 5423 (1998).

Rasmussen, K., et al., "Creation and Annihilation of Intrinsic Localized Excitations," *Phys. Rev. E* **58**, 40 (1998).

Rasmussen, K., et al., "Soliton Motion in a Parametrically AC-Driven Damped Toda Lattice" (to be published in *Phys. Rev. E*).



Time-domain transmission (TDT) response of a coplanar-waveguide device with tunable nonlinearity and dispersion. The three sets of traces represent the same pseudo-Gaussian input pulse with three different attenuation levels. At each attenuation level, three dc voltage biases were used. The simple pulse shapes at the bottom are only slight modifications of the input pulses, but the upper traces involve nonlinearities that lead to substantial modifications of the pulse shape. The ability to sharpen the leading edge of the pulse may be of value for high-speed electronics.

Cryptographic Key Generation Using Long-Base-Line Radio Interferometry

98028

Richard Hughes

As the demands for secure encrypted communications increase, there is a growing need for efficient methods to generate and distribute the secret random-number sequences, known as cryptographic keys, that are used in encryption hardware and software. Existing methods of key distribution are cumbersome, impractical for many applications, and not demonstrably secure. We are developing a new method for two parties to generate a shared secret key, on demand, from simultaneous observations of the correlated radio noise from astronomical objects. The radio emissions from the brightest radio sources can be detected with commercially available radio equipment using small antennas, and the necessary frequency and time standards are now readily available to allow the rapid generation of correlated, raw key streams by two widely separated observers. The raw key streams will then be subjected to a mathematical procedure of "advantage distillation," error correction, and "privacy amplification" to distill shorter, secret, error-free keys; the mathematical procedure secures the information contained in the final key streams. Although the mathematical formalism for key generation from partially correlated noise sources was developed several years ago, our work is the first practical demonstration of this new technique.

We are developing a proof-of-principle demonstration of key generation using joint observations from two locations of the radio emissions from bright astronomical radio sources such as Cygnus A. We have constructed antennas for the 1- to 2-GHz frequency range with low-noise preamplifiers and have developed high-frequency receivers to record the radio signals, referenced to Global Positioning Satellite (GPS)-disciplined, phase-stable quartz oscillator modules. These signals are digitized and stored on the hard disks of controlling personal computers. We are working at first with short base lines between the two observing stations and have developed the data acquisition and analysis hardware and software necessary for key generation. We expect to have demonstrated key generation by the end of this year.

High-Average-Power, Intense Ion Beam for Materials and Other Applications

96115

Blake P. Wood

The goal of this project is to develop a 1- to 2-Hz repetitive, intense ion-beam source ($E = 200$–250 keV, $I = 10$–15 kA, $t = 1$ µs). The beam will be produced in a magnetically insulated diode with an active plasma anode. Applications include (1) materials surface treatment through rapid melt and resolidification; (2) coating production at high rates through vaporization of targets and reposition onto substrates; (3) a diagnostic neutral beam for the next generation of tokamaks; and (4) an intense, pulsed neutron source for mine detection, enhanced surveillance, and neutron resonance spectroscopy of strongly coupled plasmas.

We completed construction of the active plasma anode and characterized the operation of the anode with and without the insulating magnetic field "slow coils." Operation was acceptable and as expected. We found that the main body of the plasma is produced and accelerated on the second half-cycle of the current flow in the fast (anode) coil, that the velocity of the plasma sheet reaches 2.7 cm/µs, and that the peak plasma density reaches 2×10^{16} cm^{-3}. About an order of magnitude less plasma is produced on the third half-cycle of the current flow in the fast coil. With the slow coils in operation, more plasma is produced on the third half-cycle of the current flow in the fast coil. We nearly completed construction of the accelerator before the end of the fiscal year. All components have been completed, but not entirely assembled. Because this is the final year for LDRD funding of our project, construction has been halted until a funding source can be found.

Publications

Davis, H.A., et al., "Intense Pulsed Ion Beam Target Ablation for Film Deposition" (to be published in *J. Appl. Phys.*).

Davis, H.A., et al., "Ion Beam and Plasma Technology Development for Surface Modification at Los Alamos National Laboratory," *Mater. Chem. Phys.* 54, 213 (1998).

Wood, B.P., et al., "Cratering Behavior in Single- and Poly-Crystalline Copper Irradiated by an Intense Pulsed Ion Beam" (to be published in *Surf. Coat. Technol.*).

Soliton Optical Communications

97005

Antoinette Taylor

In this project we are investigating the limits of high-bit-rate propagation of optical pulses in fiber. These pulses, called solitons, are being studied for use as communication links between computers. The experimental effort relies on a novel diagnostic that measures the exact electric-field envelope and optical phase of ultrashort optical pulses as a function of time; we can then characterize the temporal evolution of optical pulses as they propagate in fiber-optic waveguides. Using this diagnostic, we will investigate the temporal evolution of the pulse in a stretch of passive fiber as a function of initial pulse width, pulse shape, and interpulse separation and will perform a systematic investigation of techniques that ameliorate soliton instabilities that arise at high bit rates.

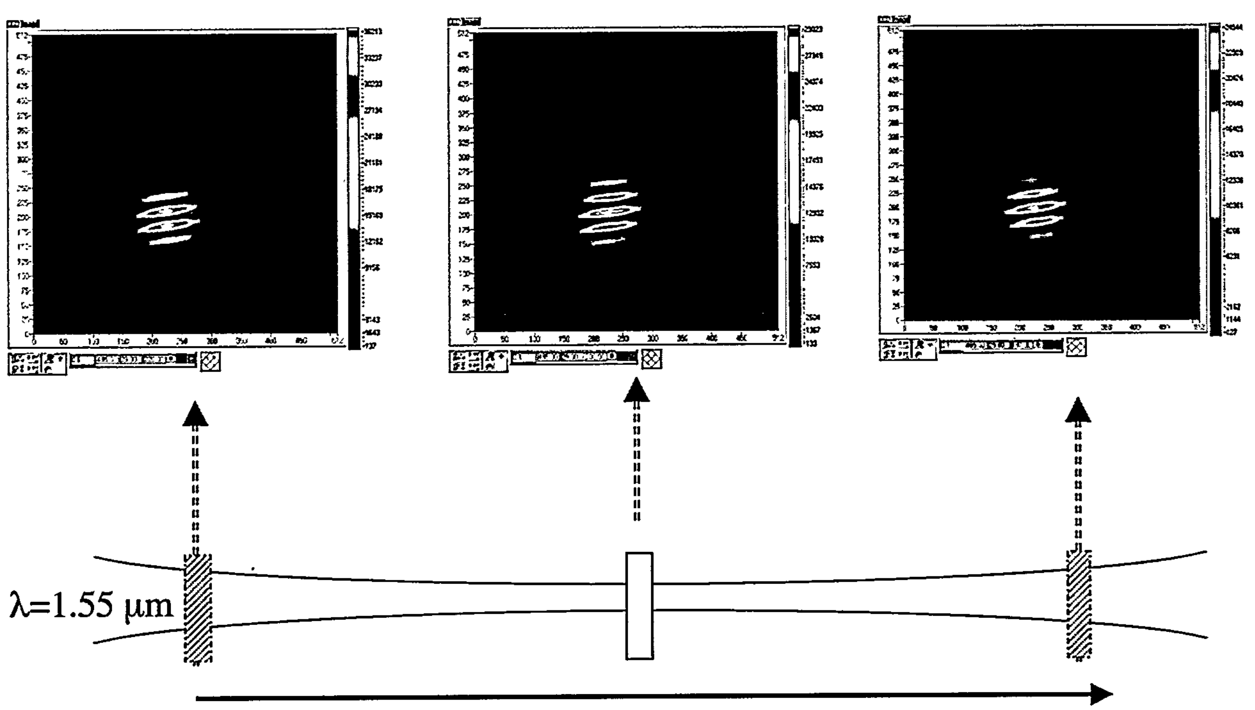
This year we set up an optical parametric amplifier to produce, propagate, and diagnose wavelength- and pulse-width-tunable pulses around 1550 nm (the optical fiber transmission window). The output pulse from an optical parametric amplifier is shaped temporally using a phase modulator. From this pulse we can generate a 16-bit pulse train with variable interpulse spacings using a high-throughput multiplexer that we invented. The pulses then traverse an optical fiber. Both the input and output of the fiber are characterized using the technique of multiple-pulse interferometric frequency-resolved optical gating (MI-FROG), where the nonlinearity used is second harmonic generation. This technique permits determination of the phase of the pulse to all orders, even for the relatively weak pulses required for fiber propagation. To demonstrate this technique, we show in the accompanying figure the evolution of the MI-FROG trace for three different irradiance pulses incident on a piece of sapphire. The highest irradiance (middle trace), close to the focus, reveals an intensity-dependent phase shift that causes the fringes to tilt and the pulse to broaden spectrally.

Publications

Taylor, A.J., et al., "Generation and Characterization of Terahertz Pulse Trains from Large-Aperture, Biased Photoconductors" (submitted to *Opt. Lett.*).

Rodriguez, G., and A.J. Taylor, "Measurement of the Cross-Phase Modulation in Optical Materials through the Direct Measurement of the Optical Phase Change," *Opt. Lett.* **23**, 858 (1998).

Siders, C.W., et al., "High-Energy Pulse Train Generation using a Michelson Interferometer," *Appl. Opt.* **37**, 5302 (1998).



The evolution of MI-FROG trace for three different irradiance pulses incident on a piece of sapphire. The highest irradiance (middle trace) close to the focus reveals an intensity-dependent phase shift that causes the fringes to tilt and the pulse to broaden spectrally. (The vertical axis is frequency, the horizontal axis is time.)

Thermal Detection of DNA and Proteins during Gel Electrophoresis

97032

Roger Johnston

Gel electrophoresis is one of the most important techniques in science and biotechnology. Applications include biological and biomedical research, polymer chemistry, forensics, law enforcement, disease control, national defense, anthropology, biometrics, genetic counseling, botany, agriculture, and animal husbandry. It is used in both low-technology laboratories and at state-of-the-art facilities.

Recent ultrasensitive refractive index measurements suggest there may be a simpler, more sensitive way to detect DNA fragments during gel electrophoresis based on thermometry and local Joule self-heating. This would permit simple, inexpensive, ultrasensitive detection of DNA and proteins in real time during gel electrophoresis. Our project involves developing the appropriate instrumentation, performing a proof of principle, optimizing the technique, and establishing the sensitivity.

In the past year we performed a series of measurements with our newly designed gel electrophoresis apparatus. These measurements led to a redesign of the apparatus. This second-generation apparatus, in turn, produced far more thermal temporal stability than we had predicted. The detected thermal effects from DNA bands, however, were a factor of 5 to 8 less than predicted. In the coming year we will use some new ultraminiature thermistors, plus a slight redesign of the gel apparatus, to try to achieve the predicted sensitivity and demonstrate the practicality of the technique.

Publication

Grace, K.M., et al., "Waveguide Zeeman Interferometry for Thin-Film Chemical Sensors," *Electron. Lett.* **33**, 1651 (1997).

Magnetic Resonance Force Microscope Development

96213

P. Chris Hammel

Our goal was to develop the magnetic resonance force microscope (MRFM) into an instrument useful for studies of scientifically and technologically important materials. This work culminated in the demonstration of our ability to provide important details about the magnetic properties of microscopic cobalt films.

Using the MRFM, we demonstrated lateral one-dimensional imaging of cobalt films by means of microscopic ferromagnetic resonance (FMR) detection. We implemented a novel approach that involves scanning a localized magnetic probe by using a spatially selective local field that was generated by a small, magnetically polarized spherical crystallite of yttrium-iron-garnet. We resolved the approximately 20-μm lateral separation between two deposited cobalt films. Data from MRFM experiments with the magnetic probe on the mechanical resonator provided insight into spurious detector responses that arise from interactions between the magnetic tip and externally applied fields. Miniature magnetically polarized $Nd_2Fe_{14}B$ particles show promise as magnetic probe tips. We demonstrated mechanical detection of FMR signals from microscopic cobalt single-layer thin films using the MRFM. Variations in the magnetic anisotropy field, as well as the inhomogeneity of the film, were clear in the FMR spectra of these samples.

This study demonstrated that MRFM detection of FMR is potentially important for microscopic characterization of the spatial distribution of internal magnetic properties in magnetic layered materials and devices.

Publications

Hammel, P.C., et al., "The Magnetic Resonance Force Microscope: A New Microscopic Probe of Magnetic Materials," in *Frontiers in Reduced Dimensional Magnetism*, V.G. Bar'yakthar, et al., Eds. (Academic Publishers, Dordrecht, Netherlands, 1998), p. 441.

Suh, B.J., et al., "Ferromagnetic Resonance Imaging of Cobalt Films Using Magnetic Resonance Force Microscopy," *J. Vac. Sci. Technol. B* **16**, 2275 (1998).

Zhang, Z., and P.C. Hammel, "Magnetic Resonance Force Microscopy with a Permanent Magnet on the Cantilever," *IEEE Trans. Magn.* **33**, 4047 (1997).

Zhang, Z., and P.C. Hammel, "Towards a Magnetic Resonance Force Microscope Employing a Ferromagnetic Probe Mounted on the Force Detector," *Solid State Nucl. Magn. Reson.* **11**, 65 (1998).

Zhang, Z., et al., "Ferromagnetic Resonance Force Microscopy on Microscopic Cobalt Single Layer Films," *Appl. Phys. Lett.* **73**, 2036 (1998).

Stable Polymeric Light-Emitting Devices

98025

DeQuan Li

Polymeric light-emitting diodes (LEDs) have attracted tremendous scientific and technological interest because they are brighter and more colorful than liquid crystal displays, they can be easily deposited onto large-area substrates, and they offer flexibility in materials and device design. Technologically, defense-related devices and instruments continue to require better and higher performance LEDs. Polymeric LEDs also can be fabricated into flexible thin films and used in many unusual places where conventional inorganic LEDs cannot be applied (e.g., a roll-up display). Using molecular self-assembly techniques and the ability to control thin-film structure and composition at the molecular level, our objective is to fabricate polymeric LEDs.

In the past year we focused on tuning the interface properties between the electrodes and polymeric layers, because interface properties are critical to the behavior and performance of the polymeric LEDs. We have fabricated and characterized a new type of diode device made of metal/organic monolayer/semiconductor heterostructures. The device consists of an organic barrier layer sandwiched between an aluminum metal contact and a p-type silicon (see accompanying figure). The barrier was formed through self-assembling (1) a monolayer of wide-band-gap poly(diallydimethyl ammonium) chloride or PDDA, (2) a bilayer of PDDA and narrow-band gap NiPc (nickel phthalocyanine tetrasulfonate), and (3) a bilayer of PDDA and donor PPP (poly p-quaterphenylene-disulfonic-dicarboxylic acid). Current-voltage (I-V) measurements show that the turn-on voltage of the devices can be tuned by varying the structure, hence electronic properties, of the organic monolayers. Unlike conventional Schottky barriers, which exhibit an exponential relationship between I and V, the new diodes have a power-law dependence of I on V, with the exponent a= 2.2 for PDDA, 2.7 for PDDA/NiPc, and 1.44 for PDDA/PPP as the barrier layer, respectively. The significance of our results is that the transport properties are controlled by the structure and electronic properties of the interface self-assembled monolayers.

Publications

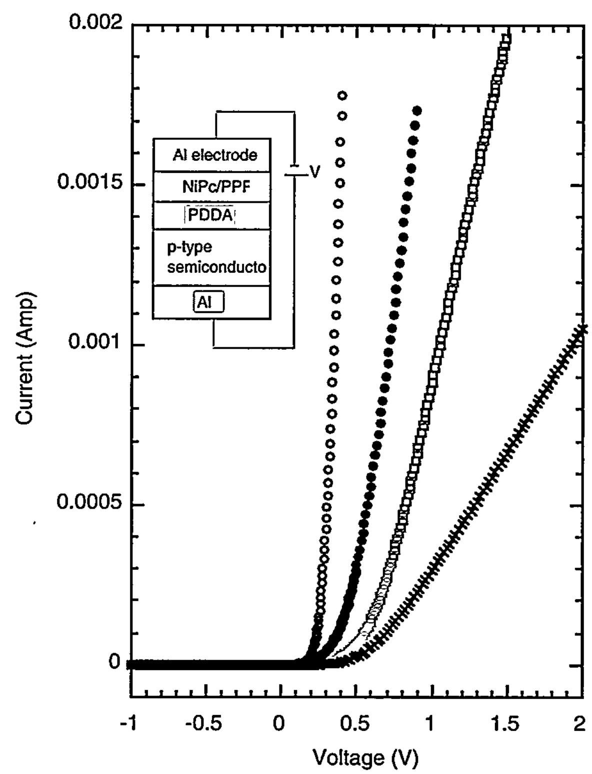
Li, D.Q., et al., "Multilayer Self-Assemblies as Electronic and Optical Materials," in *Mater. Res. Soc. Symp. Proc. on Electronic, Optical and Magnetic Properties of Org. Mater.* **488**, 401 (1998).

Li, D.Q., et al., "Preparation, Characterization, and Properties of Mixed Organic and Polymeric Self-Assembled Multilayers," *J. Am. Chem. Soc.* **120**, 1538 (1998).

Li, D.Q., et al., "Transport Properties of Metal/Organic Monolayers/ Semiconductor Heterostructures" (to be published in *Appl. Phys. Lett.*).

Mashl, R.J., et al., "Theoretical and Experimental Adsorption Studies of Polyelectrolyte on an Oppositely Charged Surface" (to be published in *J. Chem. Phys.*).

Shi, X.B., et al., "Self-Assembled Multilayers and Photoluminescence Properties of a New Water Soluble Poly(para-Phenylene)," in *Mater. Res. Soc. Symp. Proc. Electronic, Optical and Magnetic Properties Org. Mater.* **488**, 133 (1998).



Current-voltage characteristics of aluminum/organic monolayer/silicon devices with different organic interlayers. At forward bias for the aluminum/organic monolayer/p-type silicon device, the p-type silicon is positive. The diode device was tuned with functional organic monolayers, which are reference (no organic layer: open circles), PDDA/NiPc (solid circles), PDDA (open squares), and PDDA/PPP (plus signs). The inset is a diode device with an PDDA/NiPc barrier layer.

Diffusion and Quantum Mechanics

98051

David Schmidt

The objective of our six-month project was to investigate whether quantum-mechanical identical particle effects could be observed in magnetic resonance imaging (MRI) measurements of diffusion in helium-3 gas. This would be a rare example of a quantum-mechanical effect observable in a macroscopic system. The technique could prove useful for investigating other quantum phenomena.

We have attacked this problem on two fronts: a theoretical front and an experimental front. On the theoretical front, we have begun investigating the relation between diffusion and quantum-mechanical interaction cross sections in order to understand what, if any, identical particle effects will have on diffusion. An explicit goal of this work is to understand how to properly scale measurements of helium-4 diffusion with those involving helium-3. We have derived a first-order approximation to the relationship between diffusion and scattering cross sections and have written computer codes to carry out these calculations.

On the experimental front, we have designed, constructed the materials for, and carried out MRI diffusion experiments involving different mixtures of helium-3 and helium-4 gas. The results of these experiments will allow us to test if identical particle effects show up as the relative amount of helium-3 gas is increased. Preliminary analysis of these data have reproduced our earlier measurements of diffusion in pure helium-3 gas, giving us confidence in our technique. We are now beginning to analyze the data from the mixed gases.

All-Solid-State Four-Color Laser

96215

Timothy Gosnell

The goal of this project is to develop a solid-state laser that produces visible output wavelengths, including the blue wavelength that is of commercial interest. 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 leads to population of a high-lying energy level of Pr^{3+}.

This year we explored the use of direct-coated dielectric mirrors on the fiber end faces. Unfortunately, the red laser experiment failed as a result of residual absorption in the fiber coatings, a consequence of the low temperatures needed to deposit the coatings. The blue laser experiment was more successful—we measured ~1 mW of output for modest pump powers. These results were still not optimum, given that output coupling was held to about 1% in order to ensure above-threshold pumping conditions. A separate experiment involving the red laser demonstrated the highest efficiency for an up-conversion laser ever reported, namely 56%.

In a side project carried out with a collaborator at the University of the Andes, we developed a Monte Carlo model of up-conversion processes, illustrating the effects of geometric correlations in up-conversion efficiency. Such a model has wide applicability to up-conversion lasers and optical communications amplifiers.

Publications

Binder, P.M., et al., "Model Showing Effect of Impurity Sublattice Topology on Upconversion in Er^{3+}-Doped Glasses," *Electron. Lett.* **34**, 1348 (1998).

Subpicosecond Electron-Bunch Diagnostic

97009

Bruce Carlsten

The purpose of this project is to develop new, simple bunch-length diagnostics that are capable of measuring lengths below 0.25 ps. Bunch-length diagnostic techniques are falling behind the ability to create very short electron bunches. The first technique we are investigating is to use a transversely deflecting radio-frequency cavity and nonintercepting beam-position monitors (BPMs) to measure the beam's transverse second moments, which are coupled to the bunch length by the deflecting radio-frequency cavity. The second technique we want to demonstrate is based on off-axis, coherent Smith-Purcell radiation.

Last year we demonstrated the ability to measure transverse second moments of the beam without deflection. This year we performed preliminary measurements with deflection. We also established a very sound theoretical basis for the second-moment technique. The idea behind the Smith-Purcell technique is that the intensity of radiation from a beam passing over a grating will be greatly increased for wavelengths long compared with the bunch length. The angle of the radiation to the grating is related to the wavelength; by measuring the angle at which the radiation greatly increases, we can determine the root-mean-square bunch length. We have submitted an experimental plan to the Accelerator Test Facility at Brookhaven National Laboratory to do a proof-of-principle experiment, which should take place in December 1998. We submitted five papers on related second-moment and bunch-length issues to refereed journals.

Publications

Carlsten, B.E., "Characterizing the Emittance Contribution due to Rotated Quadrupoles and Canonical Angular Momentum Using the Quadrupole Scan Technique in Electron Accelerators" (to be published in *Rev. Sci. Instrum*).

Carlsten, B.E., and D.T. Palmer, "Enhanced Emittance Compensation in a High-Frequency RF Photoinjector Using RF Radial Focusing" (to be published to *Nucl. Instrum. Methods Phys. Res., Sect. A*).

Carlsten, B.E., and S.J. Russell, "Coherent Synchrotron Radiation Experiment for the LCLS" (submitted to *Nucl. Instrum. Methods Phys. Res., Sect. A*).

Russell, S.J., "Emittance Measurements of the Sub-Picosecond Accelerator Electron Beam Using Beam Position Monitors" (to be published in *Rev. Sci. Instrum.*).

Russell, S.J., "Unstable Matrix Equations and Their Relationship to Measuring the Emittance of an Electron Beam Using Beam Position Monitors" (submitted to *Nucl. Instrum. Methods Phys. Res., Sect. A*).

