LDRD

Laboratory Directed Research and Development

LAWRENCE LIVERMORE NATIONAL LABORATORY

FY2005

ANNUAL REPORT

About the Cover Image:

A transmission electron microscopy (TEM) image of germanium dioxide (GeO_2) foam, an important material in stockpile stewardship research. (The thin, gray strands are GeO_2 roughly 25 nm thick; the darker areas are overlapping strands.) Because overlap in TEM-generated images makes it difficult to determine cell sizes reliably, work supported by LDRD (project 05-ERD-003) is exploring small angle x-ray scattering to characterize sub-50-nm pore sizes and structures such as this.

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Acknowledgments

This Annual Report provides an overview of the FY2005 Laboratory Directed Research and Development (LDRD) Program at Lawrence Livermore National Laboratory (LLNL) and presents a summary of the results achieved by each LDRD project. At LLNL, Laboratory Director Michael Anastasio and Deputy Director for Science and Technology Cherry Murray are responsible for the LDRD Program and delegate responsibility for the operation of the program to the Associate Deputy Director for Science and Technology and the Director of the Laboratory Science and Technology Office (LSTO), Rokaya Al-Ayat. The LDRD Program at LLNL is in compliance with Department of Energy (DOE) Order 413.2 and other relevant DOE orders and guidelines.

The LDRD Program extends its sincere appreciation to the principal investigators of the FY2005 projects for providing the content of the annual report and to the publications team. The program also thanks the following members of the LSTO team for their many contributions to this publication: Mary Callesen, and Delores Lambert, administrators; Nancy Campos, database manager; Steve McNamara, computer specialist; and Cathleen Sayre, resource manager.

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Director's Statement

The Laboratory Directed Research and Development (LDRD) Program, authorized by Congress in 1991 and administered by the Laboratory Science and Technology Office, is our primary means for pursuing innovative, long-term, high-risk, and potentially high-payoff research that supports the missions of the Laboratory, the Department of Energy, and National Nuclear Security Administration in national security, energy security, environmental management, bioscience and technology to improve human health, and breakthroughs in fundamental science and technology. The accomplishments described in this annual report demonstrate the strong alignment of the LDRD portfolio with these missions and contribute to the Laboratory's success in meeting its goals.

The LDRD budget of $70 million for FY2005 sponsored 200 projects. These projects were selected through an extensive peer-review process to ensure the highest scientific quality and mission relevance. Each year, the number of deserving proposals far exceeds the funding available, making the selection a tough one indeed.

Our ongoing investments in LDRD have reaped long-term rewards for the Laboratory and the nation. Many Laboratory programs trace their roots to research thrusts that began several years ago under LDRD sponsorship. In addition, many LDRD projects contribute to more than one mission area, leveraging the Laboratory's multidisciplinary team approach to science and technology. Safeguarding the nation from terrorist activity and the proliferation of weapons of mass destruction will be an enduring mission of this Laboratory, for which LDRD will continue to play a vital role.

The LDRD Program is a success story. Our projects continue to win national recognition for excellence through prestigious awards, papers published in peer-reviewed journals, and patents granted. With its reputation for sponsoring innovative projects, the LDRD Program is also a major vehicle for attracting and retaining the best and the brightest technical staff and for establishing collaborations with universities, industry, and other scientific and research institutions. By keeping the Laboratory at the forefront of science and technology, the LDRD Program enables us to meet our mission challenges, especially those of our ever-evolving national security mission.

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Annual Report

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About the FY2005 Laboratory Directed Research and Development Annual Report

The FY2005 Laboratory Directed Research and Development (LDRD) Annual Report provides a summary of LDRD-funded projects for the fiscal year and consists of two parts:

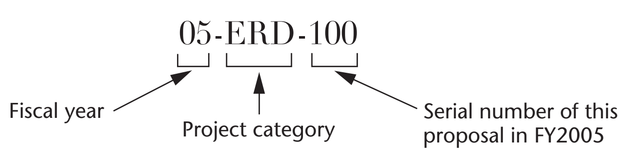
Overview

An introduction to the LDRD Program, the LDRD portfolio-management process, program statistics for the year, and highlights of accomplishments for the year.

Project Summaries

A summary of each project, submitted by the principal investigator. Project summaries include the scope, motivation, goals, relevance to DOE/NNSA and LLNL mission areas, the technical progress achieved in FY2005, and a list of publications that resulted from the research in FY2005.

Summaries are organized in sections by research category (in alphabetical order). Within each research category, the projects are listed in order of their LDRD project category: Strategic Initiative (SI), Exploratory Research (ER), Laboratory-Wide Competition (LW), and Feasibility Study (FS). Within each project category, the individual project summaries appear in order of their project tracking code, a unique identifier that consists of three elements. The first is the fiscal year the project began, the second represents the project category, and the third identifies the serial number of the proposal for that fiscal year. For example:



Program Overview: Investing in Our Nation's Future

Program Overview: Investing in Our Nation's Future

About Lawrence Livermore National Laboratory

A premier applied-science laboratory, Lawrence Livermore National Laboratory (LLNL or the Laboratory) has at its core a primary national security mission—to ensure the safety, security, and reliability of the nation's nuclear weapons stockpile without nuclear testing, and to prevent and counter the spread and use of weapons of mass destruction: nuclear, chemical, and biological.

The Laboratory uses the scientific and engineering expertise and facilities developed for its primary mission to pursue advanced technologies to meet other important national security needs—homeland defense, military operations, and missile defense, for example—that evolve in response to emerging threats. For broader national needs, the Laboratory executes programs in energy security and long-term energy needs, environmental assessment and management, bioscience and technology to improve human health, and breakthroughs in fundamental science and technology. With this multidisciplinary expertise, the Laboratory serves as a science and technology resource to the U.S. government and as a partner with industry and academia.

One of three Department of Energy (DOE)/National Nuclear Security Administration (NNSA) laboratories, LLNL has been managed since its inception in 1952 by the University of California (UC). This half-century association with UC has enabled the Laboratory to establish an atmosphere of intellectual freedom and innovation that attracts and maintains the world-class workforce needed to meet challenging national missions.

Laboratory Directed Research and Development Program

To fulfill its missions, LLNL must continually invest in the science and technology that form the foundation of its signature capabilities. The Laboratory Directed Research and Development (LDRD) Program, which was established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for fostering excellent science and technology for today's needs and tomorrow's challenges.

According to its Congressional mandate,[1] the purpose of LDRD is to foster excellence in science and technology that (1) supports the DOE/NNSA and LLNL missions and strategic vision; (2) ensures the technical vitality of the Laboratory; (3) attracts and maintains the most qualified scientists and engineers and allows scientific and technical staff to enhance their skills and expertise; (4) helps meet evolving DOE/NNSA and national security needs; and (5) enables scientific collaborations with academia, industry, and other government laboratories.

By enabling LLNL to fund creative basic and applied research activities in areas aligned with its missions, the LDRD Program develops and extends the Laboratory's intellectual foundations and maintains its vitality as a premier research institution. The present scientific and technical strengths of LLNL are, in large part, a product of LDRD investment choices in the past.

[1] U. S. Department of Energy. Order 413.2A. Laboratory Directed Research and Development (January 8, 2001).

The LDRD Portfolio Management Process

The FY2005 LDRD portfolio-management process at LLNL consisted of three major components that ensured the quality of the year's portfolio and its alignment with the DOE/NNSA and the Laboratory's missions: (1) a top-level strategic planning process to identify strategic science and technology areas for LDRD investment; (2) a call to the Laboratory scientific and technical community for innovative and relevant proposals within the DOE/NNSA mission areas; and (3) a scientific peer-review process to select the highest quality LDRD portfolio from these proposals.

In FY2005, the top-level LDRD strategic planning process was guided by the DOE Strategic Plan[2] for the next 25 years, and by the Laboratory's own long-range plan that will define the scientific and technical strategy for the coming decade. The 2004 DOE Strategic Plan articulates four strategic goals for achieving the DOE mission of advancing the national, economic, and energy security of the U.S., promoting scientific and technological innovation in support of that mission, and ensuring the environmental cleanup of the national nuclear weapons complex. In FY2005, the LLNL LDRD Program strongly supported all four DOE strategic goals:

1. *Defense*—To protect our national security by applying advanced science and nuclear technology to the nation's defense.

2. *Energy*—To protect our national and economic security by promoting a diverse supply and delivery of reliable, affordable, and environmentally sound energy.

3. *Science*—To protect our national and economic security by providing world-class scientific research capacity and advancing scientific knowledge.

4. *Environment*—To protect the environment by providing a responsible resolution to the environmental legacy of the Cold War and by providing for the permanent disposal of the nation's high-level radioactive waste.

The Laboratory's Long-Range Strategic Science and Technology (S&T) Plan continues to inform the LDRD portfolio planning process. Broadly inclusive, the Laboratory's S&T Plan is intended to elicit the most far-reaching and innovative ideas for the future shape of science and technology at LLNL. The six thematic areas of the S&T Plan are as follows:

- Stockpile science and technology.
- High-energy-density science and technology.
- Nuclear, radiative, and astrophysical science and technology.
- Science and technology at the intersection of chemistry, biology, and materials science and technology.
- Information, simulations, and systems science and technology.
- Energy and environmental science and technology (with fusion energy science and technology as a special subtopic).

[2] U. S. Department of Energy. (2003). Strategic Plan. http://strategicplan.doe.gov/ (retrieved February 5, 2005).

The NNSA oversees LLNL's LDRD Program to ensure that it accomplishes its objectives. This oversight includes field and headquarters reviews of both the technical content and management processes. As demonstrated in a memorandum (April 30, 2002) from the Secretary of Energy and the NNSA Administrator, the DOE/NNSA actively supports the LDRD Program. In the memo, Secretary of Energy Spencer Abraham writes: "I believe that all sponsors, including other Federal agencies, benefit from the strong science and technology base provided by the Department's [LDRD] programs. [LDRD] is at the core of our ability to develop research capabilities and apply advanced technologies to effectively meet the Department's and the Nation's needs."[3]

Structure of the LDRD Program

Project Categories

The LDRD Program at LLNL consists of three major project categories: Strategic Initiative (SI), Exploratory Research (ER), and Laboratory-Wide (LW) Competition. Throughout the year, the Program also funds a few projects in a fourth category, Feasibility Study/Project Definition (FS).

Strategic Initiative

The SI category focuses on innovative R&D activities that are likely to set new directions for existing programs, help develop new programmatic areas within LLNL's mission responsibilities, or enhance the Laboratory's science and technology base. Projects in this category are usually larger and more technically challenging than projects funded in other categories. An SI project must be aligned with the strategic R&D priorities of at least one of the six thematic areas of LLNL's Long-Range Strategic S&T Plan.

Exploratory Research

The ER category is designed to help fulfill the strategic R&D needs of a Laboratory directorate (ERD) or institute (ERI). In this category, researchers submit proposals to their directorates and institutes, where the proposals are screened and subsequently forwarded to the ER selection committee for review. In FY2005, LLNL's S&T Plan continued to guide directorates in evaluating the ERD and ERI proposals.

Laboratory-Wide Competition

Projects in the LW category emphasize innovative research concepts and ideas and undergo limited management filtering. The LW competition is open to all LLNL staff in programmatic, scientific, engineering, and technical support areas. Researchers submit their project proposals directly to the LW selection committee.

Feasibility Study/Project Definition

This special project category, FS, provides researchers with the flexibility to define and develop potential projects in the other three categories. To increase its responsiveness to Laboratory scientists and engineers, the LDRD Program funds FSs throughout the year.

[3] Memorandum from Secretary of Energy Spencer Abraham. 2002-0077386 (Washington, DC, April 30, 2002).

Project Competency Areas

Although LDRD projects often address more than one scientific discipline, each project is classified into one of ten research categories that is relevant to NNSA and Laboratory missions. The ten categories are:

- Advanced Sensors and Instrumentation.
- Biological Sciences.
- Chemistry.
- Earth and Space Sciences.
- Energy Supply and Use.
- Engineering and Manufacturing Processes.
- Materials Science and Technology.
- Mathematics and Computing Sciences.
- Nuclear Science and Engineering.
- Physics.

THE LDRD FY2005 PORTFOLIO

Overview of the FY2005 Portfolio

The FY2005 LDRD portfolio was carefully structured to continue LDRD's vigorous support for the strategic vision and long-term goals of DOE/NNSA and LLNL. The FY2005 projects described in this annual report underwent a stringent selection process and received ongoing management oversight.

In FY2005, LDRD funded 200 projects with a total budget of $70 million. Figure 1 shows the number of projects in each of the four categories. Figure 2 shows the distribution of funding among the four LDRD project categories.

Figure 3 shows the funding distribution by dollar amount for the 200 FY2005 projects. Eighty percent of the projects were in the $101-to-$500K range, with 6.5% falling below $100K. This lowest funding level includes all the FS projects. Seven percent of the projects were in the $501K-to-$1M funding range, and another 7% of the projects received more than $1M. The average funding level for the 220 projects was $352K. Figure 4 shows the percentage of LDRD Program funding and number of projects in each research category for FY2005.

Strategic Initiative

In FY2005, the LDRD Program funded six SI projects. Although the SI category represented only 3% of the total number of LDRD projects for FY2005, it accounted for 14% of the budget. Strategic Initiative projects ranged in funding from $346K to $2.4M.

Exploratory Research

In FY2005, 170 ER projects were funded. The largest project category, ERs accounted for 85% of LDRD projects for the fiscal year. Projects in this year's ER category ranged in budget from $9.7K to $1.4M

Laboratory-Wide Competition

Eighteen LW projects were funded in FY2005, which represent 9% of LDRD projects for the year and 4% of the budget. Laboratory-Wide projects are limited to $190K/year funding, with a few exceptions. In FY2005, LW projects ranged in funding level from $103.7 to $227K.

Feasibility Study

In FY2005, the LDRD Program funded six FS projects, or 3% of the total. Feasibility Studies are limited to $75K and a 12-month duration.

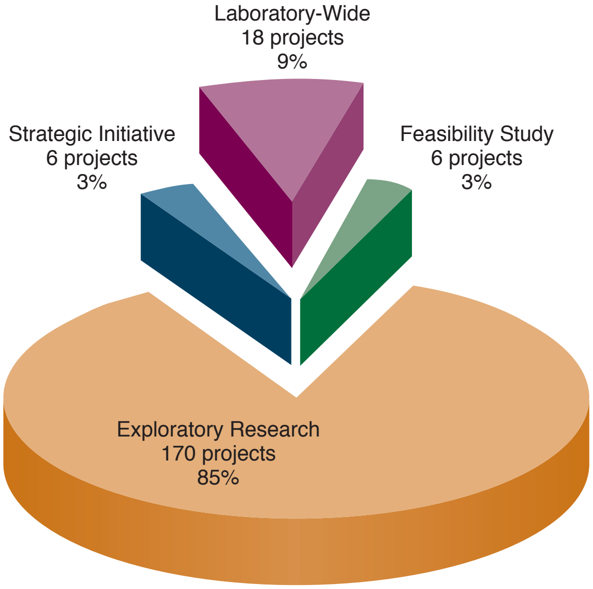


Figure 1. Number and percentage of the 200 LDRD projects in each project category in FY2005.

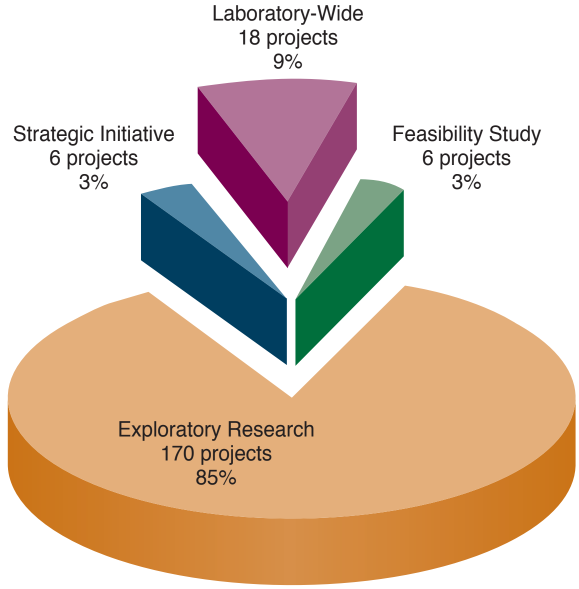
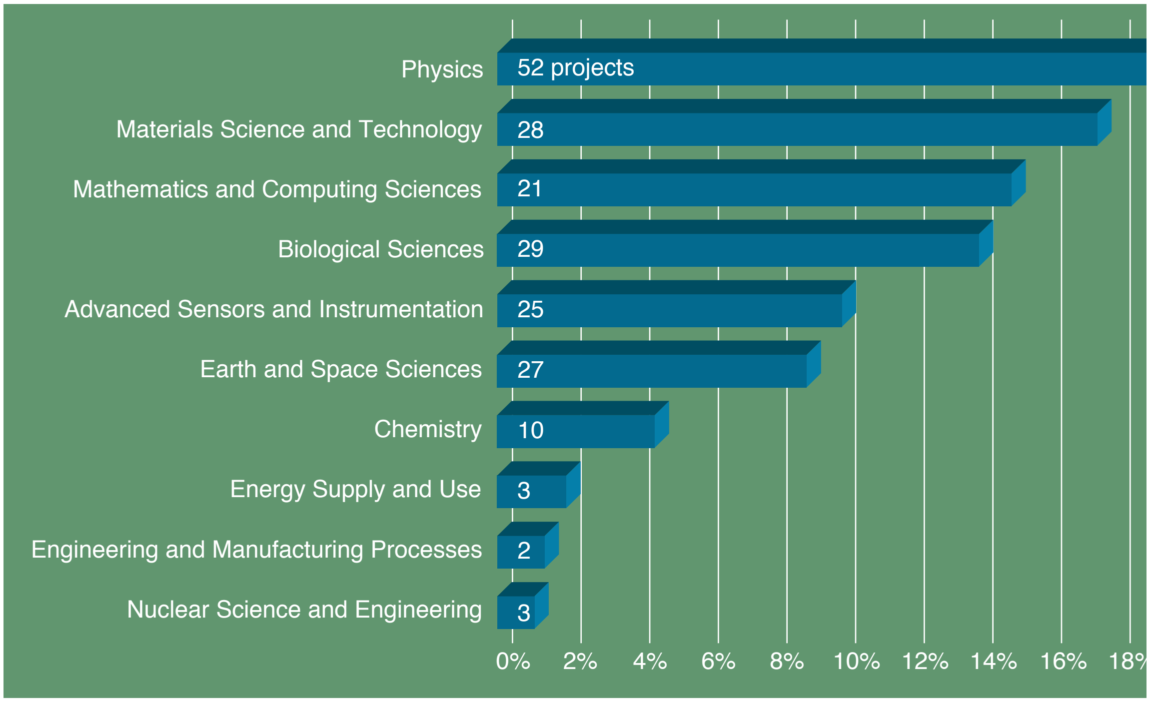


Figure 2. Distribution of funding among the four LDRD project categories. Total funding for FY2005 was $70M.

Figure 3. Number of projects and levels of funding. The average funding level for an LDRD project in FY2005 was $352K.



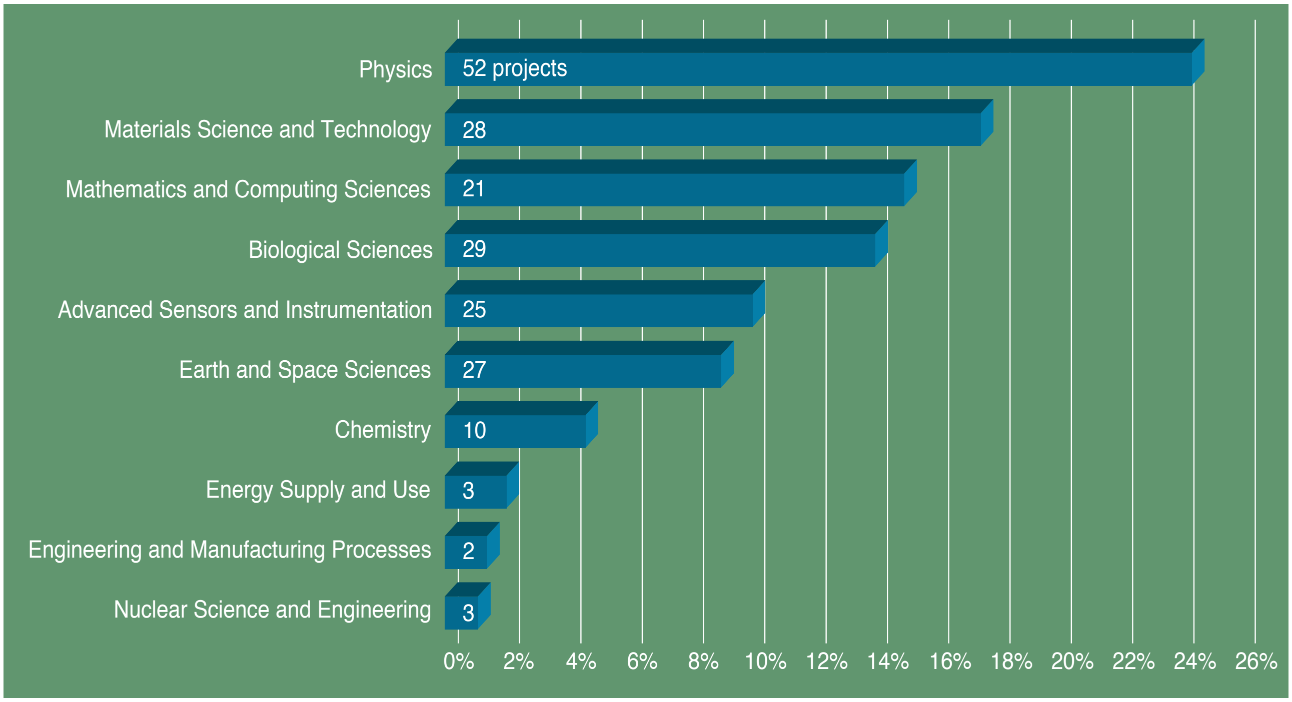


Figure 4. Percentage of LDRD funding and number of projects in each research category in FY2005.

Highlights of FY2005 LDRD Accomplishments

In FY2005, the LDRD Program at LLNL continued to be extremely successful in achieving its goals of scientific discovery, providing new concepts for core missions, and creating an exciting research environment that attracts outstanding young talent to the Laboratory. Below is a selection of FY2005 highlights that exemplify the program's noteworthy research results, timely support for critical national needs, and external recognition of Laboratory personnel.

- **Biological and Synthetic Nanostructures Controlled at the Atomistic Level (03-SI-001).**

  This project conducted combined theoretical and experimental investigations of the synthesis, characterization, and design techniques required to fabricate semiconducting and metallic nanostructures with enhanced properties. The focus was on developing capabilities with broad applicability to a wide range of materials—both to nanomaterials that are currently being developed and new, yet-to-be discovered nanomaterials. The team built expertise, computer simulation and modeling codes, chemical and vapor deposition synthesis techniques, and surface-sensitive spectroscopic characterization techniques. In addition, the project team identified nanomaterials for use in biological sensing, detection, and separation, including materials with potential use in advanced biodetection systems with national-security applications.

  In recognition of their work in unraveling the atomic structure of silicon and germanium nanoparticles, the project team received an LLNL 2004 Science & Technology Award. (Although an internal award, this is based on a recipient's national and international recognition.) The project resulted in numerous high-profile publications and was featured on the covers of *Nature* (October 7, 2004) and *Physical Review Letters*. This work also promoted collaboration with the Alivisatos Group at UC Berkeley and Lawrence Berkeley National Laboratory and helped the Lab hire many new researchers at the cutting-edge of their fields. Follow-on work has been funded by the Defense Advanced Research Projects Agency.

- **Force Spectroscopy to Study Multivalent Binding in Protein–Antibody Interactions (03-ERI-009).**

  This project uses atomic-force microscopy to measure the binding strength between the Muc1 tumor antigen and multiple Muc1 antibodies. The motivation of the project is the need to improve the binding times of cancer therapeutics that employ multiple antibodies to "hold on" to cancer cells. Determining the average bond lifetime for one, two, and three antibodies binding to their cancer targets will significantly benefit efforts to optimize therapeutics for cancer patients. The capability to measure antibody–protein interactions will enable other biological applications, including biological detectors of interest to homeland security and research in understanding drug–cell interactions.

In FY05, the team determined the kinetic rupture rate of multiple antibody–Muc1 bonds using dynamic force spectroscopy. These results confirmed a theoretical model describing multivalent binding and were published in the high-profile journal *Proceedings of the National Academy of Sciences*. (Another paper has been accepted for publication in the *Biophysical Journal*.) The team also developed a method to identify the number of interacting molecules through the force-extension profile of measurements, which greatly improves the identification of multiple bonds. This project has utilized—and expanded—LLNL's collaboration with the UC Davis Cancer Center and also stimulated further detection work with chemical and biological national security applications.

- **Hysteresis and Kinetic Effects during Liquid–Solid Transitions (05-ERD-014).**

The goals of this project are threefold: (1) gain insight into the kinetics of solid–solid and liquid–solid transition; (2) determine the shape of the high-pressure and -temperature melt line for metals in general; and (3) understand how hysteresis during multiple crossings of the melt line affect the liquid–solid transition. Acquiring this information is the first step towards developing a dynamic equation-of-state model. These goals will be accomplished by investigating the high-pressure resolidification of prototypical materials, such as iron, bismuth, tin, and water, through combinations of shock and isentropic compression in gas guns using graded-density impactors. In addition to these experiments, companion calculations will be performed at the hydrodynamic and atomic scales. This project is expected to yield increased understanding of the dynamics of materials under extreme conditions, particularly rapid resolidification in metals under conditions of high pressure and temperature, which is specifically applicable to stockpile science.

In FY05, the team performed shock experiments on the pressure-induced solid–solid transitions in bismuth. After designing a target holder to minimize the signal-to-noise ratio, they acquired very clean resistivity data, which will help confirm the results of computations. As a follow-on to this project, the principal investigator began computational work to model the same physics. As one of the very first users of Livermore's BlueGene/L computer—the fastest in the world at 280 teraflops per seconds—the investigator solved an outstanding problem in computational high pressure-high temperature materials: achieving high-fidelity, convergent results on the resolidification of tantalum. These calculations, involving billions of atoms, resulted in his team's winning the 2005 Gordon Bell Prize, which is awarded each year to recognize outstanding achievement in high-performance computing.

- **Microbial Pathways (03-ERD-062).**

Environmental remediation processes that use indigenous bacteria to reductively immobilize uranium and other metals in situ would have immediate application in the cleanup of contaminated aquifers at DOE legacy sites. This project makes use of emerging genome-enabled techniques—such as whole-genome complementary DNA (cDNA) microarrays—to explore novel, bacterial metabolic pathways and bacteria–mineral interactions that can mediate the behavior of uranium and other environmentally relevant elements in the subsurface. In addition to environmental cleanup, understanding microbial processes that mediate metal and radionuclide contamination in the environment also has relevance to national security issues, such as the intentional contamination of groundwater with radionuclides or toxic metals. Results expected in this project include enhanced understanding of the microbially mediated transformations of uranium in anaerobic soil and aquifer environments, and the genetic and biochemical basis of nitrate-dependent metal oxidation.

In FY05, the team completed a thorough, whole-genomic analysis of *Thiobacillus denitrificans*, including comparative genomic analyses, focusing on major metabolic and biogeochemical pathways. A series of cDNA microarray experiments was completed, as was computational analysis of large portions of the resultant data. Major advances were made in the development of a genetic system in *T. denitrificans* for knockout mutation (i.e., hypothesis testing) studies.

Following the sequencing of the *T. denitrificans* genome at the Joint Genome Institute, work done under this project enabled LLNL scientists to elucidate genes encoding key metabolic pathways in this bacterium and have carried out whole-genome cDNA microarray experiments to compare gene expression profiles during redox interactions between the bacterium and uranium (IV)- or iron (II)-containing minerals. For this project, LLNL scientists have also developed a system to inactivate (“knock out”) specific genes in *T. denitrificans* to investigate their roles in interactions with uranium-containing minerals.

- **Probing Other Solar Systems with Current and Future Adaptive Optics (05-ERD-055).**

Current adaptive optics (AO) systems have been barely able to detect a small number of extrasolar planets, but the planet-hunting, next-generation, high-contrast “extreme” AO systems (ExAO) being developed in this project will make it possible to probe the environments of other stars on scales comparable to the size of our solar system. This project is using existing AO to probe dust clouds and possible planets in nearby solar systems and to develop the precision optical technology needed for a LLNL-led future ExAO system for the Gemini Observatory. This project will carry out a search for young extrasolar planets orbiting nearby stars, using advanced image-processing techniques to separate planets from background noise. Direct detection of a planet orbiting a nearby star would be a major scientific achievement, opening new windows into the formation and nature of solar systems. Development of ExAO techniques, such as wavefront characterization and correction, will provide LLNL with a reservoir of skills and techniques for AO that can be applied to large optical systems and space optics that will support national security missions in remote sensing for counterproliferation and nonproliferation missions.

In FY05, using AO systems at the Keck Observatory, we observed a sample of young, dusty stars to search for planet-forming dust and possible young planets and developed new strategies to detect faint companions and circumstellar dust, such as the temporal filtering of data to enhance sensitivity by factors of 2 to 10. At UC Santa Cruz, we commissioned an ExAO test bed combining a 1024-actuator deformable mirror and LLNL's phase-shifting diffraction interferometer. Due in large part to the achievements of this project, a LLNL-led team was selected by the U.S. Gemini Observatory to construct a next-generation ExAO system for Gemini South, which will be the most advanced AO system in the world. This LDRD project team was also featured in a December 2004 *National Geographic* article on the search for extrasolar planets.

- **Target Fabrication Science and Technology: An Enabling Strategic Initiative (05-SI-005).**

This project aims to establish a new, major capability for fabricating targets for demonstrating fusion ignition under laboratory-controlled conditions. This includes developing the science and engineering for various materials and precision microassembly—and a double-shell ignition prototype target as a test bed. Specifically, the project team is developing (1) nanostructured alloys with high strength and thermal stability; (2) the capability to independently control the surface properties and pore structure of nanocellular materials and relate these to nanomechanical behavior; and (3) advanced lithographic approaches to three-dimensional (3-D) nanofabrication on large areas and complex surfaces with nanoscale-relief structures. Quantum confinement in 2-D systems will be investigated. Insight into all of these issues will lead to a new class of materials with novel electronic and luminescent properties. In addition to fusion energy, the target-fabrication technology developed in this project will also fill a need in stockpile stewardship, which requires increasingly complex targets.

In FY05 the team succeeded in fabricating high-strength nanocrystalline gold–copper alloys and demonstrating 3-D nanostructured, gradient-density materials on planar surfaces by conformal phase-mask lithography. Progress in developing the prototype ignition double-shell target included assessing the feasibility of low-atomic-number mandrels and the density requirements of nanoporous metallic foams. This work's payoffs in inertial-confinement fusion are expected to be considerable.

- **A Computational Design Tool for Microdevices and Components used in Pathogen Detection Systems (03-ERI-003)**

Microfluidics is playing a central role in the development of next-generation pathogen-detection devices for chemical and biological security efforts. This project team developed new computational models to simulate complex biological flows in integrated microsensors, and validated these models experimentally. This computational tool provides critical understanding of the fluid dynamics involved in microdevices and can shorten the design and fabrication process of microelectromechanical system devices such as the Biobriefcase by months per cycle by eliminating the need for trial-and-error design. These tools can also help optimize prototypes and provide a predictive capability for new, more advanced designs. Computational work was performed in collaboration with UC Davis, and experimental work in collaboration with UC Berkeley. By helping advance the state of the art in sensor technology, these new computational models further LLNL's national security mission in areas such as countering the proliferation and use of weapons of mass destruction.

Algorithm development in FY05 focused on physics and chemistry. A proof-of-concept calculation simulated the flow of genomic DNA in an extraction device that has possible applications in the Biobriefcase polymerase chain reaction module. In the modeling of particle-laden DNA solutions, the team devised two approaches—soft potentials and rigid constraints—to particle-interaction dynamics for polymer beads and rods. The team also demonstrated the fundamental issue of elastic wave propagation involved in the high-Weissenberg-number problem of viscoelastic flows—a problem that had been unsolved for 30 years. In addition, a model developed by the team was used to simulate biological flows in critical anatomies such as a stenotic carotid artery and a diseased trachea, demonstrating the model's broad potential application and its robustness in treating complex geometry.

Patents

Projects sponsored by LDRD consistently account for a large percentage of the patents issued for LLNL research, especially considering that the LDRD program funding represents 6% of the total LLNL budget, with the exception of FY2000, when the Program received only 4% of the budget. Table 1 shows the number of patents resulting from LDRD-funded research since 1998.

The year for which a patent is listed is the year in which the patent was granted; LDRD investment in a technology is sometimes made several years before the technology is actually patented. Furthermore, although an LDRD-sponsored project makes essential contributions to such technologies, subsequent programmatic sponsorship also contributes to a technology's further development.

	1998	1999	2000	2001	2002	2003	2004	2005
Number of all LLNL patents	78	84	93	89	97	71	95	93
Number of LDRD patents	39	45	35	42	27	29	55	51
LDRD patents as percentage of total	50%	54%	38%	47%	28%	41%	58%	55%

Table 1. Patents resulting from LDRD-funded research as a percentage of all LLNL patents from 1998 to 2005.

Awards

- **AAAS Fellow.** John F. Holzrichter, previous director of the Laboratory Science and Technology Office, which administers the Laboratory's LDRD Program, was selected as a fellow of the American Association for the Advancement of Science in the field of physics.

- **APS Fellows.** Of the five Livermore scientists who were elected as fellows of the American Physical Society in 2005, two were elected on the basis of work that was largely supported by LDRD:

   – Christina Back—For work in x-ray spectroscopy applied to high-energy-density plasmas (LDRD project 04-ERD-019).

   – Thomas Rognlien—For work in the modeling of tokamak edge plasmas and radio-frequency-excited plasmas (e.g., LDRD projects 03-ERD-009 and 99-ERD-029).

- **Edward Teller Medal.** LLNL senior scientists Max Tabak received a 2005 Edward Teller Medal from the American Nuclear Society in recognition for his seminal work in developing fast-ignition fusion. This work has been supported at important junctures by LDRD (projects 02-ERD-041 and 04-ERD-054).

- **E. O. Lawrence Award.** Livermore astrophysicist Claire Max was one of seven scientists to win the Department of Energy's 2004 E. O. Lawrence Award. Claire's award recognized her contributions to the theory of laser guide star adaptive optics, important aspects of which were supported by LDRD (projects 00-ERD-049 and 03-ERD-002).

- **Excellence in Fusion Engineering Award.** In recognition for her many technical contributions to laser systems, Livermore's Camille Bibeau received the Fusion Power Associates' Excellence in Fusion Engineering Award. Important breakthrough work performed by Camille was supported by LDRD (projects 97-SI-014 and 00-SI-009).

- **Humboldt Research Award.** A Humboldt Research Award was conferred on Livermore scientist Siegfried Glenzer in recognition for his lifetime achievements in research. Siegfried has received LDRD support for important recent work (projects 01-ERD-107, 03-ERD-070, and 05-ERI-003).

- **LLNL 2004 Science & Technology Awards.** Teams whose work was substantially supported by LDRD receive both of the FY2004 Science and Technology Awards. (Although an internal award, this is based on a recipient's national and international recognition.) The winning team leaders were:

   – Jerry Britten—For work on optics for high-energy, short-pulse lasers (LDRD project 03-ERD-059).

   – Guilia Galli—For discovery of the Bucky diamond and unraveling the atomic structure of silicon and germanium nanoparticles (LDRD projects 02-ERD-043 and 03-SI-001).

- **MSA Fellow.** Frederick J. Ryerson's work in petrology, geochemistry, and tectonics led to his election as a fellow of the Mineralogical Society of America in 2005. This work has received considerable support from LDRD over the years (e.g., projects 97-ERI-003, 00-ERI-009, and 03-ERI-001).

- **Presidential Early Career Award.** Two recipients of the 2004 Presidential Early Career Awards for Scientists and Engineers received LDRD support while working at LLNL as Ernest O. Lawrence fellows:
  - Wei Cai—For work done at LLNL (LDRD project 03-LW-027).
  - Joel Ullom—For work begun at Livermore under LDRD and continued at the National Institute of Standards and Technology (LDRD project 01-LW-054).
- **R&D 100 Awards.** In 2005, Laboratory technologies won four R&D 100 Awards from *R&D Magazine*. Of these, LDRD support directly contributed to two awards:
  - Biological Aerosol Mass Spectrometry Systems (BAMS) (LDRD projects 02-ERD-002 and 05-ERD-053).
  - NanoFoil® (LDRD project 98-ERD-044).
- **SAE Arch T. Colwell Merit Award.** Scientist William Pitz was one of two LLNL employees who were part of a team that received the Society of Automotive Engineers 2003 Arch T. Colwell Merit Award for a paper on technology for cleaner-burning diesel engines. William's work received LDRD funding (LDRD project 03-FS-020).
- **Top 100 Science Stories.** In 2005, *Discover* magazine rated two LLNL accomplishments among its list of the top 100 science stories of the year. Both projects received LDRD support:
  - Discovery of new superheavy elements 113 and 115 (LDRD project 04-ERD-085).
  - Development of active neutron interrogation techniques to scan cargo containers for hidden weapons of mass destruction (projects 02-ERD-064, 04-ERD-080, and 04-ERD-042).
- **Weapons Recognition of Excellence Award.** A team of Livermore scientists received the NNSA's Weapons Recognition of Excellence Award in recognition for their success in planning and commissioning the Joint Actinide Physics Experimental Research (JASPER) facility. One team member received LDRD support for work relevant to JASPER (project 01-ERD-098).

In addition, the contributions of LLNL will be highlighted in a hands-on demonstration at the Boston Museum of Science of the Inductrack magnetically levitated train system. LDRD supported crucial early research on Inductrack (e.g., projects 94-FS-077 and 98-ERD-035).

Advanced Sensors and Instrumentation

Development of a Quantum-Limited Microwave Amplifier Using a Direct Current Superconducting Quantum Interference Device

S. Darin Kinion 02-ERD-071

Abstract

The goals of this project are to demonstrate the first quantum-limited microwave amplifier based on a direct current (dc) superconducting quantum interference device (dc-SQUID), and to combine this amplifier with a single electron transistor (SET) to ultimately achieve a quantum-limited electrometer. This project builds on a collaboration between LLNL and the University of California, Berkeley.

With a suitable spin filter, these devices will enable rapid, single-spin measurements, which are necessary for basic science research and quantum computing. An amplifier close to the standard quantum limit (SQL) would significantly improve the sensitivity of SETs being used by a number of groups around the world. A quantum-limited electrometer using a SQUID to read out a SET would be revolutionary in the field of quantum coherence experiments. We expect to reach the quantum limit through a combination of modeling device behavior and improving device geometry.

Mission Relevance

The National Security Agency invests a substantial amount in quantum computation and quantum information processing. Both the amplifier and the electrometer are vital components in practically all viable implementations of quantum computing. Research and development in quantum information processing is vital to LLNL's national security interests because it could lead to secure information communication networks.

FY05 Accomplishments and Results

The most significant accomplishment in FY05 was the measurement of the lowest noise temperature to date—within about 10% of the SQL. After a few follow-up measurements to confirm the results, this work will be published. There was significant progress made in the fabrication of the SQUID amplifiers, which completely solved earlier problems operating them below 1 K. With a reliable supply of devices, we were able to begin using them in real measurements, including the first radio-frequency single-electron transistor measurement in collaboration with the University of New South Wales.

Considerable progress was made towards understanding the coupling between the microstrip resonator and the SQUID loop. This model will be useful for extending the frequency coverage of these amplifiers past 3 to 4 GHz.

Advancing the Technology of Tabletop, Mesoscale Nondestructive Characterization

Harry E. Martz 03-SI-004

Abstract

This project will provide state-of-the-art, tabletop technologies for nondestructive characterization (NDC) data on the structure, geometry, and alignment of mescoscale (millimeter-sized) objects at micrometer resolution. The project will combine four imaging methods: x-ray microscopy, proton microradiography, gigahertz acoustic microscopy, and nuclear magnetic resonance to achieve the spatial resolution, penetration, contrast, field of view, and acquisition times needed for NDC of mesoscale objects. The project will combine physics and engineering modeling with experiments to achieve a design and modeling tool for future instruments. As-built, two-dimensional (2-D) and 3-D models will incorporate the characterization data to bridge the gap between simulation and experimental results.

The results of this project will form the basis for a new mesoscale NDC capability with micrometer resolution, high contrast (0.999), useful fields of view (1 to 3 mm) and rapid acquisition times (seconds) that will make future NDC instruments practical in fabrication environments and enable 2-D and 3-D as-built models for simulations.

Mission Relevance

Increased understanding of radiation-matter interactions for mesoscale-object NDC may ultimately be applied to high-energy-density and inertial confinement fusion targets in support of stockpile stewardship experiments on large fusion-class lasers. In addition, mesoscale-object NDC capabilities are applicable to explosives characterization, sensor technologies for counterterrorism, advanced fuel cells, and biological research to advance human health.

FY05 Accomplishments and Results

We have integrated phase effects into HADES, a ray-tracing code for simulating radiographic projections; validated simulation codes with empirical data; and determined that diffraction effects within the object are insignificant for x-ray data for energies of ≥ 8 keV, and with ~1 µm spatial resolution, for objects of sizes up to 10 mm. Our research showed that phase recovery techniques employing differential equation solvers are limited to cases of 90% transmission or better for simple geometric objects, are sensitive to clutter and small aberrations in the detector, and are not very useful for most of our data. The “multi-slice” phantom was fabricated to contain either 6-µm-diameter C fibers or 4-µm-diameter W wires.

Publications

Aufderheide, M. B. et al. (2005). *HADES, a code for simulating a variety of radiographic techniques*. UCRL-PROC-207617.

Martz, H. E. (2006). *Validation of radiographic simulation codes including x-ray phase effects for millimeter-size objects with micrometer structures*. UCRL-JRNL-218242-DRAFT.

Pivovaroff M. J., W. W. Nederbragt, and H. E. Martz. (2005). *Lessons learned on x-ray optics fabrication*. UCRL-TR-208400.

Ultrafast Radiation Detection by Modulation of an Optical Probe Beam

Mark E. Lowry 03-ERD-007

Abstract

We will develop a novel class of ionizing-radiation detector that outputs a modulated optical beam, is capable of subpicosecond temporal response, has good sensitivity (even for hard x rays), and is scalable to imaging arrays. The detector uses an optical probe beam to detect the change in optical index that occurs when x rays impinge on a semiconducting medium and efficiently produce electron-hole pairs. This detector will enable a variety of radiography and self-emission imaging with hard x-ray energies, as well as gamma and neutron detection important for experiments at future large-laser systems. The technology will meet requirements for x-ray detection with 100-fs temporal resolution at 8 to 24 keV for the Linac Coherent Light Source (LCLS).

We expect to demonstrate a single-pixel version of this detector that will allow x rays to efficiently enter from one side and the optical probe beam to enter from the other side. This single-pixel detector will then achieve high quantum efficiency (QE) while also having a fast temporal response. These results will enable arrays of pixels to be developed for high-QE, hard x-ray imaging with picosecond to sub-picosecond temporal resolution. Such an imaging technology will have multiple applications for future large-laser system diagnostics and fundamental physics experiments at the LCLS at the Stanford Synchrotron Radiation Laboratory.

Mission Relevance

This project supports LLNL's national- and energy-security missions by developing new radiation-detector technologies that will enable a variety of inertial-confinement fusion and high-energy-density physics experiments that are planned at existing and future large-laser systems. High-energy-density experiments are a central component of the Laboratory's Stockpile Stewardship Program.

FY05 Accomplishments and Results

Budget and infrastructure resource constraints forced us to reconsider our approach. Consultation with prospective customers made it clear that the most successful strategy would be to conduct research into developing high QE x-ray imaging, leading to the key goals of (1) demonstrating single x-ray QE detection per pixel and (2) demonstrating that the technology can be applied to imaging.

For the first goal, we implemented two rounds of cavity design, packaging, and system measurements. These systems were fielded at the Titan laser facility. Our results enable us to conclude that an x-ray imager with a pixel size of 4 × 4 µm will give an optical signal-to-noise ratio of approximately 20 for a single x-ray photon. For the second goal, we conducted our first imaging experiments. Follow-on analysis will be required to determine the extent to which these experiments were successful.

DNA Detection through Designed Apertures

Sonia E. Letant 03-ERD-013

Abstract

Our objective is to demonstrate a selective Coulter counter for microorganisms by using a rigid silicon aperture that mimics an ion channel. The project has three main steps: (1) designing and fabricating single apertures with a tunable diameter from the nanometer to the micrometer range on silicon platforms; (2) functionalizing the aperture with chemical probes to provide selectivity; and (3) measuring microorganism transport through the functionalized apertures by the current blockade technique at the single-organism detection level. The challenges are to design single apertures in silicon with tunable diameter, length, and shape and to control their chemical functionalization. This will be achieved by combining nanomachining and self-assembly.

Although efforts have focused on building a single nanopore on various rigid substrates, no one has addressed either of the two key issues: finding a fast, reliable, and inexpensive technique to prepare single nanoapertures, and controlling the chemical functionality in these confined apertures. Our new approach, using nanotechnology and self-assembly, accomplishes both and will transform synthetic nanopores into extremely selective sensors able to detect and identify single organisms in real time without requiring polymerase chain-reaction amplification.

Mission Relevance

This project supports LLNL’s national-security mission by developing a new class of biosensors able to identify microorganisms with single-organism sensitivity. This technology will enable fast and sensitive sensors to improve early response to terrorist or accidental biological contamination of air or water. There are potential applications of the technique in support of homeland security and counterterrorism, preventing the proliferation and use of biological weapons, as well as attribution.

FY05 Accomplishments and Results

We demonstrated the localization of chemical functionality at the entrance of single nanopores for the first time by using the controlled growth of an oxide ring. Nanopores were fabricated by focused ion-beam machining on silicon platforms, locally derivatized by ion-beam-assisted oxide deposition, and further functionalized with DNA probes via silane chemistry. Ionic current recorded through single nanopores at various stages of fabrication showed that the apertures can be locally functionalized with DNA probes. We demonstrated selectivity by measuring the transport of functionalized beads through the apertures.

Publications

Litant, S. E. (in press). "Electrochemically nano-structured silicon platforms for sensing applications." *Review Chapter: Handbook of electrochemical nanotechnology*. American Scientific Publishers, Stevenson Ranch, CA. UCRL-BOOK-215240.

Litant, S. E. (2005). *Functional nanostructured platforms for biological and chemical sensing applications*. Nanoscience and Bionanoscience Research Mtg., Berkeley, CA, June 20, 2005. UCRL-ABS-212110.

