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G. Campbell, M. Kumar, J. Tobin, A. Noy, N. Browning

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Disclaimer

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FWP Title: Observing Fundamental Mechanisms of Transient Dynamics in Materials by Ultrafast In-Situ TEM
FWP Number: SCW0974

Lawrence Livermore National Laboratory
B&R Code: KC020101

Program Abstract:

We aim to develop a fundamental understanding of materials dynamics (from μs to ns) in systems where the required combination of spatial and temporal resolution can only be reached by the dynamic transmission electron microscope (DTEM). In this regime, the DTEM is capable of studying complex transient phenomena with several orders of magnitude time resolution advantage over any existing in-situ TEM. Using the unique *in situ* capabilities and the nanosecond time resolution of the DTEM, we seek to study complex transient phenomena associated with rapid processes in materials, such as active sites on nanoscale catalysts and the atomic level mechanisms and microstructural features for nucleation and growth associated with phase transformations in materials, specifically in martensite formation and crystallization reactions from the amorphous phase. We also will study the transient phase evolution in rapid solid-state reactions, such as those occurring in reactive multilayer foils (RMLF).

Program Impact: The LLNL DTEM possesses unique capabilities for capturing time resolved images and diffraction patterns of rapidly evolving materials microstructure under strongly driven conditions. No other instrument in the world can capture images with <10 nm spatial resolution of interesting irreversible materials processes such as phase transformations, plasticity, or morphology changes with 15 ns time resolution. The development of this innovative capability requires the continuing collaboration of laser scientists, electron microscopists, and materials scientists experienced in time resolved observations of materials that exist with particularly relevant backgrounds at LLNL. The research team has made observations of materials processes that are possible by no other method, such as the rapid crystallization of thin film NiTi that identified a change in mechanism at high heating rates as compared to isothermal anneals through changes in nucleation and growth rates of the crystalline phase. The project is designed to reveal these fundamental processes and mechanisms in rapid microstructure evolution that form the foundation of understanding that is an integral part of the DOE-BES mission.

FY 2009 Authorized Budget (New BA): $1,002,000

Program Personnel Supported in FY2009 to Nearest +/- 10% FTE (Clearly delineate status of individual: PI, PD, student. Technical Support Staff):

Dr. G.H. Campbell (PI) 30%, Dr. N.D. Browning (PI) 30%, Dr. T.B. Lagrange (staff) 80%, Dr. B.W. Reed (staff) 25%, Dr. N. Goldman (staff) 20%, Dr. J.E. Evans (postdoc) 35%, Ms. M. Bonds (student) 100% time on this project, with effort supported by a UCD scholarship.

Interactions: (limit to current interactions and collaborations)

Prof. B.C. Gates, UC-Davis – Nanoscale catalysis; Prof. W.A. Schroeder, UIC – Pulse compression; Prof. J.C.H. Spence, ASU – Photofield emission tips; P.E. Fischione, Fischione Instruments – in-situ gas stages. Prof. D. Grummon, MSU – martensitic transformations in shape memory alloys. Profs. T. Weihs and T. Hufnagel, JHU – reactive multilayer thin films. Prof. J. Wiezorek, U. Pitt. – crystallization from an amorphous phase.

FWP Title: Evolution of Grain Boundary Networks in Extreme Radiation Environments LLNL
FWP Number: SCW0939 B&R Code: KC02

Program Abstract:

Our previous work shows that grain boundary networks consisting of a high fraction of annealing twins and twin variant boundaries can be stabilized against thermal coarsening and other grain-boundary-mediated degradation. Our objective is to study whether such networks of grain boundaries can also enhance microstructural stability under irradiation. The project aims to develop the basic science needed to address the overarching question: What must a grain boundary network look like if it is to act as an effective point defect sink, not only on initial deployment, but after months or years of severe irradiation? We envision a network in which high free volume, high-energy "random" boundaries act as point defect sinks while the more stable, low energy special boundaries anchor the network, and the entire ensemble is statistically stable over the lifetime of the material. To understand whether this is possible, we are studying (1) the coupled evolution of grain boundary structure and point defects, using simulation, theory, and transmission electron microscopy of irradiated materials, and (2) what this implies about the evolution of the grain boundary network, using spatially-resolved diffraction coupled with mesoscale theory. By using a tightly coupled combination of theory (crystallography and defect dynamics), computations (molecular dynamics and phase field), and experiment (*in situ* transmission electron microscopy, electron backscatter diffraction, and three-dimensional X-ray diffraction) we are exploring the basic physics governing the evolution of microstructures under high-dose irradiation. The results are being extended into a crystallographically-sophisticated multiscale model of the interaction between radiation-induced point defects and grain boundaries in the context of a highly coordinated grain boundary network.