An Integrated Solid-State Optical Device with High-Speed Scanning, Variable Focusing, and Frequency-Doubling Capabilities

98027

Quanxi Jia

In the age of increasing demand for larger memories in smaller disk space that are accessible at speeds faster than the present magnetic storage technologies can accommodate, optical data processing and storage have arrived with the promise of high storage densities, high bandwidths for data processing, low power consumption, and potentially lower costs. The objective of this project is to design and fabricate electro-optic lenses, electro-optic scanners, total internal reflection modulators, and second harmonic generators on single-crystal lithium niobate and lithium tantalate wafers and films.

These components will be integrated on a single chip toward developing two specific devices: (1) an electro-optic scanner capable of rastering a light beam by ±2° to ±5° at frequencies approaching 1 GHz, integrated with electro-optic lenses giving variable focusing, and (2) a compact solid-state optical head for read and write applications consisting of a blue light source, lenses to collimate this light, and a scanner at the output capable of rastering the beam for nonmechanical access of data tracks. These integrated devices will act as the basis for a new generation of integrated optical devices incorporating further functions such as modulators, gratings, total internal reflection mirrors, piezoelectric actuators, pyroelectric detectors, and holographic storage components.

This year we designed and demonstrated the first integrated wafer device with electro-optic lenses and electro-optic scanners on a single lithium tantalate chip. The electro-optic scanners were capable of beam deflection up to 12.68 mrad/kV and variable focusing lenses with a power of $(1/f) = 0.233\ \mathrm{cm^{-1}kV^{-1}}$.

Publications

Gopalan, V., and T.E. Mitchell, "Wall Velocities, Switching Times, and Stabilization Mechanism of 180° Domains in $LiTaO_3$ Crystals," *J. Appl. Phys.* **83**, 941 (1998).

Gopalan, V., et al., "Ferroelectrics as a Versatile Solid State Platform for Integrated Optics," *Integrated Ferroelectrics* **22**, 985 (1998).

Gopalan, V., et al., "In Situ Video Observation of 180 Degree Domain Switching in $LiTaO_3$ by Electro-Optic Imaging Microscopy" (to be published in *J. Appl. Phys.*).

Gopalan, V., et. al., "Real-Time Study of Kinetics of 180 Degree Domains in Congruent $LiTaO_3$ under an External Field," *Integrated Ferroelectrics* **22**, 925 (1998).

Gopalan, V., et al., "The Role of Nonstoichiometry in 180° Domain Switching of $LiNbO_3$ Crystals," *Appl. Phys. Lett.* **72**, 1981 (1998).

Gopalan, V., et al., "Switching Kinetics of 180 Degree Domains in Congruent $LiNbO_3$ and $LiTaO_3$ Crystals" (submitted to *Solid State Commun.*).

Kitamura, K., et al., "Crystal Growth and Low-Field 180 Degree Domain Switching Characteristics of Stoichiometric $LiTaO_3$" (to be published in *Appl. Phys. Lett.*).

Development of Ring-Probe Instrumentation for Use in DNA Sequencing

98026

Joe Gatewood

The DNA containing the genetic code for human beings is approximately three billion base pairs in length and contains all the genes responsible for human development, life, and reproduction. The full potential of DNA sequence information will only be realized when we have the capability of rapidly sequencing individual genomes. Ultimately, DNA from millions of individuals will be sequenced to identify disease genes, locate sites of DNA damage, and provide an absolute basis for identification. The routine application of total genomic sequencing will require a "quantum" leap in sequencing instrumentation.

We are developing the foundation for a new, innovative sequencing technology that has the potential of reducing the cost and completion time of sequencing an entire human genome by orders of magnitude. Our accomplishments this year include the design, construction, and evaluation of several electrophoresis instrument and cell mockups; experimental evaluation of several insulating materials based on their electrophoresis current-blocking capability and noise characteristics; development of a new approach for sensor etching; and investigation of a new technique for pore production. We have filed a patent disclosure for our new sequencing technology, and a patent application is in progress.

Imaging Time-of-Flight Ion Mass Spectrograph

97008

Herbert Funsten

The imaging time-of-flight ion mass spectrometer uses a unique design in which an ion beam of known energy transits an electrostatic raster system, passes through a field-free drift tube, and strikes a position-sensitive detector. The trajectory followed by an ion in the drift region depends on the time (or phase of the raster) that the ion transited the raster section. From the detected position of the ion, we can localize the time that the ion passed through the raster system and can derive the distance that the ion traveled through the system. Using these measurements along with the time that the ion was detected, we obtain the time of flight, ion speed, and the ion mass.

The clear advantage of this system (as opposed to a traditional gated or pulsed time-of-flight system) is that ions can be continuously detected, enabling a fast spectrum acquisition and high degree of accuracy. The components of the system have been assembled and will be tested in the upcoming year. A patent disclosure has been filed.

In addition, this year we have demonstrated the chicane mass filter, which consists of a chicane magnet geometry—four dipole magnet sections in series. The first two magnets disperse a monoenergetic ion beam according to mass, and the second two magnets refocus the mass-separated components into a beam that is collinear with the incident beam. Between these two sections, an aperture is adjusted to physically block specific ion masses. The chicane mass filter is particularly suitable for identification of airborne chemical and biological agents using time-of-flight mass spectrometry, because it can remove the lighter constituents of air and allow the trace amounts of the agents to pass through to the mass spectrometer. An invention disclosure has been filed for the chicane mass filter.

Geoscience, Space Science, and Astrophysics

High-Spectral-Resolution Infrared Absorption and Emission Signatures as Observed against Thermal Background Sources for Selected Molecular Species

97051

Stephen Schmidt

Scientists and engineers have long been interested in the identification and measurement of concentrations of both major and minor molecular species that are either naturally present or artificially introduced into the atmosphere. Their interests range from determining molecular species for the purpose of understanding basic global atmospheric processes to monitoring effluents from local pollution sources and industrial plants. The purpose of our work has been to study in great detail the absorption/emission spectra in the 8- to 14-μm wavelength region of trace gases from plumes emitted into the atmosphere.

We used a technique called line-by-line calculations to make high-resolution spectral simulations. The effect of regions that physically differed in pressure, temperature, and constituents was handled by dividing the optical path into "homogeneous" layers and then performing the line-by-line spectral simulations in a layer-by-layer sequence. These calculations included both the relevant trace and naturally occurring atmospheric molecular species, thermal background sources (including those whose emissivity and reflectivity vary with wave number), thermal downwelling, aerosols, and convolution with appropriate instrument functions.

We used calculated spectra in simulations to demonstrate that remote sensing can be an effective technique for detecting trace species in industrial plumes. Using the results of our simulations, we envisioned two industrial applications in which we would be able to detect chemicals present in reasonable concentrations: (1) we could detect carbon dioxide, methane, and nitrous oxide that would be present in an electrical plant plume, and (2) we could detect acetone, tetrachloroethylene, and methyl ethyl ketone solvents that might appear in a reprocessing plant plume.

We also proved the value of remote sensing in a naturally occurring instance. We performed simulations of the measured spectra in the volcano plume and the adjacent sky background for Mt. Popocatepetl near Mexico City. Mt. Popocatepetl is different from most volcanoes because it sits over a layer of limestone that is deep beneath the earth's surface. Because of this limestone, it had been predicted that raised levels of carbon dioxide would be present. We were actually able to detect and measure intermittent increased levels of carbon dioxide in the plume, which heretofore were only suspected.

Regional Climate and Precipitation Variation Assessment via Integrated Global and Regional Coupled Ocean-Atmosphere Modeling

98048

Chung-Chieng A. Lai

Regional climate and precipitation are variables linked with the global climate system by way of scale interactions. Thus, an integrated global and regional coupled ocean-atmosphere model (COAM) system is the vehicle for regional climate and precipitation assessment. Our goal in this project is to make understanding and predicting regional climate and precipitation patterns easier. The specific objectives are to (1) evaluate a regional COAM system within a global-climate-model system environment and (2) use the regional COAM to show the effects of climate variability on precipitation over the southwestern United States.

This year we completed three major tasks. First, we validated the regional COAM model experiment. The experiment case is the 1997 California New Year's flood. We initialized the regional COAM with the National Center for Environmental Prediction's analysis of meteorological conditions. Two simulations were conducted, one without an active ocean component model and the other with. Comparing the results from these two simulations with observed precipitation indicates that the ocean component model plays a crucial role in obtaining the accurate precipitation forecast.

Second, we carried out the California precipitation forecast for the El Niño winter of 1997–1998. We initialized the regional COAM with the University of California at Los Angeles (UCLA) global COAM forecast. The forecast did not compare well with observed precipitation, probably because (1) the UCLA global COAM forecast itself is not good enough and (2) the interface between global and regional COAMs downgraded the accuracy.

Third, we diagnosed the multidecadal variability of summer monsoon precipitation in the southwestern United States during the past century.

Publications

Huang, Z., and C-C. Lai, "Multidecadal Variability of the Monsoon Precipitation in the Southwestern United States during the Past Century" (submitted to *J. Clim.*).

Huang, Z., and C-C. Lai, "Multidecadal Variability of the North American Monsoon Precipitation during the Past Century," in *Proceedings of the 9th Conference of Air-Sea Interactions,* Am. Meteor. Soc., 159 (1998).

Huang, Z., and C-C. Lai, "The Impact of Mesoscale Coastal Oceanic Perturbations on Pacific Winter Storm Parade over California," *EOS, Trans. Am. Geophys. Union* **78**, No. 46, 131 (1997).

A New Method for Modeling Wave Propagation in Strongly Heterogeneous Media: Applications to Seismic-Wave Propagation in the Earth

98024

Michael Fehler

The objectives of this project are to develop and test a new method for modeling seismic-wave propagation in strongly heterogeneous media. The method is called phononic lattice solid by interpolation (PLSI) and can be thought of as an extension to wave-equation modeling of the Lattice-Boltzmann approach used in hydrodynamics. The PSLI method can model wave propagation in realistic Earth models much more reliably than can be done using finite-difference solutions of the wave equation. It can correctly and easily model the effects of topographic variations of the earth's surface on seismic waveforms. Although the method was previously developed by one of our investigators, it has never been carefully evaluated or used for practical modeling of seismic-wave propagation, particularly when the rough surface topography of Earth's surface is modeled.

This year we developed and improved a method for modeling Earth's free surface that allows for rough topography. We submitted a paper that demonstrated and tested the method on several models with known solutions (i.e., we validated the method). We are currently investigating the viability of the method by modeling seismic-wave propagation in several models with strong internal heterogeneity and solutions available from other means.

Publications

Huang, L-J., et al., "Absorbing Boundary and Free Surface Conditions in the Phononic Lattice Solid by Interpolation" (submitted to *Geophys. J. Int.*).

New Windows on Gamma-Ray Bursts

98020

Edward Fenimore

Recently, it has become clear that gamma-ray bursts occur near the edge of the universe and literally flood the entire universe with gamma rays. Associated with the gamma rays is a faint x-ray afterglow that can last for days. The transition from gamma rays to x-rays is a crucial window on the mysterious process that powers these events. Our goal is to use the well-observed temporal structure to unfold what motion is occurring in the bursts.

This year we have studied the temporal and spectral evolution of the gamma rays. We have been able to successfully argue that the gamma-ray phase is not due to a single relativistic shell. We showed that any single shell would have to have a small "filling factor"; that is, only a very small fraction of the shell is capable of contributing gamma rays. Then we determined the average temporal and spectral evolution to be linear functions. Linear functions imply that we are not seeing changes in the relativistic motion in contrast to expectations from theory. Since the x-ray afterglow is a power law rather than a linear function, the afterglows must arise from a separate process (see accompanying figure).

Publications

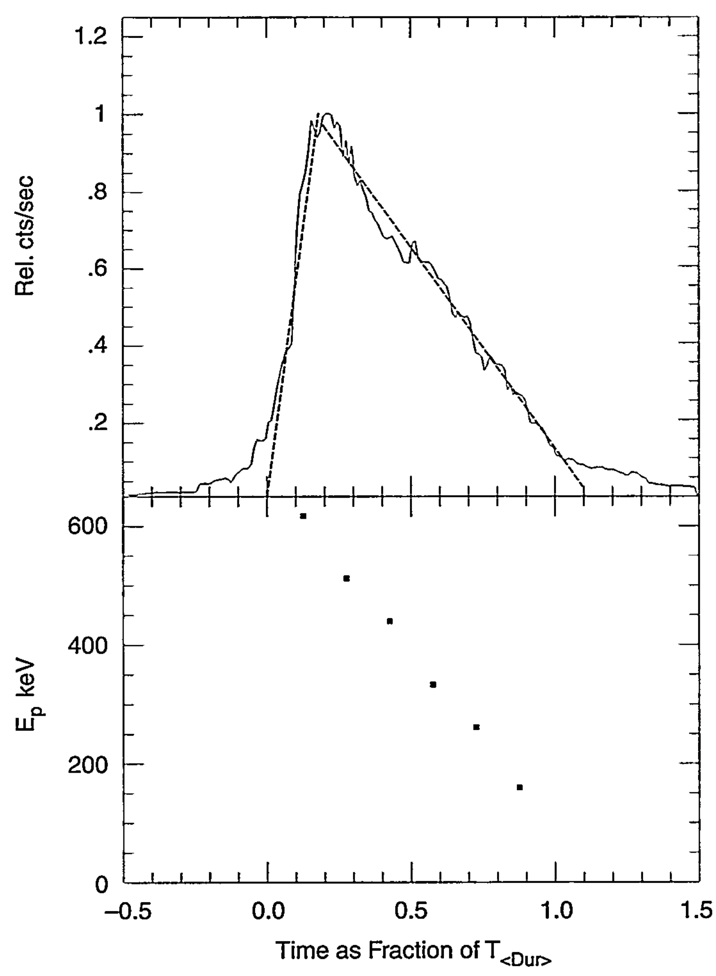
Fenimore, E.E., "The Average Temporal and Spectral Evolution of Gamma-Ray Bursts" (submitted to *Astrophys. J.*).

Fenimore, E.E., et al., "Gamma-Ray Bursts and Relativistic Shells: The Surface Filling Factor" (to be published in *Astrophys. J.*).

Fenimore, E.E., et al., "Kinematic Arguments against Relativistic Shell Models for Gamma-Ray Bursts," *AIP Conf. Proc.* **428**, 656 (1998).

Strohmayer, T., et al. "X-Ray Spectral Characteristics of Ginga Gamma-Ray Bursts," *Astrophys. J.* **500**, 873 (1998).

Sumner, M.C., and E.E. Fenimore, "Variability in Shell Models of Gamma-Ray Bursts," *AIP Conf. Proc.* **428**, 765 (1998).



The average temporal evolution of many bright gamma-ray bursts (top panel) and the average spectral evolution (lower panel). Both are linear functions implying that the cause of the variations is not due to changes in the relativistic motion.

Development of High-Durability Concrete

98043

James Carey

Concrete has a service life that can easily exceed 30 years, even under high traffic and stress. However, many concrete structures fail prematurely because of deleterious reactions that occur among cement phases, the aggregate, and the environment. Our research has focused on developing methods of preventing these reactions from occurring, and thus helping to produce next-generation, high-performance concrete.

One of the most significant of these reactions is the alkali-silica reaction (ASR), which results when cement pore fluids attack unstable silica-rich components of the aggregate to produce a swelling gel. The gel fractures the concrete, producing a maplike network of fractures on the surface that can eventually lead to structural failure. This is a national problem that has been a focus of the federally funded Strategic Highway Research Program.

We have previously developed a method for identifying and studying the extent of ASR in existing concrete structures. In this project, our objective is to find a method of preventing damage from ASR through the development of an understanding of the physical and chemical properties of ASR gel.

In studies this year of the geochemical mechanisms of ASR formation, we have investigated the conditions leading to condensation of the swelling gel. Our research suggests a more complex mechanism than previously described in which a reduction in pH must occur. We have used nonlinear ultrasound geophysical methods to analyze ASR in situ. The studies show that fracture properties of concrete with ASR are distinct and may provide a basis for nondestructive evaluation of concrete damage.

We have also investigated a possible microbial role in the generation of ASR. We have used carbon and oxygen isotope methods and biological stains to identify potential signatures of biological activity in concrete.

Publications

Byers, L.W., et al., "Application of Single-Mode Non-linear Resonant Ultrasound Spectroscopy to Identify Damage in Concrete" (submitted to *Cem. Concr. Res.*).

Carey, J.W., and G.D. Guthrie, Jr., "The Alkali-Silica Reaction in Concrete: Silica Minerals in High-pH Environments" (abstract at the 17th Annual Meeting of the International Mineralogical Association, Toronto, Canada, August 9–14, 1998).

Tsunami from Asteroid and Comet Impacts

98022

Jack Hills

We are producing a systematic study of tsunamis generated by asteroid impacts. Among this year's achievements, we were first to show that tsunamis are the most serious consequence of the impact of asteroids less than 1 km in radius.

Within the overall study, we are focusing first on the areas of Earth that are most susceptible to tsunami run-ups and then concentrating on a few to find how often they are affected by asteroid-generated tsunamis. A preliminary assessment showed that about every 3,000 to 5,000 years, a large-enough impactor falls into an ocean to produce tsunamis more than 50 m high along a sizeable fraction of the coast.

In the past year we have studied the effects of an asteroid 5 km in diameter falling in mid-Atlantic, using a tsunami propagation and run-up code. The code produces tsunamis that inundate the upper two-thirds of the eastern United States to the foothills of the Appalachians. In Europe, the damage is less dramatic: the most vulnerable area is the Spain-Portugal peninsula, where the tsunami runs up to the mountains near the coast at a height reaching 200 m. The situation in northern Europe is more favorable because the extensive, shallow continual shelf around the British Isles reflects much of the tsunami energy back into the Atlantic. Thus, the United Kingdom and the continent facing it are relatively protected.

Publications

Hills, J., et al., "Down-to-Earth Astronomy: Tsunami from Asteroid-Comet Impacts," *Bull. Am. Astron. Soc.* **29**, 1260 (1998).

Hills, J., et al., "Tsunami from Asteroid and Comet Impacts: The Vulnerability of Europe," *Ann. Geophys. IV, Nonlin. Geophys. Nat. Haz.* **16**, 1164 (1998).

Theoretical Research on Dwarf and Classical Novae

96169

Warren Sparks

The goal of this project is to understand mechanisms involved in the accretion processes in both dwarf and classical novae. We are particularly interested in thermonuclear processes as well as material capture processes.

Using new reaction rates and OPAL opacities in our model, we find that the mass being accreted onto the white dwarf is less than that for previous models. Smaller amounts of aluminum-26 are produced while the abundances of phosphorus-31 and silicon-32 increase by a factor of more than 2. The characteristics of our nova models compare favorably with observations of V1974 Cyg 1992, but the ejected mass is a factor of at least 10 less than observed.

We obtained Hubble Space Telescope phase-resolved spectroscopic observations of the white dwarf in U Geminorum. From a determination of the Einstein redshift, we obtained a carbon core white dwarf mass of 1.1 solar masses. The carbon abundance is only 0.05 times solar. The effective temperature of the white dwarf 13 days after outburst is only 32,000 K, anomalously cooler than previous early postoutburst measurements.

Spectra of AL Comae 1 year after its 1995 superoutburst show a slow decline in continuum flux. We found the cooling sequence in AL Comae to be about half as long as that of WZ Sge. This time scale is what is predicted for an outburst that was half the duration of WZ Sge, if shear mixing and angular momentum result in nonspherical accretion onto an equatorial belt.

This year, for the reference book *Astrophysical Quantities*, we completed writing a chapter that contains up-to-date information, tables, and references on classical novae, recurrent novae, nova-likes, dwarf novae, helium cataclysmic variables (CVs), magnetic CVs, D-type symbiotics, and S-type symbiotics. It is the cumulation of many years of collecting and organizing information on the objects.

Publications

Sion, E., et al., "Anomalous Cooling of the Massive White Dwarf in U Geminorum Following a Narrow Dwarf Outburst" (to be published in *Astrophys. J.*).

Sion, E., et al., "Evidence of a Thermonuclear Runaway and Proton Capture Material on a White Dwarf," *Astrophys. J. (Lett.)* **480**, 17 (1997).

Sparks, W., et al., "Cataclysmic and Symbiotic Variables," in *Astrophysical Quantities* (Springer-Verlag, New York, in press), Chap. 17.

Starrfield, S., et al., "Effects of New Nuclear Reaction Rates and Opacities on Hydrodynamic Studies of the Nova Outburst" (submitted to *Astrophys. J. Suppl.*).

Szkody, P., et al., "UV and Optical Spectroscopy of AL Comae One Year After Superoutburst," *Astrophys. J.* **497**, 928 (1998).

Evolution of Coronal Mass Ejections in the Solar Wind at Low and High Heliographic Latitudes

97006

John Gosling

Coronal mass ejections (CMEs) are among the most impressive solar phenomena. These events play a central role in the evolution of the large-scale structure of the solar corona and are the prime cause of large solar wind and geomagnetic disturbances. This project is focused on studying the evolution of CMEs at both high and low heliographic latitudes using a combination of satellite observations and numerical simulations.

This year we used a two-dimensional code to study the evolution of various sorts of CMEs impelled into a solar wind that varies with heliographic latitude in the simple manner revealed by the observations from the Ulysses spacecraft. We found that, even with these simple boundary conditions, the morphology and structure of the solar wind disturbances caused by the CMEs were highly complex. A paper summarizing these results is in progress.

We also used the two-dimensional code to map Ulysses observations made at 4.25 AU back to the vicinity of the Sun in order to relate those measurements to solar observations obtained by the SOHO satellite. We found this mapping to be significantly better than the commonly used technique of assuming that the solar wind speed is constant as a function of heliocentric distance.

Publications

Riley, P., et al., "Relationship between Ulysses Plasma Observations and Solar Observations during the Whole Sun Month Campaign" (to be published in *J. Geophys. Res.*).