Litant, S. E., A. W. van Buuren, and L. J. Terminello. (2004). "Nanochannel arrays on silicon platforms by electrochemistry." *Nano Lett*. **4**(9), 1705. UCRL-JRNL-204780.

Nilsson, J. et al. (in press). "Localized functionalization of single nanopores." *Adv. Mater*. UCRL-JRNL-215327.

Nilsson, J. and S. E. Litant. (2005). "Single aperture functionalization for bio-organism sensing." 2005 Spring Materials Research Society Mtg., San Francisco, CA, Mar. 28–Apr. 1, 2005. UCRL-ABS-209313.

Nilsson, J. and S. E. Litant. (2004). Functionalized apertures for single bio-organism identification. 2004 Fall Materials Research Society Mtg. Boston, MA, Nov. 28–Dec. 3, 2004. UCRL-ABS-207152.

Microfluidic System for Solution Array-Based Bioassays

George M. Dougherty 03-ERD-024

Abstract

We are developing an instrument for performing multiplex bioassays in the field to detect biological warfare (BW) agents. Our integrated, reconfigurable system will perform user-specified assays based on nanobarcode (NBC) solution arrays. NBCs are rod-shaped particles approximately 250 nm in diameter and 5 μm long with metal stripes for identification. A polymer-based microfluidic system will

mix the NBCs with the sample and prepare the particles for optical readout. Various other solution array approaches are under development, but none yet employs them in a way that meets the counter-BW requirements for multiplexing, user reconfiguration, cost, portability, and ease of use. To guide development, we are performing fundamental studies of NBC behavior in microfluidic systems.

Based on our experience to date with the NBCs, the fluidic self-assembly methods we have proposed appear to provide a feasible route to development of a bioassay device that meets the above criteria and will be appropriate for field use by first responders. We are also perfecting techniques for conducting bioassays in the field and methods for using NBCs as covert tags for material identification and forensics. The basic science advancements from this project will contribute to the broader field of nanotechnology and nanoparticle science.

Mission Relevance

This project supports LLNL's national-security mission by developing technology to counter weapons of mass destruction. The instrument will be used to detect BW agents in the field and can be applied to surveillance, detection to protect, and treaty verification. The project also supports the Department of Homeland Security's strategic plan for bioinstrumentation and supports LLNL's missions in homeland security, counterterrorism, intelligence, and medical technology.

FY05 Accomplishments and Results

In FY05, we leveraged the technical achievements of the prior two years to build a demonstration prototype system that performs biodetection assays in the NBC format. The system uses a low-cost polymer microfluidic card mounted on a simple, off-the-shelf laboratory microscope. Magnetic particle extraction and concentration, reagent mixing, agitation, and washing are all performed in the card under computer control. At completion, the results are automatically read by image-processing software. We demonstrated that the performance of a model four-plex biothreat simulant assay panel based on NBCs is equal to that of state-of-the-art immunoassays. We also demonstrated the ability of the system to extract and identify covert tagging particles from explosive residue.

Publications

Dougherty, G. M. et al. (2005). *Automated nanorod particle-based immunoassay system*. 6th Biennial Tri-Laboratory Engineering Conf., Monterey, CA, Sept. 12–15, 2005. UCRL-PRES-215179.

Dougherty, G. M. et al. (2005). *Zeta potential behavior of functionalized metallic nanorod particles in aqueous solution*. Particles 2005: Surface Modification in Particle Technology, San Francisco, CA, Aug. 13–16, 2005. UCRL-POST-214487.

Pannu, S. et al. (2005). *A compact system for multiplex immunoassay using bio-functionalized optically coded nanorods*. Micro Total Analysis Systems, Boston, MA, Oct. 9–13, 2005. UCRL-POST-215647.

Rose, K. A., G. M. Dougherty, and J. G. Santiago. (2005). *Flow-based detection of bar coded particles*. Micro Total Analysis Systems, Boston, MA, Oct. 9–13, 2005. UCRL-POST-213249.

Tok, J. B.-H. et al. (2005). *Multiplex immunoassays using biofunctionalized nanorod particles and automated optical readout*. NIBIB–DOE Workshop Biomedical Applications of Nanotechnology, Bethesda, MD, Mar. 17–18, 2005. UCRL-POST-210282.

Tok, J. B.-H. et al. (2005). *Nanobarcode as an efficient platform for multiplex immunoassay*. Gordon Research Conference on Chemical and Biological Terrorism Defense. UCRL-POST-209154.

Carbon-Nanotube Permeable Membranes

Olgica Bakajin 03-ERD-050

Abstract

This project will measure the motion of molecules down the axis of a carbon nanotube (CNT) by embedding ordered nanotubes in a thin film to make a semipermeable membrane with ideal pores. We will observe diffusion both by permeation and direct observation of single fluorescent molecules. The motion of molecules through pores of molecular dimensions is a crucial problem in nanoscience. Diffusion down a nanotube has been frequently simulated but never measured, so these experiments will have a significant impact on the field and enhance our understanding of molecularly confined motion, which is critical for integrated sensors and separation systems.

This project will build a deeper understanding of the physics of permeation separation through fabrication of a series of media with precisely controlled pore geometry and surface chemistry. If successful, this work could benefit other areas (including both homeland security and Genomics:GtL) and provide an avenue to address various basic- and applied-science challenges. This could include applications such as micro gas chromatography (GC), microcapillary protein-separation, and microisotope purification.

Mission Relevance

The Laboratory's national-security mission would benefit from technology that enables development of more compact, highly integrated sensors. LLNL faces the challenge of interfacing a sensitive molecular detection and identification system to a complex environment. In addition, microseparations can enable analytical science at volumes consistent with single-cell analysis.

FY05 Accomplishments and Results

The team developed a method for fabricating a membrane consisting of vertically aligned, single-wall CNTs with a pore size of less than 2.0 nm. Gas measurements carried out on single- and multi-wall

CNT membranes reveal selectivity towards hydrocarbon species. The single-wall CNT membranes show an order of magnitude enhancement in gas permeability relative to predictions of Knudsen diffusion. An enhancement in water permeability of up to three orders of magnitude is seen versus no-slip, continuum hydrodynamic predictions. In the context of slip flow, the experimental values correspond to large slip lengths of up to several hundred nanometers. The membranes exhibit permeability that is two orders of magnitude higher than commercial polycarbonate membranes despite having an order of magnitude smaller pore size.

Publications

Holt, J. (2005). *Enhanced gas and water transport through a single-wall carbon nanotube membrane*. UCRL-JRNL-216252.

Space–Time Secure Communications for Hostile Environments

James V. Candy 03-LW-005

Abstract

Communicating in complex environments, such as a hostile urban setting populated with a multitude of buildings and vehicles or a maze of military/civilian tunnels, is difficult. Here we develop multichannel time-reversal (T/R) techniques to communicate in a highly reverberative environment. We focus on two major objectives: (1) wideband acoustic communications using time-reference modulation and (2) multichannel communications in a tunnel (or cave or pipe) with many obstructions, multipath returns, background noise, disturbances, and long propagation paths with disruptions (bends). Information signals are transmitted in air with an eight-element array to two receivers, demonstrating that T/R processing can extract the transmitted code sequence with zero-bit error.

We expect to develop a T/R receiver that will enable communications in hostile environments such as caves and for critical applications such as water pipes, nuclear plant systems, and border-protection systems. We have shown that multichannel T/R receivers can be extended to the wideband designs, while demonstrating their performance in both the canonical stairwell of our previous work, as well as a tunnel-like structure. Results of the new wideband T/R processor and modulation scheme demonstrate the overall performance of both high- (24-bit) and low- (1-bit) bit analog/digital (A/D) converter designs. We validated these results by performing proof-of-principle acoustic communications experiments in air that demonstrated the T/R receiver capability of achieving zero-bit error.

Mission Relevance

The T/R receiver will greatly benefit national-security and military applications. Channel reliability in a hostile environment is a prime concern of security forces, especially in counterterrorism activities. The

T/R receiver technology also promotes protection of communication channels against intercept and thus supports LLNL's national-security mission in the areas of defense, homeland security, and intelligence.

FY05 Accomplishments and Results

In FY05 we exceeded our target milestones and (1) developed a multichannel theory for T/R communications; (2) implemented a new wideband T/R modulation scheme, executed the design and analysis, and demonstrated system performance through simulation and experiments; (3) performed controlled acoustic experiments using an array in a reverberative stairwell, a tunnel-like structure, and with electromagnetics in a hallway with multiple scatterers; (4) demonstrated T/R performance using 1-bit A/D conversion to validate performance in a stairwell and tunnel-like structure; (5) investigated an experimental design for electromagnetic hardware; (6) demonstrated T/R performance in a tunnel-like structure for a variety of LLNL personnel and potential sponsors; and (7) initiated hardware design for field-programmable gate array technology.

Publications

Candy, J. et al. (2005). "Multichannel time-reversal processing for acoustic communications in a highly reverberant environment." *J. Acous. Soc. Am.* **118**, 2339. UCRL-MI-215056.

Candy, J. et al. (2005). "Wideband multichannel time-reversal communications in a tunnel-like structure." *J. Acous. Soc. Am.* **118**, 2039. UCRL-ABS-213418.

D. Chambers (2005). "Eigenvalues of the time-reversal operator for a small ellipsoid." *J. Acous. Soc. Am.* **118**, 2039. UCRL-ABS-212815.

A Novel Antimatter Detector with Application to Dark Matter Searches

William W. Craig 03-LW-059

Abstract

We propose to demonstrate an entirely new technique, the gaseous antiparticle spectrometer (GAPS), for detecting antimatter in cosmic rays. The concept allows an orders-of-magnitude increase in searches for these elusive particles. The sensitivity of our technique has an immediate and significant impact on the detection of the mysterious dark matter component of the Universe, and could answer one of the most pressing questions of modern cosmology: what is the composition of the vast majority of gravitating mass in our Universe? In the third year of our program we have conducted antiproton beam testing to validate the physics and efficiency of the completed GAPS prototype instrument.

The GAPS approach has now been shown to provide an entirely new and very effective technique for detecting antimatter in cosmic rays—one of the key questions of cosmology. As such, it has produced novel and important results being prepared for publication, as a direct result of beamline testing.

Mission Relevance

The technologies in the GAPS prototype instrument will be directly applicable to national-security programs. The data-acquisition system developed for the prototype is being used by a large coded-aperture instrument under development for nuclear-material search programs. The GAPS approach is also being considered for use in detection of special nuclear materials in cargo containers. As a high-profile scientific achievement, GAPS will help recruit talented scientists interested in detector concept development.

FY05 Accomplishments and Results

We completed characterization of the GAPS prototype-instrument performance using the KEK accelerator facility in Japan. We obtained additional beamline data and are in the process of performing data analysis to determine absolute efficiency and background rejection capability. We can now derive the energy and time resolution requirements for space and balloon-borne implementations of the GAPS concept. Because beamline testing validated our calculations, we can now devise a plan, using follow-on funding from NASA, for a balloon-borne instrument to detect the low-energy antiproton component in the cosmic ray flux. The beamline results from FY04 and FY05 will be prepared for publication—at least two significant publications are expected.

Ultrafast Transient Recording Enhancements for Optical-Streak

Cory V. Bennett 04-ERD-025

Abstract

Several diagnostics at future large laser systems and advanced light sources such as the Linac Coherent Light Source, will require hard x-ray measurements with temporal resolution of about 1 ps or less and a high dynamic range, far beyond existing solutions. We are investigating ultrafast optical recorder technologies to develop a rugged, fiber-based temporal imaging system for optical-streak cameras. This system will (1) improve resolution and dynamic range by "stretching" an input signal in time, allowing ultrafast signal recording with slower, higher-dynamic-range instruments; and (2) convert light from 1550 to 775 nm, allowing streak cameras to record signals from detectors using 1550-nm technology.

We expect to demonstrate single-shot operation of a robust temporal imaging system and streak camera recorder that will have less than 300-fs resolution and a greater dynamic range (greater than 100,

and as high as 1000 at a slower magnified resolution) and be compatible with current and future hard x-ray detectors to produce a modulated optical carrier at 1550 nm. In addition, we expect to develop a complete ultrafast imaging system plan for implementation on large laser systems.

Mission Relevance

Our goal is to ensure delivery of the next-generation diagnostics needed for critical experiments at current and future large laser systems, which is an important improvement for stockpile stewardship applications. These diagnostics will enable high-energy-density physics and inertial-confinement fusion experiments, including measurement of reaction history, dynamic holhraum temperature, and dynamic opacity, as well as detecting sub-picosecond backscatter bursts.

FY05 Accomplishments and Results

In FY05, we (1) analyzed and compared multiple nonlinear crystal designs; (2) measured the input and pump dispersive-delay-line components and implemented aberration corrections; (3) implemented the time–lens pump pulse system and modified the laser and pulse-picking subsystems to attain the desired 320-nJ pulse, chirp rate, and pulse duration; (4) in collaboration with the University of California at Los Angeles, demonstrated a new source that should allow temporal imaging systems to be triggered off the event instead of relying on mode-locked lasers, which must be synchronized a priori to the expected arrival time. A publication on this work is in progress.

Proposed Work for FY06

In FY06, we will (1) complete a fully characterized compact temporal imaging system, which will be integrated with a streak camera and characterized (resolution, record length, dynamic range, jitter, etc.); (2) analyze technologies that can be produced in compact arrays for spatial imaging applications, as we have done throughout the project; (3) demonstrate a low-channel-count system and develop a complete ultrafast imaging system plan; and (4) prepare a number of publications on the design and performance of this fiber-based, parametric temporal-imaging system.

Publications

C. V. Bennett. (20005). *Temporal imaging: ultrafast optical waveform manipulation and recording*. UCRL-PRES-212869.

An Integrated Laboratory for the Study of Interventional Device Dynamics

Duncan J. Maitland 04-ERD-093

Abstract

We propose to develop an integrated laboratory for investigating the physics and device dynamics of endovascular interventional devices. This project will bring together four research components: novel endovascular devices, particle image velocimetry (PIV), computational fluid dynamic (CFD) models, and a core ability to generate physical and CFD models from actual human anatomies. We will apply the experimental and computational tools to a novel medical application: shape-memory polymer (SMP) foam for treating aneurysms.

If successful, the proposed research will improve the medical scientific community's understanding of endovascular interventional devices, enable novel device development, and assess impact on vascular fluid dynamics. We expect that this research at LLNL has the potential to become an internationally accepted method to perform endovascular device research and development.

Mission Relevance

This project has direct relevance to the Laboratory's mission in biotechnology to improve human health care. In addition, the proposed system would enable direct analysis and subsequent design of fluidic systems used in devices for chemical and biological detection that are under development for the Laboratory's national security and homeland security missions.

FY05 Accomplishments and Results

Detailed CFD and PIV studies on the SMP foam device were run and a prototype stent was created. The simulations and experiments are showing that a single foam device is likely to stop flow in an aneurysm, which is the desired clinical outcome. The technical results have been presented in five invited talks, three conference presentations (with proceedings), three publications in preparation, and one patent application. The foam device is currently under consideration for licensing by two medical device companies.

Proposed Work for FY06

Research in FY06 will focus on three areas: (1) adding flow nuclear-magnetic-resonance measurements to the foam device to compare with CFD and PIV measurements; (2) performing CFD simulations that include energy transport and device shape change; and (3) conducting experiments and simulations with the stent.

Publications

Small IV, W. et al. (2005). *Laser-activated shape memory polymer thrombus retrieval device for ischemic stroke treatment*. Engineering Intl. Conf. Advances in Optics for Biotechnology, Medicine and Surgery, Copper Mountain, CO, July 24–28, 2005. UCRL-POST-212919.

Small IV, W. et al. (2005). *Laser-activated shape memory polymer thrombus retrieval device for ischemic stroke treatment*. UC System-Wide Bioengineering Symp., Santa Cruz, CA, June 25–27, 2005. UCRL-POST-212919.

Tsai, W. et al. (2005). *Vascular dynamics of laser activated SMP devices*. UC System-Wide Bioengineering Symp., Santa Cruz, CA, June 25–27, 2005. UCRL-POST-212540.

Wilson, T. S. et al. (2005). "Shape memory polymer therapeutic devices for stroke." *Proc. SPIE* 6007, 60070Q. UCRL-PROC-216091.

Microfluidic Liquid Cell for Molecular Imaging in Aqueous Phase Using Atomic Force Microscopy

Todd A. Sulchek 04-FS-034

Abstract

The goal of this project is to develop a microfluidic liquid cell for in situ molecular imaging in aqueous solutions using atomic force microscopy (AFM). Achieving this goal will significantly advance the capabilities in probing protein–protein interactions and protein complex formation, exploring the physics and chemistry of biomolecular materials synthesis, and understanding the architecture of pathogens. We will accomplish this goal by fabricating a microfluid-compatible liquid cell and applying the liquid cell to crystal growth studies.

First, we expect to create an AFM liquid cell with greatly expanded capabilities. By reducing the volume contained, we enable in situ AFM experiments for a broad range of proteins. We seek to demonstrate this capability with the effect on calcium oxylate of osteopontin, a major influence of kidney stone formation. Second, the liquid cell will enable imaging at physiological conditions, including both elevated temperatures and precisely controlled solution environments. Third, this small-volume technique will enable cantilever-based sensing experiments by maintaining high concentrations of the biological or chemical signal of interest.

Mission Relevance

The success of this project will significantly advance capabilities in probing protein–protein interactions and protein complex formation, exploring the physics and chemistry of biomolecular materials synthesis, and understanding the architecture of pathogens. Probing the interactions of proteins complexes with their local environment is particularly relevant to DOE's Genomics:GtL initiative.

FY05 Accomplishments and Results

In FY05, we successfully completed a design that integrates the microfluidic channel and allows imaging of the surface. A defect in our original design that prevented the channel from sealing tightly was corrected by changing the method of fabrication. We used the new design to show force curves and to image a surface—both critical demonstrations of AFM capability—and demonstrated imaging in liquid. We filed a patent application for this work and began writing a publication on the demonstration of protein imaging in this microfluidic environment.

Developing Radiography for Advanced Radiography Capability at Future Large Fusion-Class Lasers

Hye-Sook Park 05-ERD-006

Abstract

Advanced radiography capability (ARC) is a multikilojoule, 10-ps petawatt laser under study for use in stockpile stewardship experiments at future large fusion-class lasers. Such experiments often require backlighters with an energy >20 keV and spatial resolution <10 μm. To enable such backlighters and make optimal use of ARC in these experiments, we propose to investigate the physics of high-intensity laser interactions and consequent particle and radiation transport in a small, confined volume, and develop (1) volume-localized radiation targets that reduce source size and enhance brightness; (2) high-energy, high-resolution, two-dimensional (2-D) radiography techniques; and (3) suitable x-ray detectors for these high energies.

This work will produce a petawatt radiography diagnostic that will significantly enhance our knowledge of high-intensity laser coupling efficiencies and electron transport for short-pulse lasers, benefiting the development of reliable simulation models. We will also develop a set of hard x-ray diagnostic techniques applicable to very-high-resolution x-ray radiography.

Mission Relevance

This project supports the national security mission by providing optimal radiography techniques for stockpile stewardship experiments, such as material strength experiments, high-energy-density experiments with mid- to high-*Z* capsules, and double-shell ignition experiments. Most such experiments require 2-D K-alpha backlighters in the energy range of 20 keV to 1 MeV.

FY05 Accomplishments and Results

In FY05, we (1) verified 1-D radiography at 40 keV using an edge-on foil; (2) designed and fabricated a cone-fiber and other embedded wire targets as the 2-D radiography source; (3) tested their performance by measuring their source size and brightness using the Vulcan petawatt laser; (4) designed and fabricated a multilayer mirror and utilized it as a spectrometer in the 30- to 70-keV bands; and (5) utilized implicit hybrid particle-in-cell simulation to understand electron transport and K-alpha photon generation in the cone-fiber target.

Proposed Work for FY06

Because preliminary data showed that the small wire targets are not bright enough for 2-D radiography, we will continue to study options for 2-D radiography that will give us <10-μm spatial resolution. Specifically, we will (1) enhance K-alpha yield efficiency in a small-volume target through better target design and fabrication; (2) test contact radiography in a laser experiment in which the large source size is compensated for by the proximity of the imaging screen to the target; (3) study and, if promising, test multilayer hard x-ray imaging optics to cover a large solid angle and eliminate unwanted energy bands; (4) continue utilizing integrated simulation capability to optimize target designs; and (5) continue to characterize hard x-ray detectors.

Publications

Park, H. S. et al. (2005). "Characteristics of high-energy Ka and Bremsstrahlung sources generated by short-pulse petawatt lasers." *Rev. Sci. Inst.* **75**, 4048. UCRL-CONF-203568.

Park, H. S. et al. (2005). "High-energy K-alpha radiography using high-energy, high-intensity, short-pulse lasers." 47th Ann. Mtg. American Physical Society Division of Plasma Physics. UCRL-ABS-213875.

Park, H. S. et al. (in press). "40 keV 1-D radiography using short-pulse high-intensity lasers." *Rev. Sci. Inst.* UCRL-PRES-209831.

Park, H. S. et al. (in press). "High-energy K-alpha radiography using high-energy, high-intensity, short-pulse lasers." *Phys. Plasma.* UCRL-JRNL-217367.

Park, H. S. et al. (in press). "K-alpha radiography at 20-100 keV using short-pulse lasers." *Proc. IFSA Conf.* UCRL-PRES-215011.

Town, R. et al. (in press). "LSP calculations of cone-wire experiments." *Proc. IFSA Conf.* UCRL-ABS-209626.

Wickersham, J., et al. (2005). "Imaging detectors for 20-100 keV x-ray backlighters in HEDES petawatt experiments." *Rev. Sci. Inst.* **75**, 4051. UCRL-CONF-203625.

Remote Sensing of Alpha and Beta Particle Sources

Nerine J. Cherepy 05-ERD-032

Abstract

We propose to develop a model of air ionization chemistry providing speciation and kinetics resulting from interactions with high-energy particles and radiation, and a passive remote-sensing capability, based on the luminescence of air and its radiolysis products, to detect small dispersed alpha and beta emitting sources. We will focus on the nitrogen oxide (NO) luminescence in the "solar blind" (UVc, <300 nm) region, which offers advantages of very low background ambient solar light, thus making possible low-noise, prompt detectability in full daylight. This technology would be relatively simple and usable by emergency response personnel at nuclear release incidents to aid in rapid location and containment of dispersed radiological materials, and later to guide decontamination efforts.

We expect to build a prototype viewer and demonstrate a standoff imaging capability for alpha- and beta-particle-emitting sources. If successful, this would be a completely new detection methodology for radiological materials, allowing location and imaging at a safe working distance.

Mission Relevance

LLNL currently provides support to Federal emergency responders who deal with nuclear emergencies, nuclear accidents, and nuclear terrorism. The technology we wish to develop would provide a useful tool for these applications in support of the Laboratory's homeland security mission. Application of this technology to decontamination will support the Laboratory's dedication to responsible environmental management, facilitating remediation of legacy materials handling buildings, as well as ongoing needs for assessment and cleanup.

FY05 Accomplishments and Results

In FY05 we (1) performed Monte Carlo simulations to map the energy deposition in air from a ^{210}Po source and other beta/gamma and alpha/gamma sources; (2) built a tropospheric chemistry model that predicted steady-state radiolytic speciation (e.g., NO and O_3) as a function of source strength and distance from source, using Monte Carlo model input; (3) imaged a ^{210}Po alpha source (0.5 μCi) using ultraviolet (UV) airglow, measured its airglow radius, quantified its spectrum and intensities in the 200 to 500 nm range; (4) developed and evaluated scintillating barium fluoride (BaF_2) coatings with light yields equal to single-crystal BaF_2 (600× the air signal in the "solar blind"); and (5) began work on lutetium aluminum garnet UV scintillating coatings (which enable a 15,000× signal enhancement over airglow).

Proposed Work for FY06

We will (1) image airglow from a beta source, (2) continue development of scintillating coatings, (3) complete the design and fabrication of an optimized UV viewer, and (4) demonstrate detection using airglow as well as scintillating coatings indoors and outdoors.

Publications

Cherepy, N. et al. (2005). *New technologies for standoff assessment of radiological contamination*. IEEE Nuclear Science Symp. 2005, Puerto Rico, Oct. 23–29, 2005. UCRL-PROC-212232.

Homodyne Imaging Vibrometry Experiment (HIVE)

Liesl M. Little 05-ERD-033

Abstract

We will build a homodyne imaging vibrometer using a 64- × 64-pixel focal-plane array. Current imaging vibrometers are based on heterodyne detection, where the required high-frequency sampling limits the image size to a few hundred pixels. With homodyne detection, the sampling rate can be up to a few kilohertz, allowing the use of commercial arrays with thousands of pixels. Larger arrays make vibration-signature detection more likely and may also provide information on building structure.

We expect to demonstrate the feasibility of measuring a vibrating surface with a homodyne approach. By enabling a viable image size, the homodyne approach could lead to a breakthrough in vibrometry imaging. This project will implement a model for predicting system performance, build a prototype that can be fielded, and make a direct measurement comparison with a heterodyne imaging vibrometer.

Mission Relevance

Lawrence Livermore develops remote-sensing technologies to stem the spread of chemical, biological, and nuclear weapons, as part of its national security mission. Homodyne vibrometry offers the potential for new capabilities in locating sources, determining structural properties, and identifying denied activities. This system also has applications for defense missions, including locating land mines.

FY05 Accomplishments and Results

A computer model was developed to predict system performance. Initial conclusions from the modeling included calculation of a noise-limited sensitivity, calculation of optimal beam shape for the local oscillator, and evaluation of analysis techniques for data reduction. An initial design for a proof-of-concept experiment was implemented in the lab, and a calibrated, multi-signature scattering target was designed and built. The local oscillator was shown to be shot-noise limited. Lab vibration measurements showed correct displacement values at the expected frequencies. Intensity images also appeared to be correct, but phase images showed unexpected phase distributions, including spatial symmetry, and the signal-to-noise ratio appeared higher for single channel demodulation than for quadrature demodulation.

Proposed Work for FY06

The most pressing goal for FY06 is to determine why the phase images do not have expected distributions. Proposed causes, including channel misalignment, will be modeled to evaluate their effects. Experimental goals include a direct laboratory comparison between the homodyne and heterodyne systems, and an on-site field demonstration to enable testing under realistic turbulence and speckle conditions with realistic targets. This will require building a prototype system that can be fielded. The greatest technical challenges involve size reduction of the polarization-separation optics and development of analysis software capable of handling information from thousands of pixels in quasi real-time and displaying it in a comprehensible form.

A Multiplexed Diagnostic Platform for Point-of-Care Pathogen Detection

Mary T. McBride 05-ERD-049

Abstract

The goal of this project is to develop a practical, fully validated diagnostic tool with multiplexed nucleic acid assays that can be used to simultaneously detect and identify multiple respiratory pathogens and to distinguish pathogens that cause common respiratory infections from biothreat agents. We propose to (1) develop a multiplexed polymerase (PCR) assay panel for simultaneous detection of respiratory pathogens; (2) optimize assay performance in complex sample matrices; (3) validate the multiplexed PCR assays and diagnostic platform; and (4) demonstrate an autonomous, rapid bedside diagnostic device that can process and analyze a patient sample and post a result in less than an hour.

This work will result in a prototype of a rapid, practical, and fully validated diagnostic tool with multiplexed nucleic acid assays for pathogen detection in biowarfare or bioterrorism scenarios, for civilian preparedness, and for public health. This point-of-care diagnostic tool will find application in state and local public health laboratories and hospitals, clinics, and other health-related institutions. The rapid bedside diagnostic capability of this instrument has the potential for improving patient management.

Mission Relevance

A biologically based, deployable pathogen-detection instrument has numerous potential biodefense applications for LLNL's homeland security and national security missions and supports the Laboratory's mission in biotechnology to improve human health. It also enables applications that may support Centers for Disease Control missions in public health.

FY05 Accomplishments and Results

In FY05, in conjunction with emergency room doctors at the University of California (UC) Davis Medical Center, we selected pathogens to use in our panel based on prevalence in the emergency room. We then developed nucleic acid signatures that were unique and specific for each of these pathogens. These signatures were screened against over 2500 confounder and/or interferrent signatures, to ensure a low incidence of false-positives. Changes in PCR kits and buffer concentrations allowed optimization of the assay so that influenza A and influenza B are now multiplexed. We spiked negative throat wash samples from humans, and the assays gave excellent results. We finished construction of the autonomous diagnostic device (FluIDx) and are currently testing it with a variety of well-characterized assays. In addition, records of invention were filed for our pathogen detection tool and multiplexed assays.

Proposed Work for FY06

The FY06 project activities will focus on continuing to multiplex the assay; adding signatures for respiratory syncytial virus, parainfluenza 1 and 2, and adenovirus; and developing signatures and an assay for avian influenza. We will optimize this assay so all signatures work well together, and we will validate the multiplexed assay panel by conducting a multicenter validation. Finally, fieldwork will include installing our instrument at the UC Davis Medical Center, gathering data to determine false-positive rates, and receiving feedback from users for instrument enhancement.

Rapid Screening of Human Effluents with Single Particle Mass Spectrometry for Early Detection of Respiratory Disease and Cancer

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Matthias Frank 05-ERD-053

Abstract

This project will adapt single-cell bioaerosol mass spectrometry (BAMS) to real-time analysis of human effluents, such as exhaled aerosols, and screening of those effluents for pathogens. The BAMS technique can analyze the biochemical composition of aerosol particles and single cells in real time and may be used to detect pathogens or cancerous cells. Research will begin with analyzing exhaled aerosol particles and droplets, then expand to include other human effluents, such as urine, for pathogens or cancerous cells.

If successful, the project will demonstrate that the BAMS technique can rapidly analyze human effluents and can be used in the early detection of respiratory diseases and some cancers. This will advance the biomedical diagnostics capabilities for respiratory diseases, the analysis of biological aerosols, and

the study of biochemical processes at the cellular level. This project also provides new avenues for strong collaborations with University of California (UC) campuses, (e.g., at the UC Davis Cancer Center and UC Berkeley) for emergency response to a bioterrorist attack.

Mission Relevance

This project will provide new capabilities for early detection of respiratory disease and cancer. Applications include biological weapons detection, population screening, and incident-response capabilities in support of the Laboratory's national-security and homeland-security missions. Fundamental biomedicine and public health applications also support the Laboratory's mission in biotechnology.

FY05 Accomplishments and Results

Using protein standards in the 1- to 10-kDa range, we began quantifying the sensitivity limits of our single-mass spectrometry method and achieved a new world record for matrix-assisted laser desorption/ ionization mass spectrometry (MALDI-MS) analysis of proteins: 14 zeptomoles (i.e., ~8500 molecules) of gramicidin S, a ~1-kDa peptide. We developed a method to matrix-coat single-cell aerosol particles that will allow us to perform single-cell MALDI-MS analysis. We built a new, efficient particle inlet for BAMS that is based on an aerodynamic lens focusing particles into a tightly packed beam. This new inlet allows us to introduce and focus larger particles and cells (including eukaryotic cells up to ~10 µm) into our BAMS system.

Proposed Work for FY06

In FY06, we will continue our efforts to show that unique mass signatures can be obtained with BAMS from certain respiratory pathogens and their marker molecules, demonstrating that crucial signatures are preserved when intact organisms in aerosol form are analyzed by BAMS on a single-cell level. We then plan to demonstrate the feasibility of detecting respiratory pathogens in particles in exhaled breath, breath condensate, or other complex clinical samples. In particular, the challenge of detecting characteristic marker molecules from pathogens embedded in a complex matrix present in exhaled human breath, nasal swabs, or sputum will be addressed. We will also expect to measure biomarkers from well-characterized cancer cell lines provided by our collaborators at the UC Davis Cancer Center.

Publications

Czerwieniec, G. A. et al. (2005). "Improved sensitivity and mass range in time-of-flight bio-aerosol mass spectrometry utilizing an electrostatic ion guide." *J. Amer. Soc. Mass. Spectrom.* **16**, 1866.UCRL-JRNL-207600.

Czerwieniec, G. A. et al. (2005). "Stable isotope labeling of entire bacillus atrophaeus spores and vegetative cells using bioaerosol mass spectrometry." *Anal. Chem.* 77, 1081. UCRL-JRNL-207205.

Steele, P. T. et al. (2005). "Desorption/ionization fluence thresholds and improved mass spectral consistency measured using a flattop laser profile in the bioaerosol mass spectrometry of single bacillus endospores." *Anal. Chem.* 77, 7448. UCRL-JRNL-208501.

Probing Other Solar Systems with Current and Future Adaptive Optics

Bruce A. Macintosh 05-ERD-055

Abstract

Over the past decade, Doppler techniques have allowed astronomers to discover more than a hundred planets orbiting nearby stars—discoveries of great scientific and public interest. The next step will be direct detection and characterization of extrasolar planets. Detection of a small number of such planets is barely within the reach of current adaptive optics (AO) systems. Development of a planet-hunting, next-generation, dedicated high-contrast "extreme" AO system (ExAO) is needed to probe the environments of other stars on scales comparable to our solar system. This project will use existing AO to probe dust and possible planets in nearby solar systems and develop the precision optical technology needed for an LLNL-led future ExAO system for the Gemini Observatory.

This project will carry out a search for young extrasolar planets orbiting nearby stars, using advanced image-processing techniques to separate planets from background noise. Direct detection of a planet orbiting a nearby star would be a major scientific achievement, opening new windows into the formation and nature of solar systems. We expect that LLNL will lead construction of an advanced next-generation AO system for the Gemini Observatory that would be capable of detecting planets orbiting a large sample of sun-like stars. Technologies developed for this project will be key to a wide range of future AO work in remote sensing, laser beam control, and biomedical applications.

Mission Relevance

Development of ExAO techniques, such as wavefront characterization and correction, will provide LLNL with a reservoir of skills and techniques for AO that can be applied to large optical systems and space optics that will support national security missions in remote sensing for counter- and non-proliferation missions. Similar techniques will also be used to control the wavefront in high-energy laser systems—a wavefront sensor concept developed for ExAO is now being used in Livermore's solid-state heat-capacity laser. This project also supports LLNL's mission in breakthrough science and technology.

FY05 Accomplishments and Results

Using Keck Observatory AO, we observed a sample of young, dusty stars to search for planet-forming dust and possible young planets, and developed new strategies such as temporal filtering of data that enhance sensitivity by factors from 2 to 10, to detect faint companions and circumstellar dust. At the University of California at Santa Cruz, we commissioned an ExAO testbed combining less than 1-nm internal wavefront errors, a 1024-actuator microelectromechanical system deformable mirror, and LLNL's phase-shifting diffraction interferometer. We developed algorithms to efficiently control multi-thousand-actuator AO systems and adapted existing software to carry out basic optical-propagation modeling for future ExAO systems. An LLNL-led team has now been selected by the Gemini Observatory to construct a next-generation ExAO system.

Proposed Work for FY06

In FY06, we plan to (1) observe 10 to 20 young, dusty stars with Keck AO and other AO systems such as Gemini; (2) carry out follow-up observations of targets from previous years and candidate exoplanets discovered by other groups; (3) extend our optimal wavefront reconstructor with atmosphere prediction or contrast optimization; (4) implement and test reconstruction algorithms on an ExAO testbed with simulated aberrations; (5) specify a second-generation ExAO testbed; and (6) extend our optical-propagation software to complex systems.

Publications

Farihi, J., E. E. Becklin, and B. A. Macintosh. (2004). "Mid infrared observations of van Maaen 2: No substellar companion." *Astrophys. J.* **608**, L109. UCRL-JRNL-207732.

Guenther, E. W. et al. (2005). "Low-mass companions to Hyades stars," *Astron. Astrophys.* **442**(3), 1031. UCRL-JRNL-218730.

Macintosh, B. et al. (2005). "Speckle lifetime in high-contrast adaptive optics." *Proc. SPIE* **5903**, 170. UCRL-CONF-214737.

Poyneer, L. A. and J. -P. Viran. (2005). "Optimal modal Fourier transform wave-front control." *J. Opt. Soc. Am. A* **22**, 1515. UCRL-JRNL-207495.

RadTracker: Optical Imaging of High-Energy Radiation Tracks

Mark E. Lowry 05-ERD-058

Abstract

This project proposes to develop a solid-state, uncooled radiation detector for high-energy radiation particles with a novel optical imaging approach: using ionizing radiation to generate electron-hole pairs, which in turn locally perturb the index of refraction, creating an optical-phase object track of the radiation. This can be sensed with an optical probe beam. Detector features will include (1) detection of x-rays, gammas, neutrons, and charged particles; (2) large operational energy bandwidth from 100 keV to >20 MeV; (3) high quantum efficiency of over 50% to enable longer standoff; (4) high energy resolution (>5%); (5) count rates in the megahertz range; and (6) directionality of radiation source without collimation (e.g., through electron-tracking Compton imaging for gammas).

Efficient radiation detection schemes for high-energy particles require large volumes of material to provide long particle interaction lengths. The standard radiation detection approaches requiring charge transport and collection are often degraded in performance due to charge transport limitations over

the large transport distances associated with these large volumes. We will expect to resolve many of the critical technical questions surrounding optical imaging of high-energy radiation tracks for very large detector volumes without the performance degradation of existing approaches. If successful, this research will lead to greatly improved gamma imager performance.

Mission Relevance

Radiation detection is fundamental to several major DOE and LLNL missions. Novel, large-volume, high-performance radiation detectors can address large standoff radiation detection requirements for homeland security, as well as prompt diagnostic requirements for weapons physics and experiments planned for large fusion-class lasers to support stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we worked on experimentally optimizing the radiation-induced signal-to-noise ratio (SNR) by designing and implementing a dark-field optical system as well as a phase contrast imaging system. To facilitate system design, we developed an accurate physical model for radiation optical-phase objects. In addition, we measured the background scattered light in Ga-As samples and began measuring the scattered optical power from real radiation tracks using very sensitive lock-in detection techniques. Our first radiation experiments employed a flash x-ray source. Because the background electromagnetic pulse levels from this source made it difficult to detect our radiation optical signal, we switched to a chopped gamma source for our optical setup. The chopped gamma system came on line at year's end.

Proposed Work for FY06

In FY06 we will (1) make fundamental measurements of the characteristics (i.e., angular distribution and temporal characteristics) of the light scattered from radiation optical-phase objects to enable refined optical designs and (2) enhance the SNR of the radiation optical signal by pursuing new materials with fundamentally strong radiation optical-phase shifts and developing proof-of-principal optical systems that enhance the SNR of the radiation optical images through planar waveguiding and resonant enhancement via cavities or rings. We expect the results of this work to enable a gamma camera based upon the radiation optical effect.

Amplifier and Compressor Technology for Split-Beam, High-Energy Short Pulse Generation

Raymond J. Beach 05-ERD-062

Abstract

The purpose of this project is to explore and develop the technologies required to create an integrated system for the injection, setup, compression, and monitoring of high-energy, split-beam, short pulse generation from a single aperture of a Nd:glass laser system. Production of such short pulses will require new amplifier and pulse compressor technologies that meet the stringent operating requirements and architectural constraints of large-scale amplification systems and that are capable of producing kilojoule-class, picosecond pulses in a vacuum. The concepts of split-beam amplification and compact, mixed-grating pulse compressors offer near-term technical pathways to such laser systems.

If successful, this project will develop new techniques or methods for (1) multiple, sub-aperture pulse generation, timing, and pulse-width control; (2) the precision pointing and characterization of injected, split-beam pulses; (3) the precision alignment of a folded, mixed-grating pulse compressor; and (4) the precision alignment of split-beam pulse compressors. In addition, we will evaluate the crosstalk between split-beam sub-apertures in an amplifier. This research will produce important publications and intellectual property.

Mission Relevance

Recent major advances in short-pulse laser technology have opened up opportunities to deploy powerful new radiographic diagnostics using both high-energy photons and laser-accelerated protons, which would further the LLNL mission in stockpile stewardship.

FY05 Accomplishments and Results

Detailed requirements and specifications were generated for split-beam injection into a Nd:Glass laser system. We also developed technology approaches that permit standard nanosecond and short-pulse operation on the same amplifier beam lines with only minor and easily made changes to the beam line. This should allow the implementation of a very flexible laser system in a fusion ignition facility with a capability of producing either nanosecond- or picosecond-duration pulses on demand. We also developed techniques to allow the automatic alignment and maintenance of split beams. Finally, system approaches were conceptualized that permit many beam line components to be time shared by standard nanosecond pulses as well as picosecond pulses during a single shot of the laser system.

Proposed Work for FY06

We will continue to develop detailed requirements and specifications for a split-beam backlighter system, including options for sharing beamline components between standard nanosecond and short picosecond pulses on a single shot; (2) perform an experimental demonstration of the precision metrology and beam-pointing system for the injection of chirped pulses into the front end of an amplifier chain and the alignment of the subsequent folded, mixed-grating compressor; (3) evaluate new techniques for multiple sub-aperture pulse generation, timing, and dispersion control; and (4) investigate procedures for the alignment of a folded, mixed-grating pulse compressor and the effects of vacuum isolation on optical alignment.

Publications

Barty, C. et al. (2005). *ICUIL 2004 International Conference on Ultrahigh Intensity Lasers*. 2004 Intl. Conf. Ultrahigh Intensity Lasers. Tahoe City, CA, Oct. 3–7, 2005. UCRL-PROC-213945.

Barty, C. et al. (2005). *Multi-kilojoule petawatt lasers: technology and applications at the National Ignition Facility*. UCRL-PRES-213408.

Leading the Quantum Limit Revolution

S. Darin Kinion 05-ERD-073

Abstract

The goal of this project is to utilize microstrip Superconducting Quantum Interference Device (SQUID) amplifiers to revolutionize experiments, ranging from quantum coherence to particle astrophysics, that require improved signal-to-noise ratio. The primary science deliverable will be a single-electron transistor (SET) readout that is sensitive enough to enable single spin detection in a solid-state system. We plan to develop robust packaging for the SQUID amplifier and then start using SQUIDs in experiments to read resonant-frequency SETs (rf-SETs). These experiments will be performed in collaboration with rf-SET experts located at Yale University; the University of New South Wales, Australia; and the University of Maryland.

Mission Relevance

This project will open the door to implementing quantum computing/quantum information secure communication architectures that support national-security and homeland-security missions. This work will also support the Laboratory's mission in discovery-class science, such as the dark-matter axion experiment.

FY05 Accomplishments and Results

This project had a mid-year start, beginning in May 2005. The first effort was to package a SQUID amplifier for use in a charge sensitivity measurement of a SET at the University of New South Wales. The amplifier performed as expected, but the experiment pointed out a few issues that must be addressed to achieve the quantum limit of charge sensitivity, in particular the dynamic range of the amplifier. A second experiment was performed at Yale using a shot-noise thermometer developed there. We achieved lower system noise-temperature than with traditional amplifiers, and with further improvements to the shielding, we should be ready for rf-SET measurements early in FY06.

Proposed Work for FY06

Two types of work will be performed in FY06. The first will be to transfer SQUID fabrication technology to LLNL. The University of California at Berkeley microlab will still be used for photolithography, but all material depositions, oxidations, and etching will be done at Livermore to ensure a steady supply of SQUIDs for all applications. The second will be primary research tasks that involve experiments using the amplifiers; the rf-SET charge noise measurements will be completed by the end of the year. In addition, we will begin performing 1/f-noise experiments using the amplifier to measure Josephson-junction critical current fluctuations.

Terascope: Terahertz Spectroscopic Imaging for Standoff Detection of High Explosives

Farid U. Dowla 05-ERD-076

Abstract

The purpose of this project is to investigate the viability of an approach to standoff detection and identification of high-explosives (HE) based on emerging spectroscopic and imaging technologies in the terahertz (THz) frequency regime. To this end, we will execute system-level analyses and simulations. In addition, because we believe that detectable and decipherable signatures for common HE materials of arbitrary geometrical shapes and configurations are critical elements, we will conduct an experimental program at the University of California at Santa Barbara (UCSB) free-electron laser facility, which is a well-characterized source of tunable, monochromatic THz radiation.

The THz portion of the electromagnetic spectrum is rich with information about the rotational and vibration characteristics of large molecules. We intend to demonstrate that we can exploit these signatures for identifying the materials that are detected by imaging in the THz regime, and assess the viability of the system for remote sensing. The results of this research will be published in peer-reviewed journals.

Mission Relevance

By developing THz capability for stand-off detection and identification of covert HE devices, this research supports Laboratory missions in national security and homeland security.

FY05 Accomplishments and Results

In FY05, we conducted an intense, short experimental campaign at UCSB during which materials of interest were subjected to THz radiation. Reflected signals from diverse shapes were scrutinized for distinctive signatures under a range of geometrical conditions. We conducted simultaneous systems analysis and simulation and compared the outcome to results of the experimental campaign to demonstrate that a viable capability is within reach. These activities set the stage for further characterization of solid materials in the THz regime.

Proposed Work for FY06

Having established system viability in FY05, using high-level models, we plan to continue confirming this viability with refined system simulations. Such simulation models will also be available for screening potential system improvements. Simultaneously via experiments, we will continue populating the spectral database for the THz regime, which will be crucial for material identification with our system.

Feasibility of Single Molecule DNA Sequencing using Surface-Enhanced Raman Scattering

Chad E. Talley 05-ERD-080

Abstract

This project aims to determine the feasibility of using surface-enhanced Raman spectroscopy (SERS) for sequencing DNA at the single-molecule level. This would provide a means of label-free DNA sequencing, significantly increasing the throughput and decreasing the sequencing cost per base. To establish the feasibility of DNA sequencing using SERS, we will first optimize the metal structures needed for the surface enhancement using numerical simulations. Second, we will measure the enhancement achievable by combining coherent anti-stokes Raman spectroscopy (CARS) with SERS. Finally, we will determine whether metal-molecule contact is needed to achieve single-molecule detection limits with SERS by separating the molecule from the metal surface with a layer of silicon dioxide.

This project will determine the technical feasibility of DNA sequencing using SERS. Through numerical simulations, the optimal structure providing the largest electric-field enhancement will be established. Secondly, the enhancements achievable by combining CARS with SERS will be established. Finally, we will establish whether metal–molecule contact is necessary to achieve single-molecule detection limits with SERS. These three results will provide enough information to determine the feasibility of sequencing DNA using SERS.

Mission Relevance

This project supports the Laboratory's missions in biosecurity and environmental management. If DNA sequencing using SERS is proven feasible, it will provide a platform for label-free DNA sequencing. This will dramatically reduce the cost and increase the speed of sequencing methods currently used both for identifying the presence and origin of pathogens used as weapons of mass destruction and for performing whole-colony sequencing of microbial colonies.

FY05 Accomplishments and Results

This project developed a theoretical tool that allows prediction, at the nanoscale and with arbitrary geometries, of surface plasmons on metallic structures. Calculations show that the electric field can be localized to very small volumes. We were able to tune the frequency of the surface plasmon and localized volume of the electric field by altering the geometry of the metal nanostructures. In this project, we also demonstrated that combining SERS and CARS produces enhancement of the Raman signal at a specific frequency that is greater than that obtained with standard SERS alone. Finally, SERS spectra from silver nanoparticles coated with silicon dioxide show that enhancement is still possible without metal–molecule contact, although overall enhancement was approximately an order of magnitude lower compared to bare silver nanoparticles.