Program Impact: The project objectives are not singular, with a mix of theory, simulations, and experiments, which simply means that it would be impossible for a single investigator to possess the requisite expertise to carry out all aspects of the work. LLNL has unique strengths and expertise in all of the fundamental components of the work (computational materials science, radiation effects on materials, grain boundary networks, and electron microscopy) as well as a flexible interdepartmental structure that allows rapid identification of available experts. This management structure is allowing us to steer the definition, prioritization, and execution of the individual elements with flexibility and efficiency while ensuring that every component ultimately connects back to the central goals of the project.

FY 2009 Authorized Budget (New BA): $744K

Program Personnel Supported in FY2009 to Nearest +/- 10% FTE (Clearly delineate status of individual: PI, PD, student. Technical Support Staff):
Mukul Kumar, PI, 0.3 FTE; Thomas Lagrange, 0.2 FTE; Bryan Reed, 0.2 FTE; Vasily Bulatov, 0.3 FTE; Joel Bernier, 0.1 FTE; James Belak, 0.2 FTE; Ming Tang (Lawrence Post-doctoral Fellow, no cost)

Interactions: (limit to current interactions and collaborations)

Related Project URL: (optional)

FWP Title: Investigations of Electron Correlation in Complex Systems **Lawrence Livermore National Lab**
FWP Number: SCW0289 **B&R Code: KC 0203010**