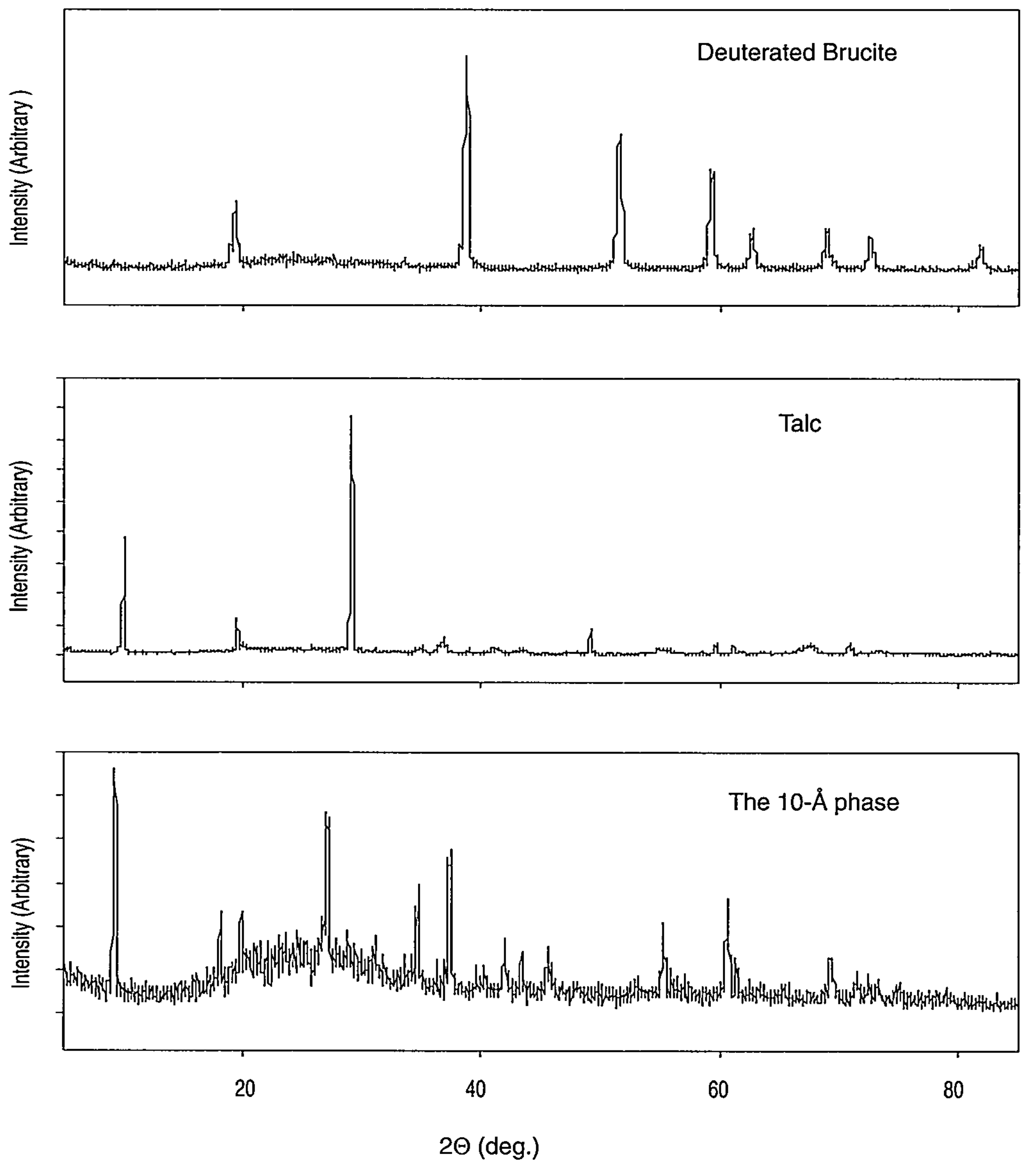
High-Pressure Crystal Chemistry of Hydrous Minerals

98019

Yusheng Zhao

We are studying hydrogen crystal chemistry and the stability of hydrogen-bearing minerals at high pressures and temperatures. As we learn more about bond lengths and vibrational features, we will be able to answer questions about the crystallographic structure of hydrous minerals at extreme conditions. One such question is, Which structures are stable, and what is the range of their abilities to retain hydrogen?

We have successfully synthesized several deuterated hydrous minerals, including brucite (MgOD, at 300°C, 12 hours), talc ($Mg_3Si_4O_{10}[OD]_2$, at 2.5 kbar, 600°C, 48 hours) and the 10-Å phase ($Mg_3Si_4O_{14}D_6$, at 70 kbar, 450°C, 10–48 hours). The deuterated samples have been analyzed by powder x-ray diffraction (see first figure), micro Raman spectroscopy (see second figure), and Fourier transformed IR absorption spectroscopy. We have also succeeded in obtaining precious neutron beam time at the Los Alamos Neutron Science Center for the coming beam cycle starting in October of 1998. Our proposal for beam time received the highest ranking for all DOE Basic Energy Sciences general user proposals, and we were allocated about one-



X-ray diffraction data for deuterated samples of brucite, talc, and 10-Å phase. They all show good crystallization under the synthesis conditions.

eighth of the total neutron time on the HIPD beam line.

We will conduct high pressure-temperature (P-T) neutron-diffraction experiments (see third figure) on simple structured hydrous phases at pressures up to 10 GPa and temperatures up to 1500 K; determine the structure response, such as lattice constants, atom positions, and thermal vibration (Debye-Waller) factors of the hydrous phases to change of pressure and temperature; and apply the results to construct a microscopic model for compression/expansion related to hydrogen bond lengths and angles. The study of high P-T crystal chemistry of hydrous minerals will help to improve our understanding of the composition and dynamics of Earth's deep interior.

Publications

Li, W., et al., "Synthesis and Characterization of Hydrous Minerals for Neutron Diffraction" (American Geophysical Union Fall Meeting, San Francisco, CA, December 7–11, 1998).

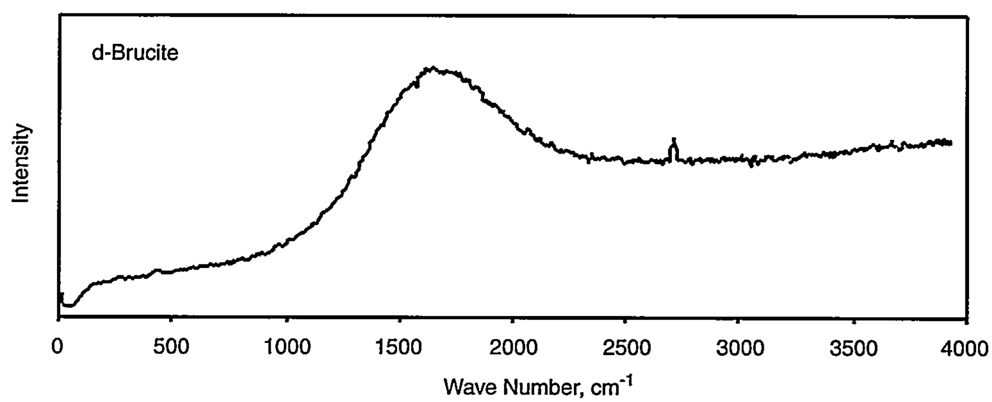
Zhao,Y., et al., "A High P-T Cell Assembly for Neutron Diffraction up to 10 GPa and 1500 K" (submitted to *High-Pressure Res.).*

Zhao,Y., et al., "Correction of Diffraction Optics and P-V-T Determination Using Thermoelastic Equations of State of Multiple Phases" (submitted to *J. Appl. Crystallogr.*).

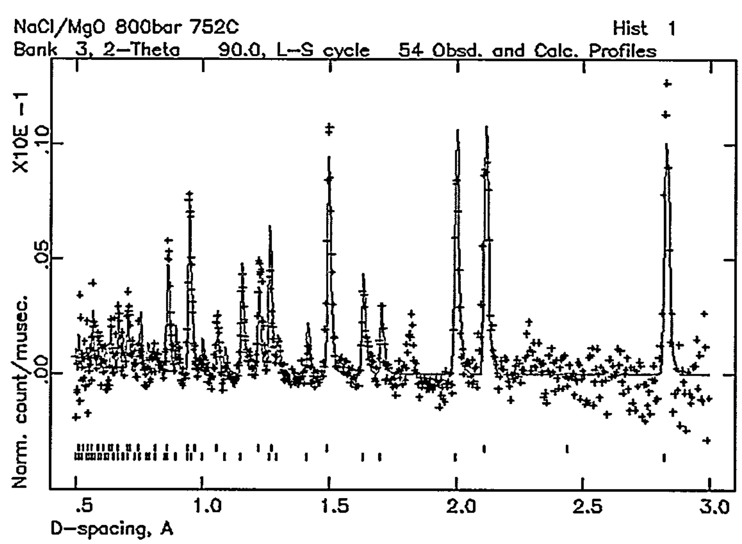
Zhao,Y., et al., "High P-T Structures and Thermoelastic Equations of State of Clinopyroxenes and Implication to Earth Mantle Modeling" (American Geophysical Union Fall Meeting, San Francisco, CA, December 7–11, 1998).

Zhao,Y., et al, "High P-T X-Ray/ Neutron Diffraction Study of Mantle Mineral Physics" (IGPP Annual Meeting, September 14–15, Los Alamos, NM, 1998).

Zhao,Y., et al., "TAP-98: A New Design of Toroidal Anvil Press for High P-T Neutron Diffraction" (IUCr-HP98, APS/ANL, Argonne, IL, November 14–17, 1998).



Raman spectra of deuterated brucite powders. A sharp peak at 2700 cm^{-1}, corresponding to the characteristic vibration of the OD stretching, can be clearly identified. There is no detectable OH vibration at 3668 cm^{-1}, implying that the substitution of deuterium by hydrogen from the water in the atmosphere was minimal.



High P-T neutron diffraction data of MgO, which is the dehydrated phase of the brucite MgOH, at P = 4.2 GPa and T = 1025 K. The high quality of the diffraction data allows the structure refinement for atomic positions as well as thermal vibrations.

Regge Geometrodynamics in Support of Gravity-Wave Astronomy

96167

Raymond Laflamme

Because gravity-wave astronomy may allow deeper probing into cosmic events than can be done with any other known means, our goal is to solve Einstein's gravitational field equations. During this effort we developed the first fully (3+1)-dimensional general relativity code based on the lattice approach known as Regge calculus (RC). In RC we seek a lattice built of internally flat four-dimensional simplexes to represent a space-time that is a solution of Einstein's field equations. This year we have made progress on three fronts.

On the foundations front, we derived the RC analogue of the Hilbert actions. We recovered the usual RC action by way of a decomposition of the simplicial geometry into four-dimensional cells defined by the simplicial (Delaunay) lattice as well as its dual (Voronoi) lattice. Within the simplicial geometry, the Riemann scalar curvature, the proper four-volume, and hence, the Regge action were shown to be exact in the sense that the definition of the action does not require one to introduce an averaging procedure or a sequence of continuum metrics that are common in all previous derivations. It appears to us that the unity of these two dual lattice geometries is a salient feature of RC.

On the numerics front, we developed and benchmarked the first discrete-time four-dimensional application of RC. We implemented a novel two-surface initial-data prescription for RC and provided the first fully four-dimensional application of an implicit decoupled evolution scheme (the Sorkin evolution scheme). Then we benchmarked this code on the Kasner cosmology—a cosmology that embodies generic features of the collapse of many cosmological models. We also demonstrated second-order convergence properties of the solution to the continuum.

On the application front, the effective energy density of a sufficiently strong set of gravity waves in a localized region of space-time can, in principle, collapse and form a black hole. The generic features of gravitational collapse (from the literature) argue that in the very early universe, the energy density of gravity waves dominated over all other sources of energy. Thus we can very well believe that gravity wave-generated black holes formed in the very early universe. These primal chaos black holes could be seeds for the formation of galaxies and could also account for some of the "missing mass" of the universe. A time-symmetric model of these gravity-induced black holes was provided by Dieter Brill of the University of Maryland, and we are now using RC to model Brill waves. We have resolved in RC the formation of apparent horizons as the amplitude of the localized gravity waves is increased. Our results agree with recent results from the Potsdam Gravity Group.

Publications

Chakrabarti, S., et al., "Geodesic Deviation in Regge Calculus" (submitted to *Class. Quantum Grav.*).

Gentle, A.P., and W.A. Miller, "A Fully (3+1) Dimensional Regge Calculus Model of the Kasner Cosmology," *Class. Quantum Grav.* **15**, 389 (1998).

Gentle, A., et al., "Apparent Horizons in Simplicial Brill Wave Initial Data" (submitted to *Class. Quantum Grav.*).

Miller, W.A., "The Hilbert Action in Regge Calculus," *Class. Quantum Grav.* **14**, 199 (1997).

Low-Energy Neutral-Atom Imager

96156

Herbert Funsten

Energetic neutral atoms (ENAs) are hot magnetospheric plasma ions that are neutralized by charge exchange with cold geocoronal neutrals of the earth's extended atmosphere. ENAs can be remotely detected and can provide information about the source plasma, such as the spatial distribution, temperature, density, and composition. The low-energy neutral-atom (LENA) imager, which was developed under this program to provide first measurements of ENAs in the energy range of 1 to 24 keV, was successfully launched in late 1997 and has measured ENA distributions over each satellite orbit.

The data have shown numerous interesting features. Of particular interest are intermittent bursts, lasting up to half an hour, that appear singly or as two or more apparently correlated events. Two bursts are observed: the first lasts approximately 30 minutes and covers all energy ranges; the second is about the same duration but is dispersed in energy (the event starts at high energies and ends at low energies). Data from the LENA imager continue to be collected and will be analyzed in considerable depth.

Determining the Mass of the Universe

97013

Michael Warren

The average mass density of the universe, parameterized by *W*, is the most sought after single number in cosmology. It determines whether the universe is open and expanding forever ($W < 1$) or closed and eventually recollapsing ($W > 1$). Unfortunately, after a half century of research, *W* 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 are testing the conventional approximations of *W* with state-of-the-art numerical simulations of gravitational clustering in the universe. The simulations provide complete dynamical information about both galaxies and dark matter; preliminary work has shown, for example, that the usual treatment of galaxies as point masses is unjustified. With the numerical work as a guide, this effort will yield methods of measuring the mass in the universe that have been optimized from a theoretical perspective and tested in numerical simulations.

We have continued to refine our data analysis capabilities in order to enable data mining from the 10-Gbyte data sets we are currently able to generate and to enable comparisons with the largest observational data sets such as the Las Campanas Redshift Survey. A paper comparing our *N*-body code with the dozen leading cosmological codes in the world shows that our numerical methods provided the highest-resolution simulation among all of the participants. One of the simulations performed for this project won the 1997 Gordon Bell Prize for superior effort in practical parallel processing research. Our tree code, which scales as $O(N \log N)$, sustained 170 Gflops over a continuous 9.4-hour 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. In addition, we have developed the capability to perform large parallel *N*-body simulations locally on the Avalon cluster.

Publications

Frenk, C.S., et al., "The Santa Barbara Cluster Comparison Project: A Test of Cosmological Hydrodynamics Codes" (submitted to *Astrophys. J.*).

Pfitzner, D.W., et al., "Halo World: Tools for Parallel Cluster Finding in Astrophysical *N*-Body Simulations," *Data Mining and Knowledge Discovery* **1**, 419 (1997).

Warren, M.S., et al., "Avalon: An Alpha/Linux Cluster Achieves 10 Gflops for $150K," in *Supercomputing '98* (IEEE Computing Society, San Jose, CA, 1998).

Lightning in the Atmosphere and in the Solar Nebula

98018

Joseph Borovsky

The objectives of this project are to develop a model for lightning stepped leaders in the earth's atmosphere and to develop a method to scale this model into the conditions of the solar nebula. Lightning is a naturally occurring phenomenon that is not at all well understood. In the earth's atmosphere, lightning leads to deaths and forest fires and produces chemical changes that can have important consequences for global warming. In the solar nebula, lightning may have been the catalyst for chemical and physical changes that had effects which are not as yet understood.

This year we derived a set of simplified differential equations that have Fourier coefficients that are similar to the Fourier coefficients of a full electrodynamic model of field propagation along lightning channels. Solutions to these equations showed that the movement of charge along the lightning channel is diffusive, rather than wavelike.

These equations were then generalized to account for the ohmic heating of the lightning channel by the flow of charge in the channel. Including ohmic heating makes modeling the movement of charge a highly nonlinear problem. In certain parameter regimes, we found that the diffusive, nonlinear movement of charge resembles a wave, which is similar to observations of the movement of charge in lightning in the earth's atmosphere.

A draft manuscript summarizing these results and their consequences for the understanding of lightning is in preparation. We plan to use our charge-propagation formulation to examine the propagation of charge and voltage globally along lightning channels that are spatially extended by stepped-leader processes.

Micro/Macroscale Coupling in Magnetospheric Plasmas

96146

S. Peter Gary

The project objective was to use plasma theory, computer simulations, and analysis of spacecraft data to develop new scaling relations for the consequences of small-scale instabilities in the magnetosphere. This year we developed a model that examined, amongst others, the whistler heat flux, the Alfven heat flux, and the electron/ion acoustic instability modes. The Alfven and ion acoustic instabilities both resonate with core electrons. Our work has led to the hypothesis that core heating by these two modes at the critical condition establishes a lower bound on beta.

We have also completed two-dimensional simulations that show that enhanced fluctuations from the firehose instability impose a bound on the proton temperature anisotropy of the same form as that of the linear theory threshold. In a study of electromagnetic proton/proton instabilities, our results indicate that the strongly unstable regime of the Alfven mode (previously unstudied in a solar wind model) has the lower threshold at sufficiently large beam density and sufficiently small core beta. The magnetosonic mode had been previously regarded by several researchers as the dominant proton/proton instability in the solar wind. Our work also implies that Alfven fluctuations are the more likely source of collisionless dissipation for long wavelength magnetic turbulence cascading to shorter wavelengths at $b_p < 1$, and magnetosonic fluctuations are more likely to dissipate such turbulence at $b_p > 1$. Two-dimensional initial-value hybrid simulations of a magnetosonic instability and an Alfven instability in a homogeneous plasma show that enhanced fluctuating fields from both instabilities yield a reduction in the relative drift speed and a characteristic heating of the more tenuous component temperature perpendicular to the background magnetic field. Ensembles of simulations yield constraints on the fluctuating field energy density, the relative drift speed, and component anisotropies.

Publications

Daughton, W., and S.P. Gary, "Electromagnetic Proton/Proton Instabilities in the Solar Wind," *J. Geophys. Res.* **103**, 20,613 (1998).

Gary, S.P., "Collisionless Dissipation Wavenumber: Linear Theory" (submitted to *J. Geophys. Res.*).

Gary, S.P., et al., "Lower Bound for Electron Core Beta in the Solar Wind," *J. Geophys. Res.* **103**, 14,559 (1998).

Gary, S.P., et al., "Proton Resonant Firehose Instability: Temperature Anisotropy and Fluctuating Field Constraints," *J. Geophys. Res.* **103**, 14,567 (1998).

High-Velocity Neutron Stars

96142

Edward Fenimore

High-velocity neutron stars were discovered as radio pulsars that were displaced from the supernova remnant where they were born. Apparently, the supernova birth process can kick the neutron star up to speeds in excess of 2000 km/s, enough to give them an escape velocity from our galaxy. Therefore, we would expect many high-velocity neutron stars to be far from the plane of the Milky Way. Neutron stars often produce large x-ray flares, and, if neutron stars were associated with gamma-ray bursts, the x-ray afterglows of gamma-ray bursts should appear to be flashes from high-velocity neutron stars.

We used the ROSAT (German X-Ray Astronomy Satellite) data to search for large flashes of x-rays that might be from high-velocity neutron stars. We did indeed discover 24 new flashes, including the largest ever observed. The source of the flash is likely to be a dwarf M star. If so, this is the most extreme type of star known to produce such large x-ray flares.

Publications

Fenimore, E.E., "Gamma-Ray Bursts Spectra and Time Histories from 2 to 400 keV," *Nucl. Phys. B* **69**, 635 (1998).

Li, H., et al., "The Expected Rates of X-Ray Flashes and Gamma-Ray Bursts in ROSAT, Preliminary Results," *Gamma-Ray Bursts: 4th Huntsville Conference* **AIP 428**, 420 (1998).

Strohmayer, T., et al., "X-Ray Spectral Properties of Gamma-Ray Bursts," *Gamma-Ray Bursts: 4th Huntsville Conference* **AIP 428**, 461 (1998).

Sun, X., et al., "X-Ray Flashes in ROSAT PSPC Data," *Gamma-Ray Bursts: 4th Huntsville Conference* **AIP 428**, 456 (1998).

Ices on Titan: Laboratory Measurements that Complement the Huygens Probe

97030

Jeanne Robinson

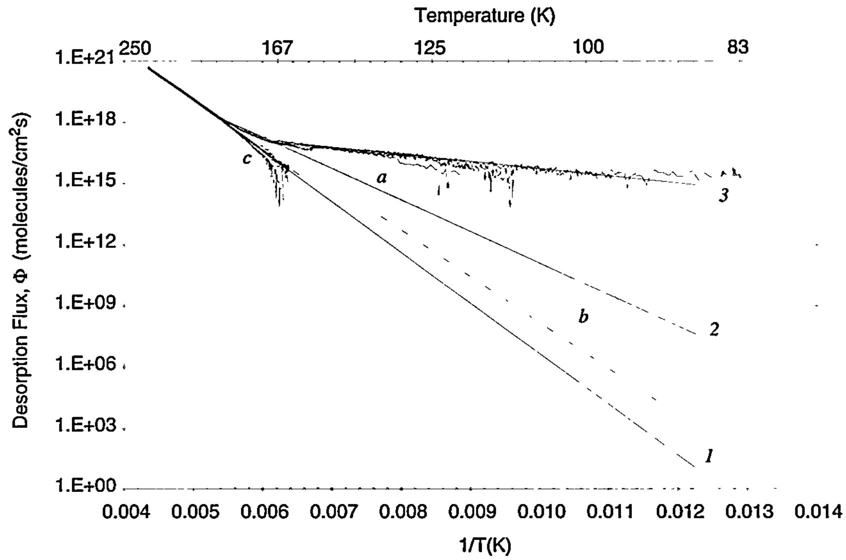
We are studying the physical chemistry of Saturn's moon Titan to address the heterogeneous processes influencing both its present state and past evolution. Laboratory-grown ice films of amorphous solid water (ASW) near 95 K are surrogates for Titan's crust. We have developed the capability to characterize the morphology, surface area, and thermodynamic properties of these complex substrates.

Specifically, we used mass spectroscopy to monitor the vapor pressure of water over the vapor-deposited ice films (see the first figure) and second-harmonic generation to monitor the phase behavior of the ice during annealing (see the second figure). The rate of desorption of water from the surface of ASW and the higher-temperature crystalline phases was determined from the vapor pressure measurements. We have observed three distinct regions corresponding to the three phases of ice over the temperature range of ~80 to 240 K, in agreement with the second-harmonic generation measurements. The rate of desorption at 95 K is about 10^{16} molecules·cm^{-2}·s^{-1}. From this analysis, it is clear that the differing pressures of the amorphous and crystalline ices are a result of well-characterizable differences in the sublimation and desorption energies, indicative of distinct stable or metastable forms.