Sensor Fusion for Regional Monitoring of Nuclear Materials with Ubiquitous Detection

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Simon E. Labov 05-ERD-081

Abstract

Detecting the unconventional delivery of a nuclear weapon is one of the most crucial and difficult national security challenges facing us today. Radiation detection systems composed of intelligent, networked devices intended for comprehensive broad-area coverage are now being developed. Cataloguing and fusing the data from these new detection systems will be one of the most significant challenges in radiation-based security systems. In the next few months, we will have access to a modest deployment of sensors with continuous measurement capability. We propose to collect these data and develop and test sensor fusion algorithms that will greatly enhance detection capability.

Mission Relevance

This project will advance our capability in radiation detection to meet future national security and homeland security mission needs.

FY05 Accomplishments and Results

In FY05 we: (1) restructured the existing database to allow both the cataloguing of radiation data and straightforward access, enabling us to produce geo-spatial radiation maps; (2) used the database approach to set up a 3- by 3-km test bed centered at LLNL and completed initial mapping of the radiation field in LLNL environs in both time and space; (3) developed algorithms to represent radiation data in a geographic information system (GIS) layer; (4) developed, in collaboration with an industrial partner, a radiation sensing and geo-location device, and collected data; (5) designed, in collaboration with industrial partner, a very small, low-power, geo-location-aware radiation-sensor "tag" device; and (6) developed first-order visualization tools for GIS layers in the database.

Proposed Work for FY06

In FY06 we will (1) simultaneously increase the fidelity of the local test bed and investigate the scalability and effectiveness of the combined layers in the database—highlights will include measurements with different detector types, improved time-selection capabilities, and exploring correlations between GIS layer features and measured radiation levels; (2) continue mapping of the local 3- by 3-km test bed to quantify the effects of bias and statistical errors in the radiation and geo-location data; (3) map a small (1- by 1-km) portion of an urban area into the test bed; (4) conduct fixed source trials within the test bed with both real and simulated sources; and (5) calculate and demonstrate the scalability of the database as a function of sensor density.

Rapid Defense Against the Next-Generation Biothreat

Raymond P. Mariella 05-ERD-084

Abstract

Bioengineered and emerging pathogens represent a significant threat to human health. The best defense against a rapidly expanding pandemic is to quickly isolate the pathogen from biological samples for analysis. The one persistent technology gap in the process of identifying and quantifying the presence of pathogenic agents has been sample handling and preparation that must precede any assay. The objective of this project is to replace burdensome, manual techniques with new automated technologies for sample handling and preparation. Specifically, we will use microfluidics with ultrasonic, electrophoretic, and dielelctrophoretic techniques to separate and purify viruses, the most transmissible and infectious agents, from biological and environmental samples.

We expect the new capabilities [e.g., surface-enhanced Raman spectroscopy (SARS)] developed in this project to reduce the time required to identify a new pathogen by up to an order of magnitude, in part by better matching methods for sample preparation with the needs of emerging assay technology. These capabilities will also be critical to developing ubiquitous, high-performance autonomous pathogen-sensing systems envisioned as sentinels that monitor for aerosol-transmitted pathogens by screening, for example, air filters or handrails at international airports.

Mission Relevance

By making it possible to rapidly isolate and detect engineered and naturally emerging biothreats, this project contributes to the nation's defense against bioterrorism, which is central to the Laboratory's homeland security mission. In addition, this project supports the Laboratory's mission in bioscience to improve human health and the NIH's mission objectives in public healthcare.

FY05 Accomplishments and Results

After a midyear start, we collected the electronics and control instrumentation to set up microfluidic test stations, as well as a selection of fluorescent beads ranging from 30 nm to 5^{-5} m in diameter, which we used to calibrate our flow system and align the microscope-based test station. Using existing microfluidics models and published data on the physical properties of human cells, bacteria, and viruses, we calculated the scales of systems (channel lengths and cross sections, electrode size, distributions, and voltages for dielectrophoresis and electrophoresis) to manipulate and separate bacteria and smaller particles from the larger particles.

Proposed Work for FY06

In FY06, we will create a capability to propagate, characterize, and purify Risk Group 1 viruses, which will be selected based on the availability of virus and detection reagents. Using simulations that will be continuously validated and improved, we will begin design and fabrication of single-function microfluidic devices to perform manipulations using ultrasonics, electrophoresis, and dielectrophoresis. We will use ultrasonics to disrupt aggregates and divert the unwanted larger particles into the waste stream. Once we have a stream that consists primarily of bacteria and viruses, electrokinetic transport will be used to trap the bacteria and to pass a stream of purified viruses for analysis.

Detection of Human Presence from Aerosol Collection

Elizabeth K. Wheeler 05-FS-004

Abstract

Developing technical means to detect the presence of a human in a designated location is a high-priority capability for the law-enforcement community. The purpose of our proposal is to determine feasibility of using epithelial cells that are constantly shed into the air as a source of DNA that could be collected and analyzed to accomplish that goal. Because extracting DNA from traditional fingerprints has already been well established and documented in the scientific literature, there is reason to believe that this approach may work.

In the proposed project, we will determine whether sufficient quantities of DNA can be collected in environmental samples to support biological assays. We will also resolve whether an aerosol collector that gathers cells from the air could yield usable DNA that would confirm a human presence. If these two steps indicate that the approach is feasible, we will then establish operational parameters such as the minimum and optimal collection times and operating distance.

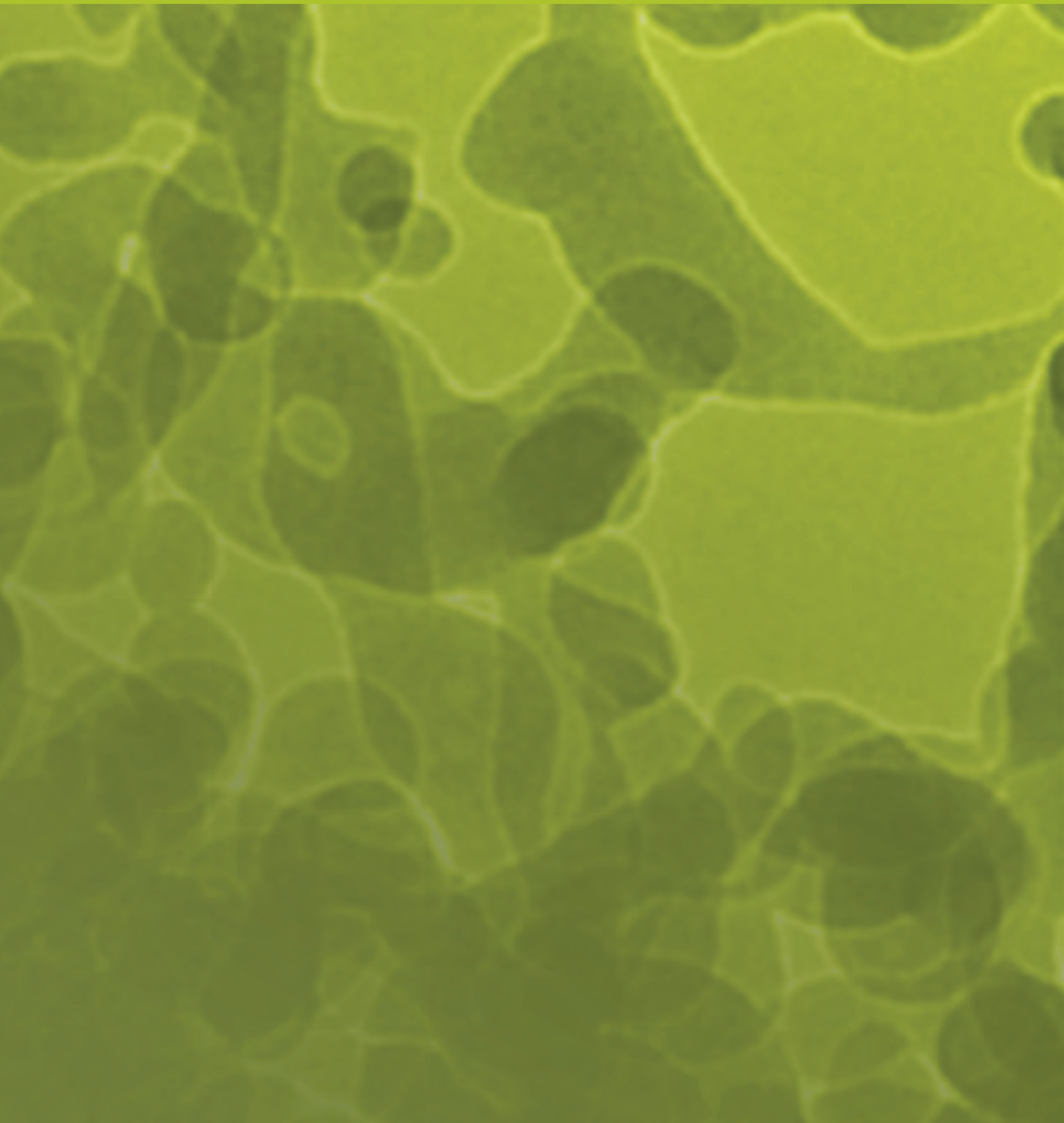
Mission Relevance

If successful, this work would support the Laboratory's national security mission, specifically in the area of detecting and preventing asymmetric threats such as terrorist attacks.

FY05 Accomplishments and Results

We have successfully proven feasibility of the proposed technique by demonstrating that an aerosol collector could be used to sample atmospherically suspended DNA that in turn could be identified as human. To further this development, more in-depth controlled studies are needed to assess the efficiency of aerosol collection of epithelial cells. These studies would focus on techniques for effectively using the limited amount of intact DNA in shed epithelial cells, determining degradation rate of DNA in the atmosphere, and improving collection efficiency.

Biological Sciences



Dynamic Simulation Tools for the Analysis and Optimization of Novel Filtration, Sample Collection, and Preparation Systems

David S. Clague 02-ERD-066

Abstract

The heightened attention to chemical and biological early-detection systems involving liquid- and gas-phase transport through sieving media creates an increased need for a high-efficiency filtration, collection, and sample-preparation system. This project will develop novel simulation tools to optimize these critical operations. This new capability will characterize system efficiencies based on the details of the microstructure and environmental effects. To accomplish this, we will develop a new, multiphysics lattice Boltzmann (LB) simulation capability that will include detailed microstructure descriptions, the relevant surface forces that mediate species capture and release, and temperature effects for both liquid- and gas-phase systems.

The project will deliver validated computational analysis tools to optimize filtration, collection, and sample-preparation systems for chemical and biological early-detection systems.

Mission Relevance

This effort will directly contribute to the Laboratory's missions in homeland defense and counterterrorism by providing simulation tools for sample collection, filtration, and preparation systems for mitigating or preventing warfare or terrorist activities involving chemical and biological agents.

FY05 Accomplishments and Results

In 6 months in FY05, we developed a novel boundary condition to enable modeling of arbitrary Knudsen number flows. In addition, we developed and integrated surface force models to predict filter species nonhydrodynamic interactions, and we improved the prediction of the dielectrophoretic force prediction for species sizes greater than a micron.

Publications

Liang, E., R. L. Smith, and D. S. Clague. (2004). "Dielectrophoretic manipulation of finite sized species and the importance of the quadrupolar contribution." *Phys. Rev. E* **70**, 066617. UCRL-JRNL-206442.

Pathomics

Kenneth W. Turteltaub 03-SI-005

Abstract

"Pathomics" is a comprehensive strategy for understanding an individual's response to infectious disease agents, particularly those that represent a threat from bioterrorism. Its overall goal is to predict possible routes of pathogenicity and enable detection systems for determining that an attack or disease outbreak is beginning prior to people becoming overtly sick. Using a vaccine strain for smallpox in humans and the infectious cowpox virus in animal models, we will demonstrate that detection of disease is possible pre-symptomatically, and molecular signatures exist that can distinguish between pathogen types. We will propose a preliminary type of diagnostic platform and demonstrate its use in public health surveillance.

By the end of the project, we plan to demonstrate that molecular changes can be detected in blood within the first few hours after exposure, and that blood-borne signatures develop within the first few hours of infection by the vaccine strain.

Mission Relevance

This project combines strengths developed through the Laboratory's Human Genome Project and the Chemical and Biological National Security programs to link the biological sciences and national security arenas. This research is needed to meet gaps in our ability to detect and mitigate the effects of infectious disease, whether natural or from acts of bioterrorism, and supports the Laboratory's national security and homeland security missions as well as missions of the National Institutes of Health.

FY05 Accomplishments and Results

In FY05 we (1) completed analysis of the spectrum of biochemical components in blood that can be used to detect cowpox infection before clinical symptoms are present in rodents; (2) completed analysis of the variables inherent in analysis of proteins by gel electrophoresis; (3) evaluated a series of antibodies for inclusion in a detection panel; and (4) completed a study in apparently healthy humans to establish baseline ranges for selected antibody and mRNA panels.

Publications

Fodor, I. K. et al. (2005). "Statistical challenges in the analysis of two-dimensional difference gel electrophoresis experiments using DeCyderTM bioinformatics." *Bioinformatics* **21**(19), 3733. UCRL-JRNL-207079.

Langlois, R. G. et al. (2004). "Serum protein profile alterations in hemodialysis patients." *Am. J. Nephrol.* **24**, 268. UCRL-JRNL-201081.

Microbial Pathways

Harry R. Beller 03-ERD-062

Abstract

This project makes use of emerging genome-enabled techniques (such as whole-genome cDNA microarrays) to explore novel, bacterial metabolic pathways and bacteria–mineral interactions that can mediate the behavior of uranium and other environmentally relevant elements in the subsurface. This work is particularly pertinent to a remediation process that is of great interest to DOE: the in situ, reductive immobilization of metals, including radionuclides. Understanding microbial processes that mediate metal and radionuclide contamination in the environment also has relevance to national security issues. This project will utilize our existing capabilities in genomics, microbiology, biochemistry, geochemistry, and computational modeling in a significantly new research direction for bioscience.

Expected results include (1) publications in high-visibility journals regarding the microbially mediated transformations of uranium in anaerobic soil and aquifer environments and the genetic and biochemical basis of nitrate-dependent metal oxidation, (2) the finished and annotated whole genome of the bacterium *Thiobacillus denitrificans* for use by the scientific community interested in anaerobic metal oxidation, and (3) a deeper understanding of nitrate-dependent Fe(II) oxidation, which is uncharacterized on a genetic and biochemical level but is a key factor in the behavior of heavy metals and radionuclides in the environment.

Mission Relevance

This project will further biogeochemical understanding that supports LLNL's mission of providing a scientific basis for environmental management and restoration. Microbial processes active in the near-surface environment are important considerations in national security, especially issues concerning radionuclide and toxic metal contamination of groundwater. The project encompasses collaborations within and between the Laboratory and the DOE Joint Genome Institute.

FY05 Accomplishments and Results

The team completed a thorough, whole-genomic analysis of *T. denitrificans*, including comparative genomic analyses, focusing on major metabolic/biogeochemical pathways. A manuscript summarizing this work was submitted for publication. A series of cDNA microarray experiments (25 chips total) was completed, as was computational analysis of large portions of the resultant data. Major advances were made in the development of a genetic system in *T. denitrificans* for knockout mutation (i.e., hypothesis testing) studies, including successful incorporation of a plasmid by electroporation and by conjugation. We also developed a colorimetric assay for anaerobic Fe(II) oxidation.

Proposed Work for FY06

Proposed work for FY06 includes (1) studying the effect of Fe(II) on nitrate-dependent U(IV) oxidation by *T. denitrificans*; (2) completing development of a genetic system for *T. denitrificans* and performing knockout mutation studies; (3) using reverse transcription, quantitative polymerase chain reaction analysis to confirm microarray results; (4) publishing the results of microarray experiments in a high-profile journal, and, if time permits; (5) conducting microarray experiments with knockout mutants (as opposed to wild type cells).

Publications

Beller, H. R. et al. (2006). "The genome sequence of the obligately chemolithoautotrophic, facultatively anaerobic bacterium *Thiobacillus denitrificans*." *J. Bacteriol.* **188**, 1473. UCRL-JRNL-215002.

Beller, H. R. (2005). "Anaerobic, nitrate-dependent oxidation of U(IV) oxide minerals by the chemolithoautotrophic bacterium Thiobacillus denitrificans." *Appl. Env. Microbiol.* **71**, 2170. UCRL-JRNL-204909.

Beller, H. R. (2005). *Anaerobic, nitrate-dependent oxidation of uraninite by the chemolithoautotrophic bacterium* Thiobacillus denitrificans. American Society for Microbiology Conf., Atlanta, GA, June 3–5, 2005. UCRL-ABS-208811.

Beller, H. R. et al. (2005). *Anaerobic, nitrate-dependent oxidation of U(IV) by the obligate chemolithoautotroph Thiobacillus denitrificans: Cell suspension and whole-genome transcriptional studies*. ISSM/ISEB 2005, Jackson Hole, WY, Aug. 14–19, 2005. UCRL-ABS-213008.

Beller, H. R. et al (2005). *The genome sequence of the chemolithoautotrophic beta-proteobacterium Thiobacillus denitrificans: A small yet metabolically versatile genome*. American Society for Microbiology Conf., Atlanta, GA, June 3–5, 2005. UCRL-ABS-208813.

Beller, H. R. et al. (2005). *Transcriptional analysis of chemolithoautotrophic S oxidation by* Thiobacillus denitrificans *under aerobic vs. denitrifying conditions using whole-genome oligonucleotide microarrays*. American Society for Microbiology Conf., Atlanta, GA, June 3–5, 2005. UCRL-ABS-208812.

The Instrumented Cell

Allen T. Christian 03-ERD-068

Abstract

Small, interfering hybrids (siHybrids), a new class of molecule developed at LLNL, are an effective gene-silencing tool. They have the potential applications in clinical medicine and may represent a new tool for high-throughput research. The instrumented cell (IC) platform developed in this project can be used to manipulate the environment of individual cells and produce analytical measurements of cells' reaction to stimuli. These capabilities will provide quantitative data on an individual cell's responses to stimuli and will be a platform for researchers to gain an improved level of precision in biological system analysis and modeling.

The joint aims of this project were to more fully explore the potential uses of siHybrids and to use the IC platform to develop models of chemical interactions within and among cells, following perturbations made within the cell and to the cell's environment. The concept was that all of the pathways of a cell could be computationally modeled, allowing creation of predictive models of cellular responses to stimuli. Accomplishing this goal at the statistically relevant level of the individual cell would revolutionize biology, from basic research to industrial pharmacology. The immediate goal was to develop instruments to allow these measurements to be made.

Mission Relevance

This project leverages LLNL's capabilities in microfabrication, analytical techniques, and cellular biology to enable single-cell characterization of biological processes. The result will be a capability that will lead to advances in bioscience and technology, in support of LLNL's mission in bioscience and technology to improve human health and of DOE's Genomics:GtL program.

FY05 Accomplishments and Results

The FY05 plan was to elucidate the mechanisms of siHybrid delivery and function to improve the efficacy of the gene-silencing technology for therapeutic and research applications. The plan also called for developing the prototype IC platform into a tool for single-cell microbial studies. The latter of these goals was achieved: we built and successfully tested a chip that captures bacterial cells in an individually arrayed manner to permit thousands of the cells to be studied singly. To date, we have been unsuccessful in determining the mechanism responsible for the action of the siHybrids, and we do not yet understand why they always seem to enter cells but do not always silence the targeted genes. Before publishing our siHybrid work, a mechanistic understanding will have to be discovered, but a manuscript describing the IC platform was submitted.

Elucidation of the Mechanism of Gene Silencing using Small Interfering RNA-DNA Hybrid Molecules

Lawrence C. Dugan 03-ERI-004

Abstract

The objective of this proposal is to answer questions concerning small, interfering hybrid- (siHybrid-) based gene silencing. This project is motivated by the improvements siHybrids have shown over siRNA, a powerful tool for silencing gene function that is being considered for use in clinical settings, including cancer and HIV therapy. In this project, we set out to determine how many molecules enter a cell and whether siHybrids function by the same mechanism as siRNA. Another motivation is to improve the stability of the siHybrids during unaided delivery. These goals will be accomplished using western and northern blot analysis of protein and messenger RNA (mRNA), respectively, from exposed cells. Also, commercial modifications will be placed on the strands to look at stability and analyzed after nuclease exposures.

Successful completion of this project will provide a population-based answer to the question of how many siHybrid molecules are required to cause an effect in a cell and evidence that siHybrids function by targeting sequence-specific mRNA, leading to its degradation. These answers will provide important clues for researchers seeking to silence genes that are responsible for disease, such as mutant p53 in prostate tumor cells, for example. Furthermore, these results will provide improvements for therapeutic delivery of siHybrids.

Mission Relevance

This research supports LLNL mission areas of (1) national security, by helping reduce the effectiveness of biological warfare agents, and (2) bioscience and biotechnology to improve human health, by providing mechanistic information for a process being tested for use in treating cancer and infectious diseases. This information will also help determine gene functions in such cellular processes as development, differentiation, cell signaling, and cell death.

FY05 Accomplishments and Results

A dose-dependent accumulation of siHybrids was shown to have occurred after aided and unaided delivery to cells, and a determination was made that siHybrids function in a manner similar to siRNA, with mRNA being the target molecule and mRNA degradation the result. Furthermore, our work in FY05 showed that modifications to the RNA or DNA strand of the siHybrids could improve the stability of the siHybrids to resist nuclease degradation in cell culture media. Collaborations were also initiated to improve delivery methods using cell-targeting antibodies.

Force Spectroscopy to Study Multivalent Binding in Protein–Antibody Interactions

Todd A. Sulchek 03-ERI-009

Abstract

This project uses atomic-force microscopy to measure the binding strength between the Mucin1 protein, an indicator of some cancers, and its antibody. The goal of this project is to determine the dynamic strength of multiple bonds. We are motivated by the need to improve the binding times of cancer therapeutics that use a multiple number of antibodies to “hold on” to the cancer cell. We will determine the average bond time for multiple antibodies through a technique called dynamic force spectroscopy.

We will determine the average bond lifetime for one, two, and three antibodies binding to their cancer targets. This result will significantly benefit our collaborators in the medical community who are optimizing cancer therapeutics for use in patients.

Mission Relevance

The capability to measure antibody–protein interactions will enable other biological applications. These include characterizing cell–pathogen interactions in support of the Laboratory’s homeland security mission. In addition, research on drug–cell and cell–cell interactions supports LLNL’s mission in bioscience to improve human health.

FY05 Accomplishments and Results

We determined the rupture rate of multiple antibody–Mucin1 bonds using dynamic force spectroscopy. These results confirm a theoretical model describing multivalent binding and were accepted for publication in a peer-reviewed journal. We also developed a method to identify the number of interacting molecules from our measurements, which greatly improves identification of multiple bonds. These results are currently being prepared for publication.

Proposed Work for FY06

We will complete publication of a second article identifying the number of bonds broken during a measurement. We also will perform measurements on live cancer cells to see whether our measurement technique can be translated to clinical research. If so, the ability to determine kinetic rate constants on live cells will be of great use to the medical community.

Publications

Sulchek, T. A. et al. (2005). “Dynamic force spectroscopy of parallel individual Mucin1–antibody bonds.” *Proc. Natl. Acad. Sci. U.S.A.* **102**, 16638. UCRL-319771.

Intracellular Chemical Measurements: A Generalized Approach with High-Spatial Resolution using Functionalized Nanoparticles

Ted A. Laurence 03-ERI-010

Abstract

The objective for this research is to develop methods to measure concentrations in chemical microenvironments in cells and tissues using recently developed, functionalized metal nanoparticles (50–100 nm in diameter). Surface-enhanced Raman spectroscopy (SERS) allows sensitive detection of changes in the state of chemical groups attached to single nanoparticles. A nanoscale pH meter will be used to image the local pH inside and outside cells in tumor tissues. A nanoscale pH meter will be tested in a cell-free medium to measure the pH of the solution immediately surrounding the nanoparticles. This will be followed with tests of the nanoscale pH meter inside cells to determine optimal conditions and methods for these measurements.

This novel measurement technique promises to provide rigorous quantitation of cellular concentrations, opening new windows to cell characteristics and behavior. In tumor cells, it may be possible to monitor the effects of the local pH gradient on uptake of various therapeutic agents while avoiding the poor spatial resolution or high cell toxicity of other methods. A decrease in pH will serve as an indicator of apoptosis; correlation with stresses or other signals will help identify events, causes, and phenomenology of apoptosis. Success with the proposed experiments will demonstrate the ability of SERS nanoparticles to measure local pH inside cells. This will encourage development of the SERS technology to measure other chemical concentrations inside microenvironments.

Mission Relevance

Measuring chemical gradients (variations in concentration) across membranes is important for understanding basic biological processes and is relevant to developing disease treatments. This research supports development of a novel method for measuring chemical concentrations. Such probes of the intracellular environment represent a key enabling capability for the DOE Genomics:Gtl program and biosecurity applications. In addition, the new measurement technique supports LLNL applications related to bioscience and technology to improve human health, as well as breakthroughs in fundamental bioscience.

FY05 Accomplishments and Results

In FY05, research focused on understanding possible causes for limited resolution (~1 pH unit). The pH was obtained from the SERS signal by calculating the ratio of intensities between a Raman line dependent on pH and one that was independent of pH. A rapid assay that uses only the relevant lines was developed, allowing higher throughput in the number of particles analyzed. By comparing variations in the ratios on intensities at the Rayleigh scattering frequency and a pH-independent Raman frequency, a heterogeneity that is independent of pH was revealed. This heterogeneity was more severe for the silver nanoparticles than with gold, which can be made more monodisperse. The research team also began pH measurements on gold nanoparticles that should have a more homogeneous distribution of plasmon resonance shapes.

Proposed Work for FY06

Proposed FY06 work will (1) test methods of incorporating the SERS nanoparticles into cells, including micro-injection, electroporation, and endocytosis; (2) prepare image-acquisition software for use in 3-D confocal imaging with a real-time display; (3) incorporate SERS nanoparticles into cells and monitor the cells over extended periods of hours to days to determine the long-term stability of the nanoparticles, as well as the viability of cells with nanoparticles inside of them; (4) measure the pH of intracellular and extracellular environments in tumor cells; and (5) monitor the effects of the local pH gradient on uptake of various therapeutic agents in tumor cells.

Publications

Fore, S. et al. (2005). “Distribution analysis of the photon correlation spectroscopy of discrete numbers of dye-molecules conjugated to DNA.” *IEEE J. Select. Topics Quantum Elect.* **11**, 873. UCRL-JRNL-215118-DRAFT.

Kapanidis, A. N. et al. (2005). “Alternating-laser excitation of single biomolecules.” *Acc. Chem. Res.* **38**, 824. UCRL-JRNL-205766.

Laurence, T. A., C. Talley, and T. Huser. (2005). *Application of SERS nanoparticles for intracellular pH measurements*. Intl. Workshop Optical Probes for Molecular and Cellular Imaging, Sonoma, CA, Dec. 7–8, 2005. UCRL-ABS-215999.

Laurence, T. A. et al. (2004). *Application of SERS Nanoparticles to Intracellular pH Measurements*. American Society for Cell Biology 44th Annual Mtg., Washington, D.C., Dec. 4–8, 2004. UCRL-ABS-208637.

Laurence, T. A. et al. (2005). “Probing structural heterogeneities and conformational fluctuations of biopolymers.” *Proc. NAS* **102**, 17348. UCRL-JRNL-208639.

Laurence, T. A., S. Fore, and T. Huser. (in press). “Fast, flexible algorithm for calculating photon correlations.” *Opt. Lett.* UCRL-JRNL-216336.

Single Molecule Techniques for the Study of Chromatin Assembly and Remodeling

Christopher Jeans 03-ERI-011

Abstract

In eukaryotic cells, DNA is tightly packaged as chromatin through interactions with histone proteins. For processes such as DNA replication, repair, and transcription, the structure of chromatin must be remodeled in such a way that necessary enzymes can access the DNA. Our understanding of the remodeling process is hindered by a lack of knowledge of the fine structure of chromatin and how it is modulated in the living cell. We propose to perform single-molecule experiments using atomic force microscopy (AFM) to determine the nature of packaging interactions in chromatin and to measure the forces involved in such interactions. The effects of histone modifications on these forces will be determined, as will the effects of damage within the DNA.

Images of native chromatin and chromatin subjected to various kinds of DNA damage will be used to gain insight into the nature of the interactions that hold the chromatin fiber together under different conditions. This will give us valuable new information about the structure of chromatin and the way in which DNA damage affects this structure. This information will improve our knowledge of the process of DNA damage in chromatin.

Mission Relevance

This work will address questions of general biological interest through development of novel techniques for the manipulation of single molecules, and will establish a new capability: the physical manipulation of individual DNA strands. This capability may provide critical data for biodefense applications in support of missions in homeland security and environmental biology.

FY05 Accomplishments and Results

Chromatin fibers have been isolated from chicken erythrocytes (CE) and Chinese hamster ovary (CHO) cells. Some of the CHO cells were treated with sodium butyrate to force a remodeled state upon the chromatin. Chromatin isolated from these three samples—CE, CHO, and CHO with butyrate—has been studied. The CHO and CE chromatin differ in their linker histones, as expected because of the greater complexity of the transcriptionally active nucleus of CHO cells. When CHO cells are grown in the presence of butyrate, there is a significant difference in the linker histone composition of the chromatin. We used AFM to study these samples and found that CE and CHO chromatin have very similar nucleosomes, but different higher order structures.

Using Femtosecond Laser Subcellular Surgery as a Tool to Study Cell Biology

Nan Shen 03-ERI-012

Abstract

Research on cellular function and regulation would be greatly advanced by new instrumentation using methods to alter cellular processes with spatial discrimination on the nanometer scale. We present a novel technique for targeting subcellular organelles or other biologically important regions in living cells using femtosecond laser pulses. By tightly focusing the pulses inside a cell, we can vaporize cellular material through nonlinear optical processes. This technique enables noninvasive manipulation of physical structures of a cell with submicrometer resolution. We propose to study the role mitochondria play in cell proliferation and apoptosis (programmed cell death) by selectively perturbing mitochondria in living cells. Our technique will provide a unique tool for the study of cell biology.

Our experimental apparatus will allow us to study cell behaviors in real time to address questions about how cells behave after laser surgery over the long term and to determine whether disruptions of different cellular structures result in different cell responses. We hope to induce apoptosis by disrupting mitochondria in a cell to study the process of cell death and to see whether the number of functional mitochondria is critical to the survival of a cell and whether mitochondria are capable of repairing structural damages. Another focus of the project is on how the cell cytoskeleton reorganizes under structural perturbation. We will follow the dynamics of cytoskeletal remodeling after laser disruption of microtubules and actin filaments.

Mission Relevance

We plan to demonstrate new LLNL capability utilizing laboratory expertise in both laser and life sciences. The resulting data will support DOE missions in understanding cellular mechanisms, such as the Genomics:GtL program. Furthermore, this understanding will ultimately prove important in helping to determine complete mechanisms for pathogenicity, in support of homeland security applications.

FY05 Accomplishments and Results

Using atomic force microscopy, we verified that cellular material was removed during femtosecond laser surgery, but the photobleaching fluorophores used to label target organelles were not. We also studied long-term cellular response to laser irradiation. Cells remained viable and continued to divide after laser surgery, thus enabling the study of cell functions that require long-term monitoring. To initiate apoptosis in human fibroblast cells, we varied the number of mitochondria in cells subjected to laser disruption. Target cells did not show signs of apoptosis or necrosis after removal of up to ~10% of the cell mitochondria. Femtosecond lasers can also create lesions in the cell microtubule network, leading to cytoskeleton remodeling without affecting cell viability.

Proposed Work for FY06

FY06 work will focus on (1) the link between laser surgery and DNA damage; (2) apoptosis after the disruption of individual mitochondria in single-cell laser surgery; and (3) the feasibility of investigating cytoskeleton remodeling in living cells by using femtosecond laser surgery to create lesions in microtubules or actin fibers in cell cytoskeleton. Results obtained in this study will support future research on understanding pathogen–host cell interaction.

Publications

Shen, N., M. Colvin, and T. Huser. (2005). “Investigate cellular functions by directly probing structures inside living cells using femtosecond laser pulses.” *Biophys. J.* **88**, 522A. UCRL-ABS-207700.

Multiprobe Investigation of Proteomic Structure of Pathogens

Alexander J. Malkin 04-ERD-002

Abstract

This project will investigate the proteomic structure, architecture, and function of human pathogens through a combination of high-resolution in vitro atomic force microscopy (AFM) with immunolabeling and mass spectrometry. A more comprehensive understanding of the architecture and properties of pathogens would contribute significantly to a general understanding of their life cycle and may lead to advances in diagnostic and immunological aspects of anthrax biodefense. We will characterize the protein structures of intact and nanodissected pathogens by AFM, correlate these structures with gene products using antibodies, and identify these protein components by mass spectrometry. This experimental approach will allow us to establish relationships between pathogen structure and function.

By elucidating the structurally related properties and function of human pathogens and modeling their architecture, this work will provide a foundation for the understanding of the structurally related properties of pathogens that could lead to the development of vaccines, detection systems, and improved decontamination procedures.

Mission Relevance

By developing AFM for the evaluation of pathogenic morphological signatures and structural attributes, this project supports the Laboratory’s national security and homeland security missions by developing techniques for identifying and characterizing pathogens that could be used by bioterrorists. In addition, this work will provide fundamental information for the modeling of pathogen architectures for use in structural biology and thereby will support DOE’s Genomics:GtL program.

FY05 Accomplishments and Results

In FY05 we (1) performed the first-ever in vitro visualization and analysis of species-specific nanometer-scale native structures of the outer spore coat of various *Bacillus* and *Clostridium* species; (2) performed the first-ever direct visualization and analysis of the dynamic structural and morphological response of individual spores to environmental changes; (3) demonstrated, through interspecies size distribution analysis, that the dimensions of individual spores differ significantly depending upon species, growth regimes, and environmental conditions; (4) produced antibodies with immunochemical specificity for protein components of the spore coats of *Bacillus* and *Clostridium* species; and (5) began in vitro molecular scale studies of germination mechanisms and immunolabeling.

Proposed Work for FY06

In FY06 we will (1) complete immunolabeling experiments for the proteomic mapping of the spore coat surfaces; (2) develop a molecular scale model of ultrastructural changes in the *C. novyi-NT* spore surface during the germination process; (3) evaluate the impact of chemotherapeutic drugs on the germination process of *C. novyi-NT* spores; (4) establish a direct correlation between the ultrastructure and the life cycle of *Bacillus* and *C. novyi-NT* spores through AFM imaging of immunolabeled dormant spores, germinating spores, and vegetative cells; and (5) characterize and map the hydrophobicity and charge distribution on *Bacillus* spore surface as a function of species and preparation by chemical force microscopy and functionalized nanogold probes, respectively.

Publications

Malkin A. J. et al. (2005). *Unraveling the architecture and structural dynamics of pathogens by high-resolution in vitro atomic force microscopy*. Third Latin American Symp. Scanning Probe Microscopy, Ouro Preto, Brazil, Apr. 18–20, 2005. UCRL-PROC-211437.

Plomp, M. et al. (2005). “Architecture and high-resolution structure of *Bacillus thuringiensis* and *Bacillus cereus* spore coat surfaces.” *Langmuir* **21**, 7892. UCRL-JRNL-209940.

Plomp, M. et al. (2005). “*Bacillus* spore coat ultrastructure, assembly and environmental dynamics.” *Scanning* **27**, 97. UCRL-ABS-210037.

Plomp, M. et al. (2005). “*Bacillus* atrophaeus outer spore coat assembly and ultrastructure.” *Langmuir* **23**, 10710. UCRL-JRNL-21726.

Electronic Polymerase Chain Reaction

Shea N. Gardner 04-ERD-030

Abstract

The polymerase chain reaction (PCR) stands among the keystone technologies of our time for analysis of biological sequence data. It is used in virtually every laboratory doing molecular, cellular, genetic, ecologic, forensic, or medical research. Despite its ubiquity, we lack the precise predictive capability that would enable detailed optimization of the dynamics of PCR reactions. In this project, we propose to develop tools to perform virtual PCR (vPCR) by building new computational methods to model the kinetic, thermodynamic, and biological processes of PCR reactions. These tools will allow us to generate precise predictive details about the effects of primers, reaction conditions, and contaminants on PCR products, and thus to optimize the selection of primers and reaction conditions.

The result of this project, a suite of programs that predict PCR products as a function of reaction conditions and sequences, will be used to address outstanding questions in pathogen detection, forensics, microarray applications, and the effects of contaminants and PCR inhibitors. Our vPCR tools should enable scientists to optimize PCR protocols with regard to time, temperature, ion concentration, and primer sequences and concentrations, and to estimate products and error rates before actual experiments. Our proposed capabilities are well ahead of all currently available technologies, which do not model non-equilibrium kinetics, polymerase extension, or predict PCR products. Licensing and publishing opportunities will be explored. A full patent application has been filed.

Mission Relevance

Applications of vPCR support LLNL missions in national security, homeland security, and human health. Any field that uses PCR, including bioforensics, biodetection, basic research in the environmental biology, and disease research (e.g., cancer), will benefit. Applications of vPCR include identification of genes and organisms present in complex microbial communities, DNA-polymerase-based gene synthesis, and forensic discrimination of closely related sequences.

FY05 Accomplishments and Results

We developed a prototype code that incorporates competition between multiple reaction pathways and several formulations of stochastic kinetic algorithms optimized to balance speed and accuracy for our library of test cases. Using more efficient data structures and faster algorithms and approximations enabled the code to handle more complex reactions than before, speeding run times by two orders of magnitude. We used the improved code to model the temperature-dependent extension of DNA and the decay of *Thermus aquaticus* polymerase, tracking all particles and their concentrations through time. We also began including DNA hairpin kinetics into the simulations. Thermal gradient PCR experiments were begun to examine the effects of primer sequence mismatches, annealing temperature, and annealing duration on the products produced.

Developing New Tools for In-Vivo Generation and Screening of Cyclic Peptide Libraries

Julio A. Camarero 04-ERD-040

Abstract

The objective of this project is to develop a new combinatorial approach for the biosynthesis and screening of small, drug-like toxin inhibitors inside living cells. Initially, this novel approach will be used for finding inhibitors against the botulinum and anthrax lethal factor (LF) bacterial toxins, respectively from *Clostridium botulinum* and *Bacillus anthracis*. This combinatorial technique combines the biosynthesis and screening of a library in the same step using a living cell as a small microchemical factory. This research will accelerate the process of discovering new ligands for any molecular target.

The success of this project would have important impacts in national biosecurity and drug discovery. It will introduce a new and generic technology that combines chemistry and biology for fast and efficient identification of high-affinity ligands for botulinum and anthrax LF bacterial toxins alike. These ligands could be used as powerful antidotes against the toxins and as biosensors attached to appropriate platforms. Furthermore, this new method can be easily generalized for finding small, drug-like effectors for any protein–protein interaction, which will have a tremendous impact in pharmaceuticals and proteomics.

Mission Relevance

By identifying ligands that have high affinity for botulinum and LF bacterial toxins, this project could lead to new antidotes for biothreats and new biosensors to detect bioterrorist attacks. The project supports the Laboratory's national security and homeland security missions by furthering LLNL efforts to counter bioterrorism.

FY05 Accomplishments and Results

Work accomplished in FY05 includes the biosynthesis of the first libraries based on cyclotides, which are small, circular peptides that are extremely stable in biological and chemical condition. We also developed a fluorogenic reporter for in vivo and in vitro screening of the anthrax LF toxin. This includes the cloning, expression, and purification of a chimeric protein composed by short peptide substrate for LF linked by the fluorescent proteins YFP (yellow fluorescent protein) and BFP (blue fluorescent protein).

Proposed Work for FY06

In FY06 we will (1) develop a method for the biosynthesis and screening of libraries of circular peptides to find high-affinity effectors and inhibitors; (2) apply the method to the anthrax LF bacterial toxin; and (3) develop the first-ever cyclic peptide inhibitors for LF toxins.

Publications

Kimura R. and J. A. Camarero. (2005). "Biosynthesis of the cyclotide Kalata B1 using protein splicing tools." *Biopolymers* **80**, 537. UCRL-ABS-209574.

Kimura R. and J. A. Camarero. (2005). "Expressed protein ligation: a new tool for the biosynthesis of cyclic polypeptides." *Protein Pept. Lett.* **8**, 789. UCRL-JRNL-214692.

A Coupled Computational and Experimental Approach to Determine Functions of Deeply Conserved "Anonymous" Human Genes

Gabriela G. Loots 04-ERD-052

Abstract

Progress in deciphering the human genome could be enhanced by improved computational tools and high-throughput experimental strategies. The aim of this program is to develop tools for determining the function of novel human genes found in all sequenced genomes. Because of its importance as a developmental model, the frog genome has been chosen for sequencing by the Joint Genome Institute (JGI). In collaboration with colleagues at University of California (UC) at Berkeley, we propose to develop methods for using frog embryos as a high-throughput system to document functions of conserved human genes with unknown functions. These methods will computationally identify and predict functions for evolutionarily conserved human genes and to test those predictions using gene manipulation strategies in frog embryos.

Because amphibian and mammalian development are very similar, the main output of this project—a robust set of computational and experimental tools and applications—can be extended to the analysis of complex eukaryotic genomes. In particular, we plan on using these resources to identify and characterize unknown, developmentally expressed human genes that are shared by all vertebrate genomes sequenced to date. These data will be critical to understanding the roles these genes play during evolution, vertebrate development, susceptibility to disease, and human health.

Mission Relevance

Results of this project have direct applications to bioinformatics, functional genomics, and other biological sciences in support of LLNL's mission in biotechnology for improving our understanding of the human genome and human health. In addition, the project fosters strong collaborations with the JGI and UC Berkeley in the field of computational biology.

FY05 Accomplishments and Results

In FY05, we (1) created a catalog of 2000 evolutionarily conserved human and frog anonymous genes based on version 3.0 of the JGI's sequenced frog genome; (2) computationally prioritized genes based on motifs and microarray expression data (of major interest were secreted molecules, transcription factors, and members of signaling pathways); (3) determined the expression patterns for over 200 novel genes, far exceeding our original 30-gene goal; (4) began using overexpression and knockdown of endogenous transcripts to analyze genes expressed in kidney, neural plate, and neural crest cells; and (5) began creating a gene-expression database.

Proposed Work for FY06

In FY06 we will (1) align the final frog genome draft assembly and create a complete catalog of conserved genes; (2) create a database that catalogs novel genes and their expression patterns, provides information on gene function, and establishes links to sequence annotation and evolutionary profiles through a Web browser; (3) determine the expression pattern and function of 50 novel genes; and (4) develop high-throughput automation to scale this process up to whole-genome analysis.

Publications

Khokha, M. K. and G. G. Loots. (2005). “Strategies for characterizing cis-regulatory elements in Xenopus.” *Brief Funct. Genomic Proteomic* **4**, 58. UCRL-JRNL-209654.

Loots, G. G. and I. Ovcharenko. (2005). “Dcode.org: anthology of comparative genomic tools.” *Nucleic Acids Res.* **33**, 56. UCRL-JRNL-209160.

Loots, G. G. et al. (2005). “Genomic deletion of a long-range bone enhancer misregulates sclerostin in Van Buchem disease.” *Genome Res.* **15**, 928. UCRL-JRNL-206198.

Ovcharenko, I. (2005). “Mulan: multiple-sequence local alignment and visualization for studying function and evolution.” *Genome Res.* **15**, 184. UCRL-JRNL-205739.

Ovcharenko, I. et al. (2005). “Evolution and functional classification of vertebrate gene deserts.” *Genome Res.* **15**, 137. UCRL-JRNL-205731.

Molecular Radiation Biodosimetry

Irene M. Jones 04-ERD-076

Abstract

Our objective is to develop valid “triage” methods to minimize the civilian health consequences of a nuclear or radiological event. These methods will assess radiation exposure dose using new technologies for measuring early biochemical changes in cells, tissues, and bodily fluids of irradiated persons within minutes to a few days after exposure. This research will lay the scientific foundations for a new class of victim-management tools and medical assessment capabilities that are needed by first responders and medical clinics to focus limited medical resources on those who will benefit most after a radiation event.

The initial phase of the project will evaluate the radiation response of DNA damage proteins and gene expression changes in human cells and the engineering criteria for field use.

This project will lay the scientific foundations for capabilities to reduce the health consequences of exposure to ionizing radiation and will have broad civilian, military, and medical applications. These studies are expected to lead to devices that require only minutes to apply and have the resolution to enable reliable triage and medical assessment to distinguish between the truly injured and the “worried well”—those who are concerned but not in immediate danger—who can be expected in very large numbers. This proposal leverages ongoing work in the areas of low-dose effects of ionizing radiation on cells, DNA repair, radiation and radionuclide dosimetry, analytical chemistry, and microbial biology.

Mission Relevance

This project is relevant to national security, especially the management of persons exposed to ionizing radiation in a significant radiological or nuclear event.

FY05 Accomplishments and Results

In FY05 we (1) optimized three sample collection and processing methods; (2) began validation of high-priority biomarkers using cells from healthy people [additional lymphocytes irradiated ex vivo and cultured for 1 or 2 days were archived for future studies of messenger RNA (mRNA) and protein biomarkers]; (3) began validation of mRNA biomarkers in buccal cells (which come from the inner lining of the mouth) taken from head and neck cancer patients receiving radiotherapy (sixteen samples from four subjects had exposures ranging from 10 to 200 cGy); (4) built and successfully tested a prototype device that collects and processes buccal cells and enables rapid chromatography assay for protein biomarkers; (5) archived samples collected from patients using a prototype triage device; and (6) submitted a record of invention.

Publications

Jones, I. M. (2005). *Challenges of validating tools for radiation biodosimetry*. UCRL-ABS-209502.

Characterizing the Regulatory Genome: Transcription Factor Proteins and Gene Regulation Networks in Living Cells

Lisa J. Stubbs 04-ERD-084

Abstract

This research seeks to establish new strategies and technologies for characterizing transcription factor (TF) proteins and the regulatory pathways in which they are involved in specific types of living cells. Understanding TFs and their regulatory "targets" is key to constructing regulatory network models, an important goal in current efforts by both National Institutes of Health and DOE biology programs, including Genomics:GtL. Beginning with characterizing a subclass of human TFs, we will develop a pipeline for characterizing TF proteins and pathways with the goal of building robust regulatory network models. All basic methods and expertise will be fully extensible to regulatory network modeling in any species, from microbes to mammals.