Principal Investigators: JG Tobin (Lead), BW Chung, S-W Yu, & GD Waddill (Missouri Inst. Sci. Tech.)
Program Abstract:
Electron correlation, the electron-electron interactions outside of simple one-electron models, is the key to resolving outstanding issues concerning the electronic structure of complex materials. These complex materials have intricate physical structures and/or multiple phases with unique electronic-structure properties. We will use cutting-edge techniques such as Bremstrahlung Isochromat Spectroscopy (BIS) and Double Polarization Photoelectron Dichroism (DPPD), a sophisticated variant of Photoelectron Spectroscopy (PES), and related techniques to determine the valence electronic structure of complex materials. These complex materials include potential spintronic spin-source materials such as Fe/GaAs; materials exhibiting correlation effects such as Kondo Shielding, e.g. the Rare Earth Ce; and the crucially important yet poorly understood actinide, Pu. These are each distinctive and separate areas of world class science, an issue at the core of the DOE Office of Science and Office of Basic Energy Research. Measurements will be performed at the Advanced Light Source (Lawrence Berkeley National Laboratory, Berkeley, CA), the Advanced Photon Source (Argonne National Laboratory, Chicago, IL) and Lawrence Livermore National Laboratory (Livermore, CA). We will solve the Pu electronic structure problem, a goal of tremendous scientific and technological importance to DOE and the USA. We will provide experimental benchmarking for the new theories of Pu and actinide 5f electronic structure, leading to the resolution of a problem that has remained unsolved for the last 60 years, the nature of the 5f electron. A proper modeling of Pu electronic structure will, in turn, lead to an enhanced capability to predict the behavior of Pu over long periods of time, an issue of tremendous importance to environmental cleanup, radioactive waste storage and science based stockpile stewardship.
Program Impact:
The key points all revolve around two parallel issues: the importance of utilizing the unique capabilities (1) onsite at LLNL for Pu and actinide research and (2) offsite at the 3rd Generation Synchrotron Radiation Sources for the pursuit of experiments with complete polarization control, i.e. high brightness circularly polarized soft x-rays. The utilization of two of our three spin resolving spectrometers at the ALS and APS addresses the second issue. Moreover, the first issue is addressed by the development of our dedicated Fano/BIS Spectrometer at LLNL. It should be stressed that the combined Fano/BIS Spectrometer, which is now operational onsite at LLNL, is a completely unique facility for Actinide Research. While other spectrometers exist at sites where similar samples are available, there is no other such facility that combines the unique and cutting edge measurements of Fano/BIS Spectroscopy with Actinide materials. The other spectrometers at other actinide facilities have no such capacity to probe electron correlation and the unoccupied electronic structure of the 5f states. If we are to solve the 5f electron correlation problem in the actinides in general and Pu in particular, it will be done with this unique experimental tool at LLNL.
FY 2009 Authorized Budget (New BA): BA $600,000
Program Personnel Supported in FY2009 to Nearest +/- 10% FTE
(Clearly delineate status of individual: PI, PD, student. Technical Support Staff):
Lead PI: JG Tobin 100%, Co-PI: Sung Woo Yu 60%, Co-PI:Brandon Chung < 1%
Interactions: (limit to current interactions and collaborations)
Advanced Photon Source and Argonne National Laboratory
We have been working closely with Dr. Lynda Soderholm at ANL regarding the potential of an Actinide Science component to the proposed APS Renewal. This began with a presentation by JGT at the 2009 APS Users Meeting and has continued through the upcoming workshop at ANL entitled "Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems." More information regarding this workshop can be found at http://www.aps.anl.gov/News/Conferences/.
Advanced Light Source and Lawrence Berkeley National Laboratory
We have continued our ingoing interaction with Dr. David Shuh at LBNL. Most recently, this involved the Co-Organization of the Actinides 2009 International Conference in San Francisco during July 2009 by LBNL and LLNL. The Actinides 2009 conference featured current discussions on research in the physics and chemistry of the actinides and the transactinide elements. Specific topics included fundamental materials science, chemistry, physics, environmental science, and nuclear fuels. Of particular importance was a focus on the key roles basic actinide chemistry and physics research play in advancing the worldwide renaissance of nuclear energy. Actinides 2009 provided a forum spanning this diverse range of scientific topics. The conference consisted of plenary lectures, invited presentations, and contributed presentations in both oral and poster formats. More information can be found at http://www.lbl.gov/actinides2009/.
We continue to interact and collaborate with personnel from the ALS/LNBL such as Simon Morton.

FWP Title: Directed organization of functional materials at inorganic-macromolecular interfaces LLNL
FWP Number: SCW0289 B&R Code: KC02

Program Abstract:
The purpose of this research is to investigate the physical controls on directed organization of complex biomolecules at surfaces by using site-specific engineering of viral proteins, synthesis of chemoselective linkers, and nanoscale patterning of those linkers to create templates for virus and biomolecules assembly. Chemical force, confocal and atomic force microscopy, and electronic device characterization are employed to characterize biomolecule-template interactions as well as the morphology and kinetics of assembly. Molecular dynamics (MD) and kMC simulations are compared to experimental results to build a physical picture of the organization process and provide design principles for the templates.

Program Impact: Resulted in interdisciplinary collaboration amongst biologists, chemists, physicists and materials scientists at two National Laboratories, and three major universities. Produced first suite of experimental and theoretical results revealing underlying physical principles controlling virus organization at chemical templates. Explored the use of DNA origami to generate hightly complex arrays that display order on multiple length scales. Enabled development of a versatile and novel nanoelectronic platform based upon lipid-protein organization on nanowire and nanotube templates and demonstrated first functional bionanelectronic devices in which proteins interact with electronic nanomaterials.