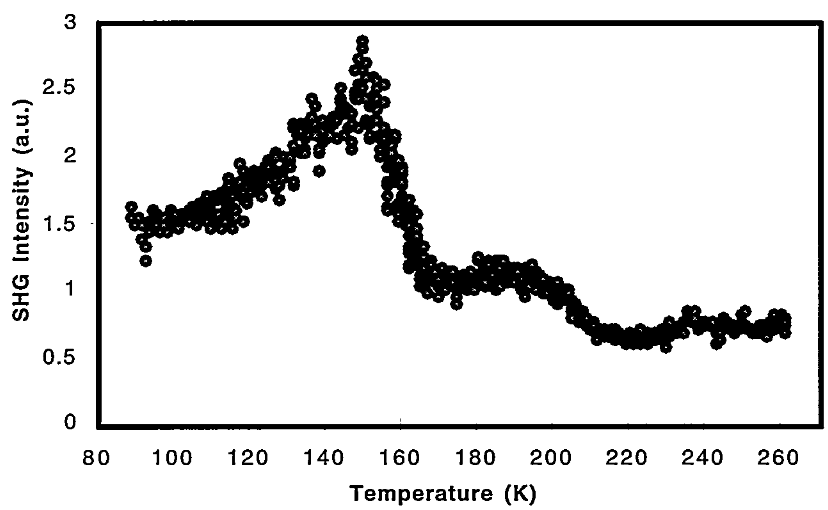
The second-harmonic measurements are the first nonlinear optical measurements of three phase transitions of water ice. Using laser excitation at 1064 nm, we were able to improve the signal-to-noise ratio by over 100, making the glass transition near 125 K, which had previously only been measured by complicated calorimetry schemes, readily apparent.

Publications

Henson, B.F., et al., "The Characterization of Porous Water Ice Films by Physical Adsorption, Mass Spectrometry, and Second Harmonic Generation: Stratospheric and Astrophysical Implications" (submitted to *J. Phys. Chem.*).



Water desorption flux as a function of inverse temperature for deposited water ice during annealing (labeled 2 and 3) and recooling (labeled 1). Three replicate measurements are shown. We calculated the flux from the measured vapor pressure over the ice as determined by mass spectroscopy. The solid lines are linear fits to the data based on zero-order kinetics with an Arrhenius temperature dependence. The dashed lines (labeled a, b, and c) are calculations using parameters reported in the literature.



Second-harmonic generation (SHG) spectrum (in arbitrary units) for 1064-nm excitation from a 0.5-g sample of neat water ice condensed at 90 K during annealing to 260 K. The glass transition (~125 K), the amorphous to cubic crystalline transition (~150 K), and the cubic to hexagonal transition (~215 K) are observed as a function of increasing temperature.

Sedimentary Basin Response to Strong Ground Motion in Populous Regions

96180

Paul Johnson

We have addressed the long-standing question regarding nonlinear sediment response in the Los Angeles region by testing whether sediment amplification was similar between the Northridge earthquake and its aftershocks. Comparing the weak- and strong-motion site responses at 15 sediment sites, we found that amplification factors were significantly less for the main shock, implying systematic nonlinearity. The difference is largest between 2 and 4 Hz (a factor of 2), and is significant at the 99% confidence level between 0.8 and 5.5 Hz.

The inference of nonlinearity is robust with respect to the removal of possibly anomalous sediment sites and with respect to how the reference-site motion is defined. Furthermore, theoretical ground-motion simulations show no evidence of any bias from finite-source effects during the main shock. Nonlinearity is also suggested by the fact that the four sediment sites that contain a clear fundamental resonance for the weak motion exhibit a conspicuous absence of the peak in the strong motion. Although we have taken the first step in establishing the presence of nonlinearity, it remains to define the physics of nonlinear response and to test the methodologies routinely applied in engineering practice. The inference of nonlinearity implies that care must be exercised in using sediment-site data to study large earthquakes or to predict strong ground motion.

Publications

Beresnev, I.A., et al., "Magnitude of Nonlinear Sediment Response in Los Angeles Basin during the 1994 Northridge, California, Earthquake" (to be published in *Bull. Seismol. Soc. Am.*).

Beresnev, I.A., et al., "Stochastic Finite-Fault Modeling of Ground Motions from the 1994 Northridge, California, Earthquake: II Widespread Nonlinear Response at Soil Sites" (to be published in *Bull. Seismol. Soc. Am.*).

Field, E.H., et al., "Nonlinear Ground-Motion Amplification by Sediments during the 1994 Northridge Earthquake," *Nature* **390**, 599 (1997).

Field, E.H., et al., "Nonlinear Sediment Response during the 1994 Northridge Earthquake: Observations and Fine-Source Simulations" (to be published in *J. Geophys. Res.*).

Field, E.H., et al., "Nonlinear Site Response: Where We're At" (to be published in *Seismol. Res. Lett.*).

Coronal Mass Ejections in the Solar Wind

96147

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 explored how three-dimensional magnetic reconnection in the solar corona can form CMEs in the solar wind that are magnetic flux ropes. We have also examined how such reconnection relates to concepts of magnetic helicity conservation. We find that the helicity associated with flux ropes in the solar wind can be the result of helicity emerging from beneath the solar surface or it can be a consequence of photospheric motions. The actual helical field lines observed in the solar wind in our model are a consequence of reconnection following eruption of a CME.

This and other results on the magnetic topology of CMEs were summarized in a paper we wrote for the first book ever devoted solely to magnetic helicity. For a book on stellar winds, we also wrote a review paper on solar wind, which included three-dimensional aspects of CMEs in the solar wind.

Publications

Gosling, J.T., "The Role of Reconnection in the Formation of Flux Ropes in the Solar Wind," in *Magnetic Helicity in Space and Laboratory Plasmas,* A.A. Pevtso, R. Canfield, and M. Brown, Eds. (Amercian Geophysical Union, Washington, DC, in press).

Gosling, J.T., "The Solar Wind in Three Dimensions," in *Cyclical Variability in Stellar Winds*, L. Kaper and A.W. Fullerton, Eds. (Springer Verlag, Berlin, 1998), p. 57.

High-Resolution Records of Global Climate Change

97014

Steven Goldstein

Paleoenvironmental research continues to indicate that very rapid shifts in climate took place during the past 100,000 years. However, the timing and origin of these shifts can only be evaluated with high-resolution dating methods. Our objective is to apply and evaluate accurate and highly precise uranium-series chronometers for dating paleoenvironmental records stored in deep-sea corals and polar ice cores. With successful development of these dating techniques, rapid variations in ocean ventilation, ocean composition, atmosphere composition, and climate can be accurately correlated. This would improve understanding and verification of models for both past and present global change.

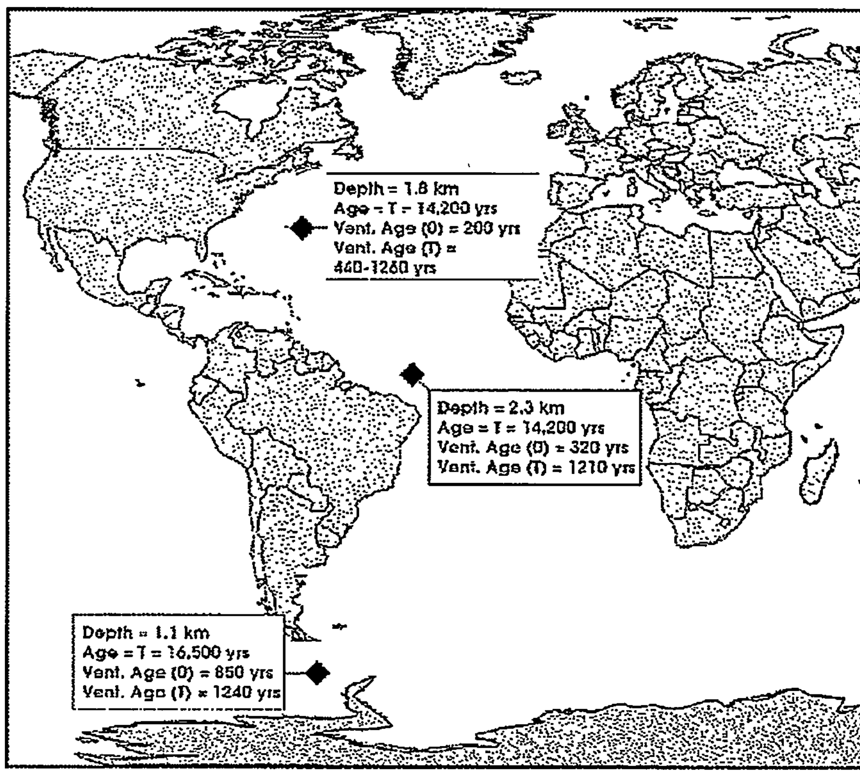
Available data for deep-sea corals (see accompanying figure) suggest a markedly different deep-water circulation pattern during the last glacial maximum ~15,000 years ago. During the past year we have obtained additional deep-sea coral samples from the South Pacific Ocean that have carbon-14 dates corresponding to this time interval. Because these new samples have significant oxide coatings, we have evaluated an acidic cleaning technique that would prevent readsorption of thorium and protactinium onto these samples during the cleaning process. We have also initiated efforts to model the available deep-sea coral ventilation data using the Parallel Ocean Program circulation model at Los Alamos.

For dating dusty polar ice, we have developed an improved method based on a solution of ethylenediaminetetracetic acid (EDTA) that effectively complexes actinides in solution while minimizing dust dissolution during ice melting and filtration. The improved processing methods for deep-sea corals and polar ice samples should permit collection of additional uranium-series dates next fiscal year.

Publications

Cheng, H., et al., "Systematics of Combined U/Th–U/Pa Dating," *Transactions, American Geophysical Union* **78**, 759 (1997).

Cheng, H., et al., "The Systematics of Combined U-230Th-231Pa Dating for Carbonates" (to be published in *Geochimica Cosmochimica Acta*).



Map showing available data for seawater ventilation from the past glacial maximum, approximately 14 to 17 thousand years ago, based on deep-sea coral dating. Our results from the Drake Passage indicate an increase in ventilation age by ~400 years relative to current conditions, consistent with increased ventilation ages for samples from the Equatorial and North Atlantic Oceans. These results suggest that deep Atlantic circulation was more sluggish, most likely because of reduced formation and/or transport of deep water in the North Atlantic.

Balloon-Based, High-Time-Resolution Measurements of X-Ray Emissions from Lightning

97029

David Suszcynsky

This project consists of a series of balloon flights to collect high-time-resolution x-ray and electric-field-change measurements in thunderstorms in order to study the validity of the runaway air-breakdown mechanism. The runaway air-breakdown mechanism is currently the leading theory to account for unexplained x-ray enhancements associated with lightning that have been recently measured from aircraft and balloons. Runaway breakdown is also believed to be the basic process responsible for recently discovered above-cloud lightning events (sprites, blue jets, etc.), the source of satellite-based observations of gamma-ray bursts of atmospheric origin, and the source of transionospheric pulse pairs measured from the ALEXIS and FORTE satellites.

No measurements with adequate temporal resolution have been made to confirm the operation of runaway breakdown in thunderstorms. The results of our experiments could have important implications for understanding fundamental issues associated with lightning initiation, above-cloud lightning, and the global electric circuit and for the design of sensors to measure the electromagnetic pulse from an atmospheric nuclear detonation.

This year we have redesigned and fabricated two lightweight payloads based on improvements from our prior work. We performed an initial test flight of one of the payloads in an active thunderstorm for engineering purposes. Results of a flight to a 22-km altitude indicate that only minor revisions are needed to complete our scientific objectives. Data from the flight are currently being analyzed. We are collaborating with the University of Oklahoma and the National Severe Storms Laboratory on the balloon flight and data analyses.

Publications

Eack, K., et al., "A High-Time-Resolution, Balloon-Borne X-Ray Detector," in *Proceedings of the American Geophysical Union Fall 1998 Meeting* (American Geophysical Union, Washington, DC, in press).

Numerical Simulations of Convection Experiments and the Earth's Interior

96149

Gary Glatzmaier

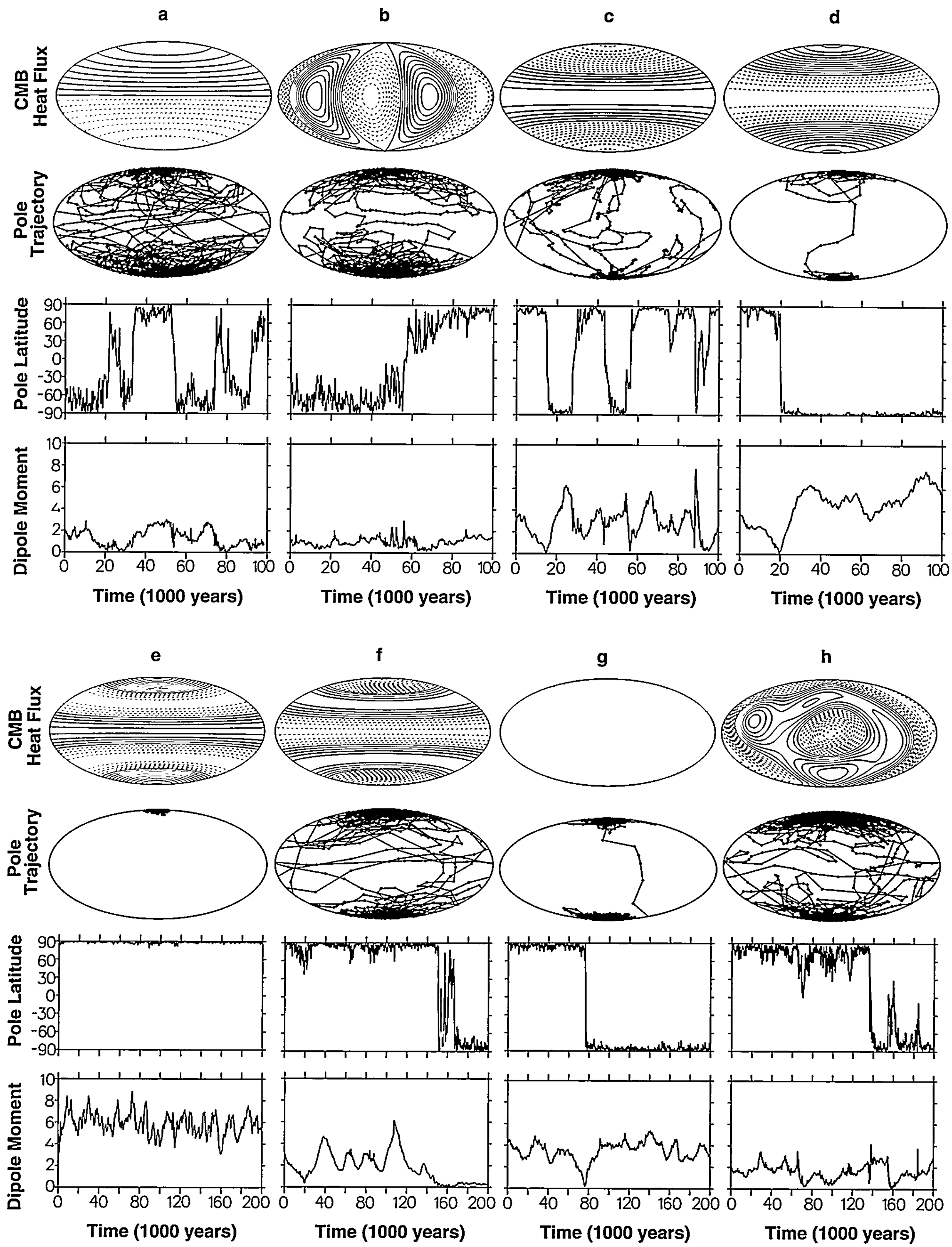
We are studying the dynamics of Earth's core by conducting three-dimensional (3-D), dynamically consistent computer simulations of convection and magnetic field generation and comparing the results with geophysical observations. We have completed eight new simulations, four that span 100,000 years each and four that span over 200,000 years each. They differ only in the prescribed pattern of heat flux being conducted out of the core into the mantle above. Spontaneous magnetic dipole reversals occurred in all but one of these cases. The accompanying figure shows, for each case, the pattern of heat flux imposed at the core-mantle boundary, the trajectory of the south magnetic pole, and the pole latitude and dipole moment versus time.

Our results illustrate how the thermal structure of the lower mantle, which evolves a million times more slowly than the fluid core below, controls the dynamics of the fluid core and the resulting structure and evolution of the magnetic field that is generated. These results begin to explain why, as seen in the paleomagnetic record, the frequency of magnetic reversals, the duration of the reversal events, and the intensity and secular variation of the paleomagnetic field can be so different for different geological epochs.

Publications

Christensen, U., et al., "Dynamo Models and Geomagnetic Field Structure," *Geophys. Res. Lett.* **25**, 1565 (1998).

Hollerbach, R., and G.A. Glatzmaier, "Mixed-Parity Solutions in a Mean-Field Dynamo Model," *Stud. Geophys. Geod.* **42**, 239 (1998).



Eight dynamo simulations (a–h) with different imposed patterns of radial heat flux at the top boundary, as illustrated in the top row. The other rows show the trajectory of the south magnetic pole, the pole latitude, and the dipole moment versus time.

Close Encounters of Asteroids and Comets to Planets

96164

Jack Hills

The objective of this research was to study the physics of close encounters of comets and asteroids to planets. Included are encounters in which the gravitational tidal field of the planet distorts and even breaks up the comet and encounters that are close enough that the intruder grazes the atmosphere of the planet. Such grazing collisions have produced some of the most spectacular and widely observed meteors of the past few decades.

This year we continued our work on grazing atmospheric encounters using our new spherical atmospheric model. We found the conditions under which asteroids of various sizes could be captured into bound orbits around Earth in grazing conditions. We found the sizes of meteorites that could survive the grazing collisions to impact Earth. We found the damage done by such grazing collisions due to blast waves, earthquakes, crater formation, and tsunami.

Our model shows that grazing collisions tend to lead to more of the kinetic energy of the meteors being dissipated in the atmosphere. This dissipation causes the damage to be more strongly concentrated as blast waves and fires generated by the bright meteor rather than as ground impact damage such as craters, earthquakes, and tsunami. In extreme cases, an asteroid a kilometer in diameter could graze Earth to produce fires over one or more continents and produce a corridor of blast damage many tens of kilometers across and thousands of kilometers long without the asteroid producing any crater or tsunami.

Publications

Hills, J., and P. Goda, "Damage from the Impacts of Small Asteroids and Comets," *Planet. Space Sci.* **46**, 219 (1998).

Integrated Systems Analysis Applied to Environmental Remediation

97062

R. Wayne Hardie

The purpose of this project was to apply and test systems-analysis approaches to explore methods of reversing the environmental decline in the Salton Sea (Sea). This decline is thought to be the result of the increased salinity of the Sea. We therefore examined four technologies/solutions to reduce the salinity: desalination; pump-in, pump-out; pump-out; and diking. Success in this project will have the multiple advantages that we will be addressing a significant national environmental problem, that our system approaches will be tested and proved, and that our capability and credibility to tackle problems of this nature will be expanded.

This year we focused on the pump-out option. The primary concerns were the effects of reducing the size of the Sea and where to put the water that was pumped out. Our analysis indicated that inserting a portion of the pumped-out water from the Sea into the geothermal aquifer near the Sea was feasible. About one-third of the required annual pump-out water could be inserted into the geothermal aquifer to replace losses due to geothermal electrical generation. There also appeared to be a sufficient reservoir in the geothermal aquifer to hold the total required annual pump-out water for 10 to 20 years.

Publications

Hardie, R.W., "Evaluation of Options for Reclamation of the Salton Sea," Testimony before the Subcommittee on Water and Power, US House of Representatives Committee on Resources, Washington, DC, March 12, 1998.

Hardie, R.W., and G.R. Thayer, "Evaluation of Options for Remediation of the Salton Sea" (Salton Sea Symposium, Rancho Mirage, CA, January 12, 1998).

Cosmology with Massive Neutrinos

96163

Wojciech Zurek

The addition of massive neutrinos to the standard cold-dark-matter cosmology was originally motivated by indications that the galactic-scale structure has less power than cold dark matter would predict. This conflict became acute after Cosmic Background Explorer (COBE) established rather stringent bounds on the large-scale power. Our goal is to systematically examine available data on galactic and large-scale powers and to explore the resulting cosmological implications, while confirming the data with various theoretical models of cold dark matter in the universe.

Massive neutrinos may have had a significant influence on structure formation in the early universe. However, with the evidence pointing to lower, cosmologically insignificant masses, we have broadened the scope of the project to include study of cosmological phase transitions with topological defect formation. The progress on this front has been significant, as our two recent publications show. Much of the cosmology work benefited from the availability of the parallel supercomputer "Avalon" in the Theoretical Division at Los Alamos.

Publications

Yates, A., and W.H. Zurek, "Vortex Formation in Two Dimensions: When Symmetry Breaks, How Big are the Pieces?" *Phys. Rev. Lett.* **80**, 5477 (1998).

Zurek, W.H., and P. Laguna, "Quenches, Dissipation, and Cosmology" (to be published in *Phys. Rev. D*).

Modeling Core-Collapse Supernovae in Three Dimensions

96166

Michael Warren

The deaths of massive stars in supernova explosions are the most violent events ever witnessed by human beings. As a consequence, understanding core-collapse supernovae represents a fascinating challenge that requires the convergence of such varied areas of expertise as nuclear physics, neutrino transport, and hydrodynamics. In addition, the extreme conditions of density, temperature, and gravity, which reign at the heart of a supernova, make such stars natural laboratories in which to test ideas about physics that are otherwise difficult to investigate on Earth.

We have previously established that the mechanism that powers supernova explosions relies on large-scale convection driven by neutrino heating. However, determining the pattern of this convection demands three-dimensional simulations. Using our combined expertise in parallel computing and supernova theory, we are beginning to carry out such simulations.

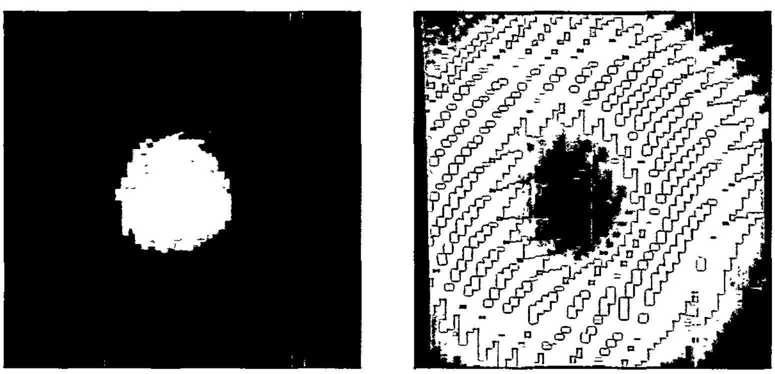
With collaborators at the University of Berne and the University of California at Santa Cruz, we have been modeling the kick imparted to a neutron star when the accretion of matter is from one side and the neutrino emission is similarly asymmetric. The resulting "neutrino rocket" in some cases accelerates the neutron star rapidly. We can then use the results to estimate the kick velocity imparted to neutron stars at birth, which has far-reaching implications for models of gamma-ray burst sources.