If successful, the project will develop novel methods for identifying TF protein binding sites and "target genes" in living cells, and cluster TFs and target genes into metabolic, morphological, and developmental pathways for robust regulatory network models. In addition, we plan to recruit strong postdoctoral scientists with state-of-the-art expertise to add depth and breadth to our research in genetics, comparative genomics, biochemical technology, and computational modeling; and develop collaborative ties to university researchers who are leaders in this field.

Mission Relevance

Accurate models of gene regulatory networks will be key to understanding biological mechanisms that govern form and function in all types of living cells. We are developing projects with direct applications in environmental microbiology, cancer biology, and the regulation of human immune response. By building expertise in regulatory biology, biochemistry, and network modeling, this project supports missions in biodefense, environmental management, and bioscience to improve human health.

FY05 Accomplishments and Results

During FY05 we (1) completed a comprehensive catalog of human "zinc finger" transcription factors and related proteins in other species; (2) analyzed the human proteins for structural differences that translate into novel regulatory functions; (3) developed a novel and robust set of complementary technologies for TF target identification and systematically applied them to analyze 20 proteins; (4) explored new high-throughput strategies to identify DNA binding motifs and measure binding affinities; (5) began work to cluster TF proteins and targets in specific regulatory pathways; and (5) initiated a focused pilot study to apply our technology to the analysis of bacterial TFs.

Proposed Work for FY06

In FY06 we will (1) continue to develop, refine, and apply technology built in FY05, analyzing proteins with relevance to cancer biology, immunology, and human brain development; (2) establish technology to validate gene targets and to link TFs and target genes in pathways and interacting functional networks; (3) explore human TF variation to identify genetic factors leading to individual differences in susceptibility; and (4) transfer methods to generate target and binding-site data for a selection of TFs in "sentinel" model microbial species to permit us to model regulatory networks for a broad range of microbes—including those with unknown organisms encountered in environmental samples—permitting prediction of their functional properties.

De Novo Identification of Regulatory Regions in Intergenic Spaces of Prokaryotic Genomes

Patrick S. Chain 04-ERD-103

Abstract

This project will implement, test, and experimentally validate the results of an algorithm for genome-wide identification of candidate transcription factor binding sites (TFBSs) in prokaryotes. Most techniques used to identify regulatory regions rely on conservation between different genomes or have a predetermined sequence motif to perform a genome-wide search. These techniques cannot be used with new genome sequences. This project will apply a de novo search algorithm to identify candidate binding-site motifs in intergenic regions of prokaryotic organisms, initially testing the available genomes of the *Yersinia* genus. We will retrofit existing nucleotide pattern-matching algorithms, analyze the candidate sites and their target genes for meaningful patterns, and test these experimentally.

If successful, this project will produce a software program capable of identifying candidate intergenic sites important for gene regulation from properly annotated prokaryotic genomes. We will demonstrate this in *Yersinia*—a model biodefense, Category A group of pathogens—and follow up with experimental evidence that these regions are indeed involved in regulation. The ability to quickly characterize TFBSs will lead to increased understanding of how virulence pathways are modulated in biodefense-related organisms, and will help our understanding and exploration of gene regulatory networks and novel pathways for metabolic processes in environmental microbes.

Mission Relevance

The novel search algorithm developed in this project will provide an important tool for genome, transcriptome, and systems biology. It will allow scientists to explore gene regulation to better understand regulatory networks in prokaryotic organisms, and to apply this pathway information to those microbes relevant to the Laboratory's missions in environmental remediation and biodefense.

FY05 Accomplishments and Results

We have developed a number of scripts to parse data generated by DNA microarrays, which are used to cluster genes into groups that have similar expression patterns. We have made use of these scripts, along with alignment and motif searching tools, to identify DNA regions that are statistically over-represented among temperature-regulated genes and thus may be involved in their regulation. We identified that motifs corresponding to the *Escherichia coli* Crp and ArcA regulators' binding sites were present in promoters of many genes strongly upregulated one hour after *Yersinia pestis* is grown at human body temperature. This fits nicely with the Crp and ArcA pathways—catabolite repression and aerobic respiration—because *Yersinia* is required to switch to a different carbon source and a microaerobic environment.

Proposed Work for FY06

We plan to (1) expand our set of known TFBSs to include a number of putative binding sites under the control of unknown regulators, and redo our analyses; (2) continue to develop methods to identify novel binding sites in prokaryotic genomes; (3) construct cloning vectors that will help us evaluate putative TFBSs; (4) confirm the regulatory function of a small number of our identified TFBSs in vivo using our reporter constructs and reverse transcriptase polymerase chain reaction; and (5) refine our motifs to improve binding site detection, and identify possible virulence targets based on shared binding site motifs with known virulence factors. These will serve as the basis for future experiments in which genes of interest have been rendered inoperative.

Time-of-Flight, Secondary Ion Mass Spectrometry Measurement of Metabolites from Single Cells

Kristen S. Kulp 04-ERD-104

Abstract

Previous studies of microbial response to environmental stress have shown that adaptive response is reflected in changes in the most abundant metabolites. We propose to use time-of-flight secondary ion mass spectrometry (TOF-SIMS) to characterize metabolites in individual bacteria in order to facilitate modeling of metabolite fluxes. TOF-SIMS is a spectrometry technique that characterizes chemical composition and chemically maps the distribution of small molecules with the resolution needed to interrogate single cells. Our goal is to enhance bio-analytical instrument capabilities, perfect sample preparation methods, and apply statistical analysis to provide the sensitivity needed to measure bacterial metabolites in single cells.

This research will advance imaging mass spectrometry to an unexplored arena: the characterization of metabolites from an individual bacterium. This proposal will result in (1) quantitative demonstration of enhanced TOF-SIMS instrument sensitivity; (2) a reproducible sample-preparation method that renders

bacterial metabolites assessable to analysis; and (3) a useful multivariate spectral pattern recognition technique that can compare metabolite production from multiple individual cells. These experiments will be the first ever to analyze bacterial metabolites in single cells and will provide preliminary data to understand pathway fluxes and population interactions of environmentally stressed bacteria.

Mission Relevance

This project will provide the foundation for single-cell metabolomics and biological imaging, in support of LLNL's mission in bioscience to improve human health. This work is relevant to DOE's Genomics:GTL initiative by developing the capability to measure metabolites in single bacterium, and complements efforts to study spectral signature definition and intracellular compound localization in mammalian cells.

FY05 Accomplishments and Results

During FY05 we (1) installed a gold primary ion source on the TOF-SIMS instrument to enhance sensitivity and mass range; (2) established a spectral library of 41 bacterial metabolites and began quantifying instrument sensitivity gains using this library; (3) developed a usable sample-preparation device that enables us to analyze the chemical contents of a single bacterium; (4) applied chemometric statistical techniques to our data sets to show capabilities for separating and identifying molecular isomers of small molecules, amino acids, and different bacterial species; and (5) established the chemical sensitivity of the method by successfully separating seven sugar isomers. These accomplishments resulted in two records of invention.

Proposed Work for FY06

In FY06 we plan to (1) explore matrix deposition methods to increase ionization yields; (2) determine fragmentation patterns of pure metabolite standards and bacterial cell homogenates spiked with known metabolites; (3) further develop our method of crushing bacterial cells to expose the metabolites for analysis; (4) develop a planar chromatography method to separate small-molecule metabolites from large cell fragments; (5) optimize statistical analysis techniques to identify the key masses in the spectra from different samples; and (6) compare these key masses with the bacterial metabolite spectral library to identify the origin of important mass fragments.

Publications

Knize, M. G. et al. (2005). *Imaging and differentiating individual cancer cells and cell extracts using time-of-flight secondary ion mass spectrometry (ToF-SIMS)*. 53rd Ann. Mtg. American Society of Mass Spectrometry, San Antonio, TX, June 6–9, 2005. UCRL-ABS-210011.

Knize, M. G. et al. (2005). *Response of single cells and cell extracts to irradiation as detected by time-of-flight secondary ion mass spectrometry*. American Association for Cancer Research 96th Ann. Mtg., Anaheim, CA, Apr. 16–20, 2005. UCRL -ABS-210011.

Kuang, J. W. et al. (2005). *Mass imaging and tissue differentiation of mouse embryo sections using ToF-SIMS*. 53rd Ann. Mtg. American Society of Mass Spectrometry, San Antonio, TX, June 6–9, 2005. UCRL-ABS-211302.

Calcium Dynamics in Human Bone

Darren J. Hillegonds 04-ERI-009

Abstract

Previous work has shown the value of calcium-41 (^{41}Ca) accelerator mass spectrometry (AMS) in tracking bone health, primarily related to osteoporosis. This proposal broadens the ^{41}Ca assay to include impact on cancer treatment and survival. Our goal is to develop a minimally invasive method to detect and quantify perturbations in the turnover of bone when cancer spreads to the skeleton. Such a method could improve patient quality of life and survival through interactive modulation of anticancer or bone-saving therapy. Ultimately, our results could result in an early detection protocol that would enable therapeutic intervention prior to significant skeletal damage, enabling more effective treatment for what is considered an untreatable disease.

Our deliverable will be a minimally invasive method to detect transition from the primary cancer site to the skeleton, as well as the ability to quantify and closely monitor the progression of skeletal tumor growth.

Mission Relevance

By developing a minimally invasive method to detect the transition from a primary cancer site to the skeleton, this project will further LLNL's mission in bioscience and technology to improve human health and will support the National Institutes of Health and National Cancer Institute missions.

FY05 Accomplishments and Results

We have validated the ^{41}Ca assay in an animal model of metastatic bone disease, and also shown that the ^{41}Ca assay is similar to, but more sensitive than, available methods of assessing clinical response to anti-resorption drugs. We furthered development of a mathematical model of Ca homeostasis, which will enable us to apply our method in both research and clinical work, and a record of invention was filed for the diagnosis and assessment of skeletal-related disease using ^{41}Ca. We have not yet assessed ^{41}Ca kinetics in human cancer patients, as planned for FY05, because of a delay in getting approval from the scientific review committee at our collaborating institution, University of California, San Diego (UCSD). The UCSD protocol was re-submitted for review—we anticipate that advanced prostate cancer patients will be dosed by summer 2006.

Proposed Work for FY06

During FY06, we intend to finish development of the assay for human subjects and determine the best way to apply the new technology to medical science. Our main goal is to assess the pharmacokinetics of ^{41}Ca in patients with cancer via measurement of serial ^{41}Ca/Ca in urine and blood—tracer abundance versus time will allow us to calculate relevant measures of calcium homeostasis in individuals, using the mathematical model we have developed.

Publications

Fitzgerald, R. L. et al. (2005). "^{41}Ca and accelerator mass spectrometry to monitor calcium metabolism in end stage renal disease patients." *Clin. Chem.* **51**(11), 2095. UCRL-JRNL-217924.

Yang, M. (in press). " The bisphosphonate, olpadronate, inhibits skeletal prostate cancer progression in a GFP nude mouse model." *Clin. Cancer Res.* UCRL-JRNL-217938.

Single-Cell-Level Investigation of Cytoskeletal Response to External Stimuli

Amy L. Hiddessen 04-ERI-015

Abstract

Our objective is to develop a new platform, based on cell micropatterning, for analyzing the response of numerous single cells to controlled environmental cues. The long-term goal is to apply the platform to determine molecular regulators of fate commitment in single stem cells. Patterned cell arrays will be used to expose numerous single cells to chemical stimuli (e.g., cytokines), while cell labeling and light/fluorescent microscopy will be used to track cell responses in real time. This work will build a new capability for systematic analysis of signaling in mammalian and bacterial cells. Furthermore, determining the roles of external and internal signals in fate commitment has important implications in regenerative medicine and understanding aberrant signaling in disease.

A new, broadly applicable platform for parallel analysis of signal transduction at the single-cell level will be developed. By enabling measurements of how single cells respond to particular environments/signals, we will obtain new data that has significance in drug development and biosecurity (identifying factors that cause disease and targets for intervention), regenerative medicine, and development of predictive cell models. The cell array platform will continue to draw new applications (and collaborations) both inside and outside LLNL. Two applications, stem-cell fate decisions in response to chemical cues and host-cell cytoskeletal response to bacterial pathogens, are subjects of particular interest.

Mission Relevance

By developing a new platform for systematically examining the behavior of single cells in response to chemical and pathogenic signals, this work will contribute to biodefense applications in support of the Laboratory's national-security and homeland-security missions, as well as its mission in leveraging Lab expertise and capabilities to solve basic biomedical problems of worldwide interest. This research will also strengthen our collaborative relationship with the University of California.

FY05 Accomplishments and Results

In FY05, we achieved our major goal of developing new technology to array single live cells in an artificial environment where they can be controllably exposed to chemical signals. Our major accomplishments were (1) development of a novel chemical patterning method for constructing regioselective microarrays; (2) use of the patterning method to construct protein microarrays and single-cell and cell-cluster microarrays; and (3) successful demonstration of delivering chemicals to, and manipulating live cells after, capture on the array. The ability to array cells and deliver chemicals that stimulate or manipulate them is an essential step toward establishing the capability to study single-cell response to specially programmed environments.

Proposed Work for FY06

The focus of FY06 work will be to further develop the new cell microarray technology to support investigations of cell response to chemical signals. We plan to (1) optimize surface chemistry to maximize viability of specific cell types, including broadening the scope of the platform to accommodate both suspension and adherent cell types and (2) continue to test and optimize the array environments for tracking and measuring cell response, including development of preliminary assays for delivering signals (cytokines and growth factors to stem cells, or pathogenic peptides to host cells) and for measuring response (cell surface marker labeling approaches to monitor proliferation and differentiation). Our FY06 goals also include preparing manuscripts for peer review.

Publications

Bearinger, J. P. et al. (2005). *Biomolecular patterning via photocatalytic lithography*. Nano Science and Technology Institute Nanotechnology Conf. and Trade Show, Anaheim, CA, May 8–12, 2005. UCRL-PROC-210154.

Bearinger, J. P. et al. (2005). *Photocatalytic patterning of biomolecules*. Materials Research Society Spring Mtg., San Francisco, CA, Mar. 28–Apr. 1, 2005. UCRL-ABS-208684.

Hiddessen, A. L. et al. (2005). "Bioengineered tools for analysis of cellular response to chemical signals." *Biophys. J.* **88**, 523A. UCRL-ABS-209314.

Understanding the Mechanism of Human P450 CYP1A2 using Coupled Quantum–Classical Simulations in a Dynamical Environment

Erik W. Draeger 04-LW-048

Abstract

The reaction mechanism of the human P450 CYP1A2 enzyme plays a fundamental role in research to understand the effects of environmental carcinogens and mutagens on humans. In this project, we propose development and use of cutting-edge theoretical computational methods to perform the first detailed study of a human P450 reaction mechanism. To perform computational simulations of unprecedented accuracy, we developed a dynamic quantum–classical technique in which ab initio molecular dynamics (MD) is coupled with classical molecular mechanics (MM) to create a quantum mechanical–molecular mechanical hybrid (QM/MM). This new tool will provide the accuracy needed to address complex, large biological systems in a fully dynamic computational environment.

The expected result of this work is to elucidate the catalytic mechanism of human P450 CYP1A2 oxidizing caffeine. The completion of this project will also result in a powerful new computational tool that will allow research groups currently using ab initio methods to study significantly larger systems over longer time scales.

Mission Relevance

This project supports the DOE mission in bioscience to improve human health by developing the computational methods and capabilities to advance understanding of complex biological systems and predict their behavior.

FY05 Accomplishments and Results

In FY05 we carried out our first QM/MM simulations and explored further algorithmic improvements to make the QM/MM tool more flexible and efficient. To this end, we coupled the ab initio molecular dynamics code JEEP to the large-scale classical MD code NAMD, using a socket interface to preserve future transferability. We implemented covalent bonding across QM/MM boundaries using constrained link atoms, and verified energy conservation to 10^{-8} eV/iteration. We performed detailed ab initio calculations of the charge transfer in a porphyrin-ring model system to quantify the size effects in QM/MM porphyrin calculations. This project has produced a new QM/MM simulation capability as well as a detailed understanding of how to accurately apply it to biological systems.

Investigation of AAA+ Protein Machines that Participate in DNA Replication, Recombination, and Response to DNA Damage

Dorota Sawicka 04-LW-049

Abstract

The objective of the project is to structurally characterize AAA+ proteins and complexes involved in DNA replication, recombination, and response to DNA damage. Because DNA carries all of the genetic information for life, understanding DNA metabolism is of utmost importance at the molecular level, where the AAA+ proteins play critical roles and function as protein machines. We expect to provide new insights into the structure, interactions, and functional mechanisms of the eukaryotic AAA+ protein families involved in DNA metabolism by (1) applying advanced sequence comparison methods; (2) comparative modeling; (3) molecular dynamics simulations; (4) and mutational analysis.

We expect to characterize all 20 currently identified eukaryotic AAA+ protein families and their complexes involved in DNA management processes. We believe the computationally obtained structural models combined with well-defined experimental verification will provide insight into structure, interaction, and dynamics of these AAA+ proteins. Because of the lack of such information, we expect our results will greatly facilitate future experimental biochemical studies.

Mission Relevance

The application of computational methods to understand some of the central processes involved in faithfully copying and maintaining genetic information supports LLNL's mission in bioscience and technology to improve human health, and may lead to approaches for mitigation of pathogen infectivity.

FY05 Accomplishments and Results

We have (1) generated a model for the Ctf18 and proliferating cell nuclear antigen (PCNA) interaction and proposed a conserved C-terminal motif of Ctf18 that mediates interaction with the dcc1-Ctf8 subcomplex; (2) tested the proposed binding motifs using rational site-directed mutagenesis; (3) studied protein–protein interactions using Ctf18 variant proteins in in vitro binding assays and in functional assays for PCNA loading onto DNA, in collaboration with the Memorial Sloan-Kettering Cancer Center; (4) confirmed that the C-terminal region of Ctf 18 is important for binding to dcc1/Ctf8 and to RFC2-5, the other clamp loader subunits; and (5) assayed peptides designed to mimic predicted parts of Ctf18 to determine the relative affinities for PCNA and to determine if these peptides block the Ctf18-dependent loading of PCNA onto DNA.

Publications

Venclovas, C., M. de Carvalho-Kavanagh, and D. Sawicka. (2004). *The alternative clamp loader subunit Ctf18: A model for its interaction with PCNA, evolutionary conservation and relationship to RFC1 and Rad17*. American Society for Microbiology Conf. DNA Repair and Mutagenesis: From Molecular Structure to Biological Consequences, Southampton, Bermuda, Nov. 14–20, 2004. UCRL-POST-207252.

A Single-Molecule Study of the Movement of a DNA Sliding Clamp

Daniel Barsky 04-LW-069

Abstract

We propose a combined experimental and computational approach to investigate the interaction between DNA and a sliding-clamp protein. By encircling DNA, sliding-clamp proteins have been thought to freely slide along DNA, a movement that defies expectations because the protein's high positive charge should create a strong electrostatic attraction with DNA. It is also mysterious that the clamp proteins can proceed past sizeable distortions in DNA. By single-molecule fluorescent energy transfer (FRET) experiments, analytical calculations, and molecular dynamics simulations, we are studying the details of the motions of the bacterial sliding clamp along DNA, including clamp speed, direction, and rotation of the clamp as well as clamp loading and unloading.

Using high-end computing, analytical theory, state-of-the-art biochemistry, and single-molecule measurements, this project endeavors to translate the atomic-scale description of biomolecular interactions to nanometer- and micrometer-scale interactions, an important feat in chemistry and material science. Moreover, this project has the potential to answer some fundamental scientific questions, such as how the replication complex holds on to and simultaneously follows two spiral helices. Many details of the interactions between two dynamically interacting biomolecules will be provided. The proposed project brings together computations and experiment in a new way that holds great promise in biology, chemistry, material science, and perhaps nanotechnology.

Mission Relevance

By answering fundamental questions in biology concerning the details of DNA replication and repair, this project supports DOE and LLNL's mission in bioscience to improve human health, and has possible applications relevant to environmental biology as well as the homeland-security mission to prevent and counter bioterrorism.

FY05 Accomplishments and Results

We re-expressed the pol III beta subunit DNA sliding clamp as an intein fusion protein, and thereby labeled it with a FRET donor dye (Alexa 488) in a highly specific, efficient process. We also redesigned our DNA substrate to have a short stretch of base-paired DNA region for the clamp motion (30 and 90 basepairs long), labeled with a FRET-acceptor dye (Alexa 647). The FRET between the clamp and the labeled plasmid were observed and analyzed, and we inferred a diffusion constant 100 to 1000 times smaller than expected for a freely diffusing clamp. Computationally, our 2-ns molecular dynamics simulations were analyzed in terms of competitive/cooperative DNA–protein interactions. This work was presented at national meetings and featured in *Chemical and Engineering News*.

Proposed Work for FY06

Fluorescence correlation spectroscopy (with FRET) will be used toward a more biologically driven set of goals: (1) monitoring the translation speed of the beta clamp on plasmid DNA in the presence of aberrant structures and (2) monitoring the loading and unloading of the clamp on DNA. The first goal may require immobilizing the plasmid substrate with a surface attachment, but we can control for changes in clamp mobility by comparison with our previous solution-phase measurements. We will also observe changes to the mobility in the presence of the polymerase with and without lesions. Finally, we will simulate longer timescales and assess correspondence to experiments. The results should produce high-visibility publications.

Publications

Barsky, D. et al. (2005). *Nanotraincars moving down DNA tracks: Single molecule FRET and simulations of a DNA sliding clamp moving along DNA*. LLNL Science Day 2005, Livermore, CA, May 23, 2005. UCRL-POST-211305.

Emerging Contaminants: Application of Microarray Technology to the Detection of Mixtures of Endocrine-Active Agents

Linda C. Hall 05-ERD-008

Abstract

The health effects of previously unidentified (or unrecognized) environmental contaminants are a prominent concern of regulatory agencies and the general public. Of particular interest are endocrine-disrupting chemicals (EDCs), including hormone agonists and antagonists, which pose a potential risk to surface water and groundwater due to wastewater treatment facility and septic system discharges. The number and structural diversity of EDCs makes it impractical to develop detection methods on a chemical-by-chemical basis. This project proposes to use DNA microarray technology and synchrotron radiation-based Fourier transform infrared (IR) spectromicroscopy to develop a biological sensing system to detect unknown EDCs present individually or as mixtures in water.

The principal result of this research will be a validated sensing system (bioassay) capable of detecting EDCs that are present in ambient water individually or as mixtures. We will (1) characterize a human

cell line and demonstrate the responsiveness of that cell line to the major categories of EDCs; (2) acquire novel and fundamental knowledge of the timing and IR spectromicroscopy-based signatures of gene alteration in response to EDCs; (3) characterize a genetic response profile for EDCs using microarray- and IR-based analyses of exposed cells; (4) define the sensitivity and dose-response of microarray and IR techniques; and (5) computationally integrate genetic responses to identify characteristic profiles of individual EDCs and mixtures.

Mission Relevance

The proposed research combines state-of-the-art technologies of gene expression microarrays and IR spectromicroscopy as the basis of a biological assay to detect mixtures of unknown EDCs in ambient water. This measurement tool is based on novel and fundamental scientific data that addresses a water contamination problem of both statewide and national interest, and supports LLNL's mission in environmental protection.

FY05 Accomplishments and Results

In FY05, we (1) used quantitative techniques to measure the presence and levels of the messenger RNA (mRNA) and protein of four steroid hormone receptors (SHRs) in three human cell lines—NIHOVCAR3, T47-D, and MCF-7; (2) established that the T47-D and MCF-7 lines contain both the mRNA and protein of four SHRs of interest: estrogen receptor (ER) alpha, ER beta, the androgen receptor, and the thyroid hormone receptor; (3) established that constitutive levels of SHRs are sufficient and will not require stable transfection with an SHR construct; (4) conducted quantitative dose and time course experiments to evaluate the functionality of SHRs; (5) conducted preliminary experiments at LBNL's Advanced Light Source; and (6) conducted two sets of microarray experiments.

Proposed Work for FY06

In FY06, we will continue time course and dose experiments to delineate IR spectromicroscopy signatures in human cells exposed to individual EDCs. We will compare IR spectromicroscopy and microarray signatures from EDC-exposed cells to establish whether qualitative agreement exists between the two techniques. If IR spectromicroscopy can detect changes in the magnitude of EDC-induced genetic response, and we can validate these data using microarray analyses, we will work to use spectromicroscopy to define the timing of the optimum genetic response caused by EDC exposure. We will also conduct microarray experiments to characterize the basic gene-expression profiles associated with the individual prototypical EDCs identified in this project.

Publications

Hall, L. et al. (2005). *Cellular and molecular biology studies of environmental contaminant toxicology*. UCRL-POST-216096.

Liu, N. et al. (2005). *Thyroid hormone-induced cell proliferation and gene expression in human breast cancer cells*. UCRL-ABS-215989.

Chemical Specific Cellular Imaging of Biofilm Formation

Julie L. Herberg 05-ERD-026

Abstract

Biofilms, colonies of micro-organisms living on surfaces, exhibit unique biochemical pathways during formation and play important roles in human health and environmental processes. The most common bacterial groundwater pollutants in the U.S. form biofilms, but little is known about the dynamics of biofilm development and metabolism. We propose to combine magnetic resonance imaging (MRI) and nuclear magnetic resonance (NMR) analysis with computer simulations of biofilm metabolite profiles to obtain unique 3-D molecular, structural dynamics, and functional information on micrometer-scale biofilms. We will apply in situ and ex situ scientific tools to quantitatively understand the morphology and chemically specific nature of the films as they form onto various surfaces, including minerals, metals, and implant materials.

If successful, this project will be the first to image the dynamic formation of a biofilm; correlate structure, dynamics, and function of metabolites in the process of biofilm formation; and develop a predictive model of biofilm formation. This research will facilitate the study of how individual cells and cell clusters react to the introduction of a pathogen or virus into the environment, or how the biochemical behavior of mutated (cancerous) cells differs from that of normal cells. The potential practical consequences of such knowledge are vast: from prevention of bacterial infection from catheters and prosthetic implants, to mediating carbon sequestration, to prevention of bacteria-induced corrosion of nuclear waste containers.

Mission Relevance

Biofilms, directly implicated in contamination, energy losses, and medical infection, are resistant to control, inhibition, and destruction. The deeper understanding of intercellular communication and signaling in biofilms developed in this project will support LLNL missions in homeland security, water security, environmental protection and remediation, and biotechnology for improving human health.

FY05 Accomplishments and Results

Durning FY05, we (1) successfully grew *Pseudomonas aeruginosa* biofilms in a reactor; (2) developed techniques to use MRI to measure both flow and diffusion through the biofilm, obtaining resolutions on the order of 20 µm square, and began working on ways to decrease the resolution even further with NMR microcoils; (3) obtained ^{1}H and ^{13}C solution state and solid state NMR spectra of supernatant and biofilm cells; (4) began using principle component analysis to identify the metabolites involved in the growth process of *P. aeruginosa*; and (5) began to establish parameters to perform in situ, time-dependent ^{1}H and ^{13}C NMR to watch the metabolite signatures during the growth stage.

Publications

Herberg, J. L. et al. (in press). "Characterization of local deformation in filled-silicone elastomers subject to high strain—NMR MOUSE and magnetic resonance imaging as a diagnostic tool for detection of inhomogeneities." *Polym. Degrad. Stabil.* UCRL-JRNL-214533.

Innovative Copolymer Complex to Inhibit the Transport of Biological Aerosols

Paula W. Krauter 05-ERD-027

Abstract

We are formulating and optimizing a polymer solution for immobilizing and suppressing hazardous particle resuspension and migration by encapsulation, chemical binding, or static attraction. Building on our development of a particle-binding solution specific to aerosolized spores, we intend to evaluate the formula for compatibility with a biocide, sporicide, or germination inhibitor; develop a copolymer formulation that specifically targets beryllium particles; and test the modified copolymer complex with hazardous particles. Preliminary tests suggest that formulation modifications (acidic copolymers with specific functional groups) may effectively encapsulate, and preferentially bind, beryllium particles.

We intend to produce (1) a copolymer-based solution formulated to be anionic to provide coulombic attraction to cationic spores; (2) publications in peer-reviewed journals regarding the development, characterization, and evaluations of the formulations; and (3) a deeper understanding of how to limit the re-suspension of contaminant particles because of application of the copolymer. We also intend to advance basic understanding of local shear stress, particle shear stress, and particle–copolymer adhesion strength under a variety of flow conditions.

Mission Relevance

This project addresses the national security mission to counter proliferation and the use of weapons of mass destruction by developing a new technology to inhibit effective dispersal and migration of biological warfare agents. This will increase the safety of emergency response personnel by abating breathing zone concentrations, as well as limit the potential for spore re-suspension and migration.

FY05 Accomplishments and Results

Accomplishments in FY05 include development of 11 film-forming copolymer formulations designed to use electrostatics to attract fluidized spores, and physical and chemical characterization of each formula. Selection of three of these formulas were based on measurements of particle/copolymer solution binding characteristics in wind-tunnel tests using highly fluidized 1- to 5-µm spores, and we determined the range of shear for different drop diameters and approach velocities by using simulations of droplets with sediments. When adhesion strength of the copolymers was challenged in several wind tunnel test configurations, it became apparent that the ability to spray a viscous formula was a limiting factor in the test methodology. Several copolymer formulas have qualities that allow for even application with a good contact angle, while others have good adhesion strength in high airflow. The spore/copolymer adhesion tests were based on particle resuspension after application of the copolymer, however, the selection criteria for the copolymer also includes other factors—the ability to mist in a spraying device, neutral pH, and electrostatically negative and low surface tension. In each adhesion test, the NS-2 formulation was one of the top performing copolymers and met all selection criteria requirements. A provisional patent has been obtained for the concept and formulations.

Proposed Work for FY06

Objectives for our work in FY06 are (1) optimizing the copolymer formulation for biological particle encapsulation; (2) testing selected formulations in a room-size test chamber; and (3) leveraging ongoing testing at the Contained Firing Facility (Site 300) to investigate candidate copolymer encapsulants formulated for beryllium and depleted uranium particulates. At the end of FY06, we will recommend a copolymer formula for the inhibition of biological particulate re-aerosolization, and we will have conducted proof-of-concept testing for a second class of hazardous particulates represented by beryllium and depleted uranium. The modeling effort will provide information on particle–copolymer droplet interactions to understand the effects of particle displacement by the copolymer droplet.

Publications

Hoffman D. M., P. Krauter, and A. Vu. (2005). *Thermal analysis of some water soluble copolymers*. UCRL-JRNL-217772.

Zalk D. M. and P. W. Krauter. (2005). *Inhibiting the transport of terrorism-related hazardous aerosols to aid decontamination efforts and prevent reaerosolization*. International Occupational Hygiene Association 6th Triennial Scientific Conf., Pilanesberg National Park, South Africa, Sept. 19–23, 2005. UCRL-ABST-216575.

Characterizing Hypothetical Proteins

Michael P. Thelen 05-ERD-064

Abstract

Nearly half the proteins inferred from microbial genome sequences are unconfirmed and bear little resemblance to any known proteins, yet many of these “hypothetical proteins” are important to natural processes in the environment. We are developing technologies for characterizing these novel proteins from natural, uncultivated microbial communities by direct isolation from the environment. Protein

distribution in a community will be determined by fractionating proteins and analyzing sequences to identify source organisms. Extracellular proteins, multiprotein complexes, and membrane-associated proteins will be purified, identified, and correlated with gene contextual information, and their functions determined using computational tools and biochemical assays.

This research will advance our understanding of how microbes affect their natural environment, particularly the molecular basis of iron oxidation, biofilm formation, and evolution of biological mechanisms such as acid tolerance. We will develop new methods to determine the function of novel proteins, which is highly relevant to genome research and to other microbial studies at LLNL. Important insights into protein function will result from both computational modeling and biochemical assays. The project will establish unique expertise in the emerging area of proteogenomic analysis.

Mission Relevance

This project will support LLNL's environmental management mission by developing new approaches to understanding natural microbial systems that influence metal-contaminated environments. It also addresses DOE missions in energy since these microbial systems exacerbate environmental problems caused by mining of principle energy sources (e.g., coal and uranium). This will provide an excellent starting point for future work supporting DOE Programs in biosciences and environmental science.

FY05 Accomplishments and Results

We examined proteins and DNA isolated from microbial biofilm communities associated with the generation of acid mine drainage. Protein sequences were correlated with mass spectrometric proteome data and genome sequences from a related community. (These findings were published in *Science*.) We detected many new proteins in addition to the 2,000 identified in our original dataset, added new DNA sequence to the *Leptospirillum* group II genome, characterized several abundant extracellular proteins, and performed computational analyses on about 200 protein sequences from the dominant bacterial species in the community. Biofilm cellular membranes were fractionated using sucrose density gradient ultracentrifugation, and membrane proteins were isolated using selective detergent extraction.

Proposed Work for FY06

In FY06, we will (1) develop chromatographic separation methods for high-molecular-mass protein complexes, and identify component proteins by N-terminal sequence analysis; (2) cultivate single bacterial and archaeal isolates to improve methods for membrane protein isolation; (3) continue to test specific detergents for efficacy in purification of membrane proteins; (4) implement a screening method to analyze many proteins in parallel, specifically for hydrolase and oxidoreductase enzymatic activities; and (5) analyze DNA sequencing data and mass spectrometry proteomic data for new hypothetical proteins and protein variants.

Publications

Beernink, P. T. et al. (2005). “Specificity of protein interactions mediated by BRCT domains of the XRCC1 DNA repair protein.” *J. Biolog. Chem.* **280**, 30206. UCRL-JRNL-218035.

Ram, R. J. et al. (2005). “Community proteomics of a natural microbial biofilm.” *Science* **308**, 5730. UCRL-JRNL-218041.

Comparative Analysis of Genome Composition with Respect to Metabolic Capabilities and Regulatory Mechanisms

Patrik M. D’Haeseleer 05-ERD-065

Abstract

Given the glut of gene sequence data, comparative genomics is essential for leveraging existing knowledge. However, most approaches are limited to closely related species. We intend to study a large collection of bacterial genomes at the level of gene content rather than precise sequence similarity, allowing us to take advantage of even remotely related species. By integrating data on gene function and species phenotype, we intend to elucidate genotype-to-phenotype mapping, with particular emphasis on metabolic processes and regulatory mechanisms. Our modeling tools to decompose the genome composition will include non-negative matrix factorization, linear, bilinear, and Bayesian models, which will be validated against published data and supplemented by targeted experimental validation.

The patterns we discover in gene composition across the spectrum of bacterial genomes will increase understanding of which genes, gene classes, pathways, etc. are associated with or required for specific bacterial phenotypes, as well as yielding computational predictions of function for many unknown genes. Based on a list of genes in a newly sequenced genome (or even an unassembled environmental “shotgun” sequence), we expect to predict the metabolic processes and regulatory mechanisms and how these regulatory mechanisms respond to perturbations, which will give us insight on modifying or exploiting the organism(s) in question. Such a predictive capability for genotype-to-phenotype mapping is crucial for analyzing the flood of new sequence data.

Mission Relevance

The predictive capability for genotype-to-phenotype mapping created in this project will address goals of DOE’s Genomics:GtL program to gain predictive mastery of the microbial world. Future applications of the capability will also support LLNL’s missions in homeland security, environmental assessment and management, and biosciences to improve human health.

FY05 Accomplishments and Results

After a mid-year start, we began gathering and organizing genotype and phenotype data on a large number of microbial species, initially focusing on integrating the European Molecular Biology Laboratory's Search Tool for the Retrieval of Interacting Genes/Proteins database (110 species by 20,000 clusters of orthologous groups) with phenotypic descriptors from the Institute for Genomic Research's genome properties. An initial decomposition of this data showed some groups of genes that are specific for (or specifically absent in) certain phylogenetic lineages. More interestingly, we also identified some components showing very strong correlations with phenotype rather than phylogeny, including growth temperature, human pathogenicity (obligate parasites), animal pathogenicity (spore-forming), methanogenesis, and photosynthesis. We also identified a number of promising additional sources for phenotype data.

Proposed Work for FY06

For FY06, we plan to (1) integrate additional genotype and phenotype datasets, and continue refining the genome decomposition method; (2) develop a statistical (Bayesian) model to capture the genotype-to-phenotype mapping in a more quantitative fashion, and analyze this with respect to the metabolism of single species and communities; (3) begin building a compendium of bacterial transcription factors and transcription factor binding sites, including evaluating existing (and possibly designing novel) algorithms to find such binding sites; and (4) analyze experimentally determined binding sites in two or three model organisms, as well as new chromatin immunoprecipitation data generated by our collaborators (starting with *E. coli,* and possibly *Rhodobacter sphaeroides*).

Chemical Synthesis of Vaccines

Lynn D. Suer 05-ERD-075

Abstract

New technology enables the construction and chemical synthesis of DNA sequences as long as 31.44 kb. Synthesizing the entire cDNA construct of viruses makes it possible to introduce defined mutations that produce an attenuated vaccine strain without sacrificing immunogenicity. This project will use chemically synthesized cDNA for in vitro production of attenuated viruses and demonstrate the effectiveness of this method by synthesizing an attenuated strain of West Nile virus (WNV). The project scope includes viral synthesis, comparison of the synthesized vaccine to several conventional vaccines, and a demonstration of vaccine efficacy.

The goal of this project is to demonstrate the efficacy of synthesizing entire cDNA constructs of viruses. If successful, this technology may be used as a more efficient method of producing influenza vaccines, such as for avian influenza, and a foot and mouth disease vaccine.

Mission Relevance

By demonstrating the effectiveness of chemically synthesized cDNA vaccines, this project supports the Laboratory's mission in biodefense and biotechnology to improve human health. More generally, this project helps to demonstrate that biology has the potential to provide a ready source of engineering designs and strategies for macromolecular machines for a wide range of national security and other mission-relevant applications.

FY05 Accomplishments and Results

We obtained U.S. Department of Agriculture permits to receive RNA of WNV and the live virus, and Livermore Laboratory Biosafety Operations Committee approval for sequencing of the DNA of WNV. We will soon have approval for virus growth and the mouse experiments, and for use of the biosafety level 3 lab at the University of California at Davis.

We obtained RNA of an unsequenced mouse-attenuated WNV virus, ETH 76b, from the University of Texas Medical Branch in Galveston, and amplified the entire genome, except for the 3-prime end. In a second approach, primers successfully amplified 4-kb segments of the viral genome by polymerase chain reaction (PCR). In a third approach, primer-walking sequenced 30 of 33 total PCR reactions.

Proposed Work for FY06

We will complete synthesis of the attenuated strain of WNV, identify virulence factors for the virus, chemically assemble its cDNA, add a cytomegalovirus promoter, and use this preparation to coat gold beads for injection into mice. The second phase of our research will consist of three mouse experiments. First, the lethal dose in 50% of subjects of WNV in BALB/c cells will be established, then the amount of ETH 76b necessary to protect mice from a WNV challenge will be established. Finally, we will compare synthesized vaccine with conventional vaccines by injecting groups of mice with the synthesized cDNA or with one of two conventional WNV vaccines and then challenging them. Serum and spleen cells from the different groups of mice will be tested for antibody and cytokine production, and the results will be compared.

Development of a Chemoenzymatic-like and Photoswitchable Method for the Ordered Attachment of Proteins to Surfaces

Julio A. Camarero 05-LW-018

Abstract

Protein arrays are ideal tools for rapidly analyzing whole proteomes as well as for developing reliable and cheap biosensors. The objective of this proposal is to develop a new entropically activated ligation method based on the protein trans-splicing process. This method will be used to generate spatially addressable arrays of multiple protein components by standard photolithographic techniques. Key to our approach is the use of protein trans-splicing, a naturally occurring process that will allow us to create a truly generic and highly efficient method for the covalent attachment of proteins through their C-terminus to any solid support.

We expect to achieve selective and efficient attachment of any protein to solid supports through its C-terminus. This highly selective process will not require the prior purification of the proteins to be immobilized. Chemical modification of the C-intein polypeptide with photocleavable protecting will allow us to use standard photolithographic techniques to create protein chips with thousands of different protein combinations. These chips could be used for proteomic analysis, drug discovery, and biosensing.

Mission Relevance

Because protein arrays are very efficient reagents for the parallel analysis of whole proteomes, the technology developed in the project will support LLNL missions in national security, homeland security, and bioscience and technology to improve human health. In addition, developing new methods for the rapid and efficient creation of protein chips is one of the top priorities of DOE's Genomics:GtL program.

FY05 Accomplishments and Results

In FY05, we (1) cloned, expressed, and purified the N-intein polypeptide fused to model proteins—the maltose-binding protein (MBP) and enhanced green fluorescence protein (EGFP); (2) chemically synthesized the C-intein polypeptide; (3) characterized, in solution, the protein trans-splicing process mediated by the DnaE intein, calculated kinetics of the process as well as the binding affinities for both DnaE intein fragments; (4) selectively attached the C-intein polypeptide to a modified glass surface; (5) synthesized the amino acid Fmoc-Gly(nitroveratryl)-OH and used it to successfully synthesize a photocaged model peptide; and (6) showed that MBP and EGFP can be selectively attached to C-intein-coated glass slides through protein trans-splicing. On the basis of this work, a patent application was filed for our method of attaching proteins to surfaces.

Proposed Work for FY06

In FY06, we will (1) clone, express, and purify several N-intein fusion proteins, including DsRed, EGFP, and several *Yersinia pestis* YOP-related proteins; (2) immobilize these proteins onto C-intein-coated glass slides; (3) modify different C-intein glycine residues with Fmoc-Gly(nitroveratryl)-OH, then test these modified inteins for their ability to trans-splice before and after the photocaged group has been removed; (4) create the first protein chips by using DNA microarrayers and photolithographic techniques; and (5) create a protein microarray containing *Y. pestis* YOP-related proteins.

Publications

Camarero, J. A. (2006). "New developments for the site-specific attachment of proteins to surfaces." *Biophys. Rev. Lett.* **1**(1), 1. UCRL-JRNL-212352.

Camarero, J. A. and A. R. Mitchell. (2005). "New methods for the synthesis of proteins by native chemical ligation using Fmoc-based chemistry." *Protein Pept. Lett.* **12**, 723. UCRL-JRNL-209192.

Camarero, J. A. and Y. Kwon. (in press). "New tools for the site-specific attachment of proteins to surface." *Proc. 19th American Peptide Symp.*, San Diego, CA, June 18–23, 2005. UCRL-PROC-213182.

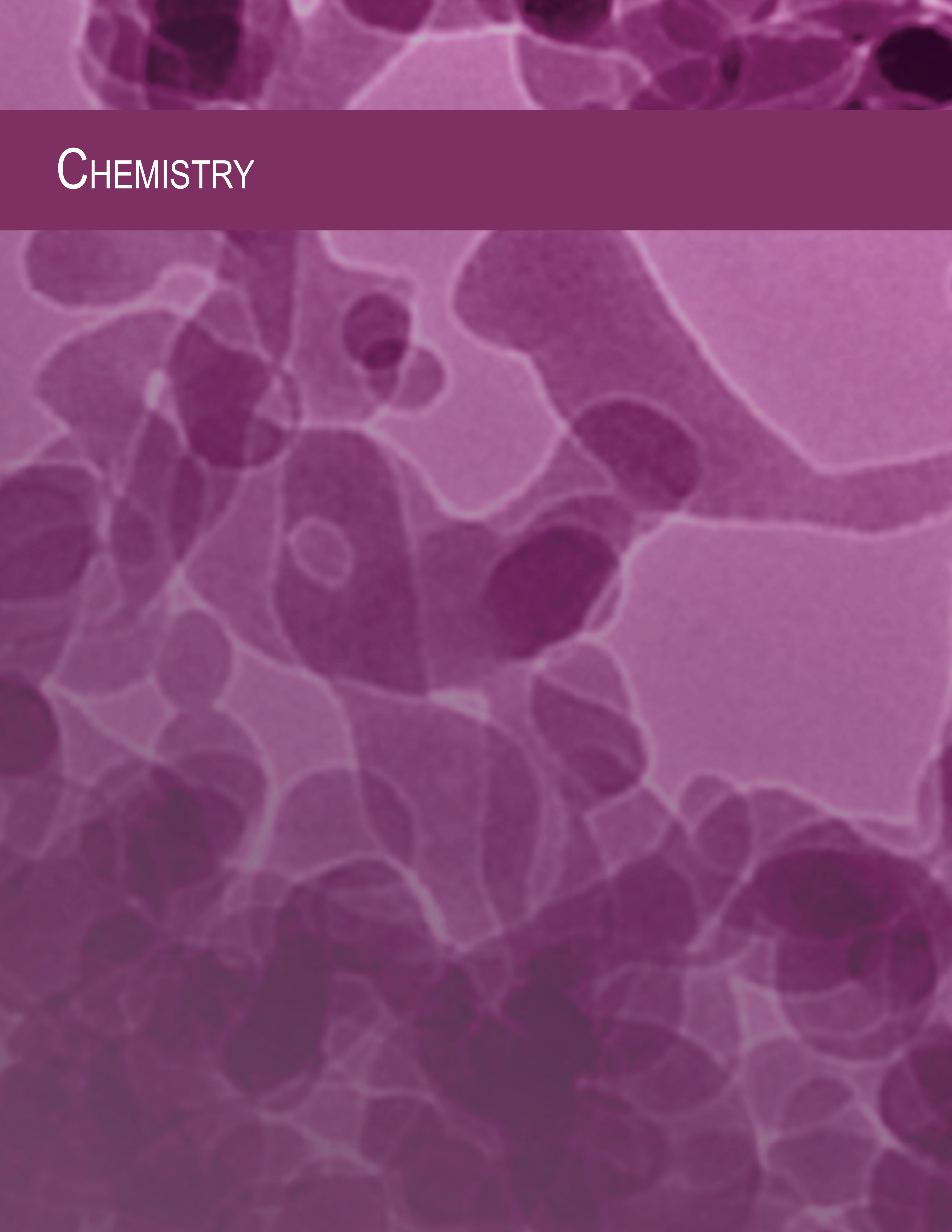
Camarero, J. A., Y. Kwon, and M. A. Coleman. (2005). "Chemo selective attachment of biologically active proteins to surfaces by expressed protein ligation and its application for protein chip fabrication." *J. Am. Chem. Soc.* **126**, 14730. UCRL-JRNL-205482.

Coleman, M. A. et al. (in press). *Applications of functional protein microarrays: current progress and future challenges*. New York: Humana Press. UCRL-BOOK-209049-DRAFT.

Kwon, Y. and J. A. Camarero. (2005). "Selective immobilization of proteins onto surfaces through split-intein mediated protein trans-splicing." *Biopolymers* **80**, 598. UCRL-POST-213225.

Kwon, Y., M. A. Coleman, and J. A. Camarero. (in press). "Selective immobilization of proteins onto solid support through split-intein mediated protein trans-splicing." *Angew. Chem. Int. Ed.* UCRL-ABS-210127.