FY 2009 Authorized Budget (New BA): $309,000 at LLNL $150K at LBNL

Program Personnel Supported in FY2009 to Nearest +/- 10% FTE (Clearly delineate status of individual: PI, PD, student. Technical Support Staff):
A. Noy (PI, 20%); J. De Yoreo (PI, 10%); G. Gilmer (Staff, 20%); R. Friddle (Postdoc, 75%); E. Guevara (Student, 100%); V. Lam (Student, 100%); N. Stephanopoulos (Student, 100%)

Interactions: (limit to current interactions and collaborations)
Internal—Computational Materials Sciences Group and Molecular Biophysics and Functional Nanostructures Groups within the BSNL, LLNL
External— Materials Science Division and the Molecular Foundry, Lawrence Berkeley National Laboratory; Department of Chemistry, University of California at Berkeley; Department of Mechanical Engineering, Colorado School of Mines; Department of Chemistry, Arizona State University (Hao Yan group), Department of Mechanical Engineering, University of California at Berkeley, Department of Chemical Engineering, University of California at Davis.

Recognitions, Honors and Awards (at least in some part attributable to support under this FWP):
J.J. De Yoreo – Elected Vice-President, Materials Research Society; Journal-sponsored Best Poster Award, Symposium KK, MRS Meeting, S2009; Associate Editor, *Journal of Crystal Growth*;
A. Noy – LLNL Excellence in Publication Award.
M. Francis – University of California, Berkeley, Distinguished Teaching Award
G. Gilmer -

Related Project URL: (optional)

FWP Title: Observing Biomolecular Materials Assembly in DTEM **LLNL**
FWP Number: SCW0951 **B&R Code:** KC020301

Program Abstract: The presence of an interface between a macromolecular component and inorganic materials is a hallmark of biomolecular materials. In many cases, the organic side of the interface plays an active role in directing the formation and organization of the inorganic materials. In others, the inorganic component becomes the substrate for assembly of macromolecules. In both, our understanding of assembly is extremely limited because until recently there was no experimental tool that possessed the spatial and temporal resolution needed to capture the formative events in the process. We believe that the time is now ripe for eliminating this obstacle by combining *in situ* fluid-cell imaging with dynamic transmission electron microscopy (DTEM). The goal of this feasibility study is to show that the DTEM can be utilized to directly image assembly in three biomolecular materials systems: mineralization in ferritin cages, lipid assembly on nano-tubes and -wires, and self-organization of protein-lipid complexes known as nano-lipid disks. In doing so, we believe we can open a new window on biomolecular materials that promises dramatic advances in our understanding of the underlying thermodynamic and kinetic factors that lead to organization of macromolecules and drive assembly at macromolecular-inorganic interfaces.

Program Impact: The LLNL DTEM has been shown to be able to uniquely capture time resolved images and diffraction patterns of rapidly evolving microstructures. As the DTEM images are formed from ~10ns pulses of electrons, typically containing ~10^9 electrons per pulse, that are spread over an imaging area of ~$10 \mu m^2$, the electron dose on a the sample is much lower than the damage threshold for damage in biological structures (nominally ~10 electrons/$Å^2$). In addition, the high speed of the imaging process means that samples can be studied in a fluid (Brownian motion does not significantly decrease the resolution on the nanosecond timescale). By constructing an in-situ fluid stage for the DTEM, this means that organic/biological structures can be studied in their live hydrated state. This means that biomolecular and in particular, biomineralization, can be studied for the first time with combined high spatial and temporal resolution. There is currently no other microscope in the world can capture images of these processes with <10 nm spatial resolution. The continued development of this innovative capability requires the continuing collaboration of laser scientists, electron microscopists, and biologists experienced in time resolved observations that exist at LLNL. This project is designed to reveal the fundamental mechanisms behind the initial stages of biomineralization that form the foundation of processes that are integral to the DOE-BES mission.

FY 2009 Authorized Budget (New BA): $292,000

Program Personnel Supported in FY2009 to Nearest +/- 10% FTE:
Dr. N.D. Browning (PI) 10%, Dr. M. Santala (postdoc) 75%, Dr. M. Wall (technical support staff) 45%.

Interactions: (limit to current interactions and collaborations)
Norman Salmon (Hummingbird Scientific) – design of the in-situ liquid stage; Dr. James Evans (UC-Davis) – design of DTEM experiments for biological systems; Dr. James DeYoreo (LBNL) – design and implementation of biomolecular experiments in DTEM to be correlated with AFM and TEM experiments.