Fundamentals of Laser Ablation in the Analysis of Geological Materials

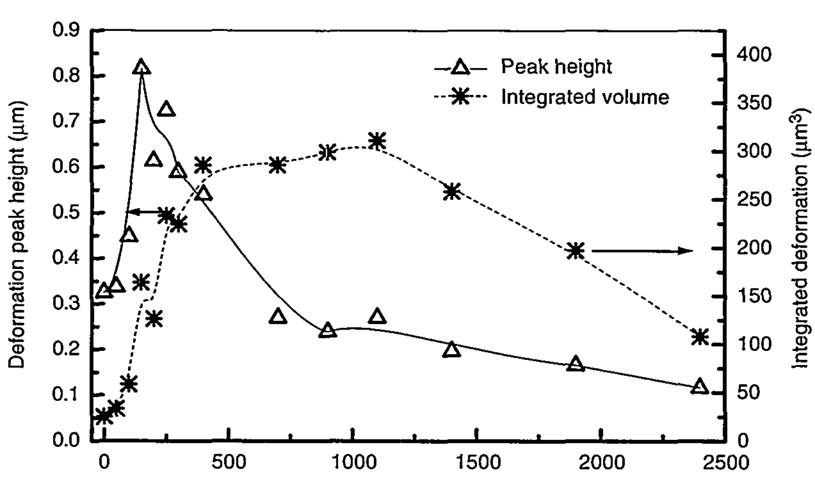
96197

Aaron Koskelo

Laser ablation underlies many of the promising methods for real-time materials recognition and rapid chemical analyses of environmental samples. Such methods could lead to faster, more effective, and cheaper means of analysis and identification than current methods. Rather than taking samples, sending them off to a laboratory, and waiting days for the results of an analysis, one could rapidly survey a site. One could then focus on the areas of greatest concern instead of wasting time and money in clean areas. Our goal is to understand how broadly laser ablation techniques can be applied. We seek to identify the most important properties of different materials that determine their response to the energy absorbed from pulsed laser light.



Transient deformation of calcite at 200 ps (left panel) and 1.4 ns (right panel) following a 60-mJ, 100-ps ablation pulse at 1.064 mm. The distance scale is approximately 50 mm × 50 mm. Lighter shades correspond to greater deformation. Shortly after the pulse, the shape of the deformation is roughly Gaussian, with a maximum deformation of about 0.8 mm. By about 300 ps, the center of the deformation has begun to decrease in amplitude, dropping below the outer regions by about 800 ps and forming a ring-like structure that can be clearly seen in the right panel.



Time dependence of the maximum surface height of deformation and the transient volume of material displaced for calcite after absorption of a 100-ps laser pulse.

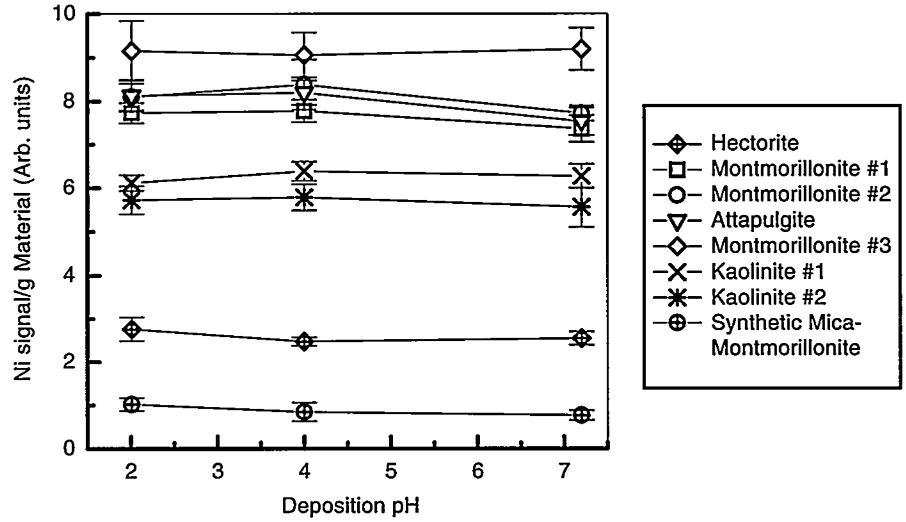
To this end, we have constructed an apparatus for examining the time dependence of the flow of energy after a laser ablation pulse as manifested in surface deformation (data for calcite are shown in the first figure). As the second figure shows, we can measure changes in the height of the surface of about 50 nm in an area as small as 50 × 50 mm. Currently, we can measure the time dependence of the energy flow from the start of the ablation pulse to 16 ns after the pulse, and we have the capability to measure time delays of up to 50 ns. This apparatus will allow us to determine the extent and time scale of absorbed energy flow in materials as a function of their material parameters.

Additionally, we studied the dependence of the laser-induced breakdown spectroscopy (LIBS) signal from heavy metals on mineral type and deposition conditions. Soils consist of a variety of minerals, the most abundant of these being clay. For the clay minerals we investigated, the acidity of the deposition conditions had no observable effect on the LIBS signal for either copper or nickel (see the third figure). Also, under the same deposition conditions, clays of the same mineral class have the same analyte signal levels, and the signal levels are different from those obtained from other mineral classes.

Publications

Scherbarth, N.L., and A.C. Koskelo, "Sensitivity of Laser-Induced Breakdown Spectroscopy to Different Contaminant Binding Sites in Basic Soil Constituents" (poster presented at the SPIE International Symposium: High Power Laser Ablation 1998, Santa Fe, NM, April 26–30, 1998).

LIBS signals for nickel in clays as a function of deposition acidity. The x-axis is the pH of the nickel solution used to make the samples. The montmorillonites group together at a higher relative signal; the kaolinites group at a lower LIBS signal. The signals have been normalized to an iron line that was used as an internal standard. Correction was made for the iron concentration in each sample.



Supermassive Black Holes and the Strong Field Limit of General Relativity

97012

Wojciech Zurek

Supermassive black holes with accretion disks are thought to exist in the center of all galaxies. It is speculated that the active galaxies (active galactic nuclei, quasars, and Seyferters) each contain a rotating black hole. These 10^8-solar-mass black holes may explain the immense power output from these active galaxies.

This year, we modeled the x-ray line profile from such a supermassive black hole in the center of the Seyfert galaxy known as MCG-6-30-15 using a general relativistic ray-tracing code. This work has provided the first model-independent measurement of the existence of the black hole and further has measured angular momentum of the black hole as at least 23% its maximum allowable rate. The photon simulations done on the Jet Propulsion Laboratory's CRAY-T3D (on 1000 × 1000 grids) agreed well with the x-ray data from the Advanced Satellite for Cosmology and Astrophysics. This work was featured on the cover of the January 1, 1998, issue of *Nature*.

Publications

Bromley, B.C., and V.I. Pariev, "Line Emission from an Accretion Disk around a Black Hole: Effects of Disk Structure" (submitted to *Astrophys. J.*).

Bromley, B.C., et al., "The Inner Edge of the Accretion Disk around a Supermassive Black Hole," *Nature* **391**, 54 (1998).

Bromley, B.C., et al., "Line Emission from an Accretion Disk around a Rotating Black Hole: Toward a Measurement of Frame Dragging," *Astrophys. J.* **475**, 57 (1997).

Striation-Image Monitoring of Plasmaspheric L-Resolved Electrodynamics (SIMPLE)

96148

Abram 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 irregularites 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.

This year our project entered its post-collection, data-analysis mode, whereby several years of continuous multisatellite collections were systematically scrutinized with a view toward extracting information on the geomagnetic-disturbance electric fields that both generate the irregularities and cause them to move. During this post-analysis we demonstrated that the simultaneous observation of radio beacons from two longitudinally separated satellites could aid in the inference of the distance to phase-refracting plasma irregularities along the lines-of-sight. It was found, however, that in most cases the two lines-of-sight saw refractive signatures that were significantly decorrelated from each other, such as to compromise the time-delay correlation's ability to infer distance.

A Large-Aperture, Wide-Angle Air Cerenkov Telescope

98023

Constantin Sinnis

Our goal is to determine the composition of the cosmic-ray background at energies between 50 TeV and 5 PeV. Although cosmic rays were discovered at the turn of the century, we still do not know their origin. The nuclear composition of the cosmic rays and its variation with energy are an important clue to their origin. Direct measurements have been made up to about 100 TeV, and there is a marked feature in the cosmic-ray energy spectrum near 1 PeV. Previous attempts to measure the composition from the ground have been plagued by systematic errors and uncertainties in Monte Carlo simulations.

Ours will be the first ground-based experiment capable of measuring the composition in an energy range where it has been directly determined. This capability will give us an absolute calibration. The technique that we developed depends on measuring the lateral distribution of the Cerenkov light generated by an extensive air shower (EAS). The heavier the cosmic-ray nuclei that initiated the shower, the flatter the lateral distribution. This information, in conjunction with information from the Milagro EAS array (muon lateral distribution, core position, and electromagnetic shower size), will provide multiple constraints on the primary composition.

We have completed the design of the instruments, obtained the six mirrors and telescope mounts, and obtained the approximately 200 photomultiplier tubes that will be used to image the Cerenkov light.

Advanced Computational Analysis of Disordered Materials and Clay Minerals

98021

David Bish

We are developing and testing a new method for diffraction analysis of the structures of disordered materials, either in single-phase form or in mixtures with ordered materials. The new method will apply wherever disordered materials are important—such as for zeolite and clay catalysts, superconductors, and nanocomposites—and represents a major advance in our abilities to analyze the structures of polycrystalline disordered materials.

Results this year include implementing and testing a realistic, single-parameter, crystallite-size distribution function derivable from first principles. We have converted equations for calculated relative intensities for each phase to absolute intensities to allow quantitative analysis of mixtures.

Our code currently calculates derivatives for the numbers of unit cells that define the crystallite dimensions along the X and Y directions. The coherence length along the Z direction is more complicated for some materials because crystals may be optically coherent along the Z direction, but not in three dimensions. Such a crystal produces two separate but additive diffraction patterns, each the result of different crystallite or domain distributions. We can now calculate derivatives for each of these patterns, allowing separate refinement in a Rietveld procedure. Because accurate values for the derivatives require very accurate intensity values, we optimized and extended intensity summations and increased the sizes of integration "windows" about calculation ellipsoids to include greater portions of the tails. This extension led to intensity discrepancies caused by overlap of the tails of the diffraction peaks at higher angle, but our method will explicitly incorporate and correct for this overlap.

Nuclear and Particle Physics

Testing the Standard Model Using Bottom Quarks

97038

Rajan Gupta

Experimental and theoretical particle physicists are extensively probing bound states involving heavy quarks. A wealth of new data has already been produced at current experimental facilities, and because heavy quarks will be the focus of new, more sophisticated B-meson factories being built (Stanford Linear Accelerator Center in the United States and others worldwide), we are anticipating further significant advances. The data from the experiments may contain the first clues to a possible breakdown of the standard model of particle interactions at very short distances, but we will need theoretical input to precisely interpret these data. Since the momentum transfers in the processes involving the decay of hadrons containing charm or bottom (b) quarks are small enough that perturbation theory cannot be used reliably, we are using large-scale simulations of lattice quantum chromodynamics (QCD) to determine a number of these desired input parameters.

The decay constants f_B, f_{Bs}, f_D, and f_{Ds}, along with the bag parameters B_B and B_{Bs}, are central ingredients needed to extract the Cabibbo-Kobayashi-Masakawa matrix elements from experimental measurements of semileptonic decays of B mesons. Because of strong interaction effects (described by QCD), we must use nonpertubative methods to accurately determine these quantities. Perhaps the most promising approach is in the use of simulations of lattice QCD. In collaboration with researchers at Ohio State University, we are carrying out a detailed study of the properties of hadrons containing b quarks using a nonrelativistic formulation of heavy quarks. This approach allows for very good control over discretization errors in both the heavy and light quark sectors.

This year we developed the codes necessary to carry out an analysis of the spectrum and decay constants of hadrons containing b quarks. These codes were optimized for the Silicon Graphics, Inc., Origin 2000 computers in the Los Alamos Advanced Computing Laboratory. The results of our first calculations of decay constants predict that $f_B = 147$ MeV and $f_{Bs}/f_B = 1.20$; further details are given in the publication listed below. These estimates represent the most complete analyses of systematic errors from lattice simulations. The rich spectrum of mesons and baryons containing b quarks is just beginning to be measured in experiments at the European Center for Nuclear Research, the Fermi National Accelerator Laboratory, Cornell University, and the Stanford Linear Accelerator Center. We analyzed hadrons composed of heavy and light quarks, and our results, which will be submitted for publication, elucidate the spectrum of S and P wave states and their radial excitations. We are developing codes to analyze the upsilon spectrum, from which we hope to provide an accurate determination of the strong coupling constant α_s.

Publications

Khan, A., et al., "B Meson Decay Constants from NRQCD," *Phys. Lett. B* **427**, 132 (1998).

Study of Parity Nonconservation in the Reaction n + p $\rightarrow$ d + γ

98040

J. David Bowman

Our goal is to measure the parity-violating asymmetry in the direction of emission of gamma rays relative to the neutron spin direction when polarized neutrons are captured by protons. The asymmetry is nonzero because the weak interaction between nucleons introduces parity impurities in nuclear levels. Seven weak meson-nucleon-nucleon couplings are needed to describe the weak force between nucleons. A measurement of the asymmetry will determine the most important of these weak couplings. The experiment will be done at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) at Los Alamos.

Because the expected asymmetry is very small (10^{-7}), it is necessary to demonstrate that we can obtain a statistical error of 10^{-8} and systematic errors small compared to statistical errors. The asymmetry is determined by the weak pion-nucleon-nucleon coupling. There is no nuclear structure uncertainty in extracting the weak coupling from the experimental asymmetry because the wave function of the two-nucleon system can be calculated exactly.

This year we carried out a test run to determine backgrounds in the experimental beam line. The backgrounds were about 1% of the signal, an acceptable level. We tested a mockup gamma-ray detector. The gamma-ray detector for the experiment will consist of forty-eight 15-cm cubes of CsI viewed by vacuum photodiodes. We measured the magnetic field sensitivity of mockup vacuum photodiodes; the gain sensitivity was 2×10^{-5}/G, five orders of magnitude smaller than for photomultiplier tubes. We also designed and constructed low-noise preamplifiers and demonstrated the ability to achieve the counting statistics limit.

Scientists from Los Alamos, University of California at Berkeley, University of New Hampshire, University of Michigan, University of Indiana, Kyoto University, National Institute of Standards and Technology, and KEK (the Japanese National Laboratory for High-Energy Physics) collaborated on the test run. Our team carefully analyzed the statistical and systematic errors. We then designed an apparatus that can achieve the desired (10^{-8}) statistical and systematic errors.

We developed a proposal for the experiment at the MLNSC; based on a favorable review, a neutron flight path has been assigned to the experiment. We also submitted a proposal to DOE for peer review. In addition, we have successfully designed and constructed a neutron spin flipper, a crucial component of the experiment.

A GaAs Detector for Dark Matter and Solar Neutrino Research

97037

Thomas Bowles

Our objective is to develop GaAs detectors that can be used in a broad research program. In searches for weakly interacting massive particle dark matter and high-resolution measurements of the solar neutrino spectrum, a GaAs detector can offer substantial advantages over other technologies. GaAs may provide lower backgrounds than are possible in other detector materials, as well as a low energy threshold and very good energy resolution at room temperature. Such detectors have potential applications in many areas of research, including medical imaging, x-ray detecting on satellites, and borehole mining.

This year we have grown more than twenty 1-kg ingots of GaAs, characterized their electrical properties, and produced a number of detectors from 400-µm-thick wafers. We have studied the GaAs and the detectors in detail using eight different types of measurements. As a result, our understanding of the limiting factors in producing GaAs crystals suitable for use in detectors is much better.

Based on our experience, we are developing a new technique for growing crystals under different conditions and have begun adapting some of the crystal-growing furnaces to accommodate the new technique. Our goal is to understand the factors that limit performance in detectors made with crystals grown using the new technique so that we can fabricate usable detectors with a thickness of 1 mm in FY99.

Exploring and Testing the Standard Model and Beyond

98038

Geoffrey West

The goal of this project is to extend and develop the predictions of the standard model of particle physics in several different directions, including 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. This year we have continued work on exploring various aspects of the standard model of the elementary particles and their interactions and have examined extensions of it in the area of unification of all of the forces of nature.

We had five major accomplishments in the past year. We provided a unified description of y-scaling phenomena in deep-inelastic, electron-scattering structure functions of nuclei; this understanding explained the gross features of the data for all nuclei in terms of simple physical effects. We made significant progress in extracting important parameters from simulations of QCD, including light quark masses and decay constants for heavy mesons. We began an effort to define an experimental signal for the new state of matter, the quark-gluon plasma, predicted to be a consequence of QCD. This understanding is very important for the new Relativistic Heavy-Ion Collider machine, which is scheduled to come online next year. Our calculations for nonperturbative phenomena derived from instanton effects showed how a confinement mechanism works in the theory. This effort was driven by the attempt to understand consequences of the duality that occurs in string theories for the inclusion of gravity. We developed a general way of viewing scaling phenomena that can incorporate classical dimensional analysis, fractals, self-similarity, and the renormalization group.

Publications

Afanasev, A., and P.R. Page, "Photo- and Electro-Production of $J^{(PC)}=1^{(-+)}$ Exotics," *Phys. Rev. D* **57**, 6771 (1998).

Burakovsky, L., and P.R. Page, "Scalar Glueball Mixing and Decay" (to be published in *Phys. Rev. D*).

Lee, W., and D. Weingarten, "Scalar Quarkonium Masses and Mixing with the Lightest Scalar Glueball," *Nucl. Phys. B* **63A-C**, 964 (1998).

Mihaila, B., et al., "The Quantum Role in d-Dimensions and the Large-d Expansion" (submitted to *Phys. Rev. D*).

Molina-Paris, C., et al., "Zeta Functions, Renormalization Group Equations, and the Effective Action" (to be published in *Phys. Rev. Lett.*).

Solar-Neutrino Physics

96349

Andrew Hime

This project is part of our collaboration in the field of solar-neutrino physics with the Sudbury Neutrino Observatory (SNO) in Canada. SNO, which is presently in the commissioning phase, will be capable of measuring both charged-current (CC) and neutral-current (NC) interactions of boron-8 solar neutrinos on deuterium with its 1000-tonne target of heavy water. At Los Alamos, efforts have focused on the research and development, design, construction, and commissioning of a discrete array of ultra-low-background helium-3 detectors to be deployed into the main SNO detector. With this array, the CC and NC signals can be measured simultaneously and independently, offering a model-independent means to resolve the long-standing discrepancies between predicted and measured solar-neutrino flux.

This year full-scale construction began on the neutral-current detector (NCD). We created a sophisticated procedure for filling and verifying counters and implemented it to establish the integrity of individual counter segments before we shipped them to Sudbury. Over the past several months, we have delivered about 20% of the NCD array to Sudbury. Upon arrival, the detectors undergo a set of verification tests before entering a cool-down phase in which we monitor their intrinsic radioactivity with analytical tools based upon pulse-shape discrimination techniques developed at Los Alamos. In addition, we have developed novel techniques with unprecedented sensitivity for monitoring radon leaching from the NCD array before its installation into the heavy-water vessel.

Extraction of the neutron-capture signal from the NCD array relies on techniques developed at Los Alamos to separate ^{3}He(n,p)T tracks from alpha background. This year, our team began to outline the calibration measurements necessary for a robust extraction of the NC signal—namely, calibration of the neutron-capture efficiency with a tagged californium-252 source and calibration of the low-energy Cerenkov background wall with beta/gamma sources. At Los Alamos, we also fabricated the required calibration sources for SNO. In addition, we have begun a program to optimize the use of such sources in SNO as well as the analysis tools required for extracting the neutron-capture efficiency and photodisintegration background.

Neutrino Physics at Fermilab

96339

Geoffrey Mills

The Liquid Scintillator Neutrino Detector (LSND) at Los Alamos has observed evidence for the oscillations of neutrinos. The measurement of neutrino-oscillation parameters is one of the paramount problems in nuclear particle physics today. The goal of this project was to explore what would be required to develop a technology that would enable neutrino physics to be carried out at higher energies. To achieve that goal, we have extended the capabilities of detectors like LSND to do neutrino oscillation physics at higher energies, for example, by using the intense 8-GeV proton beams available at Fermilab.

This year we furthered our understanding of light emission characteristics of the liquid mineral-oil material used in the detectors. It was determined that pure mineral oil has sufficient late light production from scintillation processes to allow the discrimination of muons and pi-zero events from electron events in the higher-energy neutrino interaction range of interest. We now believe that we have a complete picture on what is necessary to build a functioning liquid mineral-oil neutrino detector.

We also further improved the simulation of neutrino production with high-energy proton beams. It was determined that a magnetic horn focusing device would be the most efficient system to implement in the 8-GeV energy range of interest. We produced a more detailed simulated model of a potential enclosure for a horn system in order to understand radiation levels in a high-intensity proton-targeting hall. Neutron transport and hadron interaction points are the critical issues in these studies.

In summary, we have successfully achieved all our goals: the design-in-principle of a high-energy neutrino experiment that could significantly advance our understanding of neutrino mass and greatly improve on the LSND oscillation measurement.

Publications

Louis, W.C., III, "Searches for Neutrino Oscillations III" (Conference on Weak and Electromagnetic Interaction on Nuclei, Santa Fe, NM, June 16–19, 1998).

Mills, G.B., "Results on Neutrinos from LSND" (Stanford Linear Accelerator Center Summer Institute Topical Conference, Stanford, CA, August 13–14, 1998).

White, D.H., "LSND Neutrino Oscillation Results" (Neutrino 98 Conference, Kamioka, Japan, June 4–9, 1998).

The QCD Phase Transition in Relativistic Heavy Ion Collisions

97040

Frederick Cooper

We are developing numerical methods to study the nonequilibrium phase transition of nuclear matter following ultrarelativistic heavy-ion collisions at the Relativistic Heavy-Ion Collider (RHIC) at Brookhaven National Laboratory. These new methods, driven by advances in high-performance computing, provide much deeper levels of detail about the nature of nuclear matter and energy.

We have studied the chiral phase transition that occurs in quantum chromodynamics (QCD) during the time evolution of a heavy-ion collision. The model we have studied in great detail is the O(4) sigma model, which embodies the chiral symmetries of QCD and is in the same universality class. We have studied the dynamics of this model both for a longitudinally expanding plasma as well as for a radially expanding plasma and have determined the distortion of the pion spectra resulting from the chiral phase transition occurring out of equilibrium (quench conditions). We have also developed the formalism for determining the production of dileptons that are going to be observed by several RHIC detectors, and we are presently evaluating finite time corrections inherent in our numerical simulations. We have also made preliminary calculations in one-dimensional models on the effects of the $1/N$ corrections to our mean field results.