CHEMISTRY



Development of a Virtual Crystallizer

John Campbell 03-ERD-051

Abstract

The capability to predict how crystals grow is a valuable tool for any industry involved in commercialized crystal production. We are developing a computer model capable of predicting growth from a 1-cm-size seed to a large, 60-cm crystal using actual crystallizer system conditions. The model will use equations governing the fundamental physics and chemistry of crystal growth along with mass transfer to crystal faces determined using a computational fluid dynamics (CFD) model, coupled with a large-scale crystal growth model that calculates growth rates for the individual faces and allows them to evolve in time to form a three-dimensional crystal. We will experimentally validate the model with potassium dihydrogen phosphate (KDP) crystals grown from 1 to 60 cm.

Expected results include (1) an understanding of the sensitivity of growth to various parameters; (2) a method for optimizing growth conditions; and (3) the ability to grow reproducibly a crystal with desired dimensions and properties. The immediate impact of our investigation will be to increase the quality and yield of rapid-growth KDP and deuterated KDP crystals used for frequency conversion and large-aperture, fast optical switches in large inertial-confinement-fusion (ICF) laser systems worldwide.

Mission Relevance

Developing the scientific foundation for growing crystals of the desired size and quality will benefit the broad array of efforts worldwide to develop crystalline materials for laser systems, optical components, and photonics applications. This technology will enable advances in high-energy-density physics that will benefit LLNL missions such as stockpile stewardship and ICF.

FY05 Accomplishments and Results

For FY05, the focus was to complete experiments to elucidate kinetic versus mass-transfer contributions and incorporate these results into the virtual crystallizer growth model. The experimental effort had three major components designed to determine growth kinetics and mass-transfer coefficients: (1) NASA Ames half-scale heat/mass-transfer experiments; (2) large-scale crystal growth runs; and (3) flow-cell and 20-L-tank experiments. A 1000-L growth system was modified to allow salt addition under stable conditions during a run to grow crystals of uniform *x,y,z* dimension (controlled habit). Our ultimate goal at completion was to demonstrate habit control by reproducibly growing record-size crystals of KDP (>320 kg) using the predictive capabilities of the virtual crystallizer model. So far, the model has been tested by confirming its ability to successfully simulate the growth of nine KDP crystals that all weighed more than 200 kg.

Laser-Initiated Nanoscale Molecularly Imprinted Polymers

Bradley R. Hart 03-LW-047

Abstract

We propose to synthesize nanoscale molecularly imprinted polymers (MIPs) that will benefit applications such as chemical and biological sensing and microscale separations and catalysis. This work couples recent advances in the design, synthesis, and evaluation of MIPs with LLNL's expertise in lasers, microscopy, and advanced materials synthesis, and will establish an entirely new and versatile route to practical MIP systems. This will be accomplished by using a focused laser source to pattern polymer features on a surface covered with a solution containing polymerizable monomers, a photoinitiator, and template molecules. Progress in this research would represent significant, publishable advances in molecular imprinting. The proposed work will establish an entirely new and versatile route to practical MIP systems and also overcome the limitations associated with traditional MIP systems.

Mission Relevance

This project supports the national security mission by furthering LLNL's work in remote sensing, monitoring, and assessment technologies for detecting signatures of chemical, biological, and nuclear activities, as well as detecting chemical and biological weapons themselves.

FY05 Accomplishments and Results

We examined the layering of a thin, inert polymer on top of the selective material, as well as the use of polymer-bound fluorescent probes, as methods to prevent much of the nonspecific binding on the surface of imprinted material. While fabrication of such materials was achieved, the background signal from fluorescence was too high to achieve detection of analytes. In view of recent reports in the literature that present solutions to problems with the use of surface-enhanced Raman spectroscopy (SERS) for detection for bulk MIPs systems, we have investigated this approach, which involves surface functionalization of the SERS particles before polymerization. We have successfully fabricated nano-MIP materials on SERS active surfaces as well as nano-MIP materials that contain SERS particles.

XChem

Ricky Chau 04-ERD-024

Abstract

This project addresses the fundamental nature of dynamic chemical processes at high temperatures, very high pressures, and short timescales (10 to 1000 ps) during the passage of a strong shock wave. By exploiting the properties of a steady shock wave produced by a gas gun or laser, we can enhance our

spatial and temporal resolution using optical spectroscopy to probe the development of fast chemical processes in a simple molecular fluid. Benzene, cyclohexane, and nitrobenzene will be the prototype systems for this study, which will focus on understanding several fundamental processes such as carbon formation, dissociation, and the formation of intermediate species. Experimental data at the 10-ps timescale from this project will be compared to computational results.

The success of this project will determine fundamental properties of chemical reactions under extreme conditions of temperature and pressure. One result will be experimental data on kinetics and rates of chemical reaction at the subnanosecond timescale. In addition to improving chemical models, the data will impact many areas of science, including planetary science and energetic materials, and will enable the Laboratory to fill substantive gaps in our knowledge of high-pressure chemistry.

Mission Relevance

Results from this project will help improve the many chemical codes used to study short-timescale chemical processes involving energetic materials, which are essential to national security. Other applications of this research include the verified in situ destruction of chemical or biological weapons of mass destruction.

FY05 Accomplishments and Results

In FY05, we focused on preliminary double-pass absorption experiments on benzene using gas gun drivers. In benzene, we observed a strong dependence with pressure and temperature in the dissociation rate. For instance, photomultiplier tube data for benzene shocked at 12 and 18 GPa showed a strong dependence of the dissociation rate with the shock pressure. The time for the benzene to become fully opaque was 200 to 250 ns at 12 GPa, whereas at 18 GPa the time was an order of magnitude lower (~25 ns). Furthermore, additional structure in the dissociation of the benzene at 12 GPa was evidenced by the "shoulder" (i.e., less-steep slope) in the graph data after the initial shock breakout. This could indicate an incubation time for the dissociation or possibly further dissociation of the byproducts over time. Preliminary gas gun experiments showed that less than 10 J of laser energy is needed for laser shock experiments. Due to the high demand for JANUS and the short pulse duration of COMET, laser experiments were moved to a long-pulse 10-J laser, setup of which was begun. Absorption and Raman experiments are planned there for FY06.

Proposed Work for FY06

In FY06 we will focus on computational experiments to test the new diagnostic completed in FY05. We will field the absorption and coherent anti-Stokes Raman experiments on both gas guns and lasers, focusing on the same materials studied in the double-pass optical absorption experiments. This is because absorption experiments do not directly probe the local mechanism of dissociation and only give the average response of the sample. Consequently, absorption experiments alone cannot reveal the nature of a sample that has begun to dissociate. The computational experiments, however, will probe samples at the molecular level, giving complementary data to the absorption experiments.

Dynamic Combinatorial Libraries for Target-Driven Ligand Development

Julie Perkins 04-ERD-007

Abstract

The objective of this project is to develop disulfide- and hydrazone-based dynamic combinatorial chemistry and then enable a protein or enzyme to select ligands or inhibitors from an equilibrating pool of disulfides or hydrazone-based molecules. Ligands or inhibitors specific to a particular protein would have numerous uses in detecting biological agents in homeland-security and other potential applications. The use of antibodies is the current state of the art for detecting proteins, but more robust and reliable methods of detection are needed. The success of this project will enable the generation of ligands or inhibitors for a protein even before that protein is fully characterized. Our approach combines thermodynamically controlled chemistry with mass spectrometry and enzyme assay-based characterization.

The success of this project would allow formation of ligands or inhibitors for purified proteins without prior determination of their structure. The reaction conditions for generation and identification of ligands/inhibitors would be the outcome of our proposed work. In addition, ligands/inhibitors specific for proteins of interest to national security will be created. This would be a significant advance in the detection of proteins, because the ligands/inhibitors could be deposited on surfaces and incorporated into detection devices.

Mission Relevance

This work supports LLNL's national-security mission by advancing technology for detecting biological agents. The molecular-recognition units for sensor devices must be developed to keep pace with decreasing size and increasing complexity of the machinery for detectors. The ligands developed in this project would serve as the recognition units in detectors. This work addresses an important challenge in the development of sensor technology.

FY05 Accomplishments and Results

Numerous potential ligands were synthesized in FY05 using reversible chemistry, and conditions for establishing exchange within the library of ligands were determined. However, it turned out to be difficult to characterize which ligand from an equilibrating mixture had bound to the target protein, despite the use of several methods and observations of trends. However, results of this work will be fed into another ligand identification project for further development. In addition, hydrazone chemistry was exploited to search for inhibitors of the anthrax lethal factor. A structure–activity relationship was determined, and a variety of new hydrazone inhibitors for the anthrax lethal factor were identified using a simple ultraviolet- and fluorescence-based enzyme assay.

Bioforensics: Characterization of Biological Weapons Agents by NanoSIMS

Peter K. Weber 04-ERD-039

Abstract

The objective of this project is to identify elemental and isotopic markers in spore material that can be used for attribution of biological weapons, including markers that identify the production method, location, and date. The project is developing the capability to analyze individual bio-weapon particles for trace elements and isotopic signatures, building a library of elemental and isotopic fingerprints, and relating these signatures to specific processes, locations, and dates of production or release using the unique capabilities of the nanoscale secondary ion mass spectrometer (NanoSIMS). If successful, the project will establish a library of elemental and isotopic signatures that will aid law enforcement agencies in their attribution efforts. We will also establish the basic methodology for making these assessments.

Mission Relevance

This research project will contribute to the LLNL's national security mission by addressing the need for technologies to quickly detect, identify, and mitigate the use of chemical and biological threat agents against U.S. civilian populations.

FY05 Accomplishments and Results

Our initial data strongly suggest that elemental composition can be used to differentiate production runs and characterize the spore production process, including growth media composition and growth and washing methods. Based on this work, we have constructed a working model for determining the method of production. We initiated our elemental diffusion experiments and began isotopic analyses of spores produced at the University of Utah using sporulation media with distinct isotopic compositions. This research has been successful in attracting spin-off funding for applied work from the Department of Homeland Security and the Federal Bureau of Investigation.

Proposed Work for FY06

In FY06, we will conduct experiments to understand the reproducibility of elemental signatures in spore preparations and follow up on our diffusion rate experiments, making refinements and replicates as necessary. In addition, we plan to address the technical issues involved in making precise isotopic measurements in small samples and then return to the measurement of isotopic ratios in spores produced from isotopically distinct media. Finally, using the results of this work, we will build a library of spore-production signatures.

Ionization Chemistry of High-Temperature Molecular Fluids

Laurence E. Fried 04-ERD-069

Abstract

The purpose of this project is to investigate the phase diagram of simple polar molecular fluids (water and ammonia) and mixtures under conditions of extreme pressure (>10 GPa) and temperature (>1000 K). Under extreme conditions, the neutral molecular form of matter transforms to a phase dominated by ions, the boundaries of which are unknown. We will perform tightly coupled modeling and experiments to determine the phase diagram of simple fluids in this region. Proposed, novel "superionic" lattice states involving mobile hydrogen atoms will be investigated. Acid–base chemistry under extreme conditions will be investigated in a diamond anvil cell (DAC) for the first time, and the chemistry of synthetic planetary interiors will be addressed through molecular mixtures.

The proposed research will lead to the first understanding of ionization and acid–base chemistry under extreme conditions. We have already achieved experimental and computational results that indicate the existence of nonmolecular phases of water at pressures over 50 GPa, and that qualitatively new superionic phases may be found. By mixing water and ammonia under extreme pressure and temperature, we will understand how fundamental chemical notions such as acid–base chemistry are modified in extreme environments. This project increases our understanding of planetary interiors and will guide the modeling of high-explosive detonations. The results are of great interest to researchers in high-pressure physics, and have already generated articles in high-visibility journals.

Mission Relevance

The results of this investigation will be coordinated with Advanced Simulation and Computing efforts in high-explosive modeling. Future developments in the Cheetah and CHEQ high-explosive detonation codes will be based on new knowledge of high-pressure chemistry generated in this project. The Cheetah and CHEQ codes support the Laboratory's stockpile stewardship mission and extend scientific understanding in support of the basic science mission.

FY05 Accomplishments and Results

Experiments on the phase diagram of water under extreme conditions were successfully carried out and published this year in *Physical Review Letters*. This work has received significant scientific publicity, including news articles in *Nature and Physical Review Focus*. We measured Raman spectra of ice and liquid water obtained in situ using a laser-heated DAC over a wide range of pressure (to 56 GPa) and temperature (to 1500 K) combined with molecular-dynamics simulations under similar conditions. Our most important observation is a sudden change in the slope of the melting curve at 47 GPa and 1000 K. In agreement with these observations, first-principles simulations show an apparent transition to a superionic state above 46 GPa.

Proposed Work for FY06

In FY06, we propose to address two issues essential to understanding the prevalence of nonmolecular phases in high-explosive detonation. The first is to see if other common molecular detonation products, such as ammonia or hydrogen fluoride, could have superionic phases. We will address this problem both experimentally, through Raman spectroscopy in a laser-heated DAC, and computationally, through first-principles simulations on Thunder. The second issue is the prevalence of superionic states in mixtures of simple compounds, which is important because high-explosive detonation creates a complicated molecular mixture. We plan to study simpler acid–base mixtures, such as ammonia mixed with water.

Publications

Goldman, N. et al. (2005). “Bonding in the superionic phase of water.” *Phys. Rev. Lett.* **94**, 217801. UCRL-JRNL-209577.

Goncharov, A. et al. (2005). “Dynamic ionization of water under extreme conditions.” *Phys. Rev. Lett.* **94**, 125508. UCRL-JRNL-205550.

Goncharov, A. et al. (2005). “Polymerization of formic acid under high pressure.” *Phys. Rev. Lett.* **94**, 065505. UCRL-JRNL-206204.

New Fragment Separation Technology for Superheavy Element Research

Dawn A. Shaughnessy 04-ERD-085

Abstract

We propose to investigate the western edge of the island of stability (which describes the possibility of elements that have particularly stable “magic numbers” of protons and neutrons) to determine whether or not the closed proton shell is located at 114 protons. We will accomplish this via three different paths: (1) irradiating americium-243 (^{243}Am) with calcium-48 (^{48}Ca) ions to produce enough events to determine structure of the alpha decay spectrum of the element 115 isotope 287115; (2) irradiating californium-249 (^{249}Cf) with ^{48}Ca ions to positively identify the decay properties of the previously undiscovered element 118 isotope 294118; and (3) participating in experiments to positively identify the atomic mass of an element 114 isotope. This project will be carried out in collaboration with the Joint Institute for Nuclear Research in Dubna, Russia.

If successful, we will not only discover yet another new element (element 118), but also better establish the decay properties of the nuclides of elements 115 and 113, and determine whether the island of

stability is centered at 114 protons or at a higher atomic number. Such results would help extend and improve theoretical models used to calculate the decay properties and nuclear shapes of the heaviest elements. These results will contribute to target-preparation technology for the primary target of the Rare Isotope Accelerator fragment separator.

Mission Relevance

Efforts to synthesize new elements and measure their properties support the national security mission by furthering competencies in nuclear chemistry and radiochemistry, which are relevant to assessing nuclear device performance and countering proliferation activities involving nuclear materials. This project also supports the environmental management mission by producing technology for the safe disposal of radioactive waste.

FY05 Accomplishments and Results

In FY05, we (1) completed planning of the ^{249}Cf + ^{48}Ca experiment; (2) carried out preliminary bombardments to check and calibrate equipment and continued to analyze experimental data as it became available; (3) completed the ^{249}Cf +^{48}Ca experiment, including data analysis, with the potential discovery of element 118; and (4) prepared surrogate ceramic targets composed of zirconium, oxygen, and samarium for future in-beam testing as part of our work to develop ceramic plutonium targets for the Mass Analyzer of Super Heavy Atoms (MASHA) in Dubna.

Proposed Work for FY06

Our work plan for FY06 includes (1) participating in additional superheavy element production experiments with subsequent data analysis and publication of our results; (2) repeating the chemical separation of element 105 as the descendent nucleus of element 115, with the goal of identifying atomic numbers of the nuclei in the element 115 decay chain; (3) continuing to fabricate surrogate ceramic targets and possibly a plutonium-239 (^{239}Pu) or ^{242}Pu target for subsequent testing at a facility such as Oak Ridge or the European Center for Nuclear Research; (4) sending appropriate surrogate targets to Dubna for testing in the MASHA separator; and (5) pending the results and progress of our surrogate target tests, producing a ^{244}Pu target for element 114 production runs in Dubna.

Publications

Dmitriev, S. N., et al. (2005). “Chemical identification of dubnium as a decay product of element 115 produced in the reaction ^{48}Ca + ^{243}Am.” *Mendeleev Commun.* **15**, 1. UCRL-JRNL-215890.

Moody, K. J., et al. (2005). *Production and chemical properties of the heaviest elements.* DOE Heavy Element Chemistry—Separations Contractors’ Mtg., Rockville, MD, Apr. 27–30, 2005. UCRL-ABS-210263.

Oganessian, Y. T., et al. (2005). “Synthesis of elements 115 and 113 in the reaction ^{243}Am + ^{48}Ca.” *Phys. Rev. C* **72**, 034611. UCRL-JRNL-210965.

Oganessian, Y. T., et al. (2004). "Measurements of cross sections and decay properties of the isotopes of elements 112, 114, and 116 produced in the fusion reactions $^{233,238}U$, ^{242}Pu, and ^{248}Cm + ^{48}Ca." *Phys. Rev. C* **70**, 064609. UCRL-JRNL-206491.

Patin, J. B., et al. (2005). *Random probability analysis of recent ^{48}Ca experiments*. American Chemical Society Mtg. and Exposition, Washington, DC, Aug. 28–Sept. 1, 2005. UCRL-PRES-215891.

Shaughnessy, D. A. (2005). *Review of the synthesis of the heaviest elements and their chemistry*. American Physical Society April Mtg., Tampa, FL, Apr. 16–19, 2005. UCRL-ABS-209863.

Shaughnessy, D. A., et al. (2005). *Ceramic plutonium target development for the MASHA separator for the synthesis of element 114*. Actinides 2005, Manchester, UK, July 4–8, 2005. UCRL-PROC-213358-DRAFT.

Shaughnessy, D. A., Wilk, P. A., Moody, K. J. (2005). *Application of group five separation methods to the isolation of element 105 (dubnium)*. Sandanski Coordination Mtg. on Nuclear Science, Albena, Bulgaria, Sept. 26–30, 2005. UCRL-ABS-212207.

Wilk, P. A., et al. (2005). *A plutonium ceramic target for the MASHA mass separator*. Gordon Research Conf., New London, NH, June 26–July 1, 2005. UCRL-ABS-210964.

Heterogeneous Processes at the Intersection of Chemistry and Biology

Christopher J. Mundy 05-ERD-021

Abstract

Phenomena in heterogeneous environments are important in fields ranging from chemistry (such as the fate of aerosolized chemical weapons) to atmospheric science (such as ozone destruction). However, no computational framework exists to address such phenomena. One of the difficulties in studying heterogeneous processes is that current empirical molecular-potential models are parameterized to reproduce only bulk liquid properties. We propose to investigate heterogeneous chemistry in aerosols and enzymes using terascale ab initio methods, which are well suited to providing an unbiased representation of the force field in nonbulk environments as well as incorporating reactivity.

Using LLNL's terascale resources in conjunction with state-of-the-art software, we will conduct research on heterogeneous chemistry as applied to biological and atmospheric systems. Using these unique capabilities, we expect to solve problems with a large impact in a broad set of scientific disciplines.

Mission Relevance

The computational suite developed in this project directly benefits the national security mission by contributing to increased understanding of the physical properties of organophosphates, such as sarin and VX, which will lead to improved detection and ability to predict their fate and transport. For example, this technology can provide a model of sarin release at the city scale by incorporating our microscopic understanding of heterogeneous processes.

FY05 Accomplishments and Results

New literature suggests that one must actually include the full protein to properly stabilize the enzymatic active site. Thus, we moved directly to working on a full simulation of the ODCase protein utilizing the quantum mechanical/molecular mechanical (QM/MM) framework within the Car-Parrinello 2000 (CP2K) code, work originally planned for FY06. To this end, we completed and tested the interface of a native biomolecular simulation code to a native, very efficient, state-of-the-art QM/MM code within CP2K; simulations are currently underway. Also, we completed a simulation of the detailed mechanism of proton transfer between nitrate and chlorine ions in a stable water cluster. To obtain these results we used a state-of-the-art rare-events algorithm, referred to as metadynamics, to show that microsolvation in heterogeneous environments can alter the acidity of acids.

Proposed Work for FY06

In FY06, we plan to work in two major areas, enzymatic reactions and aerosols. To study enzymatic reactions, we will perform large-scale QM/MM calculations of model clusters of both orotidine 5'-monophosphate (OMP) and 6'-hydorxyuridine 5'-monophosphate (BMP) in ODCase. For our work on aerosols, we will continue to run metadynamics calculations on microsolvated clusters and compare them to bulk mechanisms for ion dissociation. Using metadynamics pathways, we will deduce electronic mechanisms by computing molecular states, and seek out structural moieties on large surface models to relate to recent experimental findings suggesting that halogens have larger enhancements in reactivity than previous calculations suggest.

Publications

Mundy, C. and I. W. Kuo. (in press). "First-principles approaches to the structure and reactivity of atmospherically relevant aqueous interfaces." *Chem. Rev.* UCRL-JRNL-212909.

Discovering the Folding Rules that Proteins Obey

Olgica Bakajin 05-ERD-078

Abstract

Protein folding is a fundamental cellular process: Proper folding is required for a protein to carry out its functions, while improper folding can be a source of disease. We propose to use a combination of simulations and experiments to significantly advance our understanding of the molecular mechanisms of protein folding. Our research will focus on a known set of molecules identified as “fast folders” and will attempt to discover more such molecules. We will develop a robust microfluidic mixing device, conduct long time simulations on LLNL’s supercomputers, and perform measurements on the systems that exhibit a fast folding and fast hydrophobic collapse. Our results will be used to answer the following questions: Can traps and intermediate states be observed? Why are some molecules such fast folders? Are there multiple folding pathways, a few, or just one? And is folding hierarchical?

We expect to elucidate the mechanisms of protein folding through a combination of complementary experimental and simulation studies. In addition, the equipment and technology developed for this project will provide new capabilities that will be applicable to numerous other projects at LLNL. This research will establish and strengthen collaborations between LLNL scientists and leading academic researchers. Because of the fundamental nature of this project, it should result in publications in high-impact peer-reviewed journals.

Mission Relevance

By developing the scientific basis for understanding and controlling protein function, this project is highly relevant to the Laboratory’s mission in biodefense. The knowledge base created in this project will also support the Laboratory’s mission in bioscience to improve human health.

FY05 Accomplishments and Results

In FY05, we (1) reduced the mixing time to 4 µs by optimizing the shape of the micromixer without reducing its dimensions; (2) demonstrated that direct-excitation ultraviolet (UV) measurements are feasible with our fused-silica mixers (i.e., without labels on the protein) and obtained promising results using three-photon excitation; and (3) carried out, on Thunder, the largest, longest-timescale simulations of protein folding thermodynamics to date. Our simulations are helping to resolve an experimental controversy over the folding mechanism of the 40-amino-acid protein 1BBL—whether or not the mechanism is a barrierless “downhill folder.” Our simulation on Thunder has shown one of the first examples of reversible folding in a protein this large.

Proposed Work for FY06

In FY06 we will (1) perform measurements of protein-folding kinetics on several unlabeled proteins; (2) develop three-photon excitation measurements and compare the resultant signal-to-noise ratio to that of direct UV excitation; (3) decrease mixing time further with a combination of shape optimization and feature-size reduction (our goal is a mixing time of 1 μs so we can compare the experimental results to the simulation); (4) evaluate the feasibility of fast cooling through mixing; and (5) perform simulations capable of reproducing accurate kinetic information on the folding of small proteins from experimentally relevant starting conformations, addressing proteins of a size and folding rate that can also be addressed experimentally.

Publications

Bakajin, O. et al. (in press). "Materials aspects in micro- and nano-fluidic systems applied to biology." *Mater. Res. Soc. Bull.* UCRL-JRNL-217803-DRAFT.

Hertzog, D. et al. *Optimization of a fast microfluidic mixer for studying protein folding kinetics.* UCRL-JRNL-217805.

Avoiding Surprise: Countering Novel Chem-Bio-Warfare Agent Threats

Bradley R. Hart 05-ERD-025

Abstract

Synthesis and computational modeling, when combined, can aid in characterizing the physiological capabilities and properties of candidate compounds that could be designed by an adversary to be either incapacitating or lethal chemical and biological weapons (CBW) agents. From this information, detection and countermeasure strategies can be developed. We propose to define, develop, and implement a comprehensive scientific approach that will couple cutting-edge computational chemistry and new synthetic methods to enhance our understanding of the threats posed by the development and use of novel CBW agents.

The efforts outlined here will lead to high-quality results intended to cue U.S. defensive efforts by providing guidance in creating new detection and countermeasures programs.

Mission Relevance

This research is designed to address a serious national security concern and to demonstrate a state-of-the-art capability to the broader U.S. government community. Therefore, our work directly supports the LLNL mission of enhancing national security and facilitating efforts to halt and reverse the proliferation of weapons of mass destruction.

FY05 Accomplishments and Results

In FY05, we began developing and demonstrating techniques to characterize, at the molecular level, candidate compounds that could serve as, or form a basis for, novel CBW agents. Several new compounds have been synthesized and characterized, and experiments to explore the fate, transport, and decontamination of these compounds have begun—new information in this area is of special importance for both force protection and intelligence collection. Initial synthetic routes have been developed and experimental verification of these routes is underway. Computational chemistry has been a key component of this research—for example, calculations of reaction transition-state energies have aided in determining likely reaction pathways and byproducts. Additionally, we developed four complete protein homology models of the protein targets of interest, and completed the process of optimizing models and equilibrating them in the simulated environment of water, ions, and lipid membrane. The capabilities demonstrated to date have generated great interest—we have presented results at several scientific meetings, and additionally have responded to multiple requests to submit white papers to potential sponsors.

Proposed Work for FY06

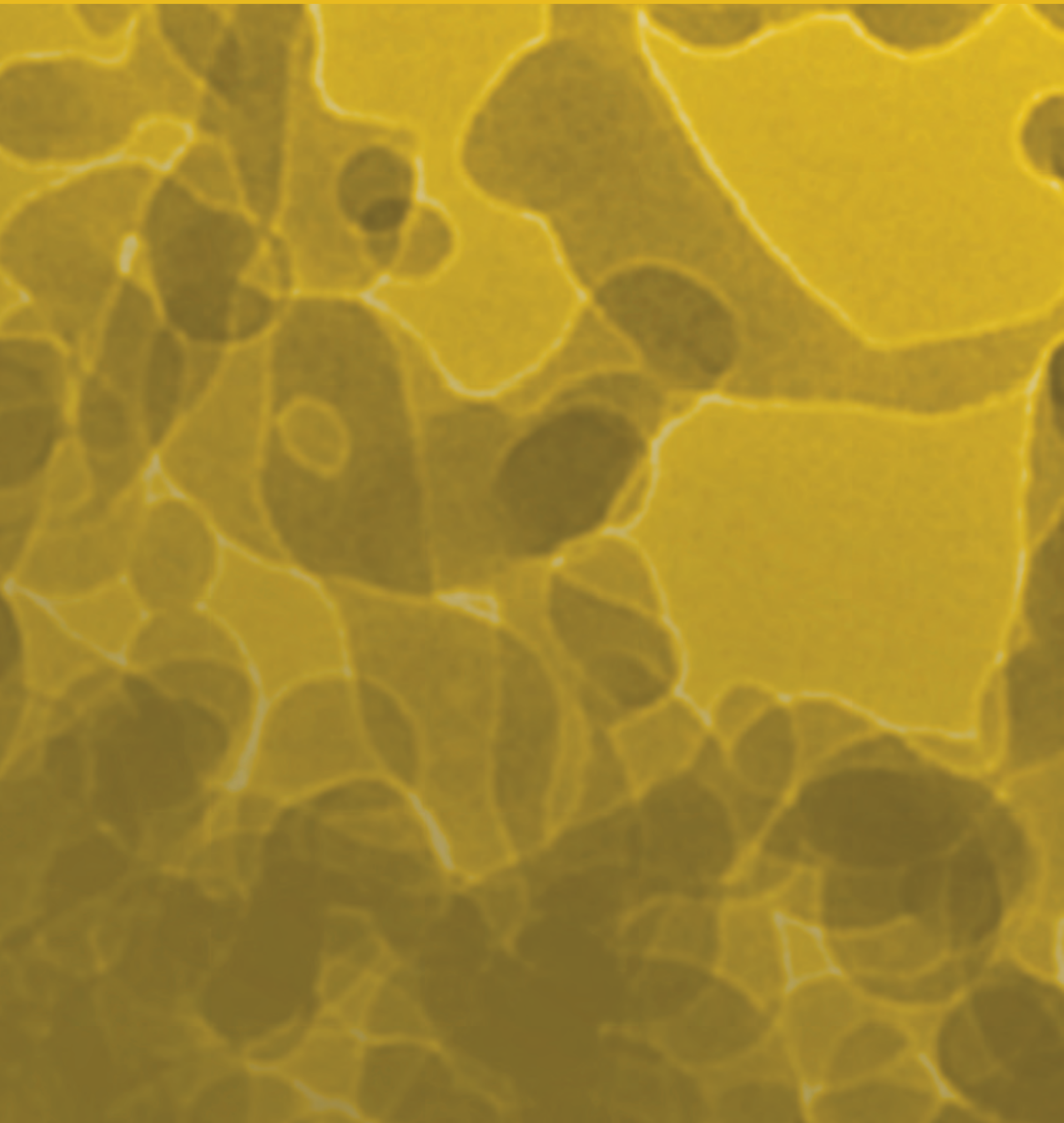
For FY06, we will continue to explore synthesis and computational modeling as methods for characterizing the physiological capabilities and properties of candidate compounds that could be designed by an adversary to be either incapacitating or lethal CBW agents. This exploration will include examining synthesis pathways with the aid of quantum calculations. Additionally, the homology models developed in FY05 will be used for molecular docking studies.

Publications

Law, R. et al. (2005). *GABA and nicotinic receptors investigated by modeling, simulation, and docking of anesthetic molecules.*, 19th Symp. Protein Society, Boston, MA, July 20–Aug. 3, 2005. UCRL-PRES-211304.

Lightstone, F. L. et al. (2005). *Application of computational biology to the analysis of future threats.* UCRL-PRES-207705.

Earth and Space Sciences



Adaptive Optics Views of the Hubble Deep Fields

Claire E. Max 03-ERD-002

Abstract

This project uses the LLNL-developed laser guide star adaptive optics (AO) systems at Lick and Keck observatories to study galaxies in the early universe. The goals are to observe large galaxies in the process of their initial assembly from subcomponents, to identify central active galactic nuclei due to black holes in galaxy cores, and to measure rates of star formation and evolution in galaxies throughout the past 10–12 billion years. The study focuses on the GOODS, GEMS, COSMOS, and Extended Groth Strip fields. These regions of sky are being intensively studied using long exposures by the Hubble Space Telescope, Chandra X-Ray Space Telescope, XMM Space Telescope, the Spitzer Space Infrared Telescope Facility, and the Very Large Array radio interferometer.

This work extends our knowledge of how galaxies formed and evolved and has detected significant numbers of active galactic nuclei at redshifts between 0.5 and 1. The project is developing new methods for efficient use of the LLNL laser guide star AO systems at Lick and Keck observatories, new methods for data analysis, and new ways to use our data to model the evolution of star formation in distant galaxies. The project is also resulting in significant improvement in the laser guide star adaptive optics system at Lick Observatory.

Mission Relevance

The AO technology and precision optics developed in this project have application to long-range surveillance for homeland security and to high-power lasers for stockpile stewardship and DoD applications. In addition, the project contributes insights into galaxy formation and evolution, which support LLNL's mission to extend scientific knowledge.

FY05 Accomplishments and Results

The success of this survey project depended on observing a significant number of galaxies at high redshifts, and we accomplished this goal: We obtained excellent data on more than 200 distant galaxies using natural guide star AO at the Keck Observatory, and obtained data on about 20 more using the laser guide star there. These include eight active galactic nuclei with redshifts between 0.4 and 1.0. In collaboration with Lawrence Berkeley National Laboratory, we observed a high-redshift supernova candidate using the Keck laser guide star. In addition, we held a workshop on extragalactic AO astrophysics and published the first paper on extragalactic astronomy using the LLNL-built Keck laser guide star system.

Publications

Gavel, D. T. et al. (2005). "Recent science and engineering results with the laser guidestar adaptive optics system at Lick Observatory." *Proc. SPIE* **4839**, 354. UCRL-JRNL-218248.

Max, C. E. et al. (2005). "The core of NGC 6240 from Keck adaptive optics and Hubble Space Telescope NICMOS observations." *Astrophys. J.* **621**, 738. UCRL-JRNL-208292.

Melbourne, J. et al. (2005). "Merging galaxies in Goods-s: First extragalactic results from Keck laser adaptive optics." *Astrophys. J. Lett.* **625**, L27. UCRL-JRNL-218256.

Predicting the Effects of Climate Change and Variability on Water Availability

Philip B. Duffy 03-ERD-042

Abstract

Global warming will affect California's hydrological cycle, with likely impacts on water availability and flood risk. Predictions of how climate change will affect California's hydrological cycle are needed to provide decision support for policymakers and water managers. Using a hierarchy of models (global climate, regional climate, and surface hydrology) we will predict how climate change and year-to-year natural climate variability will affect surface temperatures, precipitation amounts, and the amount and timing of flow through rivers. By examining results from a range of models, we will estimate uncertainties in these projections. We will also address fundamental science issues related to the theoretical potential predictability of precipitation.

This project will produce the best assessment to date of the effects of climate change and variability on the availability of water in California. Sate-of-the-art models at fine spatial resolution will be used. To help quantify uncertainties in results, the team will make multiple runs with varied input parameters and compare results with those obtained by other groups using other models to simulate the same phenomena. This work is expected to significantly advance the state of the art in the modeling of regional climates and surface hydrology. We also expect to improve fundamental understanding of the theoretical predictability of precipitation in the western U.S. and elsewhere.

Mission Relevance

This work supports multiple LLNL missions by developing prediction methodologies and evaluating and improving climate models in furtherance of DOE's long-standing commitment to understanding the environmental consequences of energy generation. This project focuses on one of those consequences—global warming—which is caused primarily by burning fossil fuels for energy generation.

FY05 Accomplishments and Results

The team completed and published the best study to date of the expected effect, including uncertainty, of climate change on river flows in California. Projections were made indicating that El Niños in a warmer climate will produce both higher flood risk and more potential water shortages in California, than in today's climate. We developed an understanding of why some El Niño winters are very wet in California while others are not. The team showed that multi-year droughts in the western U.S. result primarily from atmospheric chaos, and are thus inherently unpredictable.

Publications

Duffy, P. B. et al. (in press). "Simulations of present and future climates in the western U.S. with four nested regional climate models." *J. Climate*. UCRL-JRL-200593.

Gibbard, S. and P. B. Duffy. (2005). *Potential predictability of Pacific coast precipitation during warm-phase El Nino events*. UCRL-ABS-207832.

Maurer, E. and P. B. Duffy. (2005). "Uncertainty in projections of hydrologic impacts of climate change in California." *Geophys. Res. Lett.* **32**, L021462. UCRL-JRNL-207179.

Snyder, M. A. et al. (2004). "Climate responses to a doubling of atmospheric carbon dioxide since the preindustrial era for a climatically vulnerable region." *Geophys. Res. Lett.* **29**. UCRL-JC-145861.

Nitrate Biogeochemistry and Reactive Transport in California Groundwater

Bradley K. Esser 03-ERD-067

Abstract

Nitrate contamination is a significant threat to groundwater resources in California and numerous other states. Successful groundwater management requires modeling future impacts on affected aquifers, and assessing practices designed to reduce nitrate levels. Our research investigated the fate and transport of nitrate in groundwater at three different scales: microbial denitrification kinetics under controlled laboratory conditions, denitrification in the shallow saturated zone at the field scale, and nitrate transport at the basin scale. Research will focus on developing and applying rapid methods to quantify denitrification, assessing the role of denitrification in the shallow and deep saturated zone, and developing and validating biogeochemical reactive transport models for nitrate in the subsurface.

Motivation for this work was the need to develop science-based approaches to characterize and manage nitrate contamination in groundwater. We worked to develop a robust approach to characterizing nitrate biogeochemistry in the subsurface. When coupled with our ability to characterize groundwater flow and to model reactive transport, this approach will allow an accurate assessment of the future distribution of nitrate in California aquifers and of the impact of different management practices on nitrate input to groundwater. Such assessment is vital to making cost-effective management and policy decisions regarding land use and groundwater remediation.

Mission Relevance

This project supports the Laboratory's mission in environmental management by developing models and technologies that help assess the environmental consequences of toxic materials and manage the risk associated with these materials.

FY05 Accomplishments and Results

In FY05, we (1) validated the use of cell number as an indicator of activity by using a pure cell culture to determine correspondence between the specific denitrification rate and the messenger RNA transcript copy number of the nitride reductase gene, as determined by reverse-transcription, real-time quantitative polymerase chain reaction (qPCR); (2) used qPCR to determine denitrifier populations in soil cores taken from a dairy farm; (3) correlated cell density to sediment and water properties; (4) developed groundwater flow and biogeochemical reactive transport models to validate microbial kinetic rate expressions; and (5) developed a reactive transport model of the Llagas Basin in Santa Clara County, California, then used it to simulate the transport and distribution of shallow groundwater nitrate contamination and quantify the potential impact of different management practices.

Publications

Beller, H. R. et al. (2004). "Biogeochemistry and natural attenuation of nitrate in groundwater at an explosives test facility." *Appl. Geochem.* **19**(9), 1483. UCRL-JC-154175.

Carle, S. F., J. E. Moran, and B. K. Esser. (in press). "High-resolution simulation of basin scale nitrate transport considering aquifer system heterogeneity." *Geosphere*. UCRL-JRNL-214721.

Carle, S. F. et al. (2005). *Simulation of canal recharge, pumping, and irrigation in a heterogeneous perched aquifer: Effects on nitrate transport and denitrification*. 25th Biennial Groundwater Conf. and 14th Ann. Mtg. Groundwater Resources Association of California, Sacramento, CA, Oct. 25–26, 2005. UCRL-POST-217056.

Carle, S. F., A. F. B. Tomson, and B. K. Esser. (2004). *Modeling basin-scale nitrate transport considering alluvial and biogeochemical heterogeneity*. Geological Society of America Annual Mtg., Denver, CO, Nov. 7–10, 2004. UCRL-ABS-208496.

Carle, S. F. et al. (2004). *Simulation of nitrate biogeochemistry and reactive transport in a California groundwater basin*. Computational Methods in Water Resources International Conf. XIV, Chapel Hill, NC, June 13–17, 2004. UCRL-CONF-201876.

Esser, B. K. et al. (2005). *Characterization of saturated-zone denitrification in a heterogeneous aquifer underlying a California dairy*. 25th Biennial Groundwater Conf. and 14th Ann. Mtg. Groundwater Resources Association of California, Sacramento, CA, Oct. 25–26, 2005. UCRL-POST-216853.

Esser, B. K. et al. (2005). "Molecular and geochemical evidence of in-situ denitrification at a dairy field site in the Central Valley of California." *EOS Trans. Am. Geophys. Union* **86**(52), B31A-0972. UCRL-ABS-215237.

Esser, B. K. et al. (2005). *New approaches to characterizing microbial denitrification in the saturated zone*. 15th Ann. Goldschmidt Conf., Moscow, ID, May 20–25, 2005. UCRL-ABS-209086-DRAFT.

Esser, B. K. et al. (2004). “Denitrification in a shallow aquifer underlying a dairy farm in the Central Valley of California.” *Eos Trans. Am. Geophys. Union* **85**(47), H43. UCRL-ABS-206433.

Esser, B. K. et al. (2004). *Denitrification in a shallow aquifer underlying a dairy farm: New approaches to characterization and modeling*. 13th Ann. Groundwater Resources Association of California Mtg. and Conf.: Managing Aquifers for Sustainability, Protection, Restoration, Replenishment and Water Reuse, Rohnert Park, CA, Sept. 23–24, 2004. UCRL-PRES-207404.

Hudson, G. B. et al. (2004). *Emerging contaminants in water resources: Status, challenges, and a case study*. 2nd Intl. Conf. Soil Pollution and Remediation, Nanjing, China, Nov. 9–12, 2004. UCRL-ABS-206964.

McNab, W. W. et al. (2005). *Geochemical modeling of nitrate loading and denitrification at an instrumented dairy site in California’s Central Valley*. 25th Biennial Groundwater Conf. and 14th Ann. Mtg. Groundwater Resources Association of California, Sacramento, CA, Oct. 25–26, 2005. UCRL-POST-216428.

McNab W. W. et al. (2005). *Nitrate loading and groundwater chemistry at a dairy site in California’s Central Valley*. Intl. Conf. Safe Water 2005, San Diego, CA, Oct. 21–25, 2005. UCRL-PRES-216112.

McNab, W., J. E. Moran, and B. K. Esser (2004). *Geochemical signatures of fertilizers and soil amendments in public water supply aquifers in California*. 13th Ann. GRA Mtg. and Conf.: Managing Aquifers for Sustainability, Protection, Restoration, Replenishment and Water Reuse, Rohnert Park, CA, Sept. 23–24, 2004. UCRL-PRES-206327.

Moran, J. E. et al. (2006). *Evidence for groundwater contamination vulnerability in California’s Central Valley*. 2006 California Plant and Soil Conf., Visalia, CA, Feb. 7–8, 2006. UCRL-PROC-217713.

Moran J. E., et al. (2005). *The effects of agricultural nitrate sources on groundwater supplies in California*. Geological Society of America Ann. Mtg., Salt Lake City, UT, Oct. 15–19, 2005. UCRL-ABS-213498.

Singleton, M. J. et al. (2005). “Evaluation of nitrate sources and nitrate management strategies in California suburban growth areas.” *EOS Trans. Am. Geophys. Union* **89**(52), B43C-0295. UCRL-ABS-215210.

Singleton, M. J. et al. (2005). *Natural tracers of lagoon seepage at California dairies*. 14th Ann. Groundwater Resources Association Mtg. and Conf., Sacramento, CA, Oct. 25–26, 2005. UCRL-ABS-214160.

Singleton, M. J. et al. (2005). *Viability of intrinsic denitrification to reduce nitrate pollution at California dairies*. American Chemical Society National Mtg., Washington, DC, Aug. 28–Sept. 1, 2005. UCRL-ABS-212107.

Tectonic Morphochronology of the Southern San Andreas Fault System

Frederick J. Ryerson 03-ERI-001

Abstract

The San Andreas Fault (SAF) accommodates a major fraction of the motion between the North American and Pacific tectonic plates. The partitioning of slip among the many segments of the fault has implications for seismic risk. South of the Transverse Range, the SAF comprises strike-slip faults and thrust zones that are the focus of continuous satellite geodetic observations that track decadel deformation. However, only two millennial (long-term) slip-rate measurements exist for the SAF. We are developing a millennial slip-rate map of the western U.S. using "morphochronology," in which tectonically offset landscape features are dated using cosmic-ray surface-exposure dating at LLNL's Center for Accelerator Mass Spectrometry.

This project will add to our understanding of the interactions of fault motion within the western U.S. plate boundary region, which extends from the SAF across the Basin and Range, and from Baja to Alaska. The results have important implications for the assessment of earthquake risk and slip-partitioning. As a result of efforts at LLNL, cosmogenic dating is becoming more widely used in the western U.S. in determining millennial slip rates. Millennial slip rates help in identifying stress accumulation on the various segments of the fault, because disparities in long- and short-term slip rates influence earthquake frequency. If successful, this method will provide a new tool in evaluating crustal deformation that can be widely used in seismically active regions throughout the world.

Mission Relevance

This analysis of satellite imagery to identify tectonic features contributes to basic scientific understanding of mechanics of the Earth's crust. These topics are also related to explosion monitoring and analysis, which supports nonproliferation. In addition, structure and tectonics of the western U.S. are important in enhanced test readiness, geothermal energy, and disposal of radioactive waste. The cosmogenic dating methods developed here are also being used to date landscape features near Yucca Mountain to assess the history of strong ground motion near the Yucca Mountain Project repository site, and may influence repository design.

FY05 Accomplishments and Results

In FY05 we completed the measurement of slip rates on the Mission Creek segment of the SAF, which required detailed mapping of the offsets at the Biskra Palms sites. We also completed dating of samples from the Anza site on the San Jacinto Fault (SJF). The rate at SJF is well below that obtained from paleoseismic studies, and may indicate either the effects of inheritance on our ages, or secular changes in the slip rate. We also expanded the range of the study, obtaining new data on (1) the Dixie Valley Fault in Nevada, near an active geothermal area; (2) the range front fault on the east side of the Ruby Mountains, Nevada; and (3) the Denali Fault in Alaska. The methods developed here have enabled us to pioneer application of cosmogenic isotopes to quantify the rate of displacement along earthquake faults, which provides an independent means of tracking movement along major plate boundaries and helps discriminate between alternate models of crustal deformation.

Publications

Chevalier, M.-L. et al. (2005). "Slip-rate measurements on the Karakorum Fault may imply secular variations in fault motion." *Science* **307**, 411. UCRL-JRNL-206541.

Daeron, M. et al. (2004). "Constraints on the post ~25-ka slip rate of the Yammouneh fault (Lebanon) using in situ cosmogenic ^{36}Cl dating of offset limestone-clast fans." *Earth Planet. Sci. Lett.* **227**(1–2), 105. UCRL-JRNL-202585.

Matmon, A. et al (2005). "Dating offset fans along the Mojave section of the San Andreas fault using cosmogenic ^{26}Al and ^{10}Be." *Geol. Soc. Am. Bull.* **117**, 795. UCRL-JRNL-202584.

Meriaux, A.-S. et al. (2005). "A westward decreasing quaternary slip rate on the Denali fault, Alaska." *EOS Trans. Am. Geophys. Union* **85**, F577. UCRL-ABS-206354.

Meriaux, A.-S. et al. (2005). "The Aksay segment of the northern Altyn Tagh fault: Tectonic geomorphology, landscape evolution, and Holocene slip rate." *J. Geophys. Res.* **110**, B04404. UCRL-JRNL-204490.

Meriaux, A.-S. et al. (2004). "Rapid slip along the central Altyn Tagh Fault: Morphochronologic evidence from Cherchen He and Sulamu Tagh." *J. Geophys. Res.* **109**, B06401. UCRL-JRNL-200220.

Ryerson, F. J. et al. (in press). "Applications of morphochronology to the active tectonics of Tibet." *Geological Applications of Cosmogenic Isotopes*. Geological Society of America Books, Boulder, CO. UCRL-BOOK-209322.

Stock, G. M., R. S. Anderson, and R. C. Finkel (2004). "Pace of landscape evolution in the Sierra Nevada, California, revealed by cosmogenic dating of cave sediments." *Geology* **32**(3), 193. UCRL-JRNL-200866.