Publications

Boyanovsky, D., et al., "Evolution of Inhomogeneous Condensates: Self-Consistent Variational Approach," *Phys. Rev. D* **58**, 25007 (1998).

Cooper, F., "Unusual Dileptons at RHIC," in "Non-Equilibuium Many Body Dynamics," *Proceedings of the RIKEN BNL Research Center Workshop, Vol. 4* (Brookhaven National Laboratory, Upton, NY, 1997).

Mihaila, B., et al., "The Quantum Roll in N-Dimensions and the Large N Expansion" (submitted to *Phys. Rev. D*).

Neutrinos and Theory of Weak Interactions

96343

Joseph Carlson

We discovered a possible solution to the anomaly observed near the endpoint of the tritium beta-decay spectrum, which has persisted for 15 years through six different experiments. This solution requires recognizing the triplicity of neutrino mass eigenstates, the possibility of new interaction currents beyond those encompassed by the standard model, and the independence of the mixing between the mass and interaction eigenstates for the various interactions. At a recent international meeting covering physics beyond the strandard model, our "pedagological example" solution matched closely the structure of expermental results reported by others in the field.

The pi+ decay of Lambda hypernuclei is a rare process, observed so far only in the mesonic decay (about a 5% branching ratio) of 4Lambda He. (In free space, the Lambda does not decay to pi+.) Others have found a theoretical branching ratio contribution of no more than 1%. We have re-examined the first- and second-order decay processes and have shown that Lambda-Sigma coupling leading to an s-wave pi+ nn three-body decay can provide a satisfactory explanation of the branching ratio and the resulting pi+ decay spectrum. The work has inspired interest in a new experiment.

Publications

Carlson, J.A., "Progress and Prospects in Quantum Monte Carlo Studies of Light Nuclei," in *Innovative Computational Methods in Nuclear Many-Body Problems*, H. Horiuchi, et al., Eds. (World Scientific, Singapore, 1998).

Gibson, B.F., and R.G.E. Timmermans, "The pi+ Emission Puzzle in 4Lambda He Decay," *Nucl. Phys.* **628**, 417 (1998).

Herczeg, P., "Exotic Muon Decays and Neutrino Oscillations," in *Proceedings of the First International Conference on Particle Physics Beyond the Standard Model*, H.V. Klapdor-Kleingrothaus and H. Pas, Eds. (Institute of Physics Publishing, Bristol, UK, 1998), p. 124.

Schiavilla, R., et al., "Weak Capture of Protons by Protons," *Phys. Rev. C* **58**, 1263 (1998).

van Kolch, U., et al., Electromagnetic Corrections to the One-Pion-Exchange Potential," *Phys. Rev. Lett.* **80**, 4386 (1998).

Helium-3 Magnetometry for a Neutron EDM Measurement

97041

Martin Cooper

We are investigating a physical measurement that may challenge the most fundamental scientific explanation of the origins and nature of matter: the Standard Model. The Standard Model predicts a value for the matter/antimatter ratio in the universe that contradicts some ideas in cosmology, making tests of the Standard Model potentially fruitful for discovering new physics. The challenge to the model would come from the precise observation of a non-zero value for the neutron's electric dipole moment (EDM), a very small spacial separation between the positive and negative charges inside the neutron. Improving measurement sensitivity requires a new experiment, the ideas for which have been worked out in the literature.

This proposed technique can enhance sensitivity by 3 orders of magnitude to around 10^{-28} e·cm (electron charges times centimeters). The experiment's technical feasibility depends on obtaining a uniform spacial distribution of helium-3 in a low-temperature bath of helium-4. Our objective is to measure the distribution of small concentrations of helium-3 in superfluid helium-4. We are measuring the distribution for concentrations from 10^{-3} to 10^{-7} and for temperatures in the range 0.3 to 0.7 K, using neutron tomography developed in gases during the first phase of this project. The neutrons are produced by the accelerator at the Los Alamos Neutron Science Center (LANSCE).

The LANSCE accelerator has not operated this fiscal year, so we have used this time for construction work. Modifications to the target cell, to match the needs of the experiment, are well under way. In the laboratory that we established, we refurbished the dilution refrigerator and provided adequate space for it by expanding the shielding house on flight path 11. Our group is currently scheduled for the first beam in December 1998. Depending on the accelerator performance, we should have several data runs to answer the questions we have posed. Studies of polarized helium-3 will follow. The data gathered will be invaluable in determining the feasibility of a design for a new search for a neutron EDM.

Chiral Symmetry in Finite Nuclei

96346

David Madland

We are calculating the properties of finite nuclei over a wide mass range using quantum chromodynamics (QCD) scales and chiral Lagrangians together with dimensional-power-counting techniques to test for naturalness in the various in-medium N-body nuclear forces that we consider. Good evidence has been found for naturalness, implying that QCD and chiral symmetry apply to finite nuclei, but the evidence at this time remains 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 relativistic mean-field Lagrangian (Nikolaus, Hoch, Madland).

This past year, we used our Dirac single-nucleon wave functions to demonstrate the origin of (approximate) pseudo-spin symmetry in nuclei; namely, the conserved pseudo-spin of an observed pseudo-spin doublet is the orbital angular momentum of the lower component of each of the corresponding Dirac wave functions. We then introduced the least-bound proton and neutron experimental pseudo-spin doublets in lead-208 as two new observables in our chi-square minimization program for testing various model Lagrangians and their corresponding coupling constants.

We hoped that these new observables would be particularly sensitive to the lower components of the corresponding wave functions in comparison to the other observables that we use. Unfortunately, this does not appear to be the case. While it is true that new minima were located, none of them resulted in enhanced predictive power. Currently, we are adding axial vector and tensor interactions, both isoscalar and isovector, and have excluded all pseudoscalar interactions on the basis of nucleon fields only and no explicit meson fields in our Lagrangians.

Publications

Friar, J.L., et al., "Chiral Symmetry and Three-Nucleon Forces" (submitted to *Phys. Rev. C*).

Ginocchio, J.N., and D.G. Madland, "Pseudo-Spin Symmetry and Relativistic Single-Nucleon Wave Functions," *Phys. Rev. C* **57**, 1167 (1998).

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Determination of the Neutron Lifetime and Ultracold-Neutron Source Development

98039

Steven Lamoreaux

The objective of this project is to develop a new experimental technique to measure the beta-decay lifetime of the neutron with over an order-of-magnitude improvement in accuracy as compared with the accuracy of current experimental techniques. In addition, we are developing a new type of ultracold-neutron (UCN) source that will give several orders-of-magnitude increase in the UCN densities available for a number of experiments.

In collaboration with Harvard University, we have constructed the experimental system for the magnetic trapping of UCNs produced by scattering cold neutrons in superfluid helium-4 and for observing the decays of the trapped neutrons by scintillation light produced in the superfluid helium-4. This apparatus has been installed on a cold-neutron beam line at the National Institute of Standards and Technology reactor, and we have been studying background signals that are due to the choice of materials used for internal neutron shielding.

We have directly measured the UCN production rate from inelastic scattering of cold neutrons in solid deuterium at temperatures near 4 K. This measurement employed a cryogenic system built at Los Alamos that cooled a 1-L sample of solid deuterium and had provisions for separating and collecting the UCNs from the incident cold-neutron beam.

In addition, we developed a crude UCN energy spectrometer to measure the kinetic energy spectrum of the produced UCNs. This apparatus was operated at the Hahn-Meitner Institut in Berlin, Germany. The experimental results agreed well with results of Debye model calculations.

Finally, we invented a new type of UCN source that is based on scattering of cold neutrons in solid deuterium but employs its own dedicated low-duty-factor neutron spallation source. The basic idea is to enclose a tungsten spallation target within a polyethylene/beryllium flux trap/moderator assembly that contains a 1-L sample of solid deuterium coupled to a UCN extraction pipe. Initial calculations show that an increase in UCN density of up to several orders of magnitude is possible as compared with the UCN density of currently operating sources.

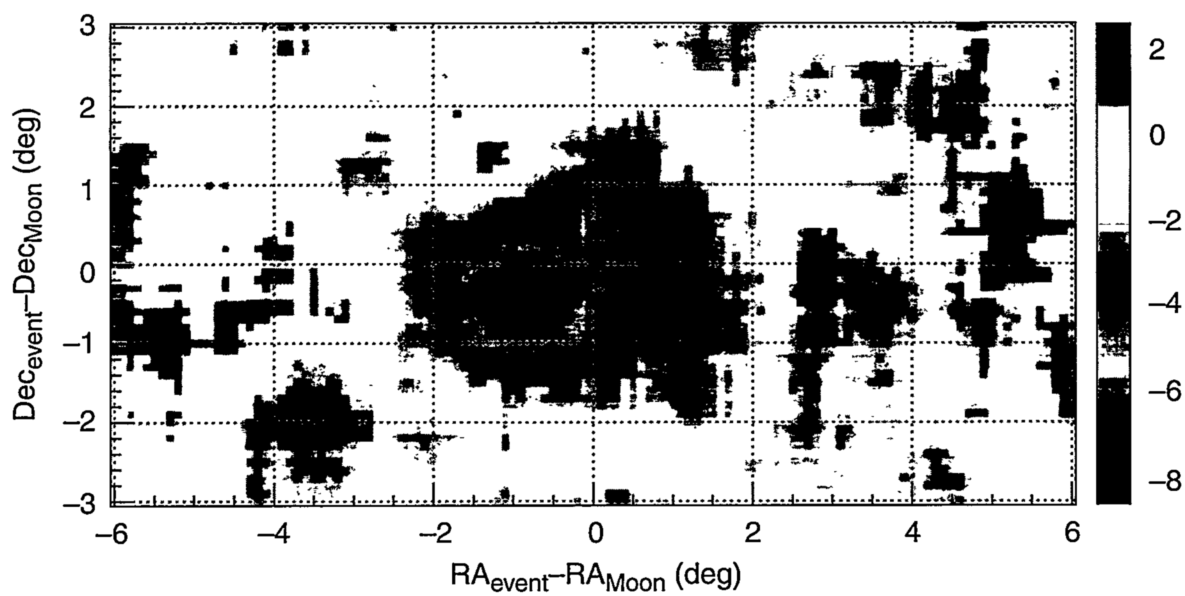
Search for Cosmic Antimatter with Milagrito

97004

Cyrus Hoffman

It is important in both cosmology and particle physics to understand whether the universe is composed entirely of matter or of equal amounts of matter and antimatter. The limits on the existence of antimatter beyond our galaxy are quite poor. We are searching for energetic cosmic antiprotons by studying their deflection in the earth's magnetic field using the shadow of the moon. The moon's shadow due to 1-TeV cosmic protons is deflected somewhat toward the west of the true position of the moon by ~30 mrad; cosmic antiprotons would produce a shadow deflected in the opposite direction. Our project will use data from the Milagrito air-shower detector.

Milagrito collected data in 1997 and 1998, recording over 9 billion events. This year we honed the algorithms that were used to reconstruct the data and improved the calibration of the detector. After Milagrito was turned off, we reconstructed all the data—a monumental task that included reading in over 24 Tbytes of data. The accompanying figure, based on the Milagrito data, shows the density of events from the vicinity of the moon. A clear deficit resulting from protons is obvious to the left (west) of the true moon position (at 0,0); no deficit is apparent on the opposite side.



Event density from the vicinity of the moon, in units of standard deviations, from Milagrito. A clear deficit caused by protons is obvious to the left of the true moon position (at 0,0).

A Search for Superradiant Emission in a Nuclear Isomer Crystal

98041

Robert Rundberg

Our goals are to synthesize an oriented crystalline film of lithium niobate containing an inverted population of excited nuclear states (i.e., niobium-93m) and look for evidence of superradiant emission states by studying the photon emission rates along Bragg angles. Specific objectives for this year were to (1) assay the number of atoms of niobium-93m produced in the irradiation of molybdenum-isotope production targets at the Los Alamos Neutron Science Center (LANSCE), (2) develop a separation procedure for efficiently extracting submilligram quantities of niobium-93m from 600-g molybdenum targets, and (3) begin developing a procedure for growing epitaxial films of lithium niobate on single-crystal sapphire substrates.

For our first objective, we found the yield of niobium-93m to be approximately 150 µg, or 36 mCi of niobium-93m per molybdenum target. This was a factor of 3 to 10 less than we previously estimated. The amounts produced in the LANSCE target are more than two orders of magnitude more than our alternate supply, which is produced in a reactor by neutron irradiation of molybdenum followed by 10 years of ingrowth. The niobium isotopes are in a solution containing several curies of arsenic-73, selenium-75, rubidium-83, zirconium-88, and yttrium-88. Therefore, we must perform the chemical procedure in a hot cell.

To meet our second objective, we developed a procedure that removes 99% of niobium from macroscopic quantities of molybdenum. This procedure uses a ferric hydroxide precipitation with careful control of pH (at about 6). The ferric hydroxide coprecipitation was efficient enough for the purification of the molybdenum for later use as a generator of niobium-93m from the decay of molybdenum-93 (estimated to yield 100 µCi of 90% isomer per year). We also demonstrated that niobium can be readily extracted with a high degree of specificity into isobutyl ketone from hydrofluoric/hydrochloric acid mixtures. This process allows the recovery of niobium-93m from the ferric hydroxide precipitate.

For our third objective, we constructed a Schlenk line and synthesized lithium niobate double-salt sols. We attempted the epitaxial growth of lithium niobate without spin coating to conserve niobium-93m and minimize the potential for radioactive contamination. These attempts were not successful. Without spin coating, the films were unoriented but crystalline. The spin coater we purchased to solve the problem was delivered at the end of the fiscal year. We will continue developing a procedure for growing radioactive epitaxial films next fiscal year.

Publications

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High-Energy Cosmic Transients

98042

Todd Haines

The goal of this project is to acquire and analyze data from the Milagro gamma-ray observatory in the search for transient (time-varying) sources of high-energy gamma rays (tera-electron volts or TeV). Milagro is the world's only air-shower array capable of making observations of cosmic gamma rays in the few hundred giga-electron volts to several tens of TeV energy range. Particular sources of interest include gamma-ray bursts, cosmic strings, active galaxies, and primordial black holes.

This year we began searching the data taken with a gamma-ray observatory that we developed, called Milagrito, which operated from February 1997 through May 1998. Over 10 billion air showers were recorded during this time, yielding over 10 terabytes of data. These data have now been completely reconstructed, a necessary task that preceeds the search for high-energy transients. This search has now begun.

We also completed the full Milagro gamma-ray observatory. Milagro offers substantially higher sensitivity to transients because of its larger area and background rejection capabilities.

Weak Interaction Measurements with Optically Trapped Radioactive Atoms

96351

David Vieira

Our primary goal in this work is to undertake high-precision tests of electroweak interaction in atomic nuclei. We are currently concentrating on measuring the beta-decay asymmetry of trapped and highly polarized rubidium-82 atoms. Such a measurement will search for the existence of right-handed and tensor interactions, thereby testing the maximal parity-violating nature of the electroweak interaction in a pure Gamow-Teller transition to a new level of precision. The key technology needed to undertake new measurements of this type is the efficient optical and magnetic trapping of selected radioactive species.

During the past year we have made excellent progress in trapping and manipulating radioactive rubidium-82 with a double magneto-optical trap (MOT) system coupled to a mass separator (see accompanying figure). In particular, we have been successful in (1) demonstrating the first MOT trapping of rubidium-82 in quantities of 6 million atoms, 100 times more atoms than in any previous radioactive atom-trapping work, (2) measuring the rubidium-82 hyperfine structure of the D_1 transition for the first time and remeasuring the D_2 transition with higher precision, (3) transferring ~50% of the radioactive atoms from one MOT to a second MOT by using a "push" laser beam and magnetic guide approach, (4) constructing a time-orbiting-potential (TOP) magnetic trap, and (5) constructing and testing the positron-detection system. Very recently we have trapped stable rubidium-82 in the TOP trap, and we are looking forward to beginning our initial beta-asymmetry measurements with rubidium-82.

Publications

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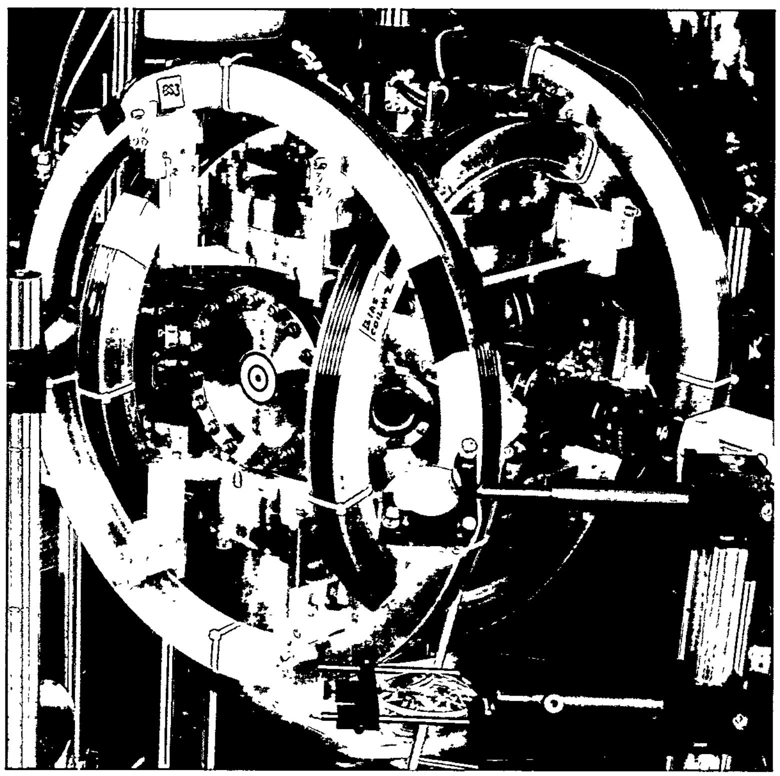

Photograph of the magneto-optical trap (MOT) plus the time-orbiting-potential (TOP) trap setup for measuring the rubidium-82 parity-violating, beta-decay asymmetry. The second MOT and TOP trap chamber are surrounded by large-bias coils. A beta detector will be installed on the flange marked with a bullseye.

Instantons and Duality in Strongly Coupled Quantum Theories

97039

Michael Mattis

We are studying the properties of strongly coupled gauge theory in a completely novel way—by a direct, dynamic nonperturbative calculation of the instanton series. Instantons are the Euclidean-space "particles" associated with a tunneling between distinct ground states in the theory. In a significant preliminary result, we have recently calculated all instanton and 2-instanton effects in the Seiberg-Witten model, which has independently confirmed the relevant parts of their solution.

The principal goals of this project are (1) extension of this work 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 because instantons generically exist even in theories (e.g., QCD itself) that admit neither duality nor supersymmetry. Using these techniques, we hope ultimately to develop a compelling physical picture of the vacuum in strongly coupled gauge theories. Especially, we hope to be able to shed light on the long-unsolved problem of confinement in QCD.

This year we have presented a compendium of results for Atiyah-Drinfeld-Hitchin-Manin (ADHM) multi-instantons in SU(N) supersymmetric gauge theories, including applications to N=2 supersymmetric models. Extending our earlier SU(2) work and treating the N=1 and N=2 cases in parallel, we constructed (1) the ADHM supermultiplet, (2) the multi-instanton action, and (3) the collective coordinate integration measure.

Specializing to $N = 2$, we then gave a closed formula for F_k, the k-instanton contribution to the prepotential, as a finite-dimensional collective coordinate integral. This result amounts to a weak-coupling solution in quadratures of the low-energy dynamics of N=2 supersymmetric QCD without appeal to duality.

As an application, we calculated F_1 for all SU(N) and any number of flavors N_F; for $N_F < 2N - 2$ and $N_F = 2N - 1$, we confirmed previous instanton calculations, and our results agreed with the proposed hyperelliptic curve solutions. For $N_F = 2N - 2$ and $N_F = 2N$ with $N > 3$, we obtained new results, which in the latter case we do not understand how to reconcile with the curves.

Publications

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New Paradigms in Simulating the Prediction, Intervention, and Control of Infectious Diseases

98009

James Hyman

Mathematical models based on the underlying transmission mechanisms of a disease can help the medical and scientific communities (1) understand and anticipate the spread of an epidemic and resistant strains in different populations and (2) evaluate the potential effectiveness of different approaches for bringing an epidemic under control. Even more important than the successes with these specific diseases has been the development of frameworks and concepts for understanding epidemiology.

The primary goal of our modeling effort is to understand the spread of infectious diseases, including influenza, hepatitis, HIV, and other sexually transmitted diseases, and to be able to estimate and subsequently predict the impact of control measures on their spread. Modeling can reduce the uncertainty of the estimates of disease prevalence and aid in the development of scientific understanding of the mechanisms of the disease and of the epidemic. Modeling can also estimate the benefits and the costs of projected interventions and project the requirements that an epidemic will place on the health care system. Thus, the modeling techniques can join with biological, epidemiological, behavioral, and social science studies to produce better projections and better understanding of the epidemic.

We have initially focused on diseases whose spread is strongly influenced by the fact that people have contacts based on age, ethnic, social groups, or geographic location. This is true for most diseases. Childhood infections are spread primarily at school and to siblings at home. The spread of influenza is determined by work, travel, and other patterns of life. Sexually transmitted diseases are spread by contacts that are biased by behavioral factors, age, and other considerations. Working with epidemiologists, we began developing and comparing forecasts based on deterministic models, stochastic differential equation models, and Monte Carlo Markov-Chain models. The Monte Carlo Markov-Chain model is capable of incorporating more detail in the diseases and social structures than the other approaches, so that new data or concepts can be quickly tested in the appropriate model.

Publications

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Hyman, J.M., et al., "The Differentiated Infectivity and Staged Progression Models for the Transmission of HIV" (to be published in *Math. Biosciences*).

Multiplex Polymorphism Analysis by Flow Cytometry for High Throughput Screening

98006

John Nolan

Single-nucleotide polymorphisms (SNPs) are sites in the genome where a different nucleotide may occur on the two chromosomes of an individual or within a population of individuals. Our goal is to use flow cytometry (FC) to develop new approaches for analyzing SNPs.

To analyze the frequency of each base at polymorphic sites, we have developed a novel microsphere-based method employing fluorescently labeled oligonucleotides. Primers designed to bind adjacent to an SNP are immobilized on microspheres and bind to target DNA from solution. We will design fluorescent reporter-oligonucleotides that anneal upstream from, and vary at, the site of interest. We will then add ligase to covalently attach reporter-oligonucleotides that are correctly matched to the immobilized primer. After we heat the sample to dissociate template and unannealed reporter, we will use flow cytometry to measure the microsphere fluorescence. We will multiplex this oligo ligation assay (OLA) by using different-sized or different-colored beads to carry different primers. Thus, we will simultaneously measure the SNP frequency at multiple sites in a single individual or pool of individuals.