Van der Woerd, J. et al. (in press). "Long-term slip rate of the southern San Andreas Fault, from ^{10}Be–^{26}Al surface exposure dating of an offset alluvial fan." *J. Geophys. Res.* UCRL-JRNL-218149.

Nonaqueous-Phase Liquid Dissolution in Porous Media: Multi-Scale Effects of Dissolution Kinetics on Cleanup Time

Walt W. Mcnab 04-ERD-001

Abstract

The goal of this project is to identify and quantify, in unprecedented detail, the factors controlling the rate at which dense, nonaqueous-phase liquids (DNAPLs) dissolve from source areas. The rate is critical to understanding and predicting contaminant flow and transport. We will experimentally and numerically investigate the multiphase physics of dissolution of DNAPL at the microscopic level, then scale up this formulation for field use. Experimental results will be used to calibrate a numerical pore-scale model that incorporates Navier-Stokes equations and multiphase physics. The pore-scale modeling

will yield an interphase mass-transfer term to be incorporated into an existing field-scale, solute-transport model. This field-scale model will be validated through comparison to historic groundwater data from sites such as LLNL.

By directly addressing the scaling up of DNAPL dissolution, this project will yield a greatly improved understanding of the behavior of groundwater plumes and will benefit remediation efforts at Lawrence Livermore and other sites across the DOE complex.

Mission Relevance

This project leverages environmental-restoration and groundwater-modeling capabilities at LLNL to improve our understanding of the behavior of groundwater contaminants in source areas. If successful, this project will have a direct effect on cleanup efforts at many sites across the DOE complex, in support of DOE's environmental-management mission.

FY05 Accomplishments and Results

Accomplishments for FY05 include (1) constructing the mesoscale experimental apparatus from materials provided by Sandia National Laboratories; (2) conducting microscale and mesoscale experiments for single- and multiple-component DNAPL-water systems; (3) constructing an improved microscale conceptual model of multiple-phase dissolution in a DNAPL-water domain (yielding good agreement with experimental results); (4) defining a time-dependent, mass-transfer term that represents the evolution of the entire source body for application in large-scale models; and (5) developing a set of coupled, scaled-up equations from the microscale that describe transient dissolution and aqueous-phase transport of DNAPL.

Proposed Work for FY06

During FY06, we will conduct experiments of multi-component DNAPL dissolution with and without the coexistence of a co-solvent at the mesoscale. This will be accomplished using the apparatus acquired from Sandia. The major objectives for FY06 will be to develop a set of coupled equations, scaled up from the microscale, that describe transient dissolution and aqueous-phase transport of DNAPL. These equations will be validated under mesoscale dissolution conditions to (1) define a time-dependent, mass-transfer term that represents evolution of the source body (to be used in our large-scale model); (2) solve these coupled equations at the mesoscale and field scale; and (3) validate the scaled–up microscale model via analysis of data from mesoscale experiments and the field.

Publications

Ezzedine, S., R. Detwiler, and W. McNab. (2005). *An integrated numerical and experimental study of DNAPL dissolution and mass transfer in porous media*. 2005 American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-215074.

McNab, W. W., S. M. Ezzedine, and R. L. Detwiler. (2005). *Quantifying an intrinsic mass transfer rate for TCE dissolution via pore-scale experiments and simulation*. DNAPL Source Zone Characterization and Remediation Symp., San Francisco, CA, Dec. 7–8, 2005. UCRL-PRES-217179.

Magnetic Dynamos and Stars

Peter P. Eggleton 04-ERD-027

Abstract

This project is upgrading Djehuty, a code that models stars in three dimensions (3D), by incorporating magnetohydrodynamics (MHD). Djehuty already includes an accurate equation of state, radiative heat transport, a full network of nuclear reactions, self-consistent gravity in the spherical approximation, Lagrangian hydrodynamics, and an algorithm for adjusting severely distorted meshes. We will add the magnetic-force term to the hydrodynamics and the induction equation in the frozen-in approximation, enabling the code to model a wide range of inherently 3-D problems currently beyond computational possibility, including dynamo activity in stars and planets, and ultimately the sun's well-known but poorly understood magnetic cycle.

By the end of this project, we expect to produce code that incorporates MHD and has improved abilities to model a broad class of astrophysical phenomena. This code will be the first to tackle and understand, for example, (1) the generation of huge but transient (1 to 3 year) star spots observed on some red giant stars; (2) similar, shorter-time-scale phenomena in red dwarf stars; (3) magnetic behavior in gaseous planets such as Jupiter and many recently discovered extrasolar planets; (4) energy transport in contact binaries comprising two red-dwarf stars; and (5) the solar cycle. These highly significant advances will give LLNL a leading role in the astrophysical world and thus help recruit new talent to the Laboratory. The project has also attracted collaboration with the University of California.

Mission Relevance

Because the thermonuclear phenomena found in stars are the same as those that occur in man-made nuclear explosions, this project will benefit numerical simulation and fundamental research used in stockpile assessment and in thermonuclear experiments at future, large fusion-class lasers, in support of the Laboratory's national-security mission. Our results will also benefit atmospheric modeling and the exploration and use of space, which will benefit LLNL missions in national security and fundamental science.

FY05 Accomplishments and Results

In FY05, we moved the Djehuty code from the TeraCluster 2000 (TC2K) supercomputer to the Multiprogrammatic and Institutional Computing Capability Resource and Frost machines, in response to the termination of TC2K. We have also computed models of a new kind of supernova explosion, and the helium flash in low-mass stars. These are major scientific achievements and are close to publication. We have also completed a book, which is in press, describing the astrophysical situations where work by Djehuty will be important. The introduction of the induction equation into Djehuty is close to completion.

Proposed Work for FY06

In FY06, we will begin to incorporate convection and electromagnetic induction into our models. As a first test of the MHD capability, we will consider how it affects results of our previous simulations of the helium flash and the simulation of rotation. Two other testbeds of the MHD capability will be the convective core of a middle-main-sequence star and the convective envelope of a red subgiant. We will also investigate non-MHD convection in subgiant envelopes.

Publications

Dearborn, D. S. P, J. C. Lattanzio, and P. P. Eggleton. (in press). "3D numerical experimentation on the core helium flash of low-mass red giants." *Astrophys. J.* UCRL-JRNL-213547-DRAFT.

Dearborn, D. S. P., J. R. Wilson, and G. J. Mathews. (2005). "Relativistically compressed exploding white dwarf model for Sagittarius A East." *Astrophys. J.* **630**, 309. UCRL-JRNL-208008.

Eggleton, P. P. (in press). *Evolutionary processes in binary and multiple stellar systems*, vol I. New York: Cambridge University Press. UCRL-BOOK-209818.

Eggleton, P. P. (2005). "Evolution in low-mass binary systems." *Astrophys. Space Sci.* **296**, 327. UCRL-CONF-204638.

Eggleton, P. P. and L. Kisseleva-Eggleton. (2005). *A mechanism for producing short-period binaries.* Close Binaries in the 21st Century: New Opportunities and Challenges, Syros, Greece, June 27–30, 2005. UCRL-CONF-216093.

Gualandris, A., S. Portegies Zwart, and P. P. Eggleton. (2004). "Simulations of stars escaping from the Orion Nebula Cluster." *Mon. Notic. Roy. Astron. Soc.* **350**, 615. UCRL-JRNL-202279.

Yakut, K. and P. P. Eggleton. (2005). "Evolution of close binary systems." *Astrophys. J.* **629**, 1055. UCRL-JRNL-209496.

Dynamic Data-Driven Event Reconstruction for Atmospheric Releases

Branko Kosovic 04-ERD-037

Abstract

Atmospheric releases of hazardous materials can have a powerful and rapid impact on large populations. In the case of an atmospheric release, event reconstruction determines how much material was released, where and when it was released, and potential consequences of the release. Accurate estimation of the source term is essential to accurately predict plume dispersion, effectively manage the emergency response, and mitigate consequences. For emergency response, forensic, and sensor network design needs, we are developing a flexible and robust capability that integrates observational data streams with predictive models and provides probabilistic estimates of source-term parameters consistent with both data and model predictions.

Event reconstruction tools developed for this project will provide a unique capability for interpreting and responding to atmospheric releases including (1) quantitative probabilistic estimates of the principal source-term parameters (e.g., the time-varying release rate and location); (2) predictions of increasing fidelity as an event progresses and additional data become available; and (3) analysis tools for sensor network design and uncertainty studies. The final goal is a multi-resolution capability for both real-time operational response and high-fidelity applications. The results of this project will enable optimal siting of sensors, and facilitate uncertainty analyses that will be used in studies to assess vulnerability and protection of critical infrastructure.

Mission Relevance

This project directly contributes to the Laboratory's homeland and national security mission by addressing a critical need for atmospheric release event reconstruction tools to support the rapidly growing number of operational detection, warning, and incident characterization systems being developed and deployed by the Department of Homeland Security and Department of Energy. This capability will transform the way we respond to terrorist attacks, industrial accidents, and military engagements by reducing situation-awareness uncertainties and facilitating informed decision-making for more effective response. The event reconstruction tools developed by this project are targeted for integration into the National Atmospheric Release Advisory Center (NARAC) at LLNL.

FY05 Accomplishments and Results

In FY05, we: (1) demonstrated the efficiency and robustness of a Markov Chain Monte Carlo (MCMC) capability with the NARAC three-dimensional Lagrangian Operational Dispersion Integrator model using concentration measurements from an approximately 10-km-scale tracer field experiment; (2) developed a hybrid MCMC and sequential Monte Carlo (SMC) methodology and demonstrated its effectiveness in characterizing releases from complex, multiple sources; (3) used optimization methods to develop a prototype sensor network design tool; (4) implemented a computational fluid dynamics model for the simulation of urban dispersion into the MCMC capability and tested it using data from the tracer field experiment; (5) developed a computational framework including MCMC, SMC, and hybrid algorithms on massively parallel platforms; and (6) explored methods for incorporating alternative input data types.

Proposed Work for FY06

In FY06 we will: (1) extend the event reconstruction capability to handle complex continental-scale atmospheric releases; (2) implement data, input parameter, and internal model error quantification procedures in the computational framework; (3) implement a multi-resolution capability for more efficient source characterization; (4) continue developing and testing efficient stochastic sampling and convergence algorithms; (5) demonstrate methods for incorporating alternative input data types (e.g., remote sensing and imagery); and (6) continue performance enhancement of the computational framework on the range of platforms for efficient event reconstruction of complex atmospheric releases.

Publications

Chow, T., B. Kosovic, and S. Chan (2005). *Source inversion for contaminant plume dispersion in urban environments using building-resolving simulations*. 9th Ann. George Mason University Conf. Atmospheric Transport and Dispersion Modeling, Fairfax, VA, July 18–20, 2005. UCRL-PRES-213634.

Johannesson, G. and W. Hanley. (2005). "Sequential Monte Carlo based framework for dynamic data-driven event reconstruction for atmospheric releases." *Proc. SPSS Joint Statistical Mtgs.*, Minneapolis, MN, Aug. 7–10, 2005. UCRL-PRES-214316.

Kosovic, B. et al. (2005). *Stochastic source inversion methodology and optimal sensor network design*. 9th Ann. George Mason University Conf. Atmospheric Transport and Dispersion Modeling, Fairfax, VA, July 18–20, 2005. UCRL-PRES-213633.

Coupling Micromechanics and Reactive Fluid Flow in Fracture Networks

Russell L. Detwiler 04-ERD-046

Abstract

The combined influence of mechanical stresses and geochemical reactions leads to alterations in fracture permeabilities that are difficult to predict using currently available models. We are developing and applying coupled computational models of micromechanics and reactive fluid flow in fracture networks that explicitly incorporate the controlling, small-scale physical processes within individual fractures. A parallel implementation of these coupled models will allow us to simulate these processes in networks composed of hundreds of fractures, in which small-scale surface roughness and it's influence on mechanical deformation and geochemical reactions are represented explicitly. These efforts will bridge a critical gap between our current knowledge of small-scale processes and field-scale behavior.

The proposed fracture-network model will explicitly take account of small-scale processes in individual fractures within fracture networks. This will complement existing large-scale dual-continuum models (e.g., the LLNL-developed reactive transport model, NUFT) by providing physically based, quantitative descriptions of the subgrid-scale response to coupled geomechanics and geochemistry. This modeling approach will enhance our ability to predict complex field-scale behavior based on laboratory measurements of the properties of fractured rock. Comparing our approach to efforts using continuum models to predict cap rock integrity at a carbon dioxide (CO_2) injection site will lead to improved understanding of the importance of the competing geomechanical and geochemical processes.

Mission Relevance

Improved understanding of the long-term evolution of permeability of fractured rock under changing mechanical and chemical stresses is critical to a broad range of problems related to DOE's energy and environmental missions. For example, effective implementation of long-term CO_2 sequestration strategies requires a quantitative understanding of how alterations in the stress field caused by CO_2 injection and alterations in fluid chemistry interact to alter storage capacity and potential leakage pathways. Other applications that will benefit from improved understanding of these coupled processes include enhanced oil recovery, engineered geothermal systems, and radioactive waste isolation.

FY05 Accomplishments and Results

In FY05, we coupled a single-fracture reactive transport model with Flex, a model of mechanical deformation of single fractures. In addition, we conducted a systematic parametric and scaling investigation of the single-fracture reactive transport model. These efforts help us understand mechanisms that lead to alteration of fracture permeability. Additional enhancements were made to our model, including incorporating a higher-order advection scheme that allows effective use of coarser grids in simulations. We recently parallelized our flow and reactive transport models by incorporating LLNL-developed sparse-matrix solvers. This allows us to explicitly incorporate the physics represented in the single-fracture model in large-scale fracture networks.

Proposed Work for FY06

We will extend the capabilities of the parallelized fracture-network reactive transport model by coupling it with the geomechanical model LDEC. The mechanical response of individual fractures within the network will be calculated by Flex and passed to LDEC to calculate the network-scale mechanical deformations. Subsequently, flow and transport through the stressed fracture network will be calculated until geochemical alteration of fracture surfaces requires a recalculation of the stresses in the network. Concurrent efforts will be the design and implementation of representative network-scale test problems, such as pressurized CO_2-rich fluid flowing through a fractured cap rock. This will involve defining geochemistry, fracture geometries, and boundary conditions that are representative of CO_2 injection sites.

Publications

Detwiler, R. L., H. Rajaram, and R. J. Glass. (2005). "Satiated relative permeability of variable aperture fractures." *Phys. Rev. E* **71**, 031114. UCRL-JRNL-205167.

Carbon Sequestration and Transport in Natural Environments: The Role of Organic Carbon and Microbial Processes in the Ocean—Observations and Modeling

Thomas P. Guilderson 04-ERD-060

Abstract

The fixation of inorganic carbon by phytoplankton in the ocean and its subsequent export out of the surface ocean (the "biological pump") is one of the fundamental controls on atmospheric carbon dioxide (CO_2) . Production in the ocean is expected to change over a variety of time scales as a result of future climate change and nutrient loading. This project will analyze samples collected in a sediment trap at

the compound class and molecular level and dissolved organic carbon samples to provide a unique perspective on the mechanisms controlling the remineralization of organic carbon in the ocean, including the role of microbial mediation and processes.

This project will provide basic data to elucidate transformation pathways of organic carbon in the ocean. In turn, these data will provide a unique diagnostic suite for enhancements to ocean-biology or ecosystem models that are incorporated into coupled ocean global climate models. It is expected that detailed data-model comparisons will increase the accuracy by which we can predict future changes in the ocean carbon cycle. In particular, this project will facilitate the study of future climate change scenarios and the efficacy of iron-fertilization strategies for carbon sequestration.

Mission Relevance

Fundamental research on the ocean carbon cycle supports DOE environmental and climate change missions. Because this project has a focus on the role of microorganisms in determining the ocean carbon budget, it is also relevant to DOE's Genomics:GtL program.

FY05 Accomplishments and Results

We found that less than 1% of the surface ocean carbon export is permanently sequestered in deep-sea sediments, with 99% being remineralized via microbial processes into various constituents (e.g., amino acids and lipids). Over 50% of the oceans' organic carbon is molecularly uncharacterized carbon (MUC). Isotope and nuclear magnetic resonance analyses on sediment trap organic carbon and dissolved organic carbon indicated that different major constituents have very different cycling rates that are not reflected in the analysis of the bulk material. In particular, lipids appear to have a kerogen-type source. Different biochemical classes were found to dominate MUC in the Cariaco and Santa Barbara Basins, indicating different biogeochemical pathways. Our results were presented at the American Geophysical Union Conference and the 10th International Conference on Accelerator Mass Spectrometry.

Publications

Bratcher, A. J. et al. (2005). "Radiocarbon in surface waters of the Gulf of Mexico and Caribbean as recorded in hermatypic corals." *Eos: Trans. Amer. Geophys. Union* **85** (47), OS13B-0537. UCRL-ABS-206482.

Roland, L., T. P. Guilderson, and M. D. McCarthy. (2005). *Investigating the composition of the molecularly uncharacterized component of sinking POM: stable and radiocarbon comparisons between Cariaco and Santa Barbara Basins.* 10th Intl. Conf. AMS, Berkeley, CA, Sept. 5–10, 2005. UCRL-ABS-215843.

Voparil, I., M. D. McCarthy, and T. P. Guilderson. (2005). *Radiocarbon suggests independent cycling of biochemical constituents of ultrafiltered DOM*. 10th Intl. Conf. AMS, Berkeley, CA, Sept. 5–10, 2005. UCRL-ABS-215845.

Wagner, A. J. et al. (2005). *Radiocarbon in Gulf of Mexico and Cariaco Basin surface waters as recorded in hermatypic corals during the pre- to post-bomb era*. 10th Intl. Conf. AMS, Berkeley, CA, Sept. 5–10, 2005. UCRL-ABS-215846.

Woodworth, M. et al. (2005). *Radiocarbon measurements of bulk organic carbon in sinking particles from the Cariaco Basin*. 10th Intl. Conf. AMS, Berkeley, CA, Sept. 5–10, 2005. UCRL-ABS-215842.

Creating the Core Conditions of Extrasolar and Solar Giant Planets in the Laboratory

Peter M. Celliers 04-ERD-065

Abstract

The goal of this proposal is to determine the properties of planetary materials in a pressure–density–temperature regime that is critical to models of giant planets. The key to a credible planetary model is an accurate equation of state (EOS) for its constituents at extreme conditions. We have developed a method that combines dynamic and static compression techniques using diamond anvil targets to reach relevant conditions. The technique will enable us to create material states that exist in planetary cores. We will load diamond anvil cells with hydrogen (H_2), helium (He), and H_2–He mixtures at an initial pressure of 1 to 5 GPa, and perform EOS (single and double shock) and transport measurements using a large laser as the shock driver.

We expect to characterize the insulating–conducting transition on the He phase diagram at high density and electronvolt temperatures, and to generate similar data on the phase diagrams of H_2–He mixtures. The data will provide important constraints on theoretical models of the EOS and conductivity of these fluids. The results may provide an unambiguous indication of He phase separation in H_2–He mixtures, and thus lead to a new understanding of the internal structure and evolution of Saturn and Jupiter. New high-pressure data on the melting curve of H_2 will further constrain theoretical understanding of high-pressure H_2, particularly in relation to the metal–insulator transition.

Mission Relevance

Basic understanding of the properties of materials at high pressures and high temperatures, especially hydrogen isotopes, is key to the Laboratory's stockpile stewardship mission. This multi-institutional collaborative project also supports LLNL's basic science mission by furthering theoretical understanding of the interiors of giant planets.

FY05 Accomplishments and Results

We extended and consolidated our data sets on dense He and H_2. The measurements produce accurate Hugoniot data (pressure, density, and energy) as well as temperature and optical reflectivity. We measured the Hugoniot of He to 2 Mbar, approximately 4 times higher pressure and 2.5 times higher density than previous shock experiments. We also produced Hugoniot data for H_2 and deuterium (D_2), which can be compared to the existing data on cryogenic D_2. Several papers are in preparation detailing our measurement techniques and our data on He and H_2. Recently we performed the first two experiments on He–H_2 mixtures. The experiments were successful and indicate that the characteristics of the mixture (reflectivity and temperature) are intermediate between those of the pure phases.

Proposed Work for FY06

For the third year of the project, we will collect a few more data points on the pure phases of He and H_2, which are needed to fill gaps in our database. Following this, we will focus our investigation on He–H_2 mixtures. Because they are immiscible at cryogenic ambient pressures, and because these mixtures have never been studied at high pressures and temperatures, all the data we collect will be unique and unprecedented. Our studies of He–H_2 will begin to address the issues of He–H_2 miscibility, the transition to conducting states in the mixture, and the effect of the mixture on the EOS.

Publications

Celliers, P. M. et al. (2005). *Approaching metallic states in dense helium*. 14th APS Topical Conf. Shock Compression of Condensed Matter, Baltimore MD, July 31–Aug. 5, 2005. UCRL-PRES-214199.

Celliers, P. M. et al. (2005). *Equation of state and transport properties in dense helium*. Intl. Workshop Warm Dense Matter, Vancouver, Canada, Oct. 5–8, 2005. UCRL-PRES-216085.

Eggert, J. H. et al. (2005). *Equation of state and transport measurements under astrophysical conditions*. Intl. Conf. High Pressure Science and Technology, Karlsruhe, Germany, June 27–July 1, 2005. UCRL-PRES-214591.

The Large Synoptic Survey Telescope and Foundations for Data Exploitation of Petabyte Data Sets

Kem H. Cook 04-ERD-070

Abstract

The next generation of imaging surveys in astronomy, such as the Large Synoptic Survey Telescope (LSST), will require multigigapixel cameras that can process enormous amounts of data read out every few seconds. This huge increase in data throughput over current capabilities calls for a new paradigm for extracting the knowledge content. We will develop the foundations for this new approach. In the process, we will produce significant scientific breakthroughs by developing new methods to probe both the elusive time and spatial variations in astrophysics data sets from the SuperMACHO (Massive Compact Halo Objects) survey, the Lowell Observatory Near-Earth Object Search (LONEOS), the Livermore Optical Transient Imaging System (LOTIS), and the Taiwanese American Occultation Survey (TAOS).

This project will develop the scientific foundations for future wide-field, time-domain surveys, while our algorithm and pipeline development will pave the way for the LSST science software system. If this project is successful, LLNL will play a major role in the LSST, which will have significant applications for LLNL imaging and data-mining activities. We have already published many significant scientific results and expect to publish more.

Mission Relevance

This project supports the national security mission by developing the LSST camera and software for solving a variety of technical problems in imaging and data mining that are directly applicable to surveillance for nonproliferation. It also supports the DOE goal of understanding the nature and distribution of dark matter and the nature and evolution of dark energy.

FY05 Accomplishments and Results

In FY05 we (1) continued to detect microlensing in real time in the SuperMACHO survey and won Hubble Space Telescope time to support SuperMACHO; (2) analyzed unusual B-type emission-line stars and active galactic nuclei discovered with SuperMACHO; (3) discovered three new supernova light echoes in the Large Magellanic Cloud; (4) discovered two new populations in the Milky Way's halo using LONEOS data, leading us to a better understanding of the halo's formation; (5) identified and analyzed new sets of eclipsing binaries, R Coronae Borealis stars, and long-period variables; (6) refined the SuperMACHO reduction pipeline, using it to prototype an LSST pipeline system; and (7) designed a database schema for the SuperMACHO data as an LSST prototype, using it to extract new scientific information and investigate scaling and use scenarios.

Proposed Work for FY06

We will continue to support SuperMacho plans to assist a Deep Impact spacecraft extended mission to measure microlensing parallax and determine the location of a lens. Specifically, we will (1) re-reduce MACHO image data using the SuperMACHO pipeline; (2) load LONEOS, MACHO, and SuperMACHO data into the prototype LSST database; (3) continue to develop data-mining tools and expertise using MACHO, SuperMACHO, and LONEOS data; (4) develop new algorithms for finding unusual objects and test new algorithms for characterizing light curves; (5) analyze LONEOS data from the new camera; (6) extend our analysis of the Milky Way halo; and (7) finish the development of the LSST prototype pipeline and run it on SuperMACHO and LONEOS.

Publications

Cook, K. H. et al. (2005). *LSST operational cadence simulation and design*. American Astronomical Society 205th Mtg., San Diego, CA, Jan. 11, 2005. UCRL-POST-208955.

Fraser, O. J. et al. (2005). "Long-period variables in the large magellanic cloud: results from MACHO and 2MASS." *Astron. J.* **129**, 768. UCRL-JRNL-205457.

Kunder, A. M. et al. (2005). *Metallicity of galactic bulge RR Lyrae*. American Astronomical Society 205th Mtg., San Diego, CA, Jan. 11, 2005. UCRL-POST-208954.

Ngeow, C.-C. et al. (2005). "Further empirical evidence for the non-linearity of the period-luminosity relations as seen in the Large Magellanic Cloud Cepheids." *Mon. Not. Roy. Astron. Soc.* **363**, 831. UCRL-JRNL-213034.

Popowski, P. et al. (2005). "Microlensing optical depth toward the galactic bulge using clump giants from the MACHO survey." *Astrophys. J.* **631**, 879. UCRL-JRNL-213625.

Rattenbury, N. J. et al. (2005). "Determination of stellar shape in microlensing event MOA 2002-BLG-33." *Astron. Astrophys.* **439**, 645. UCRL-JRNL-214971.

Seppala, L. G. et al. (2005). *Optical design for the 8.4-m Large Synoptic Telescope*. American Astronomical Society 205th Mtg., San Diego, CA, Jan. 11, 2005. UCRL-ABS-207882.

Thomas, C. L. et al. (2005). "Galactic bulge microlensing events from the MACHO collaboration." *Astophys. J.* **631**, 906. UCRL-JRNL-213632.

Mission to Very Early Earth

Ian D. Hutcheon 04-ERI-004

Abstract

This project conducts a combined analytical and experimental program to study mineral and melt inclusions in ancient zircons (age >4.0 billion years) to determine when conditions suitable for life first emerged on Earth. After gathering the geochemical data as a function of zircon age, we will relate these data to the respective parent materials. The work involves dating and isotopically and chemically analyzing the zircons and the mineral and melt inclusions in them. The experimental petrology portion focuses on the partitioning of trace elements, notably uranium and actinides, between zircon and melt. The investigation uses a broad array of new microanalytical facilities that also support a variety of Laboratory nuclear forensic applications.

The most important scientific output of this project is the chemical and isotopic characterization of Jack Hills zircons as a function of their age. Based on these new data we will (1) understand how the fossil record of ancient life is preserved today; (2) determine the time when conditions suitable for supporting life first emerged on Earth; (3) understand the evolution of the atmosphere and hydrosphere during the Hadean period, the earliest epoch of Earth's history; and (4) evaluate the evidence for magmatic activity and the formation of evolved felsic rocks (granitoids) very early in Earth's history, as far back as 4.4 billion years.

Mission Relevance

The project develops and enhances advanced microanalytical capabilities in support of LLNL's national security missions and advances its mission in basic science. In addition, the knowledge of the Earth's atmosphere and hydrosphere will benefit NASA and DoD space missions.

FY05 Accomplishments and Results

Work in FY05 focused on measuring trace element partitioning between zircon and silicate melt, measuring trace element abundance patterns in Jack Hills zircons, and examining the evidence for the biogenic origin of graphitic inclusions in 3.7-billion-year-old apatite on Akilia Island, Greenland. Laser ablation and inductively coupled plasma mass spectrometry yielded positive cerium (Ce) and negative europium anomalies and heavy rare-earth element (REE) enrichment. Noteworthy was significant scatter in light REE abundance with chondritic samuirium/lanthanum ratios from 1 to 600. We found no correlations between REE and age or oxygen (O) isotopes. Nano-secondary-ion mass spectroscopy analyses of synthesized zircons showed that barium is highly incompatible and that more compatible elements (e.g., Ce and uranium) are inhomogeneously distributed within individual zircons.

Proposed Work for FY06

The proposed work for FY06 is divided into analytical and experimental components. Analytical activities will focus on retrieving chemical and isotopic information preserved in Jack Hills zircons, while experimental work will focus on phase and isotopic equilibria studies at elevated pressure and temperature. We will test the "cool early Earth" hypothesis by analyzing titanium (Ti) and REE contents of zircons from ancient tonalites. If the Ti content of zircons from tonalites is similar to Jack Hills zircons, the link between apparent low temperatures, sediment melting, and the presence of a hydrosphere is no longer unique. We will determine the fractionation of O isotopes and the partitioning of the REE between zircon and granitic melts to elucidate the provenance of Jack Hills zircons.

Publications

Hutcheon, I. D. et al. (2005). "NanoSIMS Mg isotope analyses of refractory inclusions in metal-rich CB chondrites." *Geochim. Cosmochim. Acta* **69**, A524. UCRL-ABS-202615.

Iodine-129 Accelerator Mass Spectrometry for Earth Science, Biomedical, and National Security Application

Gregory J. Nimz 04-ERI-013

Abstract

This project will enable iodine-129 (^{129}I) analysis by accelerator mass spectrometry (AMS) and build a foundation for its scientific application by developing chemical processing methods for environmental and nonproliferation use. During nuclear reprocessing, ^{129}I escapes to the environment, becoming an indicator of clandestine nuclear activity. Creating an AMS analysis capability requires optimizing interrelated instrumental parameters and developing methods for low-level iodine chemical extraction. As proof-of-principle of our new capabilities, ^{129}I in soil, groundwater, and plants will be analyzed. In FY04 we focused on AMS instrumental development; in FY05 and FY06 we are focusing on processing techniques, proof-of-principle demonstrations, and collaborative research.

This research is expected to result in (1) the ability to analyze small quantities of materials with extremely low $^{129}I/I$ ratios; (2) creation of a new generation of iodine-sampling techniques; (3) methods for iodine extraction from a wide range of natural materials (soil, groundwater, plants); and (4) an understanding of labile ^{129}I migration in the environment. It permits detection of clandestine nuclear activity and facilitates basic research in ecology, hydrology, agriculture, oceanography, and human nutrition. Publishable results include newly developed AMS procedures, characterization of ^{129}I migration in soils (relevant to determining moisture flux, colloid transport, and nutrient cycling), and ^{129}I hydrologic tracing methods (relevant to projecting effects of climate change on water resources).

Mission Relevance

This project provides techniques for nuclear forensics and nonproliferation in support of LLNL's national-security mission; develops technology for contaminant characterization in support of LLNL's mission in environmental management; and develops methods for ocean circulation analysis and biomedical research in support of LLNL's commitment to breakthroughs in fundamental science.

FY05 Accomplishments and Results

This year we focused on capabilities for natural matrices. Many important applications provide only small iodine samples, so we reduced AMS requirements from >1 mg to <200 µg, and characterized backgrounds at 10 µg. We developed quantitative iodine extraction from soils, employing both furnace combustion and organic extraction of complex leachates. We established gas- and amperometric ion-chromatography to measure iodine concentration. Technique verification employed International Atomic Energy Agency and National Institute of Standards and Technology standard reference soils. We began analysis of soil bomb-pulse ^{129}I to determine moisture flow and iodine migration. Collaborative ^{129}I hydrology research on climate change on California water resources was initiated with University of California (UC) at Merced, and we began investigating ^{129}I as a means of dating oceanic methane hydrates.

Proposed Work for FY06

The focus in FY06 will be proof-of-principle demonstration projects for developing national security capabilities. The soil bomb-pulse ^{129}I project will be completed and submitted for publication. The collaborative California hydrology project will be furthered and initial ^{129}I results submitted for publication. Final completion will be a UC Merced Ph.D. thesis. A third project will develop techniques for analyzing ^{129}I in methane hydrate sediment pore waters to determine their age and stability. The advanced ^{129}I methods developed in this project for soils, surface waters, and small-scale samples are fundamental to ^{129}I national-security applications.

Carbon Flux in a California Grassland Soil Sequence: The Role of Dissolved Organic Carbon in Carbon Sequestration

Christopher W. Swanston 04-ERI-014

Abstract

Soils have the capacity to stabilize large amounts of carbon. A thorough understanding of the factors that control this capacity is critical for defining input parameters used by models that predict climate change. The role of dissolved organic carbon (DOC) in carbon sequestration in soils is currently poorly understood. We propose to measure the carbon isotopic composition of DOC, bacterial biomass, and soil organic matter (SOM) below annual grasslands on California coastal terraces to identify important transport pathways and carbon cycling rates. The project will document compositional changes in DOC and SOM through the soil profile, which will complement the isotopic measurements. This research will result in accurate and comprehensive soil carbon cycling models at the site level.

We expect to quantify production and stabilization of DOC, identify important soil carbon cycling rates and pathways, measure mineralogical and seasonal effects on these cycling rates and pathways, and integrate flux measurements with a site-level soil carbon model. Insights into the role of DOC in transport and sequestration of soil carbon will improve the accuracy of input parameters needed by global carbon-cycling models.

Mission Relevance

The proposed research will enhance understanding of soil carbon cycling in support of LLNL's mission in environmental management, and it will provide valuable input for various DOE programs investigating the effects of increased levels of atmospheric carbon dioxide on the Earth's climate. The project will also strengthen LLNL collaborations with researchers at two University of California (UC) campuses and the Lawrence Berkeley National Laboratory.

FY05 Accomplishments and Results

We continued with root, soil, and DOC sampling and analysis to measure natural variability in samples and incorporate preliminary results and additional pathways into the soil model as required. We performed soil separation and characterized the particulate carbon cycle using radiocarbon values and steady state flux models. These data, and data showing mineral attenuation of DOC flux, were used in the broader terrace studies. We found that overall, the mean residence time of carbon in the upper soils of the terraces was similar (~6 years) but that the controls of carbon stabilization varied by terrace age. As terrace age increased, the primary mechanism of stabilization shifted from structural protection through soil aggregation to mineral protection through organo-mineral interactions. This coincides with increased attenuation of DOC flux with increasing terrace age, presumably due to more effective mineral stabilization, as well. In practical application, these results suggest that the younger terraces are more susceptible to rapid carbon loss resulting from changes in land management or climate. These initial findings confirmed our original hypotheses regarding basic trends with terrace age, and subsequent measurements in FY06 will further test the relationship between particulate and dissolved carbon fractions.

Proposed Work for FY06

In FY06 we intend to (1) finish sample collection, preparation, and analysis; (2) incorporate data into a site-level biogeochemistry and carbon cycling model; (3) submit two manuscripts to peer-reviewed journals; and (4) investigate (in collaboration with the U.S. Geological Survey and UC Merced), the interaction of fire with the carbon cycle using a controlled burn at one of the terraces.

Publications

Swanston, C. W. et al. (2005). “Initial characterization of processes of soil carbon stabilization using forest stand-level radiocarbon enrichment.” *Geoderma* **128**, 52. UCRL-JRNL-201893.

Constraints on the Nature of Terrestrial Core-Forming Melts: Ultrahigh-Pressure Transport Property Measurements and X-Ray Computed Tomography

Jeffery J. Roberts 04-LW-077

Abstract

This project is an integrated study of the electrical conductivity, texture, and permeability relationships of silicate and sulfide partial-melt samples to provide much-needed constraints on models of planetary core formation. Tasks include synthesis and characterization, electrical conductivity measurements, and x-ray computed microtomography (CMT) three-dimensional (3-D) imaging. The goal is a unified model of the bulk conductivity and permeability of a partial melt that incorporates the melt geometry. Material synthesis will occur in piston cylinder and multi-anvil devices at pressures up to 20 GPa. Recovered samples will be characterized by CMT.

We expect to obtain experimental measurements of electrical conductivity and 3-D images of wetting and nonwetting silicate sulfide partial melts and to form the basis for determining melt extractability based on CMT. We hope to have advanced microscale imaging using x-ray CMT to near-micrometer

spatial resolution. Successful development of a cell for in situ measurement of electrical conductivity in LLNL's multi-anvil device will be a significant advance, setting the stage for measurements made on high-pressure silicate phases, such as perovskite in contact with melt. These results will constrain models of core segregation and compositions of the melt phase. This work will result in several papers.

Mission Relevance

In addition to basic science applications, the development of microscale imaging techniques and the distribution of melt or minority phases on a microscale is relevant to obtaining a better understanding of weapons performance issues, consistent with several DOE national security missions, including stockpile stewardship.

FY05 Accomplishments and Results

Silicate iron sulfide partial melts with a range of compositions, wetting and non-wetting, were synthesized with melt fractions ranging from 2 to 12 vol%. Each of these samples was imaged at the Advanced Light Source (Beamline 8.3.2). A key finding is the significantly different percolation behavior of the nickel-bearing (wetting) and nickel-absent (non-wetting) samples. A number of experimental problems were solved that answered questions raised by conflicting results from other researchers. We have calculated permeability using an implicit lattice Boltzmann flow simulation. The results are the first estimates of permeability in these systems that are not inferred from empirical relationships based on microstructure. These estimates of permeability provide important constraints on models of planetary core formation.

Publications

Du Frane, W. D. et al. (2005). *Anisotropic and isotropic effective medium olivine models*. American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-214900.

Du Frane, W. et al. (in press). "Anisotropy of electrical conductivity in dry olivine." *Geophys. Res. Lett.* UCRL-JRNL-211347.

Roberts, J. J. et al. (2005). *Electrical conductivity and tomographic imaging of olivine-FeS partial-melts*. Joint American Geophysical Union Assembly, New Orleans, LA, May 23–27, 2005. UCRL-ABS-209514.

Roberts, J. J. et al. (2005). *Permeability of olivine-FeS partial-melts based on tomographic x-ray imaging*. American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-214980.

Wright, H. M., J. J. Roberts, and K. V. Cashman. (2005). *Pore structure of pumice: comparison between laboratory measurements and x-ray tomographic image analysis*. American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-214899.

The Innermost Inner Core: Fact Or Artifact?

Hrvoje Tkalcic 04-FS-019

Abstract

There is considerable debate in the earth sciences over the composition and dynamics of Earth's inner core. Because of the uneven distribution of seismic receivers around the globe, the details of structural models are based on limited seismological observations of the travel times of primary waves traversing the core (PKP waves). Using state-of-the-art signal-processing techniques to measure and compile a unique set of global travel-time data of surface-reflected seismic waves that propagate through the center of Earth (*P'P'* waves), we propose to investigate the existence of the innermost inner core. We will carry out a systematic investigation to determine the configuration of inner core anisotropy, which is currently biased by a limited spatial sampling of the inner core by PKP waves.

We expect to demonstrate if the existence of the innermost inner core can be supported by seismological data and provide major constraints on the amount of inner core anisotropy. This is a topic of very high interest and active research in the earth science community, and the results would be of great interest in seismology and other areas of earth science. We expect to publish our results in peer-reviewed journals.

Mission Relevance

This project enhances and extends a critical Laboratory core competency in seismology for national security, particularly for ground-based nuclear explosions monitoring. This high-profile science project also supports LLNL's mission in basic science by leading to further significant contributions in deep-earth structure, physical properties, and models of core evolution.

FY05 Accomplishments and Results

We observed hitherto unobserved P'P' waves and precursors to P'P' at very small epicentral distances. The P'P' phases provide new sampling of the inner core and their travel times argue against the existence of an innermost inner core. The observation of precursors is unexpected and provides new constraints on Earth's structure. The precursors illuminate the scattering properties of the Earth's upper mantle, and we interpret them as a back-scattered energy from horizontally connected small-scale heterogeneity, in a zone between 150 and 220 km beneath Earth's surface. These results are not only of interest to a wide range of scientists including mineral physicists and mantle convection modelers, but the existence of a newly discovered structures beneath the oceans also represent a new challenge for ground-based nuclear explosion monitoring.

Publications

Flanagan, M. P. , H. Tkalcic, and V. F. Cormier. (2005). *Evidence for back scattering of near-podal seismic P'P' waves from the 150–220 km zone in Earth's upper mantle*. 2005 American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-214997.

Tkalcic, H. and M. P. Flanagan. (2004). *Structure of the deep inner core from antipodal PKPPKP waves*. 2004 American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 13–17, 2004. UCRL-CONF-206394.

Tkalcic, H., M. P. Flanagan, and V. F. Cormier. (in press). "Evidence for back scattering of near-podal seismic P'P' waves from the 150-220 km zone in Earth's upper mantle." *Geophys. Res. Lett.* UCRL-JRNL-213682.

Geophysics Experiments on High-Powered Lasers

John J. Zucca 04-FS-031

Abstract

An area of research important for monitoring underground nuclear explosions is cavity decoupling. Using actual nuclear explosions for such research is precluded, and tests using high explosives produce inadequate data because pressure from the high-explosive gases confuses the results. We propose to investigate the feasibility of using high-power laser-driven experiments to create scaled explosion systems to study cavity decoupling. We have the opportunity to demonstrate this concept of scaled geophysics experiments at the Z-Beamlet Laser at Sandia National Laboratory. This would demonstrate the feasibility of conducting such experiments on high-powered laser systems.

This study will demonstrate that such scaled experiments are possible, paving the way for large-scale experiments to explore the critical issues in cavity decoupling using fusion-class lasers.

Mission Relevance

This study supports LLNL's national-security mission by exploring the feasibility of using fusion-class lasers to gain information on cavity decoupling, a high-priority topic in the science of global nuclear-explosion monitoring.

FY05 Accomplishments and Results

In FY05, we (1) conducted laser shots at the Z-Beamlet Laser; (2) inspected the targets at LLNL, reduced and interpreted the data, and completed the final report; (3) using our detailed computational work, determined the ratio of peak pressures recorded at the surface of the fully coupled and decoupled targets, as well as the increase in radius of the two targets; and (4) based on this demonstration of the capability to conduct cavity-decoupling experiments using high-powered lasers, devised plans that could be used to guide follow-on experiments.

Publications

Dunlop, W. and J. J. Zucca. (2005). *Final report for geophysical experiments using high-power lasers*. UCRL-TR-217864.

A New Capability for Regional High-Frequency Seismic Wave Simulation in Realistic Three-Dimensional Earth Models to Improve Nuclear Explosion Monitoring

Arthur J. Rodgers 05-ERD-019

Abstract

We propose to perform a proof-of-concept simulation that will extend model-based signal processing to lower thresholds and improve confidence for seismic nuclear explosion monitoring (NEM). Currently, NEM is highly dependent on empirical observations of explosions and earthquakes and therefore is spatially limited. In addition, inherent noise at seismic recording stations limits signal detection. Earth models derived from diverse data sets that include realistic stochastic heterogeneity will be used to compute theoretical seismograms and represent an observed seismogram using a subspace methodology.

The proposed research addresses several challenges to current NEM technologies. Enabling the generation of Earth models for areas with few or no earthquakes or explosions will allow for monitoring in broad, uncalibrated areas. We expect the use of coherent signal processing will drastically lower detection thresholds.

Mission Relevance

If successful, this project would provide new capabilities to support LLNL's national security mission in ground-based nuclear explosion monitoring.

FY05 Accomplishments and Results

In FY05 we (1) developed the basic technical infrastructure for the project; (2) learned how to use the CalTech-developed seismic wave simulation code (SPECFEM3D) on Livermore Computing and modified it to implement long-wavelength (>220 km) Markov-Chain Monte Carlo (MCMC) models of seismic velocities and densities in the Korean Peninsula; (3) computed model-based signals for eight different MCMC models of the region for our test event; (4) developed the signal-processing software to represent the observed signals in terms of an optimal singular-value decomposition of the model-based signals; and (5) developed software to generate small-scale (<100 km) random heterogeneity that will be used in FY06 to increase the frequency content of the model-based signals.

Proposed Work for FY06

In FY06 we propose to increase the frequency content of the model-based signals to investigate the applicability of this technology to smaller events. This will require using a different version of the SPECFEM3D seismic wave simulation code, or the LLNL-developed WPP elastic finite difference code. These codes will allow for calculation of higher frequencies (>1 Hz). We will also develop improved MCMC models, going from 220 to 110 km resolution for the Korean Peninsula. Finally, small-scale (<100 km) random heterogeneity will be included to represent scattering of high-frequency seismic waves.

Publications

Rodgers, A. and J. Tromp. (2005). *Modeling nuclear explosions and earthquakes with the spectral element method and high-performance computing*. Seismological Society of America Annual Mtg., Incline Village, NV, Apr. 27–29, 2005. UCRL-ABS-208922.

Rodgers, A. et al. (2005). *A model-based signal-processing approach to seismic monitoring: stochastic Earth models and spectral element method synthetics*. 2nd SPICE Workshop, Smolenice, Slovak Republic, Sept. 4–10, 2005. UCRL-ABS-213711.

CHEMTREAT: Accelerated Remediation of Contaminated Fine-Grained Sediments by a Chemical Clay Cracking and Co-Solvent Flushing Process

Ananda M. Wijesinghe 05-ERD-028

Abstract

Contaminants trapped in low-permeability, high-sorptivity, fine-grained clay sediments are inaccessible to advectively delivered treatment fluids. This is a hitherto unsolved, multibillion-dollar problem in groundwater remediation. We will investigate a process for chemically shrinking and cracking the sediments, and then flushing out the trapped contaminants using an effective chemical cracking agent and co-solvent such as ethanol. Laboratory experiments will be performed to determine the constitutive properties, crack propagation velocities, crack spacings and patterns, and to test methods of cost-effectively delivering the chemicals and extracting the contaminants. Using these results, we will develop and verify predictive models and codes for designing the field tests to follow.

This project will (1) develop models to reliably predict the performance of this novel chemical clay-cracking process and (2) explore and define the design limits of the process. We will build on results from a previous project that demonstrated feasibility of the basic concept by showing that cracks can be created under confining stress by chemically induced shrinkage, and that the speed of cracking is not limited by the slow rate of diffusion of the cracking agent into the clay. This project will develop a method of overcoming contaminant entrapment by fine-grained impermeable sediments that is the primary barrier to cost effective cleanup of contaminated groundwater. No other technology is currently available for this purpose.

Mission Relevance

This project supports the Lab's environmental management mission by developing an effective method of remediating contaminated fine-grained sediments that is not possible with currently available technology. This addresses a serious threat to the national water resources and furthers the remediation of intractable contaminant sources at several DOE sites. Enhanced understanding of chemically induced cracking will also be relevant to many other applications, including the design of waste containment facilities, and predicting the performance of nuclear waste repository seals.

FY05 Accomplishments and Results

We (1) performed a series of constitutive experiments to define the shrinkage, deformation, species diffusion, and fracture properties of clays; (2) conducted crack breakthrough experiments to measure chemical crack initiation and crack propagation under confining stress; (3) developed theoretical models for elastoplastic deformation, a microstructural Derjaguin-Landau-Verwey-Overbeek model for shrinkage, and a hydraulic-fracturing critical stress intensity factor model for clays; (4) programmed finite-element codes, and designed and analyzed the experiments; (5) acquired a triaxial loading system and built test cells for automated deformation, shrinkage, and permeability testing; and (6) developed a multichannel fiber-optic Raman spectroscopic system for automated real-time measurement of chemical concentrations.

Proposed Work for FY06

In FY06, we will (1) build and perform a fluorescent dye imaging experiment on an areally extensive clay layer that is contacted and cracked by exposure to ethanol, to determine the crack density, crack spacing, and crack patterns as functions of confining stress, ethanol concentration, fluid chemistry, and clay type; (2) develop modeling concepts, including a theoretical model and finite-element codes to predict the crack-spacing/density characteristics of the experimental crack patterns; (3) continue and extend the constitutive experiments of FY05 to real clays from LLNL's Site 300 having both clay and sand components; (4) complete the x-ray imaging crack-velocity experiments that could not be performed as scheduled in FY05 due to breakdown and unavailability of the x-ray scanning equipment.

Controls of Fluid Chemistry on Fracture Growth

Carol J. Bruton 05-ERD-035

Abstract

The role of water and its dissolved content in fracturing and rock deformation is poorly understood. The controls of surface chemistry on fracturing and the relation between fracturing and breakage of bonds during mineral dissolution suggest new ways to quantify the impact of fluid chemistry on fracturing. We are quantifying these effects to advance our understanding of the role of solute-bearing water on rock deformation and applying this knowledge to predict and engineer fracture growth in subsurface materials as a function of their environment. The hydrothermal atomic force microscope (HAFM) is being used to measure the velocity of fracture growth in single crystals and rocks as a function of fluid chemistry and pH at temperatures up to 150°C.

The expected outcome of this project is a validated predictive capability for describing subcritical crack growth to 150°C in crustal rocks bonded by quartz or calcite. This model is required in fossil fuel production and geothermal energy industries to control fracturing during drilling, hydrofracturing, and injection. We will break through the 80°C barrier that has limited previous experimental efforts, and

link surface chemistry and rock mechanics to enable significant interdisciplinary advances. The development of a HAFM incorporating a sample bending apparatus will allow the imaging and direct measurement of fracture growth velocity in any material of interest with control of fluid chemistry at temperatures up to 150°C. Research results will be published in peer-reviewed journals.

Mission Relevance

This work supports LLNL missions in energy security and carbon management through its applications to DOE programs in fossil and geothermal energy. Project results can be used to evaluate the feasibility of manipulating the chemistry of fluids to control fracture growth and change permeability during drilling, hydrofracturing, and injection, which will enhance energy production, increase resource lifetime, and reduce the cost of energy production. Our HAFM-based technology can also be used to quantify the impact of the thermal and chemical environment on stress corrosion cracking (SCC) of materials such as laser glass, and on material lifetimes for application to repository and materials science. The HAFM can detect slower rates of SCC than other technologies.

FY05 Accomplishments and Results

We built a specially designed sample bending apparatus (jig) to fit within the HAFM sample chamber. A Vickers indenter was used to initiate a 5-µm-long crack in float glass, our initial test material. When the glass plate was positioned in the bending jig so that the induced crack was directly over the pivot point, we applied bending stress to grow the crack to about 30 µm in the HAFM while tracking fracture growth. We identified a means of calculating stress intensity at a crack tip using the curvature of the surface (measured with a vertical scanning interferometer) and the material properties assuming ideal geometry. Finite element modeling was used to calculate the macroscopic stress and deformation caused by bending the sample, for comparison with experimental results.

Proposed Work for FY06

During FY06, we expect to (1) complete fracture propagation studies for float and silica glasses under hydrothermal conditions and varying pH and fluid composition; (2) compare results to published data for macroscopic samples; (3) initiate and complete fracture propagation measurements for quartz under pH, fluid composition, and temperature regimes appropriate for testing a published model of fracture growth kinetics; (4) identify other minerals to study as single crystals and as polycrystalline materials; (5) approximate more realistic boundary conditions in our finite-element models; (6) calculate the stress intensity at the crack tip, taking into account the geometry of the crack; and (7) also model the impact of thermal perturbations on the elastic moduli and thus, the stress field.

Publications

Bruton, C. J. et al. (2005). “Controls of fluid chemistry on subcritical crack growth.” *EOS Trans. AGU* **86**(52), Fall Mtg. Suppl., Abstract MR33A-0140. UCRL-ABS-215306.

Integration and Codevelopment of a Geophysical CO_2 Monitoring Suite

Samuel J. Friedmann 05-ERD-038

Abstract

As geological carbon sequestration increases in importance as a carbon management strategy, an outstanding need exists for subsurface monitoring, measurement, and verification (MMV) tools. In particular, inexpensive, non-intrusive, and well-calibrated approaches are required. We propose using the Lab's Stochastic Engine (a Monte Carlo, Markov-Chain algorithm) to compare and simultaneously invert data from three orthogonal geophysical approaches. This will provide alternative approaches to subsurface CO_2 monitoring as well as oil field flow-front management.

If successful, we expect to be able to determine the distribution and concentration of subsurface CO_2 using these combined, jointly inverted data. This would greatly improve risk characterization associated with CO_2 storage and provide potential operators, regulators, and petroleum producers with information to manage CO_2 injection and storage. Additional significance would come from an improved scientific understanding of key subsurface processes and uncertainties (e.g., subsurface heterogeneity).

Mission Relevance

The research supports a variety of DOE geologic carbon-sequestration efforts and is strongly aligned with areas of the Lab's environmental management mission, including carbon management and environmental risk reduction.

FY05 Accomplishments and Results

In FY05, we (1) built a Teapot Dome shared-Earth model (ShEM); (2) developed ShEM-based forward models for each geophysical tool; (3) developed the interface between the ShEM and the Stochastic Engine; (4) migrated our codes onto a Lab supercomputer, validated their individual performance, and began staged runs on synthetic data to verify the codes; and (5) found and began assessing two potential test sites.

Proposed Work for FY06

In FY06 we will (1) identify an industrial site for monitoring suite development; (2) collect and interpret data from the industrial site; (3) develop and initiate plans for automation of electrical resistance tomography, tilt-meter, and microseismic tools; (4) complete the industrial site ShEM; (5) improve the microseismic location, characterization, and mapping algorithms; (6) collect data over two to three months of injection; (7) reduce the injection signal data into mutually usable forms; (8) use the Stochastic Engine to compare tool data sets and validate the Earth model; (9) write up and present our results in journals and at conferences; and (10) license one or more of our MMV approaches to a commercial entity.

Detection and Attribution of Regional Climate Change

Govindasamy Bala 05-ERD-042

Abstract

This project will perform high-resolution coupled atmosphere/ocean climate simulations that (1) enable regional-scale climate change detection over a time scale of up to centuries; (2) facilitate understanding of how the spatial resolution of climate model affects our ability to detect climate change at regional scale; and (3) drive studies of potential climate change on regional policy issues. This project will use LLNL's Thunder machine to perform first-ever numerical experiments of high-resolution coupled climate models with and without anthropogenic forcings and use climate-change-detection software developed at LLNL. We will collaborate with the San Diego Supercomputing Center (SDSC) and the University of California at San Diego (UCSD) to use this high-resolution data to investigate related hydrological changes.

The consequences of global energy production and use appear first at the regional level. We expect to gain insight into regional-scale climate change detection and impact and understand how uncertainties in anthropogenic forcings in our regional climate models affect our ability to detect regional climate change. Results of these studies will provide a suite of simulations for continued analysis at a regional scale of phenomena that may have a great societal impact (e.g., water resources, agriculture, and storms). Papers describing our results will be submitted for publication in peer-reviewed journals.

Mission Relevance

The simulations performed in this study will form the foundation for regional-scale analyses of carbon and air chemistry that support LLNL's energy and water security missions and the DOE's missions in environmental management and climate change.

FY05 Accomplishments and Results

Tasks completed in FY05 include (1) identifying climate variables that provide information on expected regional climate changes based on existing climate model runs; (2) identifying sources of high-quality observational data for the proposed detection variables; (3) performing long, high-resolution, coupled climate-model control simulations for present-day and pre-industrial conditions (the simulated climate was validated in collaboration with the National Center for Atmospheric Research); (4) began further diagnostic work to assess the ocean and ice simulations; and (5) started transferring data to collaborators at SDSC.

Proposed Work for FY06

In FY06, we will perform an ensemble of historical high-resolution coupled global climate simulations on Thunder. In this simulation, we will downscale the pre-industrial control and one of the historical ensemble using the WRF (Weather Research Forecasting) regional climate model, and perform global simulations with a 1% increase of CO_2. We will also use our pre-industrial and historical simulations from both global and regional models to perform simulations aimed at detection and attribution of regional climate change in the western U.S. We will also continue transferring simulation data from global and regional climate models to SDSC, where UCDC hydrology modelers will use our climate data in the Variable Infiltration Capacity (VIC) hydrology model to assess the historical changes in hydrology in the western U.S.

Publications

Bala, G. (2005). *Atmospheric model diagnostics*. UCRL-WEB-206543.

Mirin, A. and G. Bala. (2005). *Data from simulations of community climate system model*. UCRL-MI-215754.

Oliker, L. et al. (2005). "Leading computational methods on scalar and vector HEC platforms." *Proc. Supercomputing 2005 Conf.*, Seattle, WA, Nov. 12–18, 2005. UCRL-CONF-212184.

Rood, R., G. Bala, and A. Mirin. (2005). *A curious discussion of sea ice*. 10th Annual CCSM Workshop, Breckenridge, CO, June 21–23 2005. UCRL-PRES-218024.

Wehner, M. et al. (2005). "Towards a direct simulation of human induced changes in the hurricane cycle." *Proc. Supercomputing 2005 Conf.,* Seattle, WA, Nov. 12–18, 2005. UCRL-POST-216579.

A Dynamically-Coupled Groundwater, Land Surface Water, and Regional Climate Model to Predict Seasonal Watershed Flow and Groundwater Response

Reed M. Maxwell 05-ERD-043

Abstract

This project will develop a new model that couples three distinct submodels for climate, land surface hydrology, and groundwater flow that are traditionally used independently with oversimplified boundary conditions. It is hoped that the new model will yield a better understanding of important mass and energy couplings and afford more accurate predictions of seasonal watershed flow and groundwater response to precipitation. We expect to determine (1) whether the simplified lower boundary condition currently used in the regional climate model is reasonably accurate; (2) whether interactions among the atmosphere, land surface, and subsurface have a substantial impact upon mass, energy, and momentum balances; (3) the extent to which practical and stability considerations affect the dynamic coupling of these models; and (4) how well the models represent observations of physical processes within a watershed.

We expect that the new model will provide (1) an improved capability to simulate the physical processes through which the subsurface and precipitation are coupled; (2) a new tool for predicting surface water and groundwater hydrology at the watershed scale on a seasonal timeframe; (3) a better understanding

of the hydrologic response of watersheds that will provide insight into hydrologic forecasting; and (4) a more realistic representation of weather predictions at the regional scale.

Mission Relevance

This project will contribute to the evolution of regional climate, land surface water, and groundwater models in support of DOE's environmental management and climate change missions, and lead to breakthroughs in energy and water security research.

FY05 Accomplishments and Results

In FY05, regional reanalysis data for the Southern Great Plains (SGP) site in the central U.S. obtained from the National Centers for Environmental Prediction (NCEP) were used to initialize and to drive the regional climate model. We also acquired test data that are needed for surface water and groundwater model calibration, including water levels and subsurface lithology from groundwater wells, and weather information from seven meteorological stations within a portion of the SGP site (i.e., the Little Washita watershed). A subsurface geologic description and topographical and surface cover/land-use descriptions of the test watershed have been generated from these data, and land, surface, and groundwater flow models have been set up and tested. The Advanced Regional Prediction System (ARPS) climate model was run over the SGP domain, driven by NCEP reanalysis data.

Proposed Work for FY06

In FY06, we will (1) dynamically couple the ARPS and land surface water and groundwater models; (2) test the coupled model to ensure mass and water balance between the submodels; (3) run the fully coupled model over the SGP domain driven by the NCEP data with a focus on the Little Washita watershed; (4) compare the code results with available data, the uncoupled control run done in FY05, and other models run over the same watershed; and (5) evaluate seasonal prediction capabilities of the new model.

Publications

Kollet, S. J. and R. M. Maxwell (in press). "RM integrated surface–groundwater flow modeling: A free-surface overland flow boundary condition in a parallel groundwater flow model." *Adv. Water Res.* UCRL-JRNL-211468.

Kollet, S. J. and Maxwell, R. M. (2005). "Distributed watershed modeling: Incorporation of a free-surface overland flow boundary condition and land surface parameterization scheme into a parallel, variably saturated groundwater flow model." *Eos Trans. AGU* **86**(52), Fall Mtg. Suppl., H21L-05. UCRL-ABS-215079.

Maxwell, R.M. et al. (2005). *Simulating water and energy fluxes using a coupled groundwater, surface water, land surface and regional climate model*. 5th Intl. Scientific Conf. Global Energy and Water Cycle, Costa Mesa, CA, June 20–24, 2005. UCRL-POST-218459.

Developing a Reactive Chemistry Capability for the NARAC Operational Model (LODI)

Philip J. Cameron-Smith 05-ERD-050

Abstract

Recent work has demonstrated the potentially large effects of atmospheric chemistry on the impact of inadvertent or intentional chemical releases of gaseous compounds such as chlorine and nerve agents. To address the current imbalance between the dispersion and chemistry capabilities of LODI, LLNL's National Atmospheric Release Advisory Center (NARAC) operational response model, this project proposes research that will lead to a full reactive chemistry and aerosol capability for LODI that will account for an arbitrary network of chemical reactions and the evaporation and condensation of aerosols. The upgrade will include the ability to read time- and space-dependent ambient concentrations of relevant species from IMPACT, LLNL's global atmospheric chemistry model.

The addition of a full reactive chemistry capability to LODI, plus the interfacing with the IMPACT global atmospheric chemistry code, will create a unique emergency-response capability that could be used to enhance NARAC's ability to respond to terrorist attacks and industrial accidents that involve reactive chemistry, including many chemical agents and toxic industrial chemicals. The resulting dual-use model can also be used for air-quality studies.

Mission Relevance

By improving NARAC's operational emergency response capability, this project supports a range of national security and homeland security missions. Chemical dispersion and fate are key components for managing response to terrorist attacks, and chemical fate also has an important role in understanding proliferation signatures as well as pre- and post-strike consequence evaluation of military targets.

FY05 Accomplishments and Results

In FY05, we upgraded LODI to handle time- and space-dependent first-order losses. The loss rates are read from a file, created from any appropriate source such as observations or the IMPACT model. This is sufficient to model the loss of volatile nerve-agents, such as sarin as it reacts with the hydroxyl (OH) radical (which is naturally occurring). We also developed a fast parameterization for the highest, lowest, and most-likely concentration of OH for any time and place, which will facilitate integration of this capability into NARAC's emergency response framework. To provide a more accurate estimate of OH prediction, we also developed a forecast capability for IMPACT, which we successfully tested against data collected during a DOE field campaign conducted near Pt. Reyes, California, this summer.

Proposed Work for FY06

To provide a more general reactive chemistry capability, we will implement a sophisticated "GEAR" solver into LODI that can handle arbitrarily complex sets of chemical reactions. Such a general reactive chemistry capability requires a Eulerian framework, but LODI works in a Lagrangian framework. This necessitates Eulerian-Lagrangian remapping. We will develop a semi-Lagrangian advection scheme for background species that is compatible with the existing Lagrangian scheme in LODI to reduce numerical diffusion and increase computational efficiency. During the final year of this project, we will focus on testing and validating the new LODI capabilities.

Publications

Atherton, C. S., D. J. Bergmann, and P. Sheridan. (2005). *Prediction and diagnosis of aerosol species using a global model: analysis of the MASRAD and MASE 2005 aerosol campaigns*. American Geophysical Union Fall 2005 Conf., San Francisco, CA, Dec. 5–9, 2005. UCRL-ABS-215278.

Atherton, C. S., D. J. Bergmann, and P. Sheridan. (2005). *Non-sulfate aerosol modeling near Pt. Reyes, CA for 2005: Forecast, hindcast, and ARM-MASRAD and ASP-MASE observations*. Atmospheric Chemistry Gordon Conf., Big Sky, MT, Sept. 4–9, 2005. UCRL-POST-216160.

Enhanced Isolation Performance of Geologic CO_2 Storage Sites through Mineral Trapping: Experimental and Field Confirmation of Model Predictions

James W. Johnson 05-ERD-054

Abstract

Our recent reactive transport modeling of engineered geologic CO_2 storage predicts that influx-triggered carbonate precipitation may ultimately lead to significantly reduced permeability within typical shale cap rocks, thereby significantly improving their long-term hydrodynamic seal capacity—the single most important constraint on long-term isolation performance. This project proposes to perform a multiscale confirmation of this key model prediction by conducting integrated reactive-transport modeling and batch-reactor experiments on the laboratory benchtop scale and by modeling the formation of the best-characterized natural CO_2 reservoir (McElmo Dome) on the field scale.

Our recent modeling work on geologic CO_2 storage in saline aquifers demonstrates unparalleled expertise in this emerging simulation arena. Experimental and field confirmation of our most significant model predictions regarding mineral trapping—which have crucial implications for the isolation performance of geologic CO_2 storage sites—will provide a critical measure of confidence in our unique reactive transport modeling capabilities. Hence, success in this project will further establish LLNL as the leader in reactive transport modeling applied to geologic CO_2 storage.

Mission Relevance

Engineered geologic CO_2 sequestration represents the most promising near-term emissions-reduction strategy for carbon management. The multiscale confirmation of key model predictions for CO_2 sequestration performed in this project supports the LLNL mission in energy security and is well aligned with DOE programs in CO_2 sequestration.

FY05 Accomplishments and Results

In FY05, we (1) completed simulations of the baseline batch-reactor experiment at field pressure and temperature; (2) identified optimal conditions for this experiment at elevated pressure and temperature; (3) designed, manufactured, and tested a new experimental sample chamber; (4) obtained detailed stratigraphic and structural data for the McElmo Dome work, as well as data on locations, logs, and fluid and gas chemistry; (5) developed a preliminary EarthVision geologic model for McElmo Dome; (6) obtained McElmo reservoir and cap-rock core samples; (7) initiated the McElmo modeling work; and (8) presented our progress to date at the Fourth Annual Conference on Carbon Capture and Sequestration.

Proposed Work for FY06

Proposed FY06 work includes (1) completing batch-reactor experiments for baseline conditions and for conditions that determine key physical and geochemical parameters; (2) conducting a post-mortem analysis of initial and baseline variant experiments; (2) simulating initial and secondary baseline variant experiments, comparing experimental and simulation results, and improving experimental–simulation agreement by model refinement; (3) completing McElmo Dome natural–analog modeling work; and (4) publishing the results of baseline and variant experiments and modeling and natural–analog modeling.

Publications

Johnson, J. W. et al. (2005). *Enhanced isolation performance of geologic CO_2 storage sites through mineral trapping: Experimental and field confirmation of model predictions*. 4th Ann. Conf. Carbon Capture and Sequestration, Alexandria, VA, May 2–5, 2005. UCRL-ABS-218391.

ULTRA-PRECISION $^{14}CO_2$ MEASUREMENTS IN AIR SAMPLES

Thomas P. Guilderson 05-ERD-082

Abstract

The primary goal of this project is to use the advanced analytical capabilities of LLNL's Center for Accelerator Mass Spectrometry (CAMS) to make ultrahigh-precision (<2 parts per million) measurements of carbon-14 (^{14}C) levels on a unique set of archived atmospheric samples. The scientific interest in analyzing these samples stems from their potential to place quantitative constraints on dynamics of the season-dependent and spatially variable flux of carbon dioxide (CO_2) between various sources and sinks in the natural environment. Air samples from several sampling stations stretching

from the South Pole to Point Barrow are archived at UC San Diego. These samples present a unique opportunity to define seasonal and spatial variations in the $^{14}CO_2$ cycle.

A geographically distributed dataset of accurate atmospheric $^{14}CO_2$ measurements would significantly improve estimates of the distribution of fluxes in and out of the land biosphere. Ratios of $^{14}C/^{12}C$ will be used to explicitly distinguish fossil fuel CO_2 from other sources of CO_2 and to provide constraints on the mass and turnover times of carbon in land ecosystems and exchange rates of CO_2 between air and sea. By measuring $^{14}C/^{12}C$ ratios at <2 per million archived samples collected as part of the ongoing air-sampling program at the Scripps Institution of Oceanography, this project will lay the foundation for a more expanded effort, involving collaborations with other air-sampling programs and providing valuable constraints that would enhance coupled carbon-climate modeling efforts.

Mission Relevance

The proposed work is directly related to LLNL's missions in obtaining breakthroughs in basic science as well as in understanding the impact of increased CO_2 concentrations on the global climate. Carbon management data generated by this research will be of considerable interest to DOE efforts in carbon management and global and regional carbon cycle modeling.

FY05 Accomplishments and Results

We documented the spatio-temporally varying $^{14}CO_2$ content at a series of "clean air" stations as determined from the Scripps CO_2 flask archive with unprecedented precision and accuracy. Interlaboratory comparisons were made with the Keck carbon cycle accelerator mass spectrometry facility at UC Irvine. Comparisons with other institutions' data, such as data from the National Oceanic and Atmospheric Administration's widespread measurement program, would also be useful. Results from this project were included in a white paper for DOE on biogeochemistry science at LLNL and were presented at the 10th International AMS Conference and the 7th International Carbon Dioxide Conference.

Publications

Graven, H. D. (2005). *Precise measurement of background $^{14}CO_2$*. 7th Intl. Carbon Dioxide Conf., Boulder, CO, Sept. 26–30, 2005. UCRL-ABS-215801.

Guilderson, T. P. et al. (2005). *$^{14}CO_2$ measurements on the Scripps flask archive: A status report*. 10th Intl. AMS Conf., Berkeley CA, Sept. 5–10, 2005. UCRL-ABS-215817.

Energy Supply and Use

Molecular Engineering of Electrodialysis Membranes

William L. Bourcier 03-ERD-060

Abstract

We will develop energy-efficient and element-selective membranes for use in the treatment of impaired waters. These membranes will allow cost-effective treatment of waters for which there is no currently available treatment technology. Our initial target species will be nitrate, a major water impurity in California. Project success will make available more water for public use, potentially from 3000 California wells that have been abandoned because of high nitrate levels. The nitrate-selective membrane will be synthesized by combining first-principles modeling of ion transport through charged membranes with membrane synthesis and testing. The membranes will then be installed in a modified commercial electrodialysis unit for testing.

Our goal is to develop molecular separations methods based on a molecular-level understanding of membrane transport processes. When fully developed, our methodology will be applicable to a variety of separations needs in water treatment as well as in industrial applications, such as processing "produced waters" that derive from extraction of fossil fuels and water used in power plants. Our target application for this project is nitrate-specific and energy-efficient membranes used to remove nitrate from contaminated water supplies. We plan to synthesize membranes whose design is based on our models of ion transport through metallized charged membrane pores. We will test the membranes in laboratory and field settings, and then license the technology to the water treatment industry.

Mission Relevance

In addition to supporting the Laboratory's role in environmental management, this effort will contribute to LLNL's homeland security mission by developing a means to protect water, a resource that is vital to national security. To meet the goals of a safe, economic supply of water, it is important that clean water be generated in a way that consumes the least possible amount of energy. Technologies developed under this research project would provide clean water in a cost-effective manner for the U.S.

FY05 Accomplishments and Results

Test results proved the concept of using nanoporous polymer sheets as cation–anion selective electrodialysis membranes. The track-etched polycarbonate membranes outperformed the best commercial cation membranes used in electrodialysis, thus satisfying the milestone on membrane energy efficiency. We also were able to develop a useful anion membrane but were not able to complete the work needed to make it selective for nitrate. This important portion of the project remains the goal of future work. Modeling capabilities were developed, including linking our code for numerical solution of the Poisson-Boltzmann equation with a Navier-Stokes flow code to model the trajectories of ions in a fluid moving between parallel charged plates. A prototype ion pump was developed and used successfully to desalinate test solutions. Work in FY05 resulted in a record of invention being filed.

Publications

Letant, S. E. et al. (2005). "Evidence of gating in hundred nanometer diameter pores: An experimental and modeling study." UCRL-JRNL-218429-DRAFT.

Environmental Consequences of Large-Scale Deployment of New Energy Systems

Thomas J. Phillips 05-ERD-047

Abstract

The prospect of steadily increasing emissions of carbon dioxide (CO_2) makes future global warming likely. We proposed to address critical U.S. energy-security needs by using a suite of environmental simulation tools to assess the feasibility of large-scale deployment of those energy technologies that may mitigate global climate change. Results from FY05 efforts, however, have led us to focus on assessing the feasibility of implementing large-scale forestation as a possible strategy for mitigating anticipated climate change. While new forests will store part of the increasing CO_2 as biomass, their overall climatic impact also depends on physical changes that may result from altering existing land cover: In some locations, these physical effects may overwhelm the carbon-storage benefits. We are employing a suite of climate simulation models of different complexities to gain greater scientific insight on the physical versus biophysical effects of forestation, and to evaluate their respective magnitudes and spatial dependencies.

We will assess the range of expected climatic effects from pursuing different large-scale forestation strategies (e.g., growth of forests in tropical, mid-latitude, or high-latitude zones). This work will increase scientific understanding of the climatic effects of land-cover change, identify needed additional climate-carbon research studies, and provide useful guidance for the developers of climate mitigation policies.

Mission Relevance

This project supports LLNL's energy-security and environmental-management missions by providing assessments relevant to the national/international formulation of long-term climate mitigation policies.

FY05 Accomplishments and Results

The original scope of this project included not only simulations of forest growth and destruction, but also assessment of high-altitude wind energy extraction techniques, evaluation of an engineered stratospheric aerosol strategy, and coupling energy-system and environmental modeling. Results of simulations obtained in early FY05 led us to concentrate on the impacts of forestation and deforestation. We have run several simulations with models of varying complexity to investigate the climatic effects of vegetation. These involved covering bare soil with different vegetation types in off-line runs of a land model driven by a prescribed atmosphere, and in runs of an interactive land–ocean–atmosphere model. In the off-line runs, all vegetation types induced lower-latitude cooling and higher-latitude heating associated with spatially varying changes in evaporation and surface albedo. The heating magnitudes depended on vegetation type, however, and were largest for forests. In the interactive model, the heating from forestation increased markedly in both magnitude and spatial extent. Analysis of hydrological sensitivities to vegetation changes is also in progress.

Proposed Work for FY06

In FY06, we will investigate both physical and biophysical effects of vegetation by building on our previous work with the LLNL Integrated Climate Carbon (INCCA) model. In addition to providing a fully interactive physical climate system, INCCA also accounts for dynamical uptake of CO_2 by vegetation, soil, and oceans. In a pilot multi-century simulation of CO_2 release from all sources, INCCA projected a large (+8ºC) global-mean warming, with substantially increased forest biomass and extent. In FY06, we will run a complementary simulation with dynamical vegetation switched off to isolate the effects of forest carbon-storage and expansion. We also will modify INCCA's dynamical vegetation model, which will allow us to follow up with simulations that evaluate the possible impact of different forestation strategies.

Publications

Bala, G. et al. (2004). *Multi-century changes to global climate and carbon cycle: Results from a coupled climate and carbon cycle model*. American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 13–17, 2004. UCRL-PRES-213277.

Bala, G. et al. (in press). "Multi-century changes to global climate and carbon cycle: Results from a coupled climate and carbon cycle model." *J. Climate* **18** (21), 4531-4544. UCRL-JRNL-209851.

Gibbard, S. et al. (in press). "Climate effects of global land cover change." *Geophys. Res. Lett.* UCRL-JRNL-215046.

Persistent Monitoring Platforms

Charles L. Bennett 03-ERD-076

Abstract

The objective of this project is to build and test the power plant for a stratospheric aircraft powered by thermal energy from the Sun. Such an aircraft could maintain station over a designated ground location almost indefinitely, since it would not need fuel. A thermally coupled system with an efficiency nearly an order of magnitude better than the state of the art (Helios) is being developed by creating the technology for a Sun-tracking solar-heat collector, thermal storage reservoir, and a direct-drive high efficiency heat engine. The project will also develop physics models for thermal transport, materials interactions, loss mechanisms, and engine performance in the stratospheric environment.

Physics models to prove the principles involved in a solar thermal-powered aircraft will be developed and validated. This work is in preparation for constructing a prototype scale-model aircraft to demonstrate station-keeping capability at sea level. Successful demonstration of this technology would set the stage for construction of a stratospheric-altitude prototype capable of circumnavigating the globe.

Mission Relevance

Persistent surveillance, having essentially unlimited dwell time over a region of interest, would enable the acquisition of a qualitatively new type of intelligence information for various national-security applications, such as countering the proliferation of weapons of mass destruction. Furthermore, inexpensive persistent surveillance has direct utility in border monitoring for homeland-security missions.

FY05 Accomplishments and Results

In FY05, we demonstrated, with a prototype, that a lithium-hydride–lithium (LiH–Li) thermal "battery" can deliver a specific thermal power of 7500 W/kg and store a specific thermal energy of 1300 W·h/kg. These performance levels are well in excess of the minimum required to sustain a solar powered aircraft in overnight flight. In addition, we invented a new type of heat engine that may achieve, in practice, a higher efficiency for conversion of heat to power than the well known Stirling engine. From this work, two patent applications were filed, and one record of invention was submitted.

Proposed Work for FY06

In FY06 we will focus on the research and development involved in the new heat engine concept, as well as on a ground-based quantification of the engine efficiency when coupled to a high-temperature thermal energy storage reservoir. We will initially use an innocuous thermal energy storage medium (e.g., hot sand) and will follow this work up with the LiH–Li "thermal battery" developed in the earlier phases of this project, once the engine characteristics, especially the potentially hazardous features, are sufficiently well understood. Thus in this project we have laid the groundwork for the eventual development of operationally useful systems.

ENGINEERING AND MANUFACTURING PROCESSES

Large-Aperture Diffraction Gratings: The Enabling Technology for High-Energy Petawatt Lasers

Jerald A. Britten 03-ERD-059

Abstract

The objective of this project is to develop a state-of-the-art capability for producing meter-scale, multilayer dielectric diffraction gratings for pulse compression in high-energy, petawatt-class laser systems. We plan to pattern dielectric coatings on monolithic substrates with grating lines approximately 0.2 µm wide, ~0.7 µm high, and ~0.5 µm apart, uniformly over areas of 91 by 45 cm. Methodologies will be developed for cleaning these optics after fabrication to optimize laser damage resistance, and for measuring their performance at full aperture. This effort requires development of tooling and processes applicable to large, heavy substrates, involving laser interference lithography, reactive ion-beam etching, and, in particular, wet-chemical processing.

Mission Relevance

This research effort will enable new and enhanced studies relevant to NNSA's Stockpile Stewardship Program on LLNL's Janus Intense Short-Pulse, University of Rochester's Omega, Sandia's Z-Beamlet, and Los Alamos National Laboratory's Trident lasers. It will also enable academic research into high-energy-density physics at the University of Texas Petawatt Laser, an NNSA-funded activity.

FY05 Accomplishments and Results

We (1) constructed a unique, semi-automated wet processing tool for ultraclean safe priming and photoresist coating of meter-scale substrates, which is now in routine use; (2) fielded a vibration-insensitive interferometer for measuring full-aperture diffracted wavefront of meter-scale gratings at use angle and wavelength; (3) developed automated tooling for final cleaning of meter-scale gratings with hazardous chemicals—the tooling was made to minimize operator exposure and waste volume; (4) demonstrated production of 807- × 417-mm, 1780-line/mm diffraction gratings made by ion-assisted e-beam multilayer deposition, that exhibit flat diffracted wavefront and uniform diffraction efficiency >95%; and (5) filed two records of inventions for dielectric gratings.

Publications

Britten, J. A. et al. (2004). *Demonstration of meter-scale, high laser damage*. 1st Intl. Conf. Ultrahigh Intensity Lasers, Lake Tahoe, CA, Oct. 4–9, 2004. UCRL-ABS-206926.

Britten, J. A. et al. (2005). *Status of large-aperture multilayer dielectric grating production capability at LLNL*. 4th Intl. Conf. Inertial Fusion Sciences and Applications, Biarritz, France, Sept. 4–9, 2005. UCRL-ABS-210667.

Nguyen, H. T. et al. (2005). *Gratings for high-energy petawatt lasers*. Annual Symp. Optical Materials for High Power Lasers, Sept. 19–21, 2005, Boulder, CO. UCRL-ABS-211646.

Acoustic Characterization of Mesoscale Objects

Diane J. Chinn 04-ERD-013

Abstract

We hope to achieve micrometer-resolution characterization by extending the range of laser-acoustic testing to gigahertz frequencies. Materials and the geometry of components in many LLNL mesoscale objects necessitate use of a non-contacting technique at frequencies from 100 MHz to 10 GHz. This frequency range is required to acoustically characterize features from 0.5 to 5 µm in size. To be applicable to mesoscale objects, the gigahertz acoustic waves must propagate sufficient distances into materials of interest—for LLNL applications, mesoscale structures are on the order of 25 to 200 µm thick.

The expected results of this research will have implications for acoustic characterization of high-energy-density physics (HEDP) and cryogenic target components. The understanding gained by this research will broaden the field of acoustic testing by filling the existing gap in acoustic characterization capabilities. Work in these areas has never been performed.

Mission Relevance

This promising technique will provide an acoustic characterization tool for many mesoscale applications. In particular, the work will benefit HEDP experiments in support of LLNL's stockpile stewardship mission. Other applications could support the Laboratory's energy-security mission by enabling developments in fuel cells or providing understanding of geochemical processes. Using this technique to study tissue and cell abnormality would support LLNL's mission in bioscience to improve human life.

FY05 Accomplishments and Results

Major accomplishments for FY05 included validation of acoustic and laser-acoustic models with experimental data, and the assembly of a prototype gigahertz laser-ultrasonic testing system. The prototype system was initially assembled at Boston University and transferred to LLNL. In the summer of 2005, a graduate student from Boston University worked at Livermore to assemble, calibrate, and operate the system. A paper describing this mesoscale acoustic research was presented at an annual meeting for quantitative nondestructive evaluation, and a record of invention has been filed.

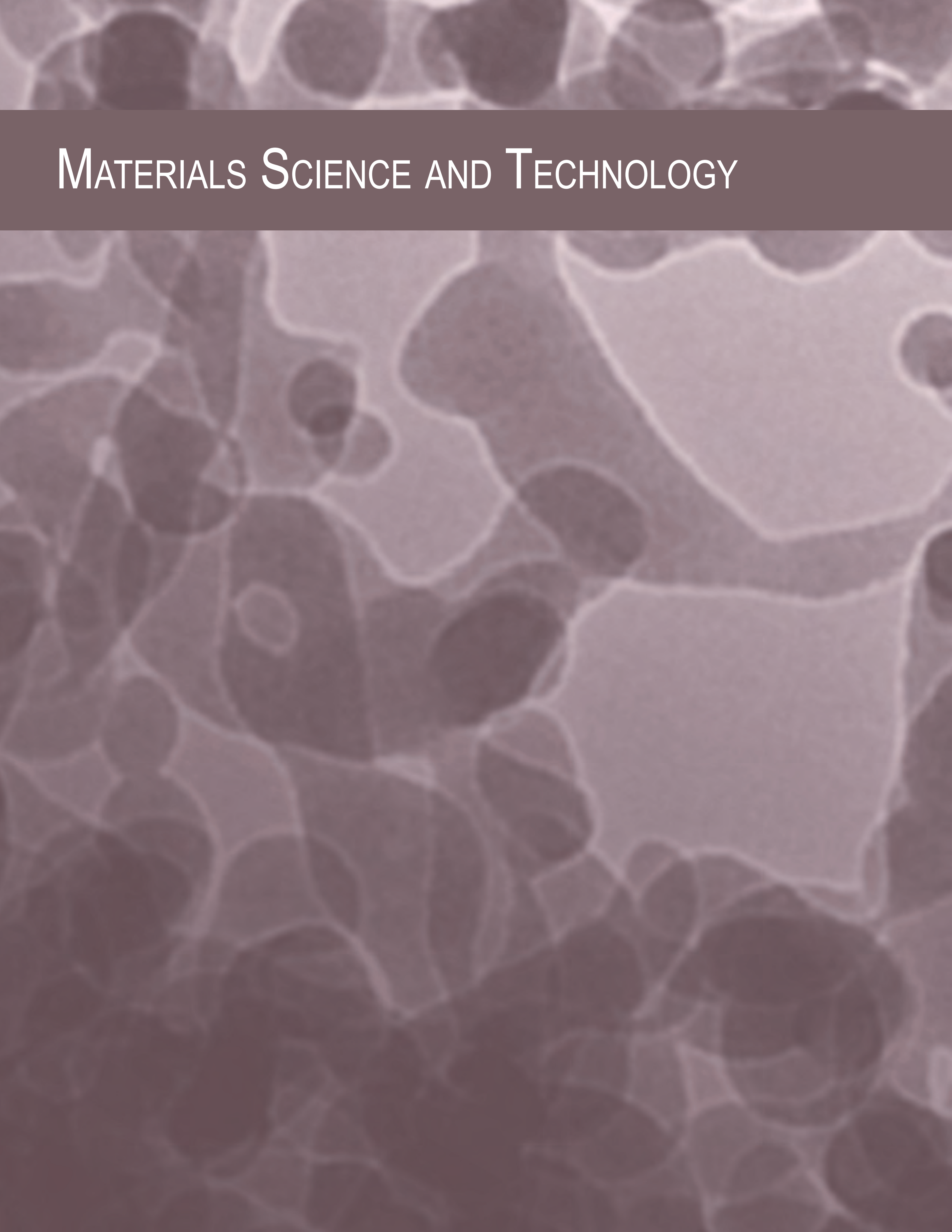
Proposed Work for FY06

Our work to date has enabled new research on acoustic wave generation, scattering mechanisms, and interface characterization in the gigahertz regime. Research in FY06 will cover three specific areas: (1) gigahertz laser-acoustic wave generation with advanced laser-interaction models to identify the thermoelastic–ablative threshold for materials; (2) gigahertz wave propagation and scattering mechanisms from anisotropy and microporosity; and (3) use of models and experiments to study the capabilities of gigahertz characterization of interfaces with micron-sized gaps and repeating structures.

Publications

Huber, R. et al. (2005). *High frequency laser-based ultrasound*. 32nd Ann. Review of Progress in Quantitative Nondestructive Evaluation, Brunswick, ME, July 31–Aug. 5, 2005. UCRL-PROC-215718.

Materials Science and Technology



Magnetic Transition Metals and Oxides at High Pressures

Valentin Iota-Herbei 02-ERD-046

Abstract

We investigated the electronic and magnetic properties of three-dimensional (3-D) transition metals and oxides at high pressures and focus on spin transitions and the determination of the magnetic phase diagrams of iron (Fe), cobalt (Co), and nickel (Ni). To obtain electronic structure information under pressure, we have adapted for use in a diamond anvil cell (DAC) two new x-ray spectroscopy techniques sensitive to the configuration of the valence band: x-ray emission spectroscopy (XES) and x-ray magnetic circular dichroism (XMCD). The experiments have been carried out at Sectors 16 and 4 of the Advanced Photon Source (APS) at Argonne National Laboratory.

This study resulted in detailed understanding of the pressure-induced changes in the electronic structure of transition metals and oxides, including pressure changes in the valence band spin polarization (spin state). By monitoring changes in the spin state at high pressure, we determined a magnetic phase diagrams for each of the three magnetic elements studied (Fe, Co, and Ni) and proposed a framework for describing magnetism suppression in these 3-D transition metals. We observed a correlation between magnetism suppression and the pressure-induced broadening of the 3d bands, suggesting electron delocalization as the primary mechanism for spin quenching in 3d magnetic metals.

Mission Relevance

This project has developed two new techniques for electronic structure measurements for use at high pressures: XES and XMCD. Both rely on third-generation synchrotron x-ray spectroscopy to yield element and orbital-sensitive information on the electronic configuration of the valence band. These experimental techniques are crucial to future studies of highly correlated systems such as rare earths, actinides, and alloys, systems of critical importance to LLNL's stockpile stewardship mission.

FY05 Accomplishments and Results

In FY05 we completed our XMCD measurements on the 3-D magnetic metals under pressure. Our measurements helped establish the phase boundaries for the magnetic phase diagrams in these elements, and show in detail the mechanisms responsible for the loss of magnetism under pressure. We submitted two manuscripts for publication that are currently under review.

Determination of the Microstructural Morphology of Shock-Induced Melt and Resolidification

Jeffrey D. Colvin 03-ERD-018

Abstract

The principal objective of this project is to determine what characteristics of a shock-induced solid-to-liquid (melt) phase transformation affect the microstructure upon resolidification. Potential characteristics include the initial crystal ordering at the microscale, how fast the solid disorders, how long it is in an amorphous (disordered) phase, and how rapidly it resolidifies. The project has four major phases: (1) computational design of laser targets and drive pulses to produce the desired shock morphologies; (2) development of specially tailored target materials that have, for example, specific initial microstructures; (3) pre- and post-shot analyses of the target samples to determine the microstructural morphology; and (4) theory and simulation at the atomistic scale.

Results will increase scientific understanding of the dynamics of metals by answering the question of how shock-melted pure metals and metal alloys structure themselves upon refreezing at a very high cooling rate. It is the microstructure of the material—ordering on the scale of grains—as much as its lattice arrangement—ordering on the atomic scale—that determines its constitutive properties, particularly its strength. These data will thus aid in the development of advanced constitutive material models.

Mission Relevance

This project will provide data needed for the development of advanced constitutive models for hydrocodes and a proven technique to determine target specifications for planned solid-state materials strength experiments in support of the Laboratory's stockpile stewardship mission.

FY05 Accomplishments and Results

The team fabricated several sets of custom-tailored four-layer targets for these experiments, completed experiments using LANL's Trident laser and a smaller laser at LLNL, and performed microscopic analyses of the recovered samples. We met all the project milestones. Estimated effective superheating and undercooling rates on the order of 10^{10} K/s were obtained in the experiments, a rate that is orders of magnitude faster than any achieved before in bulk material. The melt was shown to be very far from homogeneous, and the material failed in the liquid state. Equally remarkable is that the material grain structure that remained solid or re-solidified was completely altered, retaining little or none of its original microstructure or crystal orientation.

Publications

Colvin, J. et al. (2005). *Microstructure morphology of shock-induced melt and super-rapid resolidification in bismuth*. UCRL-JRNL-216516.

A Two-Particle Formulation of Electronic Structure

Antonios Gonis 03-ERD-064

Abstract

The problem of correlated electrons in elemental solids and alloys figures prominently in the Laboratory's efforts to understand the behavior of plutonium (Pu) metal and its alloys, and to predict such properties as phase and dimensional stability under self-irradiation and as a function of time. We have introduced theoretical and computational methods for studying the electronic structure of matter based on two-particle states (and in general *n*-particle states) rather than single-particle states as is current practice. The codes we develop will thus simulate the Coulomb interaction and its effect on materials properties with greater accuracy and provide deeper insight into the physics of materials than is possible with traditional methodology.

This work could provide theoretical and practical electronic structural information about important materials and systems for which conventional methods are currently inadequate, particularly complex systems such as Pu. In the long term, we will develop realistic models for the study of heavy elements and their alloys and apply them to materials central to the Laboratory's mission. The methodology developed will not only allow the simulation of materials properties but also provide insight into the basic mechanisms driving these properties.

Mission Relevance

Understanding the electronic structure and properties of materials, especially actinides, is crucial to nuclear science and relevant to areas such as stockpile stewardship, in support of the Laboratory's national-security mission.

FY05 Accomplishments and Results

Our most striking accomplishment in FY05 was finding that, contrary to conventional wisdom, the valence and conduction bands in strongly correlated materials have different shapes. The conduction band—the excitation part of the spectrum—carries the signature of the two-particle density of states, in contrast to the valance band, which resembles the densities of state of the non-interacting spectrum. This is because the conduction band arises from the scattering of two particles rather than just single-particle ionic potentials.

Proposed Work for FY06

In FY06, we will use model systems to compare the proposed method with the results of recently developed methodologies such as the dynamical mean field theory. We will also finalize the construction of the infrastructure for calculating two-particle exchange and correlation potentials for two-particle state, and extend the formalism to the case of chemically disordered alloys.

OPTICS PERFORMANCE AT 1 OMEGA, 2 OMEGA, AND 3 OMEGA

John Honig 03-ERD-071

Abstract

The interaction of intense laser light with dielectric materials is a fundamental applied science problem that is becoming increasingly important with the rapid development of ever-more-powerful lasers. To better understand the behavior of optical components in large fusion-class laser systems, we are systematically studying the interaction of high-fluence, high-power laser light with high-quality optical components, with particular interest in the effects of polishing/finishing, stress-induced defects, and surface contamination. We will focus on obtaining comparable measurements at three different wavelengths: 1ω (1053 nm), 2ω (527 nm), and 3ω (351 nm). Modeling at both microscopic and macroscopic scales will support interpretation of experimental results.

Materials research that enhances our understanding of complex mechanisms describing the response of optical materials to intense laser irradiation will benefit worldwide efforts to field ever more powerful, fusion-class laser systems and allow them to operate efficiently and reliably at or above their design specifications. Our modeling efforts will, in turn, allow operators of fusion-class laser systems to predict optics lifetime for upcoming experimental campaigns, and enable optimized campaign planning by minimizing incremental costs of replacing optical components. Additionally, this effort will help advance and ensure LLNL's continued leadership in the fields of optical materials science and laser technology.

Mission Relevance

This work will add substantially to the knowledge base of optical materials under the intense laser illumination (i.e., fusion-class lasers systems) and allow us to validate theoretical and stochastic models of optical damage. This work supports NNSA and Laboratory missions in stockpile stewardship by providing data that can be used to validate computer simulations related to the safety and reliability of the nuclear stockpile.

FY05 Accomplishments and Results

The effects of 1ω and 3ω light and of 1ω and 2ω light on damage growth rates were measured and reported. The growth rate coefficient for combined 1ω and 3ω light was found to be the same as that for pure 3ω light of equal fluence. In contrast, the growth rate coefficient for combined 1ω and 2ω light was found to be the same as that for pure 1ω light of equal fluence. The defect density/cm^2 in fused silica for fluences above 14 J/cm^2 was measured and reported, as was the wavelength and pulse-length dependence of laser conditioning and bulk damage in potassium-dihydrogen-phosphate second harmonic crystals. A protocol using 3ω light in 300-ps pulses gradually ramped to full fluence gave the best performance.

Publications

Adams, J. J. et al. (2005). "Pulse length dependence of laser conditioning and bulk damage in KD_2PO_4." *Proc. SPIE* **5647**, 265. UCRL-PROC-207946.

Carr, C. W. et al. (2005). "Effect of scattering of complex morphology of DKDP bulk damage sites." *Proc. SPIE* **5647**, 532. UCRL-PROC-207879.

Carr, C. W. et al. (2005). "The nature of emission from optical breakdown induced by pulses of fs and ns duration." *Proc. SPIE* **5647**, 494. UCRL-PROC-207943.

DeMange, P. et al. (2005). "A new damage testing system for detailed evaluation of damage behavior of bulk KDP and DKDP." *Proc. SPIE* **5647**, 343. UCRL-CONF-208281.