We will test these methods on sequences with known polymorphic features and on samples from patients with beryllium sensitivity, a condition associated with a specific polymorphism. These methods will have significant advantages in speed and sensitivity over existing methods for detecting and analyzing SNPs, providing powerful genomics tools for exploring genetic diversity and disease.

In the first year of the project, we established a one-color, single-site, FC-based OLA and demonstrated its accuracy in analyzing SNPs. We also developed a complementary approach to SNP scoring based on minisequencing. We have used these new methods to develop a rapid and inexpensive screening assay for genetic susceptibility to chronic beryllium disease.

Publications

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Cai, H., et al., "Flow Cytometry Based Mini-Sequencing: A New Platform for High Throughput Single Nucleotide Polymorphism Analysis" (submitted to *Nat. Biotechnol.*).

Targeted in Vitro Evolution of Protein Ligands

98007

Bruce Lehnert

We are developing a procedure capable of selectively evolving proteins to bind to molecular targets in vitro. To accomplish this goal, we have designed a degenerative cDNA library that directs the expression of RNAs that contain a domain that encodes random protein sequences linked to a second domain that encodes the bacteriophage MS2 coat protein. The mRNAs transcribed from the cDNA library are translated in vitro to generate random peptides. These peptides are tethered to their parental mRNA molecules via the interaction of the MS2 coat protein chimera with the MS2 viral operator located at the 5' end of the RNA molecule. Hybrid molecules that bind to a target molecule will be isolated, the mRNA will be reverse transcribed, and polymerase chain reaction (PCR) will be amplified to enrich for the cDNAs that encode protein domains that were bound by the target molecule. Multiple rounds of this selection strategy will be employed to evolve peptide domains that bind to the target molecule with high affinity.

Major accomplishments during the past year include generation of the MS2 coat protein dimer expression cassette. We have cloned a mutant MS2 coat protein dimer into the c-myc epitope expression cassette. Another achievement was confirmation that the MS2 fusion is recognized by the anti c-myc monoclonal antibody and is capable of being PCR amplified. We also began making constructs to alter the spacing between the MS2 domains and the c-myc peptide domain to optimize ribosome clearance during in vitro translation. In addition, we produced bacterial lysates that are capable of translating T7 RNA polymerase-transcribed mRNAs in vitro.

Numerical Simulation of Biochemical Self-Organization: Calcium Wave Propagation and Microtubule Dynamics
96025

John Pearson

Calcium is stored in the endoplasmic and sarcoplasmic reticula. It is released at discrete sites where protein receptors and channels reside. Most modeling of the release processes to date assumed that the release sites are smeared out in space. In fact, this assumption is not always valid.

This year we constructed and analyzed numerous models to elucidate the effects of the discrete distribution of the release sites. We showed that to determine whether the effects of discreteness are important, it is necessary to estimate the following quantity: DT/d^2, where D is the diffusion coefficient of calcium, T is the average time required for a site to release its calcium, and d is the mean distance between sites. In other words, the ratio of the release time (T) to the intersite diffusion time (d^2/D) is the relevant quantity to examine. This analysis is useful in that these quantities are generally available to experimentalists by means of high-resolution confocal microscopy.

In addition, we developed the beginning of a theory that classifies the excitable reaction-diffusion systems. This is a problem relevant to excitable media that produce calcium waves and nerve impulses and to numerous other chemical systems. The main result is that if a system is excitable, then generically one should expect a Takens-Bogdanov bifurcation to exist in the parameter space of the system. The dynamics depend on the diffusion coefficients and on how close the system is to the Takens-Bogdanov point.

We believe that these results are important and that they will lead to further generalizations, perhaps making it possible for us to understand the dynamics of reacting and diffusing chemicals without first resorting to numerical simulations.

Publications

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Engineering the Specificity of xylR, a Bacterial Protein that Detects Chemicals in the Environment
96017

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 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. This project has three parts: mutagenesis of a gene encoding a toluene-binding domain from such a regulator protein, determination of the structure of this domain, and selection of variants of the protein specific for detecting trichloroethylene.

This year we isolated a series of variants of a bacterial detector protein that can partially differentiate among a group of related toluene-based compounds. These variants as a group could be useful for identifying trace amounts of these compounds in soil or water. During the process of developing detector proteins, we also developed a novel approach for using bacteria to produce domains of proteins in a soluble form. This technology allows us to modify protein domains so that they can be readily produced in bacteria, often without disturbing their functions.

Predictive Models for Transcriptional Enhancers

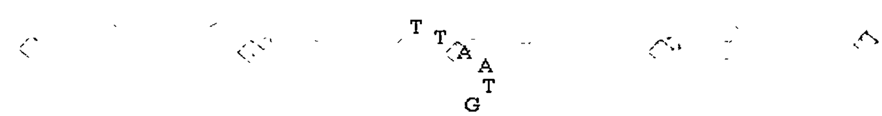
97022

Goutam Gupta

Being able to identify control elements such as transcriptional enhancers is an important step toward the understanding of gene expression. This process requires the knowledge of how these elements interact, specifically with the corresponding proteins. The homeodomain is a small, conserved protein motif that binds specifically to DNA and plays a central role in gene regulation. Different homeodomain proteins (for example, engrailed, antennapedia, and ultrabithorax) control the development of different body parts. The homeodomain/DNA complex provides an ideal system for studying the interactions between gene control elements and their binding proteins. To accomplish this goal of being able to identify transcriptional enhancers, we have developed an algorithm for predicting structures of both protein and DNA using a homology modeling approach.

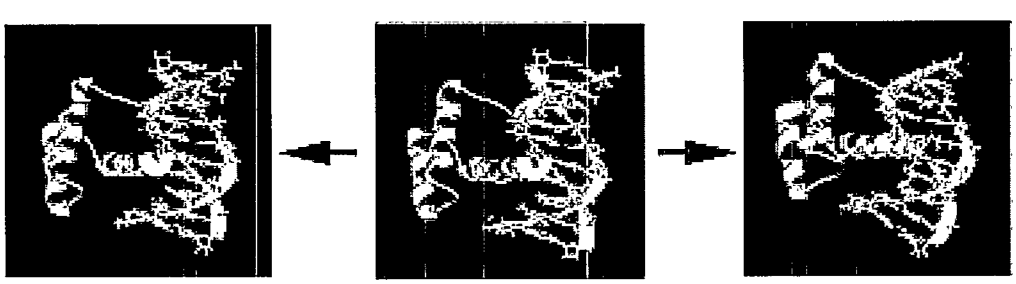
Based on the crystal structure of the engrailed homeodomain protein-DNA complex, we model the binding of the protein to different locations of the DNA. The molecule in the center of the first figure shows that the protein binds to its recognition sequence, TTAATG. The DNA duplex is moved up to two base pairs in both directions with respect to the protein while maintaining the same binding configuration. In each of the binding modes, the structure is energy minimized using AMBER, a widely used energy-minimization software package. The resultant structures are shown in the first figure. While the total number of hydrogen bonds between the DNA backbone and the protein remains unchanged, the number of hydrogen bonds between DNA bases and protein is reduced by two to three when the protein is moved from its optimum DNA binding position. This allows the sequence discrimination of the DNA binding for the protein.

Using the crystal structure of the engrailed homeodomain protein-DNA complex as the template (central molecule in the second figure), we model the structures of both the antennapedia (left molecule in second figure) and the Mat-alpha-2 (right molecule in second figure) homeodomains. The sequence identity between the engrailed and the antennapedia homeodomains is 50%, while that between the engrailed and the Mat-alpha-2 homeodomains is 30%. The root-mean-square errors for the antennapedia homeodomain are 2.8 Å and 1.7 Å for the protein and DNA, respectively, while the corresponding values are 3.0 Å and 2.0 Å for the Mat-alpha-2 homeodomain. The results of our model compare favorably with those determined experimentally.



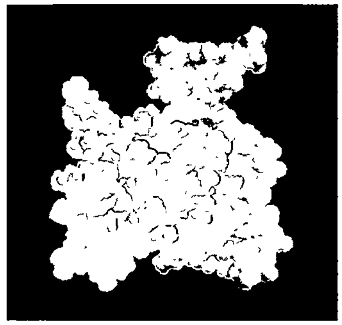
5(1) 5(1) 7(3) 5(1) 5(1)

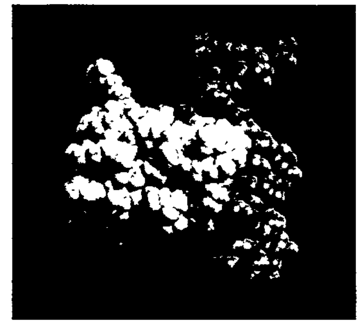
Engrailed homeoedomain and DNA bind in different modes. The total number of hydrogen bonds between the protein and the DNA, as well as those between the protein and the DNA bases (in parenthesis), is listed below each of the complexes.



The engrailed homeodomain protein-DNA complex (the template), as well as the modeled antennapedia and Mat-alpha-2 protein-DNA complexes, is shown as the center, the left, and the right molecules, respectively.

Finally, we predict structures of deformed and ultrabithorax homeodomain protein-DNA complexes. The two modeled structures are shown in the third figure. These two homeodomains are highly homologous in sequence (73% identity) and yet bind to different DNA sequences. The modeled structures will be used to study the sequence specificities of the two homeodomains.





The predicted structure of the ultrabithorax homeodomain-DNA complex is shown on the left, and the predicted structure of the deformed homeodomain-DNA complex is shown on the right. While the two homeodomains share a high sequence homology, they recognize and bind to different DNA sequences.

Publications

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Next Generation of Molecular Dynamics: Implicit-Solvent/Langevin Models for Folding of Peptides and Proteins

98008

Gerhard Hummer

Molecular dynamics (MD) simulations are widely used in structural biology, with applications to experimental protein-structure refinement and theoretical studies of protein dynamics and function. MD studies of proteins are computationally demanding because of the explicit treatment of solvent water and the short time steps imposed by fast bond vibrations. To overcome these limitations, we are developing effective, solvent-averaged potentials (thereby eliminating explicit treatment of water) and a stochastic Langevin dynamics with constrained bonds and bond angles. With these improvements, we expect up to three orders of magnitude gain over conventional MD simulations, allowing us to study microsecond dynamics of peptides and proteins.

This year we made major advances in two areas: (1) folding/unfolding kinetics and thermodynamics of alanine and glycine-based peptides in explicit solvent and (2) developing effective, solvent-averaged potentials from simulations of small molecules in water. In the first area, we could, for the first time, exhaustively sample the conformational space of several penta- and hexa-peptides in explicit-water simulations at temperatures from 275 to 400 K covering about 10 nanoseconds each. We observed stretched-exponential folding kinetics and "folding funnels" (enthalpy-entropy compensation) with sequence-dependent roughness. In addition to forming the basis for the ongoing potential development, these results have an impact on our understanding of early events in protein folding and the dynamics seen in single-molecule experiments.

In the second area, we have tested and implemented code to produce effective pair potentials between solute atoms. Production simulations are currently being performed to investigate neutral and pairwise neutral solute pairs, effective screening, and collective diffusion of solutes in water.

Publications

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Structure and Function of Nucleosomes Positioned on Repeated DNA Sequences Associated with Human Disease

96440

E. Morton Bradbury

In this project we are (1) investigating the unusual DNA structures formed by polynucleotides that contain triplet microsatellite and insulin minisatellite sequences, (2) developing an in vitro replication assay to demonstrate the presence of these unusual DNA structures during replication, and (3) reconstituting nucleosomes with DNA containing CTG/CCG triplet repeats as positioning sequences (C is cytosine, T is thymine, and G is guanine). Completion of these tasks will help us understand the structural basis of the length polymorphism and genomic instability exhibited by various micro- and minisatellites in the human genome. Such understanding is important for developing DNA fingerprinting and in studying degenerative diseases such as Friedrich's ataxia.

We have determined with high resolution the structure of the hairpin formed by the fragile X repeat, $(GCC)_n$. Using ^{15}N-edited nuclear magnetic resonance (NMR) spectroscopy, we have shown that the Cs and CpG sites of the hairpin are C-C paired. This feature of the $(GCC)_n$ hairpin makes it an excellent substrate for the human methyltransferase, the enzyme that methylates the Cs at the CpG sites. The hairpin formation by the fragile X repeat explains its expansion and the hypermethylation associated with fragile X syndrome.

We have also utilized ^{15}N-edited NMR experiments to analyze the folding of the TTC strand of the triplex formed by the Friedrich's ataxia repeat, GAA/TTC (A is adenine). We have shown that formation of the GAA/TTC triplex inside the first intron (coding segment) of the fratixin gene may cause repeat expansion and abnormal gene expression.

Publications

Mariappan, S.V.S., et al., "Fragile X DNA Triplet Repeats, $(GCC)_n$, Form Hairpins with Single Hydrogen-Bonded Cytosine-Cytosine Mispairs at the CpG Sites," *J. Mol. Biol.* **283** (1), 111 (1998).

Mariappan, S.V.S., et al., "The High Resolution Structure of a Unimolecularly Folded Triplex Formed by the GASS/TTC Triplet Repeat Associated with Friedrich's Ataxia" (submitted to *J. Mol. Biol.*).

Extending the Capabilities of Optical Biosensors

98049

Byron Goldstein

Optical biosensors offer ways to determine equilibrium and rate constants that are rapid, make measurements in real time, and require no labeling of the interacting proteins. One of these instruments, BIACORE, has gained wide use among experimental biologists. In a BIACORE flow cell, one of the reactants is coupled to a sensor chip (the receptor) while the other reactant (the analyte) flows past the chip. Detection of binding is based on the optical phenomenon of surface plasmon resonance (SPR). Biologists use the SPR response to detect index-of-refraction changes caused by mass changes at the surface of the chip. These changes are brought about by the binding of the analyte to the receptor. Continuous monitoring of the SPR signal allows the kinetics of binding to be followed in real time. However, there are concerns that the present data analysis methods yield poor estimates of the rate constants that characterize the interaction when the reaction is rapid.

To address these concerns, we have developed a computer model of a BIACORE flow cell that we can use to simulate experiments and test any data analysis method. We have used this model to simulate BIACORE experiments in which the binding kinetics are influenced by transport (diffusion and flow). These simulations have, in turn, been used to test a newly proposed method, based on a two-compartment model, for analyzing BIACORE data. We have demonstrated that the method does determine accurate rate constants, even when the binding kinetics are influenced by transport.

We have also developed ways to use soluble receptors to partially block rebinding in the dissociation phase of a BIACORE experiment and determine dissociation rate constants. We expect that this approach will allow us to determine the dissociation rate constant even when we cannot achieve high enough concentrations of soluble receptor to completely block rebinding. We are presently testing this approach in a joint theoretical-experimental study.

The Role of Low-Frequency Collective Modes in Biochemical Function: Ligand Binding and Cooperativity in Calcium-Binding Proteins

97020

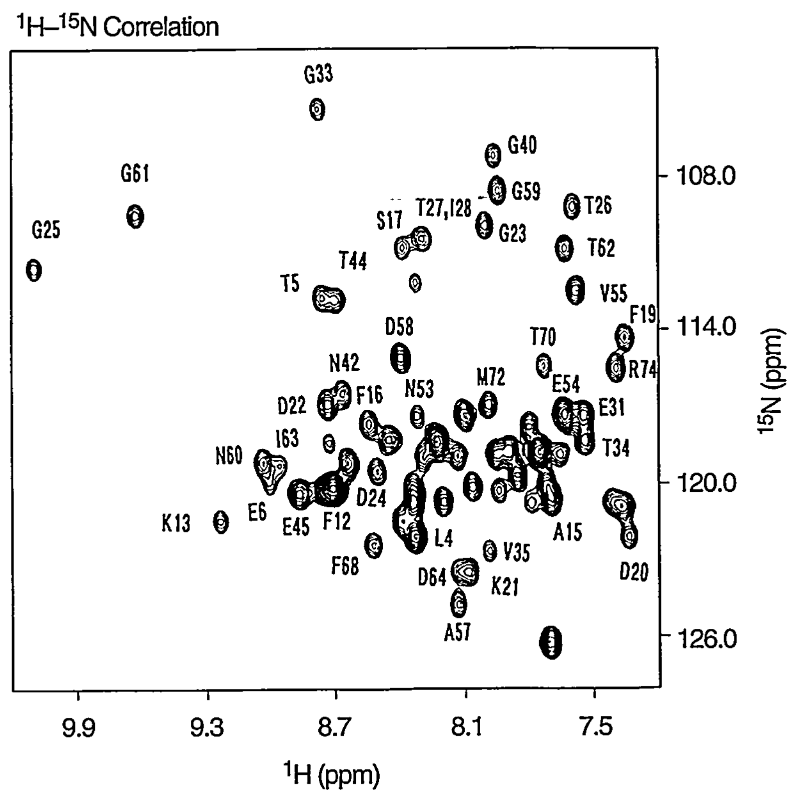
Jill Trewhella

We are studying the low-frequency collective modes in proteins that have been widely discussed as potential means for direct control of biochemical processes. We are using a combination of isotope labeling with nuclear magnetic resonance (NMR) relaxation and vibrational spectroscopy to probe the dynamic fluctuations within individual domains of the calcium-binding protein calmodulin (CaM). CaM represents a large class of calcium-binding proteins that share the same basic structural motif for calcium binding; the details of the protein scaffold and specific side-chain interactions are used to tune these binding properties for specific functions in the cell.

Our experiments are aimed at obtaining quantitative information on the dynamic fluctuations widely speculated to be important in target enzyme-binding and calcium-binding in CaM, and on how dynamic fluctuations influence calcium-binding. We are using optical spectroscopy to characterize low-frequency collective modes in CaM and to study the time evolution of these dynamic fluctuations. In parallel studies, we are using NMR relaxation measurements to obtain information on the motions of individual nuclei that are related to collective modes in the protein. By using the novel isotope labeling schemes and combining the NMR and vibrational measurements applied to the same system, we will be able to obtain quantitative dynamic information at a level of detail not previously achieved.

This past year, we have continued to make good progress on the NMR component of the project. We have completed the resonance assignments for the N-terminal fragment of CaM using spectra from a ^{15}N and 75% ^{2}H labeled sample. We have completed NMR relaxation measurements on one of the two alternate ^{13}C labeled samples (both apo and calcium loaded forms), and have begun acquiring relaxation data on samples with the second labeling pattern. The ^{13}C labeling patterns were obtained by using chemically synthesized, specifically labeled glycerol and an *E. coli* strain we constructed bearing the metabolic mutations required to generate the desired labeling pattern.

In the vibrational spectroscopy component, we have recorded time-resolved FTIR spectra using a caged calcium that can be released by a laser pulse. These data have yielded information on the kinetics of calcium binding in the N-terminal fragment of CaM as well as in the intact protein (see figure). We have initiated instrument modifications to begin the Raman experiments. Finally, we have completed molecular dynamics simulations of the apo N-terminal CaM fragment that will be tested against the experimental data.



Two-dimensional ^{1}H-^{15}N correlation spectrum of the N-terminal fragment of calmodulin with uniform ^{13}C and ^{15}N labeling and with no calcium bound. This spectrum provides the basis for assignments of the NMR signals to specific chemical groups in the protein, indicated by the one-letter amino acid code with sequence numbers. In addition, the sequential connectivities along the polypeptide backbone are identified using this spectrum. This high-resolution spectrum was acquired on the DRX 500 MHz Bruker spectrometer at Los Alamos National Laboratory.

The Molecular Basis of Universal Scaling Laws in Biology

98056

William Woodruff

Virtually every aspect of an organism's biology is affected by its size. This is evident in the empirical allometric scaling laws that compare a biological quantity with its mass (M). One of the best-known of these scaling laws, one that governs metabolic rate or power $P = P_0 M^{3/4}$, applies to all organisms, from bacteria to vertebrates, and is related to other laws that involve the scaling of drug doses, the effects of exposure to toxic substances or radiation, and longevity ($L = L_0 M^{1/4}$). The principles underlying these laws are fundamental to biology and need to be understood. We have made the following recent advances that put us in a unique position to make fundamental contributions toward understanding these principles.

We have developed a theoretical model that shows that the $M^{3/4}$ dependence of the metabolic rate for most of the members of the animal kingdom (metazoa) can be understood from fundamental physical principles that govern the transport of an organism's essential resources by biological distribution networks.

We have shown that the allometric relationship for the metabolic rate spans more than 26 orders of magnitude for mass and that it not only applies to the largest and smallest animals but also to single cells, mitochondria, and the enzyme molecules of the respiratory complex. This provides a molecular basis for understanding allometry. Furthermore, the expressions for mass-specific metabolic rate (proportional to $M^{-1/4}$) and longevity (proportional to $M^{1/4}$), when combined, show that all aerobic organisms are allocated the same amount of metabolic energy per unit mass per life span.

Theoretical and experimental approaches are helping us understand these issues by examining (1) how the theoretical model may be generalized so we can understand cellular and subcellular processes; (2) the molecular and cellular basis of the variations in mass coefficients P_0 and L_0 among taxa, and (3) the reasons for the correlation between the laws governing oxygen metabolism and longevity. Our results will advance the understanding of the molecular, cellular, and higher-order phenomena that are responsible for the allometric laws as well as provide insight into their implications for human health and quality of life. In the two months of this year that our project was in force, we searched the relevant literature in chemistry, biology, medicine, and theory to determine the most productive experimental and theoretical approaches for the forthcoming funding period.

Identification and Characterization of a Human DNA Double-Strand Break Repair Complex

96030

David Chen

Our objectives are to use atomic force microscopy (AFM) to characterize the assembly and structure of the macromolecular assemblies involved in DNA repair. The AFM is uniquely suited to biological studies and provides a means by which macromolecular interactions can be visualized under native conditions. Our studies have provided a better understanding of the structure, function, and dynamics of protein-DNA complexes formed by DNA damage response proteins. Using AFM, we have demonstrated that the DNA-dependent protein kinase can play a structural role in the repair of DNA double-strand breaks (DSBs) by physically holding DNA ends together. Presumably, this behavior aids in the correct covalent repair of the DSB by tethering the broken ends until covalent repair can be accomplished.

We have extended these studies to include other DNA damage response proteins and more complex DNA-based substrates. These efforts have resulted in important and novel findings regarding the ATM protein, the protein that is defective in patients suffering from the disease known as ataxia-telangiectasia, a condition characterized in part by increased sensitivity to ionizing radiation and a predisposition to cancer. Specifically, our work has demonstrated, for the first time, that the ATM protein binds directly to DNA. Moreover, our ability to directly visualize the protein in association with DNA using AFM has allowed us to conclude that the ATM protein binds with specificity to the DNA end. This finding is the first to implicate the ATM protein in the detection of DNA damage by direct physical interaction with DSBs.

Publications

Smith, G.C.M., et al., "DNA Binding Properties of the Ataxia-Telangiectasia Gene Product ATM" (submitted to *Mol. Cell*).

Development of a Human Artificial Chromosome

97019

Norman Doggett

Chromosomes contain specialized structures—including telomeres, centromeres, and origins of replication—that are responsible for maintaining chromosome integrity and functionality. The aim of this project is to develop a human (mammalian) artificial chromosome (HAC) that contains the minimal number of essential chromosomal components that are necessary for the stable maintenance of the HAC in the nucleus of a mammalian cell.

A HAC would permit us to clone fragments of DNA that would be large enough to contain whole genes with their controlling elements. When introduced into a desired cell type, the HAC would be stably maintained along with the chromosomes already existing in that cell and would also express the introduced gene under the appropriate genetic and physiological conditions. Thus, a HAC will be an important tool for human gene therapy by allowing the introduction of an intact gene to correct certain genetic defects, and it will be an important research tool for investigating how chromosomes function and how genes are controlled.

In collaboration with researchers from the National Institute of Environmental Health Sciences, we have used a new transformation-associated recombination (TAR) cloning approach to accomplish the directed cloning of a human centromere region of a chromosome. We have recovered several clones of the centromere region with this approach, using specific sequences (hooks) adjacent to the chromosome-16 centromere to drive the recombination. We are currently analyzing these clones for the presence of centromeric DNA, and we intend to incorporate the centromeric sequences into HAC cloning vectors developed by another collaborator at the National Institutes of Health. Although the current version of his cloning vectors contains the elements that are essential for propagation in yeast cells and in human telomeres and also contains a human origin of replication, it lacks the human centromere. We hope that with our TAR cloning of intact centromere sequences we can add functional centromeres to his vectors to bring about the generation of a HAC.

Publications

Her, C., and N.A. Doggett, "Cloning, Structural Characterization and Chromosome Localization of the Human Ortholog of the Saccharomyces Cerevisiae MSH5 Gene," *Genomics* **52**, 50 (1998).

Noninvasive Techniques for Genetic Analysis

98005

Jonathan Longmire

From its extensive work on the Human Genome Project, the Laboratory's Life Sciences Division has developed extraordinary expertise in the molecular analysis of genetic variations within a species (called polymorphisms). The existence of such variations is an important indicator of the viability of a species. Drawing on the Lab's expertise, we are developing noninvasive analytical techniques that will allow us to examine the genetic structure and demographics of the Mexican Spotted Owl (*Strix occidentalis lucida*), a bird on the government's list of threatened species. Our technique is to analyze DNA from molted feathers collected from owl nests in the Jemez Mountains.

During this first year of our project, we cloned DNA from a blood sample taken from an owl in captivity and made a cosmid library, that is, a library of all the DNA sequences within the species. To our knowledge, this is the only cosmid library for Spotted Owl DNA in existence. We then screened the library to identify clones that contain microsatellite repeats—characteristic sequences of DNA that indicate species variability. We also worked on developing techniques to isolate DNA from tissue in the feathers that were collected. Once we have isolated feather DNA, we will then be able to use information gained from the library microsatellites to index the amount of genetic variability in the Jemez owl population.

This project is expanding the Laboratory's core competency in bioscience and biotechnology and will significantly increase our understanding of a local threatened species. It will also address important scientific questions concerning the usefulness of noninvasive techniques such as ours for genetic analysis of threatened or endangered species in general and for studying the genetic viability of isolated Mexican Spotted Owl populations in particular.

Publications

Maltbie, M., et al., "Molecular Genetic Analysis of Mexican Spotted Owl Populations" (The Wildlife Society 5th Annual Conference, Buffalo, NY, Sept. 22–26, 1998).

Substrate-Dependent Cell-Cycle Disturbances in Response to Ionizing Radiation

97021

Donna Gadbois

We are investigating the effect of the extracellular matrix (ECM) environment on a cell's response to radiation exposure. The ECM is a meshwork of proteins that surrounds the cell to provide mechanical and elastic properties to tissues, provide a diffusion barrier for signaling molecules, and dictate whether a cell will proliferate or differentiate. A cell's replication and subsequent function are stimulated, in part, by the ECM environment; thus, changing the ECM within the body alters cell development and function. These alterations influence, for example, the healing of wounds and the course of disease, and they condition the body's response to implanted materials such as artificial joints, biosensors, or heart valves. The ECM environment is particularly important in radiation cancer therapy because a dose-limiting side effect of radiation therapy is the generation of radiation fibrosis. In the lung, radiation-induced fibrosis involves the accumulation and differentiation of fibroblast cells, which deposit copious amounts of aberrant ECM, resulting in loss of lung elasticity and often death months after the initial radiation treatment.

The cell cycle is divided into four parts: G1, S, G2, and M phases. G1 and G2 phases are the areas where proliferation control mechanisms are in place. S phase is the DNA synthesis period, and cells divide in the M phase. In the case of radiation exposure, noncancerous cells delay temporarily in the regulatory phases, G1 and G2, in order to repair damaged DNA, thus preventing gene alterations that can lead to cancer. Once the DNA damage is repaired, the cells can reenter the cell cycle and proliferate in a normal manner. However, we find that a particular cell type, human fibroblasts, do not reenter the cell cycle and remain arrested indefinitely in G1 phase after radiation exposure, even after DNA damage has been repaired.

This past year we have completed experiments that show that an extracellular factor, which is present in the culture medium of irradiated fibroblasts, is required for the prolonged radiation G1 phase arrest. We have begun work, using gene-array technology, to identify candidates for extracellular factors that cause the arrest. We have also shown that fibroblasts arrest when grown on certain types of ECM but not on others, and have begun characterization of cell-cycle protein expression when fibroblasts are irradiated on the different types of ECM. Finally, we have shown that the radiation G1 phase arrest is accompanied by differentiation of fibroblasts to the myofibroblast phenotype only on certain types of ECM. These observations can contribute significantly to improved outcomes in cellular responses during wound healing and radiation cancer therapy.

Role of New Cancer Gene in Environmental Carcinogenesis

98047

Min Park

Mutations in a new cancer gene (FHIT) have been implicated in a high incidence (>50%) of esophageal, colon, and lung cancers in humans. These organs are primary targets of environmental carcinogens and are therefore vulnerable to mutations that cause defects in cellular response to DNA-damaging agents. The predicted protein sequence suggests the FHIT gene product is an enzyme that degrades a dinucleotide (Ap4A) induced by DNA damage and environmental stress. The main objective of this study is to determine whether the FHIT gene product is a sensor molecule that detects the environmental carcinogens.

To test this idea, we used molecular biological approaches to produce the purified FHIT protein, and then initiated biochemical characterization of its enzymatic activities in order to understand its regulation. We successfully introduced a copy of the FHIT gene into a human cell line lacking FHIT. This new system allows us to study dynamic changes of Ap4A level in vivo using various environmental carcinogens. It also provides an opportunity to identify a novel signaling pathway that is responsible for the prevention of cancer development in the lung, the esophagus, and the colon. Immediate benefits of this research will be a better understanding of this new molecular mechanism of environmentally induced carcinogenesis, with future applications likely in the areas of cancer risk assessment, diagnostics, and therapy.

Theoretical Studies of the Allosteric Changes and DNA Binding of cAMP-Dependent, Transcription-Activation Protein

96013

Angel 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. The mechanism by which cAMP effects allosteric control over CRP activity remains unclear. We study the structure and dynamics of CRP by computer simulations. These studies provide information from which to predict currently obscure elements of cAMP-mediated activation pathways of CRP.

This year we developed new methods for correctly treating the electrostatic effects during computer simulations. These methods include the finite-size corrections to the electrostatic free energy resulting from simulating small systems. We developed a new free-energy-calculation approach that relies on a multistate Gaussian model for the distribution of the electrostatic solvation energies of simple solutes in aqueous solution. We studied the dynamics and physical properties of short (7 to 14 base pairs) DNA sequences by exhaustive molecular dynamics (MD) simulations in aqueous solution. The sequence dependence of the persistence length of the DNA and its propensity to bend have been studied within the context of protein DNA complex formation.

In addition, we studied a model of a nucleosome by means of detailed MD simulations (see the accompanying figure). The simulated system consisted of the protein octamer forming the histone core and 149 DNA base pairs. The system was immersed in an aqueous solution. The systems consisted of approximately 90,000 atoms. A 1.2-ns simulation of this system was performed. We analyzed this simulation in terms of the roles of ionics, hydrophobic effects, and DNA bending in the stability and fluctuations of the system.

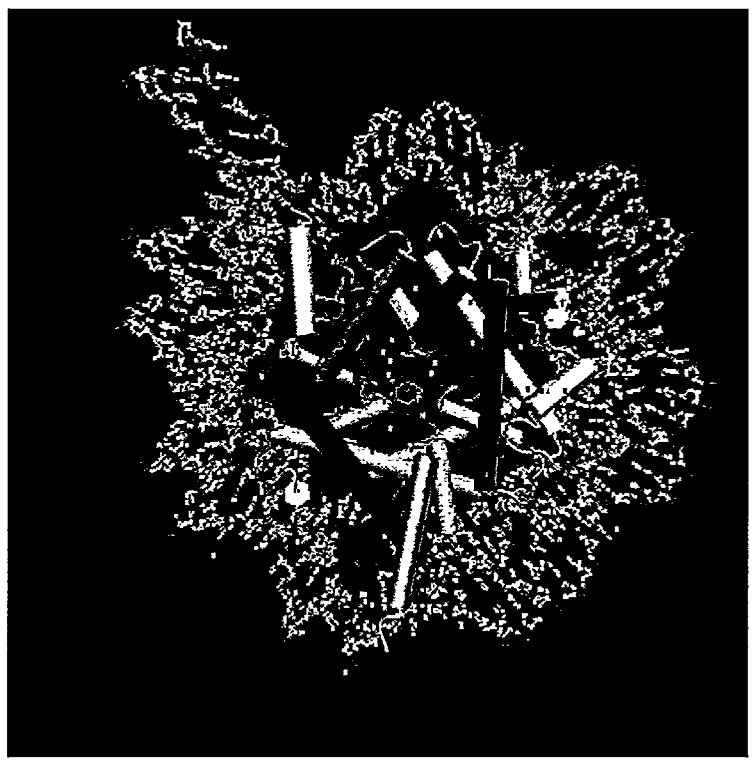
Publications

Garde, S., et al., "Temperature Dependence of the Solubility of Nonpolar Gasses in Water" (to be published in *Biol. Chem.*).

Hummer, G., et al., "Hydrophobic Effects on a Molecular Scale" (to be published in *J. Phys. Chem.*).

Hummer, G., et al., "Molecular Theories and Simulation of Ions and Polar Molecules in Water," *J. Phys. Chem.* **102**, 7885 (1998).

Hummer, G., et al., "The Pressure Dependence of Hydrophobic Interactions is Consistent with the Observed Pressure Denaturation of Proteins," *Proc. Natl. Acad. Sci. U.S.A.* **95**, 1552 (1998).



The nucleosome is the basic protein-DNA structural unit responsible for DNA compaction in the cell nuclei. An atomic model for the structure of the nucleosome has been constructed from the histone core-particle crystal structure. The position of the DNA relative to the histone core is determined from constraints on protein DNA contacts obtained from available nuclease digestion, cross-linking data, and hydroxil radical footprinting studies. We employed molecular modeling tools to generate an atomic model of the super-helical DNA (149 base pairs) double helix. The resulting nucleosome model system in aqueous solution consists of approximately 90,000 atoms. We performed a 1.2-ns MD simulation on this system. This simulation revealed the roles of ions and hydrophobic contacts in stabilizing the nucleosome.

Protein Motions That Determine the Efficiency of Photosynthesis

96032

Joel Berendzen

The photosynthetic reaction center from purple bacteria is a standard model system for studying the remarkably efficient process of photosynthesis. The goals of this project were to (1) experimentally determine structures of photosynthetic reaction centers in dark- and light-adapted states of bacteria and (2) apply theoretical methods in order to understand the changes and their effects on reaction rates. Working with collaborators from the Max Planck Institute in Germany, we are measuring atomic motions that take place during the process of charge separation. Our collaborator from the Laboratory's Theoretical Biology and Biophysics Group performed the theoretical calculations that allow us to interpret the results.

This year we investigated correct crystal-freezing conditions for photosynthetic reaction centers. Cryofreezing is required for obtaining data of sufficiently high resolution to build a structural model for electron transfer. Although our Max Planck collaborators have not yet succeeded in reliably growing crystals in the tetragonal space group needed for high-resolution diffraction studies, we have taken spectra of dark- and light-adapted reaction centers and of other heme proteins with our completed microspectrophotometer. Recent research emphasizes the need for high-resolution x-ray data on photosynthetic reaction centers; with relatively poor resolution, one study showed that the structure of the protein changes upon illumination, but that the motions are rather ill-defined and do not appear to agree with previous results. We are continuing our collaboration with the Max Planck Institute and are pursuing active collaborations on other charge-transfer membrane-bound proteins.

Full-Length Sequencing of Human cDNAs

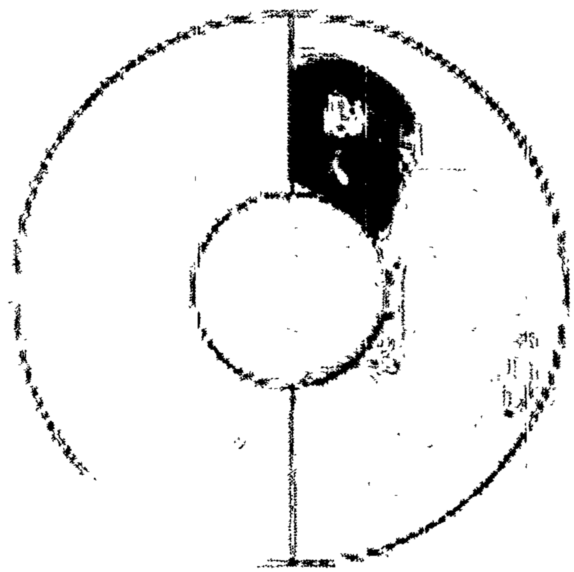
98052

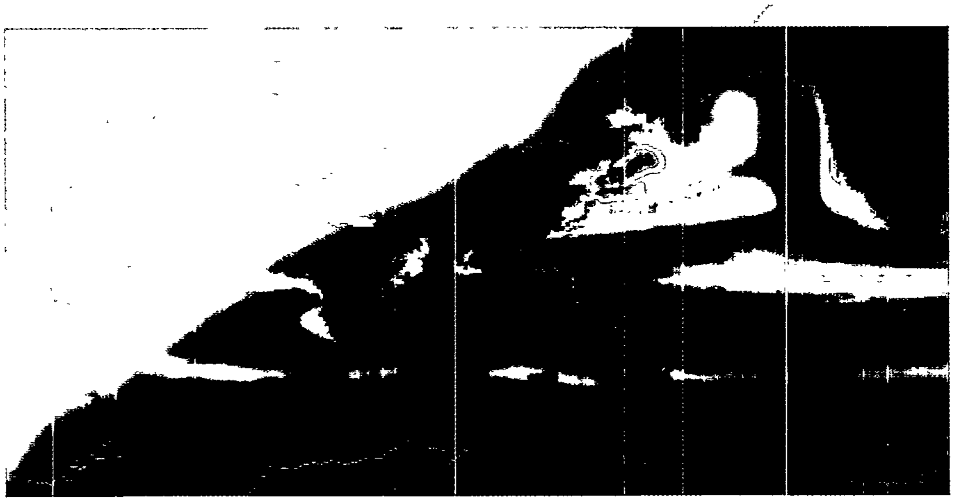
Michael Altherr

The ultimate goal of this project is to establish a high-throughput cDNA sequencing process capable of generating cDNA sequences that represent complete transcription units (the segments of genomic DNA that encode proteins). To initiate full-length sequencing, we are using the simultaneous shotgun sequencing (SSC) method.

With the SSC method, we purified cDNA inserts in the 1- to 5-kbp range following restriction digestion. The fragments were pooled on an equal molar basis, concatenated by ligation, sheared, randomly cloned into M13-like vectors, and sequenced. The sequence of individual clones is ultimately obtained in the assembly process, using the concatenation sites as tags to identify the ends of the individual cDNAs.

The SSC method holds promise for rapidly generating sequence information for a large number of clones simultaneously. The specific aim of this LDRD project is to generate sequencing-ready substrates by developing and demonstrating expertise in the associated processes required for using the SSC method. Toward this end, approximately 500 cDNAs identified in the IMAGE set that have been mapped to chromosome 16 were subjected to the process and used to generate subclone libraries ready for sequencing. IMAGE stands for integrated molecular analysis of the human genome and its expression.





1998 LDRD

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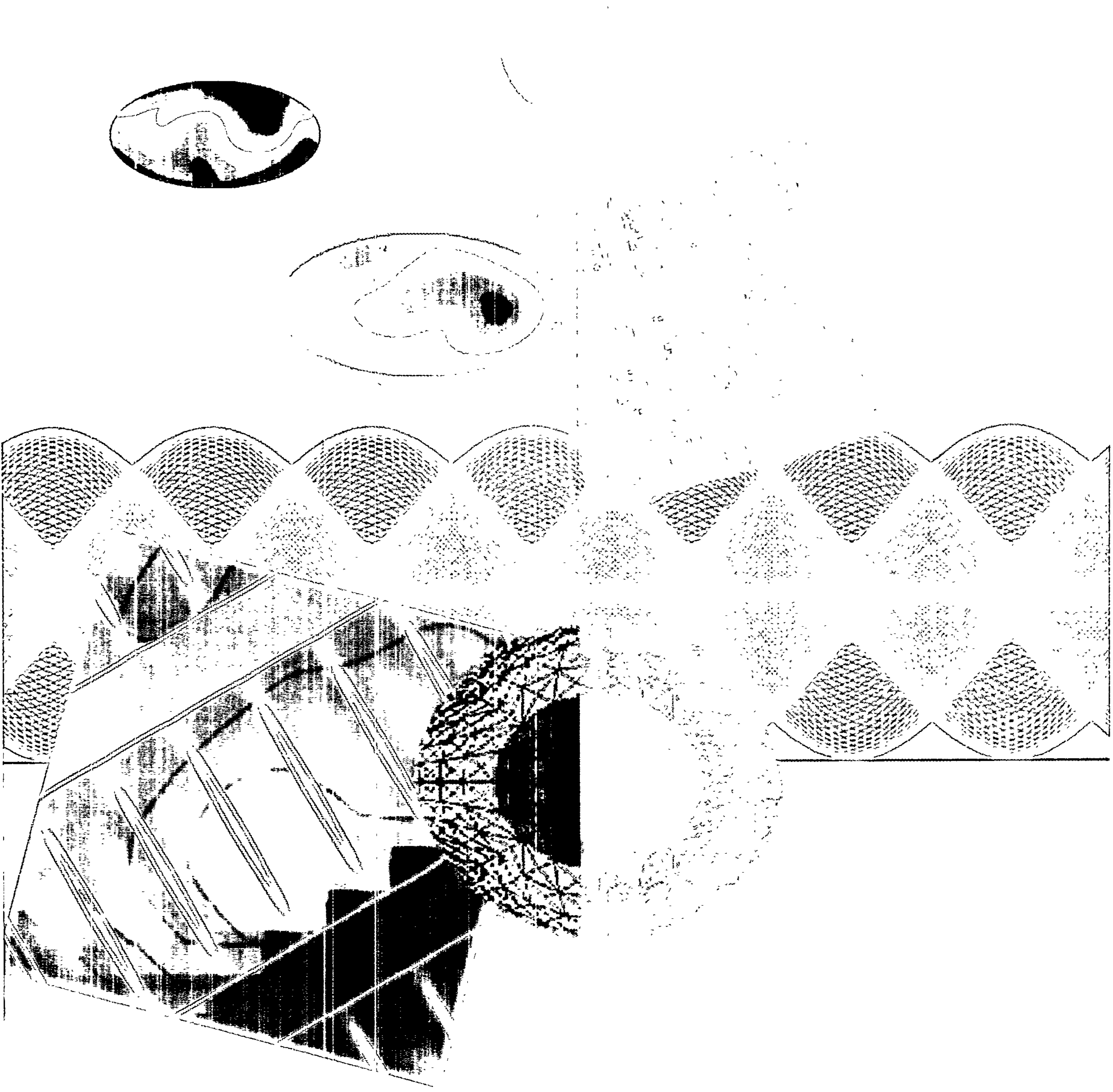

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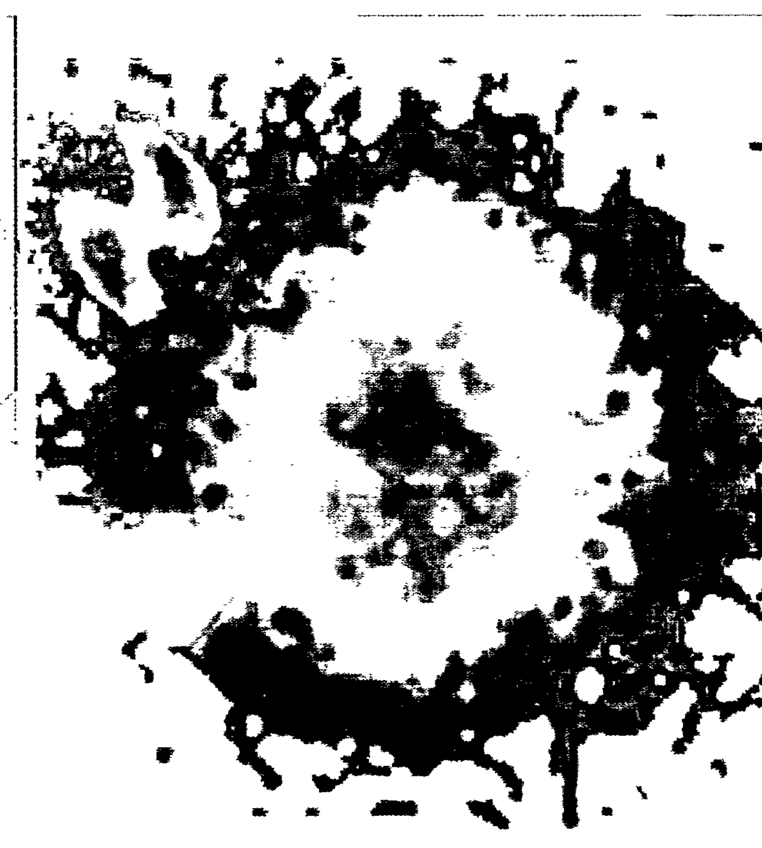

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