Honig, J. et al. (2005). "Experimental study of 351-nm and 527-nm laser-initiated surface damage on fused silica surfaces due to typical contaminants." *Proc. SPIE* **5647**, 129. UCRL-PROC-207942.

Negres, R. A. et al. (2005). "Stoichiometric changes to KH_2PO_4 during laser-induced breakdown." *Proc. SPIE* **5647**, 306. UCRL-PROC-208139.

Norton, M. A. et al. (2005). "Growth of laser initiated damage in fused silica at 1053 nm." *Proc. SPIE* **5647**, 197. UCRL-PROC-207855.

Nostrand, M. C. et al. (2005). "Correlation of laser-induced damage to phase objects in bulk fused silica." *Proc. SPIE* **5647**, 233. UCRL-PROC-207944.

Novel Methods for Bonding Disparate Materials

Michael W. Mcelfresh 03-ERD-074

Abstract

This project will use a systematic strategy to develop new processes for bonding together disparate materials for fabricating experimental samples (e.g., laser targets). The project will focus on methods that yield extremely thin, uniformly thick, strong bonds that have minimal impact on the properties of the materials to be bonded. We will focus on long-chain, polymer-tethered molecules covalently attached as monolayers to the surfaces to be bonded. Force spectroscopy will be used to study bond strength and the ability for tethers to span gaps that would be associated with surface roughness in real materials. Patterned, tethered molecular systems will be used to study chemically directed assembly.

The expected result is a more scientific understanding of possibilities for bonding between disparate materials. This study will allow us to identify bonding systems and strategies for using these systems with materials common to laser-target fabrication. These strategies may involve using appropriate

solvents to modify the tethered molecule systems so they will bridge the gaps that exist when bonding together pieces of material. Chemically directed assembly will offer a new strategy for aligning small parts during fabrication. This research will result in publications in refereed journals and provide new concepts for mission-relevant work.

Mission Relevance

If successful, this project will support the Stockpile Stewardship Program (SSP) by providing targets for SSP experiments that study materials under precisely controlled shock conditions driven by a gas gun or laser. In these experiments, the bonding must be sufficiently strong so as not to delaminate prematurely, and the bonding layer must be extremely thin to meet specifications.

FY05 Accomplishments and Results

Using force spectroscopy, we characterized the strength and frequency of tethered molecular systems as a function of tether length and active bonding group. An exciting observation was the difference in the force required to remove a long-chain molecule that is bound to a surface with one thiol linkage from the same polymer chain bound via two thiol linkages. The polymer chain with two surface links was actually found to be weaker for reasons relevant to surface-coating technology for biological applications. Also using force spectroscopy, we showed how solvents can affect the way polymers attached to surfaces extend into the solvent. We found distinct differences in the stiffness of the polymer chains between different solvents. This has important implications for bonding disparate materials.

Publications

Langry, K. et al. (2005). “The AFM measured force to rupture the dithiolate linkage of thioctic acid to gold is less than the rupture force of a simple gold-alkyl thiolate bond.” *Langmuir* **21**, 12064. UCRL-JRNL-208819.

Ratto, T. et al. (in press). “Nonlinearly additive forces in multivalent ligand binding to a single protein revealed with force spectroscopy.” *Langmuir*. UCRL-JRNL-214218.

Ratto, T. et al. (2005). *Force spectroscopy of single-molecule adhesive interactions in three polymer-tethered multivalent systems: Protein carbohydrate, thiol/thiolate-gold, and shal-protein bond pairs*. 229th Natl. Mtg. American Chemical Society, San Diego, CA, March 2005. UCRL-PRES-210520.

Ratto, T. et al. (2004). *Mono and multivalency in tethered protein-carbohydrate bonds*. 227th Natl. Mtg. American Chemical Society, Anaheim, CA, March 2004. UCRL-PRES-205468.

Ratto, T. et al. (2004). *Mono and multivalency in tethered protein-carbohydrate bonds*. Biophysical Society Mtg., Baltimore, MD, Feb. 14–18, 2004. UCRL-CONF-202145.

Ratto, T. et al. (2003). *Force spectroscopy of the double-tethered concanavalin-A mannose bond*. Seeing at the Nanoscale Conf., Santa Barbara, CA, Aug. 24–27, 2003. UCRL-PRES-210520.

Plutonium and Quantum Criticality

Michael J. Fluss 03-ERD-077

Abstract

Understanding the anomalous properties of plutonium (Pu) that arise because the 5f-electrons are poised between localized and itinerant behavior is a science challenge with important technological implications. By exploring the possibility of a second-order phase transformation at 0 K, which is a quantum critical point (QCP), we hope to explain the origin of Pu's complexity. We will "tune" alpha- and delta-Pu using disorder, alloying, pressure, and magnetic fields, while characterizing its physical properties at low temperature. This will accomplish our primary goal of identifying the QCP. We will then further characterize the physical properties of Pu in the vicinity of the QCP, which will allow us to map its low-temperature phase diagram.

Both the Pu phase diagram and knowledge of its physical properties are needed to validate theories on how to introduce many-body effects into the first-principles description of Pu and Pu alloys. We also will develop evidence for the existence or absence of local moments in the Pu system through experiments with point defects and muon relaxation. This work will provide new insight into the organizing principles of Pu, and a fundamental understanding of the origin of its six solid-state phases and its many anomalous physical properties, including large volume changes and extraordinary phase sensitivity to impurities.

Mission Relevance

By contributing to a better understanding of the physical properties of Pu, this project explores the foundational science of stockpile stewardship. In addition, this research supports LLNL's mission in breakthrough science and technology.

FY05 Accomplishments and Results

In FY05, we determined that, for temperatures less than 30 K, the magnetic susceptibility of alpha- and delta-Pu increases with time, and appears to saturate over long periods. The source of this temperature dependence was attributed to two separate sources; local perturbations of the magnetization from defects, and a long-range magnetic "bubble" with a damage cascade at the core. Ongoing muon spin-relaxation studies for alpha-Pu confirm that there are no local moments and that the material exhibits Pauli susceptibility from 300 to 3 K, validating results of our earlier magnetization experiments. We used magnetometry to examine a series of $Pu_{1-x}Am_x$ specimens in search of a QCP. The specimens measured were found to exhibit time-dependent susceptibility similar to alpha- and delta-Pu. New plutonium-cobalt-pentagallium muon experiments were performed on year-old, self-damaged material.

Proposed Work for FY06

In the last year of this project, our experimental focus will be the $Pu_{1-x}Am_x$ system, where we plan to move across the hypothesized QCP observed previously as a magnetic and resistive discontinuity by our colleagues in CEA Valduc (France) and the Institute for Transuranics (Karlsruhe, Germany). If successful, this will represent a breakthrough in Pu science.

Publications

Heffner, R. H. et al. (in press). “New limits on the ordered moments in alpha-Pu and Ga-stabilized delta-Pu.” *Physica B*. UCRL-CONF-215129.

McCall, S. et al. (in press). “Magnetic properties of radiation damage in Pu and Pu alloys.” *Adv. Actinide Sci*. UCRL-PROC-211062.

McCall, S. et al. (2005). “Magnetic properties of radiation damage in Pu.” *Materialovedenie* **4**, 35. UCRL-JRNL-207582.

Morris, G. D. et al. (in press). “The symmetry of the superconducting order parameter in $PuCoGa_5$.” *Phys. Rev. Lett*. UCRL-JRNL-208925.

Development of Sample Handling and Analytical Expertise for the Stardust Comet Sample-Return Mission

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John P. Bradley 03-ERI-007

Abstract

NASA’s Stardust mission will result in analysis of “fresh” cometary particles collected from a young Jupiter family comet. Sampling occurred during the spacecraft flyby of Comet 81P/Wild-2 in January 2004. Eight Livermore scientists will be part of the preliminary examination team that will investigate Stardust samples for the first nine months after the spacecraft’s return. The scientists will characterize particles using state-of-the-art electron microscopes and ion microprobe facilities at Livermore. Because comets are small bodies that spend most of their lifetimes at extreme heliocentric distances, they are believed to contain preserved interstellar dust, as well as the first solids condensed in the (inner) solar system. Study of Stardust samples should give new insights into early solar system processes.

Collection of cometary particles occurred through impact with low-density silica aerogel tiles and aluminum foils on a sample tray assembly at approximately 6.1 km/s. Recovery of particles from the aerogel is extremely challenging—the hypervelocity capture could result in material embedded up to 2 mm deep. The overall aim of this project is to develop enabling technologies to isolate and extract cometary material. Techniques to be developed include use of focused ion-beam microscopy and laser-cut diamond micro-blades under ultrasonic oscillation.

Mission Relevance

The proposal aligns with the Laboratory's fundamental science and applied technology mission goal with specific reference to astrophysics and space science. The scientific findings resulting from detailed laboratory investigations of Comet 81P/Wild-2 particles will give new insights into the properties of small solar system bodies. The ultrasonic diamond-blade technology that will be employed to extract impact tracks has potential application to fusion-class laser experiments seeking to recover, from aerogel, particulate hypervelocity ejecta generated during laser shots. The integrated analysis approach developed to investigate Stardust samples should have application to the interrogation of interdicted materials for forensic studies in support of national security objectives.

FY05 Accomplishments and Results

We have continued to investigate new emerging technologies for extraction and manipulation of small particles from silica aerogel and metallic foils. This has lead to the a capability of for rapidy recovery of impact tracks from aerogel using laser-cut diamond micro-blades under ultrasonic oscillation. This micro-blade system has been duplicated by NASA and will be used to extract some of the first impact tracks when the Stardust sample-return canister is opened in January 2006. High-precision focused ion-beam microscopy has been utilized to recover impact residue material, only a few micrometers in diameter, from the interior walls of micro-craters generated by laboratory simulations of Stardust impacts. These accomplishments will significantly improve the ability to analyze Stardust samples.

Publications

Bajt, S. et al. (2005). *Amorphization of forsterite grains due to high energy ion irradiation—implications for grains processing in ISM*. 36th Lunar and Planetary Science Conf., League City, TX, Mar. 14–18, 2005. UCRL-ABS-209107.

Bradley, J. et al. (2004). "An astronomical 2175 angstrom feature in interplanetary dust particles." *Science* **307**, 244. UCRL-JRNL-205703.

Graham, G. A. et al. (2005). *Electron beam analysis of micrometeoroids captured in aerogel as Stardust analogues*. 36th Lunar and Planetary Science Conf., League City, TX, Mar. 14–18, 2005. UCRL-ABS-209123.

Graham, G. A. et al. (2005). "Recovery of isotopic hotspot in interplanetary dust particles by focused ion beam microscopy: Bridging the gap between NanoSIMS and (S)TEM." *Microscop. Microanal.* **11**(2), 1390. UCRL-ABS-214009.

Graham, G. A. et al. (2004). *Great expectations—dust recovered from aerogel*. Workshop Dust in Planetary Systems, Kauai, HI, Sept. 26–30, 2004. UCRL-ABS-215065.

Ishii, H. A. et al. (in press). "Rapid extraction of dust impact tracks from silica aerogel by ultrasonic micro-blades." *Meteor. Planet. Sci.* UCRL-JRNL-209950.

Ishii, H. A. et al. (2005). *Hard x-ray spectro-microscopy techniques at SSRL for astromaterials analysis*. 36th Lunar and Planetary Science Conf., League City, TX, Mar. 14–18, 2005. UCRL-ABS-208887.

Ishii, H. A. et al. (2005). *Ultrasonic micro-blades for the rapid extraction of impact tracks from aerogel*. 36th Lunar and Planetary Science Conf., League City, TX, Mar. 14–18, 2005. UCRL-ABS-208888.

Ishii, H. A. et al. (2004). *Synchrotron x-ray fluorescence analysis of dust particles*. Workshop Dust in Planetary Systems, Kauai, HI, Sept. 26–30, 2004. UCRL-ABS-213668.

Sheffield-Parker, J. et al. (2005). "High resolution phase contrast imaging of cosmic dust captured in aerogel." *Microscop. Microanal.* **11**(2), 1396. UCRL-ABS-214015.

Infrared Diagnostics for Dynamic Events

Michael W. Mcelfresh 04-ERD-005

Abstract

How mechanical work is converted to different forms of energy during deformation of a material is not well understood. For instance, there is disagreement over the amount of work dissipated as heat during uniform plastic deformation. Our exploratory research is using high-speed, spatially resolved infrared (IR) measurements of temperature to investigate thermodynamics of the dynamic deformation and failure of materials. The goals of this project are to develop dynamic IR temperature measurement techniques and then apply them for a better understanding of dynamic failure processes in materials.

This project will produce a more thorough understanding of the thermomechanical behavior and failure of materials under dynamic loading. The experimental results will provide an excellent dataset for testing existing models and may lead to new discoveries in material response. The dynamic IR temperature measurement capability developed will have applicability to other problems involving dynamic temperature gradients, such as the interaction between lasers and materials. This project addresses fundamental problems in thermomechanics and will lead to several journal publications and conference presentations.

Mission Relevance

The Stockpile Stewardship Program will benefit from the development of high-speed IR imaging capabilities and improved understanding of materials undergoing dynamic processes. For instance, this technology could reveal why the material properties of uranium vary so greatly with temperature. Other applications with national-security relevance include high explosives, target modeling for future large-scale lasers, and hydrodynamics.

FY05 Accomplishments and Results

Experiments to discern whether or not the fractional conversion of mechanical work to heat (β) equals one under uniform strain were undertaken using thermocouples welded to the samples. The results of these experiments on aluminum show that β can be significantly less than one under uniform loading conditions, and that it is strain dependent. Other experiments with thermocouples on tantalum samples showed evidence of shear banding, indicating that some energy dissipation may occur through mechanisms other than heating. An IR imaging system incorporating Cassegrain optics and a linear IR imaging system based on a high-speed array was constructed. The emissivity showed changes from before to after deformation, leading to complications in data interpretation. Studies of appropriate surface finishes were made to ameliorate this issue.

Time-Resolved Dynamic Studies using Short-Pulse X-Ray Radiation

Art J. Nelson 04-ERD-010

Abstract

This project proposes to continue development and demonstration of real-time, in situ interrogation of the dynamic response of materials under extreme condition—for example, response to femtosecond (fs) laser heating—together with simulations that include the time-dependent electronic structure during equilibration between electrons and ions, and the resulting motion of the atomic nuclei. Short-pulse (subpicosecond) x-ray radiation will track changes in the physical properties in tandem with measurements of the atomic and electronic structure of materials undergoing fast laser excitation and shock-related phenomena. Time-resolved x-ray photoelectron spectroscopy, x-ray absorption, and x-ray scattering will be used to study phase changes in materials with subpicosecond time resolution.

The results of laser pump–x-ray probe and x-ray pump–laser probe interrogation of ultrathin metal foils will be compared with the first-principles electronic structure methods and with atomistic and molecular dynamics simulations of materials under extreme conditions. The simulations include the time-dependent electronic structure during equilibration between electrons and ions, and the resulting motion of the atomic nuclei. Thus we will introduce the notion of a phase diagram based on electron temperature rather than the conventional phase diagram, in which ion temperature is used. The combination of modeling and experiment developed in this project will be used to answer important scientific questions about the dynamic response of materials under extreme conditions.

Mission Relevance

Ultrafast materials response under extreme conditions, together with state-of-the-art models and computational facilities, are directly related to LLNL's missions of national security and stockpile stewardship.

FY05 Accomplishments and Results

We performed the first measurements of the disassembly dynamics of femtosecond-laser-heated ultrathin copper (Cu) foil and the first time-resolved photoemission measurements of laser-heated ultrathin Cu foil, showing changes in electronic structure. We completed custom fabrication of an experimental chamber and new magnetic-bottle electron time-of-flight spectrometer. The latter has improved time response and low energy sensitivity for x-ray pulse measurements by chirped pulse laser-assisted Auger decay performed on the Sub-Picosecond Photon Source (SPPS) at the Stanford Linear Accelerator Center.

Proposed Work for FY06

In FY06, we will further develop the pump-probe technique with experiments and modeling extended to other metals of interest and to vacuum ultraviolet (VUV) heating. Specifically, work will be directed at (1) demonstrating the pump-probe technique on actinide foils (e.g., uranium-238; (2) further utilizing the SPPS for time-resolved studies and two-photon ionization of the Cu K-edge, with subsequent observation of the Auger cascade of the excited state; (3) performing experiments on the Femtosecond Phenomena Beamline at the Advanced Light Source; and (4) performing experiments on the VUV free-electron laser at Deutsches Elektronen-Synchrotron.

Publications

Dunn, J. et al. “Picosecond 14.7 nm interferometry of high-intensity laser-produced plasmas.” *Laser Part. Beams* **23**, 9. UCRL-JRNL-207241.

Nelson, A. J. et al. “Advancements in time-resolved x-ray laser induced time-of-flight photoelectron spectroscopy.” *Proc. SPIE Int. Soc. Opt. Eng.* **5919**, 139. UCRL-PROC-214158.

High-Strain-Rate Deformation of Nanocrystalline Metals

Eduardo M. Bringa 04-ERD-021

Abstract

We are performing experiments and simulations to validate constitutive models for plastic behavior and to determine the deformation mechanism of nanocystalline materials of different grain sizes (<500 nm) at high strain rates (>10^6/s), which could lead to the design of improved materials. The experiments use both laser-induced shocks and isentropic compression to study, for the first time, the high-strain-rate deformation of nanocrystalline nickel (Ni). Samples are characterized using electron microscopy, nanoindentation, profilometry, and x-ray diffraction before and after loading. We are validating current constitutive models using both atomistic molecular dynamics and continuum simulations performed at the boundary of their current computational possibilities to match experimental scales.

Improved understanding of the deformation mechanisms in nanocrystalline metals could lead to the design of new, improved materials with higher or lower resistance to plastic deformation. Nanocrystals are useful materials for laser targets and other applications because of their high hardness. Our simulations and experiments suggest a novel way to obtain even harder nanocrystals both during and after shock loading. The results to date are unique because these strain rates have never before been experimentally attained in nanocrystals and because experiments and atomistic simulations cover the same length and time scales. Our use of local facilities—such as the Janus laser, electron microscopy and characterization facilities, and massively parallel computers—is increasing in-house expertise.

Mission Relevance

We proved that nanocrystals are still stable under high-strain-rate conditions, making possible multiple applications, from fusion pellet covers to piercing-resistant armor. Work at Janus will prepare for future research on fusion-class lasers, where all materials science experiments will operate at strain rates of 106/s or higher, where the deformation mechanisms are not well understood. This project will (1) validate continuum models by establishing boundaries between different deformation mechanisms as a function of grain size at these strain rates, which is relevant to the problem of melt resolidification and (2) develop capabilities in the simulation of polycrystalline metals using molecular dynamics applicable to realistic experimental conditions such as those expected in fusion-class lasers. Therefore, the proposal is well coupled to the Laboratory's strategic plan for materials under extreme conditions.

FY05 Accomplishments and Results

A total of 20 nanocrystalline Ni samples between 9 to 50 nm in size were loaded at pressures of 20 to 72 GPa and recovered at the Janus/Omega lasers. Transmission electron microscopy (TEM) and hardness studies show that the nanocrystalline structures survive the large plastic deformation. Twins appear at 30 GPa for regular Ni, but they are suppressed for nanocrystalline Ni (>20 nm). Massively parallel molecular dynamics simulations uncovered a novel mechanism for reduction of grain boundary sliding, which leads to ultrahard nanocrystals with multiple possible applications. We have implemented a new continuum model based on these atomistic simulations, including grain boundary sliding, while the predictions of standard CALE (C-language-based arbitrary Lagrangian-Eulerian) simulations of laser-induced deformation using current strength models show severe departure from experiments.

Proposed Work for FY06

We plan to complete laser loading, TEM, and hardness measurements of nanocrystals of different grain sizes at different pressures. Some samples will be sent off-site for positron emission tomography studies, to measure nanometer-scale porosity. We will perform molecular dynamics simulations of shocks for the 50- to 70-nm grain size (for possible detection of homogeneous nucleation inside grains) and controlled grain-boundary sliding simulations. Continuum simulations will be performed for laser-induced cratering with different strength models, including those with stress-dependent grain-boundary sliding, to compare with the results of atomistic simulations and experiments. In addition, we plan to study the pressure dependence of hardness that is the basis of many potential applications for nanocrystalline materials.

Publications

Bringa, E. M. et al. (2005). “Ultra-hard nanocrystaline metals by shock loading.” *Science* **309**, 1838. UCRL-JRNL-211450.

Bringa, E. M. et al. (2005). “Wave propagation in polycrystals.” *J. Metals* **57**, 67. UCRL-JRNL-213433.

Remington, B. A. et al. (in press). “Material dynamics under extreme conditions of pressure and strain rate.” *Mater. Sci. Technol.* UCRL-JRNL-215701.

Fission Fragment Sputtering

Bartley B. Ebbinghaus 04-ERD-026

Abstract

Fission fragments born within 7 µm of the surface of uranium (U) metal can eject a thousand or more atoms per fission event. Existing data in the literature show that the sputtering yield ranges from 10 to 10,000 atoms per fission event near the surface, but nothing definitive is known about the energy of the sputtered clusters. This project combines experimental data with molecular-dynamics simulations to develop a continuum model for this internally initiated sputtering process. The study will investigate the amount of material removed per fission event and its dependence on the depth in the material of the nucleus that fissioned, the kinetic energy distribution of the sputtered atoms, and the effects of temperature, and the surface condition of the material on the sputtering process.

The primary goal of this project is to provide a model that can accurately predict the amount, size, and energy of clusters of atoms that are sputtered per fission event in U. The model that is generated from the study will also include best estimates of the effects of surface condition, gallium content, and temperature, but accurate quantification of these effects will likely require follow-on work.

Mission Relevance

By developing a continuum model of U fission events, this work directly supports the Laboratory’s stockpile stewardship mission. By helping to define the physical parameters that cause U to migrate during handling, this project will contribute to safer handling practices for U.

FY05 Accomplishments and Results

Specially designed experimental packages containing U foils, transmission electron microscope (TEM) collection grids, and varying pressures of stopping gas were irradiated. Analysis of these grids using

TEM yielded typical fission-induced fragment size, yield, and energy. Simulated fission fragments of accelerator-driven xenon in tin (in both transmission and grazing geometries) were also carried out. Collected under controlled conditions, these data will aid in connecting the experimental and modeling efforts. Initial modeling using TRIPOS-E indicated that the transfer of the momentum of fragment-driven electrons to local nuclei is not responsible for fragment formation. In conclusion, we theorized that excited electrons change the bonding potential, causing repulsion between the atoms and thus producing fragment ejecta.

Nanosecond Ultrasonics to Study Phase Transitions in Solid and Liquid Systems at High Pressure and Temperature

Brian P. Bonner 04-ERD-033

Abstract

We will combine ultrasonic and multi-anvil technology to make precise measurements of elastic moduli, attenuation, and dispersion at pressures up to 40 GPa and temperatures up to 2200 K on samples with volumes up to 10 mm^3. We will advance our existing expertise in static high-pressure methods by incorporating higher pressures and recent innovations in high-frequency contact ultrasonics and wave-propagation analysis methods developed for geophysical applications. Materials with large grains and fabrication-dependent textures will be studied, as well as time-dependent behavior in melts and phase transitions. We are adapting the gigahertz interferometer developed by our university collaborators to LLNL needs and using these methods in our multi-anvil apparatus for polycrystalline and liquid samples.

A capability for studying time-dependent material properties under extreme conditions will be developed—enabling, for the first time, direct determination of the viscoelastic properties of liquids and crystalline mush, and the kinetic phase boundaries of materials. We will be able to (1) measure precisely, at high pressure and temperature, the properties of polycrystalline materials with grain sizes larger than a few hundred microns and (2) make high-temperature and high-pressure measurements of strength-related properties such as the frequency-dependent shear modulus, and test equation-of-state (EOS) assumptions about the thermal independence of the Gruneisen parameter γ. This is of scientific interest for material characterization problems in stockpile stewardship and earth sciences.

Mission Relevance

This work supports the Stockpile Stewardship Program by providing improved adiabatic EOS information, such as precise velocity measurements of γ and data for shear strength estimates, to validate computer simulations of explosion phenomena. This work also supports LLNL's strategic plan in materials under extreme conditions by providing scientific breakthroughs with high-pressure and high-temperature ultrasonic measurements that address important questions about the composition and thermodynamic state of the Earth and other planetary bodies.

FY05 Accomplishments and Results

In FY05, we accomplished all major milestones and demonstrated the viability of high-frequency ultrasonics in the Laboratory's deformation DIA-type multi-anvil device through successful measurements on tantalum at 6 GPa and 700ºC. We completed the gigahertz apparatus for measurements on gallium alloys and also completed a new opposed-anvil apparatus for ultrasonics at pressures to 30 GPa and at 1000ºC. We adapted and applied the E3D elastic wave propagation code to cell design and analysis of wave propagation.

Proposed Work for FY06

In FY06, we will complete investigations of the frequency dependence of ultrasonic velocity (i.e, dispersion) in liquid gallium alloys, above and below the melting transition. The objective is to determine the transition from relaxed (weak) to unrelaxed (strong) behavior in the liquid phase and to probe the kinetics of melting. We will use the newly completed opposed-anvil cell to characterize frequency dependent velocity and attenuation in the vicinity of the bismuth I–bismuth III phase transition. We will study the volume dependence of velocities to determine the γ in the vicinity of melting and solid–solid phase transitions. The results will be published in appropriate peer-reviewed journals.

Multiscale Characterization of Body-Centered-Cubic Crystals Deformed to Large Extents of Strain

Jeffrey N. Florando 04-ERD-036

Abstract

The objectives of this proposal are to provide large-strain data on body-centered-cubic (bcc) crystals for the validation of dislocation dynamics (DD) simulations and the development of crystal plasticity models. The accuracy of such simulations is based on the ability of the underlying theory to capture the necessary physics, but the large-strain deformation behavior of bcc single crystals is currently not well understood. Because most materials of interest are deformed to large strains, the need exists to develop new experimental techniques to measure the deformation behavior of materials to large extents of strain. The new experimental design will include using a non-contact method to measure strains, in conjunction with a stress analysis of the deformed sample, and multiscale characterization.

The insight gained from the results of the large-strain experiments has the potential to impact crystal plasticity theory and advance predictive modeling capabilities. Crystal plasticity models are an intricate part of a multiscale modeling effort, in which information is passed between simulations conducted at different length scales, with the eventual goal of predicting the deformation response of complex materials under extreme loading conditions.

Mission Relevance

Material-strength models that are accurate under extreme conditions of high pressure, high strain rate, and large extents of strain are a major component of the nation's efforts in stockpile stewardship. Such models will also play a role in the simulation of future ICF experiments. Little work has been done to develop large-strain models, and computer code simulations require experimental data to develop and validate the multiscale crystal plasticity models currently used.

FY05 Accomplishments and Results

In FY05, we validated the testing technique using zinc crystals, which were found to behave according to traditional theory. Samples of molybdenum (Mo) were remachined and tested to achieve 5% strain. The results, which were significantly different than those predicted with conventional theories, showed the underlying deformation mechanisms before and after remachining to be similar. We proposed a theory to explain this anomalous behavior. We also performed experiments on symmetric oriented Mo samples, which qualitatively agreed with the DD simulations; experimentally verified a recently discovered four-node junction; and characterized the deformed samples using x-ray microdiffraction at the Advanced Light Source.

Proposed Work for FY06

Building upon our accomplishments from FY05, we will (1) continue to remachine asymmetrically oriented Mo samples to increase strain to 15%; (2) conduct experiments on tantalum single crystals; (3) carry out large-strain Mo experiments on different orientations and compare the results to our simulations and theories; (4) continue characterization of deformed material; (5) use optical microscopy and atomic force microscopy to search for any changes in the slip-system activity in the deformed samples; and (6) use transmission electron microscopy to characterize the internal dislocation structure of the deformed material, then compare the results to our DD simulations.

Publications

Florando, J. N. et al. (2004). *Shape change studies of bcc single crystals*. Second Intl. Conf. Multiscale Materials Modeling, Los Angeles, CA, Oct. 11–15, 2004. UCRL-PROC-206316.

Magid, K. R. et al. (2004). "X-ray microdiffraction characterization of deformation heterogeneities in bcc crystals." *Proc. Mater. Res. Soc.* **840**, Q7.2. UCRL-PROC-208332.

NanoBIS Determination of the Unoccupied Electronic Structure of Plutonium

James G. Tobin 04-ERD-105

Abstract

The unoccupied conduction band electronic structure of plutonium (Pu) is the missing set of data needed to experimentally validate models of its electronic structure. This project is using nanofocused bremsstrahlung-isochromat spectroscopy (nanoBIS) to obtain direct measurements of the conduction band of Pu, other actinides, and rare earths. By using a scanning electron microscope (SEM) as the source in nanoBIS, we will be able to probe single-crystal microcrystallites in polycrystalline samples. Furthermore, by working at sufficiently high energies, we will preferentially sample bulk properties, thus reducing the impact of surface and band effects. Studies will begin by using rare earths such as cerium (Ce), an analogue of Pu with a similar multiphase electronic structure, and the less-radioactive actinides such as depleted uranium (U).

We expect to determine the unoccupied conduction-band electronic structure of Pu, other actinides, and rare earths in a phase-specific fashion, with emphasis on bulk contributions. Results of this project will resolve controversies about the 4f and 5f electronic structure through direct comparison between our data and predictions made with dynamical mean field theory.

Mission Relevance

In this project, we will validate Pu electronic-structure models and provide experimental benchmarking of modeling. This is foundational science for stockpile stewardship in support of the national security mission.

FY05 Accomplishments and Results

FY05 was the first complete year of funding, and though many of the equipment purchases and setup activities needed to conduct the proposed research were completed in FY04, a significant portion of these activities continued into FY05. Namely, we (1) completed setting up an initial laboratory framework and requisite room modifications; (2) installed and successfully tested the SEM; (3) ordered the last component for the photon detector, which will be a soft x-ray spectrometer; and (4) hired a summer student with computer-aided design experience to help us back-engineer modifications to the SEM.

Proposed Work for FY06

Our original timeline called for instrumentation purchases and lab set up to take place throughout most of FY05, with Ce-nanoBIS experiments scheduled to begin before the end of FY05. However, budget constraints and purchasing difficulties resulted in unexpected delays. Our FY06 plan now calls for

(1) finishing the laboratory setup, including the high-vacuum sample chamber; (2) completing the Ce-BIS calibration; and (3) conducting both U- and Pu-nanoBIS experiments, with the Pu experiments scheduled to begin late spring of FY06.

Publications

Tobin, J. G. et al. (2005). *Using nano-focussed bremsstrahlung isochromat spectroscopy (nBIS) to determine the unoccupied electronic structure of Pu*. 8th Zababakhin Scientific Talks, Snezhinsk, Russia, Sept. 5–10, 2005. UCRL-ABS-212808.

Tobin, J. G. et al. (2005). *Using nano-focussed bremsstrahlung isochromat spectroscopy (nBIS) to determine the unoccupied electronic structure of Pu*. Actinides 2005, Manchester, UK, July 4–8, 2005. UCRL-PRES-211497.

Mapping Phonons at High-Pressure: Phase Transformation, Phase Stability, and Elastic Anisotropy

Daniel L. Farber 04-ERD-106

Abstract

This proposal represents LLNL's participation in an international collaboration to develop new techniques for mapping phonon dispersion curves (PDCs) of cerium—an element often used as a surrogate for plutonium—at high-pressure in the diamond anvil cell using high-resolution inelastic x-ray scattering (HRIXS). The project will focus on probing PDCs at or near electronic transitions at moderate pressure and temperature, and measuring PDCs at ultrahigh pressure (~1 Mbar) and in high-pressure phases. Our ultimate goal is to apply these techniques in U.S. facilities when they become available.

If successful, our results will open a new field of research directed at probing the dynamics of systems at extreme conditions with HRIXS. Understanding the role of phonons in phase transitions is critical to our ability to describe the underlying physics that controls phase stability and a range of transport properties in lanthanide and actinide systems. This work will likely result in publications in high-profile journals.

Mission Relevance

The study of physical properties of cerium promises to provide the key to understanding other lanthanide and actinide systems of importance to the stockpile stewardship mission. We will also investigate the pressure evolution of the elasticity tensor in vanadium, another material relevant to stockpile stewardship.

FY05 Accomplishments and Results

We measured the longitudinal acoustic (LA) phonon branches across the alpha-to-gamma transition in cerium. The LA phonon energies in the alpha phase at 8 Kbar were higher than at lower pressure, consistent with the higher density. However, substantial changes occur along the [110] direction. Phonons between q = 0.4 and 0.6 showed a large energy increase with pressure, while the energy decreased at the zone edge. The shape of the LA phonon branches in alpha-cerium were close to those measured in thorium at ambient conditions, while the gamma-phase phonon dispersion resembled face-centered-cubic metastable lanthanum. This behavior could signify substantial changes in the Fermi surface topology, which would lead to significant changes in the electron–phonon coupling mechanism.

Nanomaterials for Radiation Detection

Tzu F. Wang 04-ERD-107

Abstract

We propose to fabricate three-dimensional (3-D) nanomaterials for radiation detection as improvements over current semiconductor- and scintillation-based technologies. This work is a significant leap beyond the current 2-D nanotechnology and has the potential to significantly improve radiation-detection technologies, which are based on crystal growth. There are two important components of radiation detection: gamma rays and neutrons. For gamma-ray detection, our first goal will be to use porous nanomaterials to achieve scintillation output that is improved several times over that achievable with thallium-doped sodium iodide. For neutron detection, we will construct a 3-D structure using a doped nanowire “forest” supported by a boron matrix.

We expect improvements over semiconductor-based technology in flexibility, cost, durability, surface area, and uniformity in charge collection. Scintillation detection will be improved in output and wavelength. If successful, this project will also enable future development in wide-area radiation monitoring because both neutron energy and neutron direction can, in principle, be determined. Determining both quantities with current radiation-detection technologies is not possible.

Mission Relevance

This work will have significant impacts on nuclear science, especially in radiation detection, in support of the missions in nonproliferation and homeland security. In addition, the neutron spectroscopy capability may have applications to (n,n') (n,2n) and cross-section measurements for the stockpile stewardship mission.

FY05 Accomplishments and Results

For gamma-ray detection, we (1) infiltrated the prepared nano-porous glass matrix with quantum dots and also with laser dyes, characterized the emission and absorption, and studied the effect of the Stokes shift; (2) studied both alpha-particle and gamma-ray interaction with dyes and cadmium selenide quantum dots embedded in nano-porous silica matrices; and (3) used Monte Carlo simulation codes, TRIM and DETECT 2000, to understand the photon transport. For neutron detection, we (1) simulated the detector geometry to determine its neutron detection efficiency; (2) successfully demonstrated the fabrication of a 3-D nanometer-sized pillar system of various diameters using deep reactive ion-etching and nanosphere lithography; and (3) using this method, fabricated pillars with sub-micron size and 1:10 aspect ratio.

Publications

Cheung, B. et al. (2005). *Fabrication of nanopillars by nanosphere lithography*. 2005 Materials Research Society Spring Mtg., San Francisco, CA, Mar. 28–Apr. 1, 2005. UCRL-ABS-207641.

Welty, R. J. et al. (2005). *Roadmap for high efficiency solid-state neutron detector*. SPIE Optics East Conf., Boston, MA, Oct. 23–26, 2005. UCRL-PROC-213583.

Welty, R. J. et al. (2005). S*olid-state pillar structured thermal neutron detector*. Institute of Electrical and Electronics Engineers Nuclear Science Symp. and Medical Imaging Conf., Fajardo, Puerto Rico, Oct. 23–29, 2005. UCRL-ABS-211991.

New Shape-Memory Polymers for Actuators

Thomas S. Wilson 04-LW-054

Abstract

We proposed to develop new shape-memory polymers (SMPs) that bridge the gap in mechanical properties between current SMPs and shape-memory alloys. The new SMPs will have recovery forces up to 10× (50 MPa modulus) over current commercial SMPs and will have new applications in biotechnology and defense. This will be accomplished by the design and synthesis of acrylic and polyurethane network polymers with high crosslink density, and additional modification of these materials by nanoscale modifiers such as single-walled carbon nanotubes. In this two-year project, the first year's focus was on development of new materials, and the second year's focus was on demonstration of the materials as micro-grippers, stents, and expandable foams for applications in defense, biomedicine, and space science, respectively.

We expect that the novel class of actuating materials based on SMPs with significantly increased recovery forces that are created in this project will expand the possibilities for the use of SMPs in many engineering and biomedical applications. This work will attract and retain young scientists and lead the way to new materials development. The intellectual property developed will increase LLNL's portfolio in shape memory polymer technology.

Mission Relevance

The new nanotechnology-based SMPs created in this project have applications in microelectromechanical components for weapons-detection and inspection systems in support of the Laboratory's homeland security and stockpile stewardship missions. In addition, SMPs developed here are applicable to biomedical microdevices for stroke, dialysis therapy, and implants in support of LLNL's mission in bioscience to improve human health.

FY05 Accomplishments and Results

We further investigated extending the new SMPs created in FY04 by modifying SMP chemistry through the use of additional comonomers and by modifying the SMPs with carbon nanotubes and other nanoparticle fillers. We created new SMPs with rubber plateau moduli up to 50 MPa based on new SMPs modified with silica nanoparticles, as well as SMP foams with densities as low as 0.024 g/cm^3. We then demonstrated these to be suitable for aneurysm devices and potential aerospace applications. Additional devices demonstrated include an SMP micro-gripper, SMP optical fiber with diffusing tips, and an SMP stent. These results were accepted for three conference presentations and have to date resulted in two patent applications.

Publications

Wilson, T. S. et al. (2005). *Network polyurethane shape memory polymers for therapeutic devices*. UC Bioengineering Symp., Santa Cruz, CA, June 25–27, 2005. UCRL-ABS-213062.

Wilson, T. S. et al. (2005). *Shape memory polymer therapeutic devices for stroke*. SPIE Optics East, Boston, MA, Oct. 23–26, 2005. UCRL-PROC-216091.

Wilson, T. S., J. P. Bearinger, and D. J. Maitland. (2004). *New shape-memory polymers for actuators*. Polymer Networks 2004, Bethesda, MD, Aug. 15–19, 2004. UCRL-PRES-206090.

Application of Light-Emitting Polymers to Detect Pathogen DNA in Blood

Rodney L. Balhorn 04-LW-065

Abstract

This project proposes to determine whether light-emitting polymers (LEPs) can be used to detect bacterial and viral pathogen DNA as well as various types of damaged DNA containing specific adducts produced by chemical exposures at concentrations that enable their use in clinical and biodefense applications. In this study, we will develop and test the sensitivity of two related methods for DNA detection and analysis. We will also examine the possibility that LEPs may be used to increase the sensitivity of protein activity assays by as much as 100 times.

Successful application of LEPs for detecting pathogen DNA and DNA adducts can result in patentable, highly sensitive chemical sensors for homeland-security and medical applications. Uses include cancer detection, customized treatment for cancer radiation and chemotherapy, more sensitive protein activity assays, and the development of methods not based on the polymerase chain reaction (PCR) for pathogen detection.

Mission Relevance

Numerous applications in national security and medical surveillance can benefit from developments that enable identification of specific DNA sequences in real time and with high sensitivity. Applications include forensic and genetic mutation analysis, detection and monitoring of food-borne pathogens, and biodefense. A 100-fold increase in the detection sensitivity of PCR and protein activity assays would have a direct impact on the development of next-generation biosecurity monitoring systems.

FY05 Accomplishments and Results

A key aspect of this project was the design and synthesis of two new types of LEPs: a water soluble, positively charged LEP that binds to double-stranded DNA, and a LEP with N-hydroxysuccinimide (NHS) groups that could be used to attach peptide substrates. Four LEPs for DNA binding were synthesized and tested. The uncharged LEP109 increased the adriamycin adduct signal four-fold, but only weakly bound to DNA. Positively charged LEPs 180 and 181 bound tightly to DNA. The LEP180 increased fluorescent resonant energy transfer (FRET), but it was not sufficiently soluble. The LEP181 was very soluble (0.8 g/ml); it exhibited suboptimal spectra overlap with adriamycin, but is ideal for ALEXA430 probes. Results showed DNA binding LEPs increase FRET and identified the structures required to optimize signal enhancement. Efforts to synthesize NHS LEPs are ongoing, and a record of invention has been filed based on our research.

Target Fabrication Science and Technology: An Enabling Strategic Initiative

Peter A. Amendt 05-SI-005

Abstract

This project will establish a new, major capability in target fabrication for demonstrating fusion ignition with applications in national security and technology development. To this end, we will develop the science and engineering for (1) nanocrystalline grains; (2) graded-dopant materials; (3) high-strength diamond shells; (4) nanoporous metallic foams; (5) high-strength aerogels; (6) atomic-layer deposition on aerogels; (7) interference and advanced lithography; and (8) joining techniques for precision microassembly. We will develop a double-shell ignition prototype target as a test bed for the elements we develop and integrate.

This project will advance nanoscience techniques for fabricating targets and other structures by developing (1) nanostructured alloys with high strength and thermal stability; (2) the capability to independently control the surface properties and pore structure of nanocellular materials and relate these to nanomechanical behavior; and (3) advanced lithographic approaches to three-dimensional (3-D) nanofabrication on large areas and complex surfaces with nanoscale-relief structures. Quantum confinement in 2-D systems will be investigated. Insight into all of these issues will lead to a new class of materials with novel electronic and luminescent properties.

Mission Relevance

This work supports LLNL's missions in national security and energy resources by developing target-fabrication technology for the increasingly complex targets needed in stockpile stewardship and fusion ignition.

FY05 Accomplishments and Results

In FY05 we (1) fabricated high-strength nanocrystalline gold–copper alloys; (2) produced 80-µm-thick, spherical, nanocrystalline diamond shells by chemical vapor deposition; (3) produced nanoporous gold with a density of 1.8 g/cm^3; (4) demonstrated 3-D nanostructured, gradient-density materials on planar surfaces by conformal phase-mask lithography; and (5) assessed the feasibility of low atomic-number mandrels and the density requirements of nanoporous metallic foams for the prototype ignition double-shell target. Low atomic-number mandrels such as beryllium and glass were found to be an acceptable feature in the double-shell ignition designs, provided the mandrel thicknesses remained below 10 µm. A density scan of the intra-shell nanoporous metallic foam also revealed a favorable tolerance in the ignition designs of three to four times higher density (up to 100 mg/cm^3), thereby significantly facilitating the path to prototype delivery of a double-shell target.

Proposed Work for FY06

In FY06 we will (1) measure tensile properties, strain-rate sensitivity, and activation volume of the target materials; (2) produce a bimetallic graded inner shell of gold–copper; (3) produce nanoporous copper with a density below 100 mg/cm^3; (4) demonstrate—with conformal phase-mask lithography—3-D nanostructured, gradient-density materials on a 500-µm-scale, spherical surface; (5) demonstrate a sub-100-nm density-matched, void-free joint; (6) establish upper bounds on allowable foam porosity; (7) complete the study of atomistic binary transport in double-shell ignition-relevant plasmas and its impact on stability; and (8) complete a design study of diamond ablators for the prototype ignition double shell.

Publications

Amendt, P. A. et al. (2005). "Hohlraum-driven ignition-like double-shell implosions on the Omega laser facility." *Phys. Rev. Lett.* **94**, 0650041. UCRL-JRNL-207191.

Baumann, T. F. et al. (2005). “Facile synthesis of a crystalline, high-surface-area SnO_2 aerogel.” *Adv. Mater.* **17**, 1546. UCRL-JRNL-208832.

Biener, J. et al. (2005). “Nanoporous Au: A high yield strength material.” *J. Appl. Physics.* **97**, 024301. UCRL-JRNL-204561.

Biener, J., A. M. Hodge, and A. V. Hamza. (2005). “Microscopic failure behavior of nanoporous gold.” *Appl. Phys. Lett.* **86**, 121908. UCRL-JRNL-209008.

Hodge, A. M. et al. (2005). “Monolithic nanocrystalline Au fabricated by the compaction of nanoscale foam.” *J. Mater. Res.* **20**, 554. UCRL-JRNL-205649.

Kucheyev, S. O. et al. (2005). “Atomic layer deposition of ZnO on ultralow-density nanoporous silica aerogel monoliths.” *Appl. Phys. Lett.* **86**, 083108. UCRL-JRNL-206530.

Kucheyev, S. O. et al. (2005). “Surface electronic states in three-dimensional SnO_2 nanostructures.” *Phys. Rev. B.* **72**, 035404. UCRL-JRNL-210129.

Kucheyev, S. O. et al. (2005). “Synthesis and electronic structure of low-density monoliths of nanoporous nanocrystalline anatase TiO_2.” *J. Electron Spectrosc. Relat. Phenom.* **144**, 609. UCRL-JRNL-206134.

Kucheyev, S. O. et al. (2005). “Ultrathick, low-stress nanostructured diamond films.” *Appl. Phys. Lett.* **86**, 221914. UCRL-JRNL-209012.

The Structure and Properties of Nanoporous Materials

Anthony W. van Buuren 05-ERD-003

Abstract

Although a great amount of study has been devoted to the physical properties of porous structures, it is not clear if the theoretical models developed to date can be extended to the nanoscopic scale. In this project, we are measuring how the structure of highly porous metal and metal oxide foams change with temperature, pressure, and surface environment. How these porous materials react under such conditions (e.g., how they deform with thermal and mechanical stresses) will be examined with a combination of small-angle x-ray scattering (SAXS) and high-resolution synchrotron-radiation-computed tomography (SRCT). Input from both SAXS and SRCT will be used to generate microstructural models of the foams for analysis with finite-element modeling.

We expect to develop critical technologies for characterizing a new class of high-energy-laser target materials. A key deliverable will be the ability to characterize gradient-density foam microstructures for future laser targets. We propose the following specific aims: (1) perform high spatial resolution measurements of the pore structure in metal and metal-oxide aerogels; (2) use extremely high-resolution diffraction imaging to determine structure of the lattice of a select foam; (3) create finite element modeling, using experimentally measured structures, to study the effects of mechanical loading on the cell structures, and to map out relationships between processing, density, and strength; and (4) determine the extent of any anisotropy in lattice architecture and characterize graded density structures.

Mission Relevance

The ability to characterize and model nanoporous materials will further applications in stockpile stewardship and high-energy-density science, in support of the national security mission.

FY05 Accomplishments and Results

In FY05 we met all the project milestones. We used SAXS on tantalum (Ta) oxide foams as a function of preparation conditions. For the 100-mg/cm^3 foam, x-ray scattering is consistent with a random orientation of 40-nm rod-like structures. Also, images were obtained on the 100-mg/cm^3 foam using a lensless diffraction technique. At 500-nm length scales, the structure appears to be anisotropic with most of the mass oriented in a plane. This is an important new result, for it has been assumed that these aerogels are isotropic at all length scales. The modulus (about 16.5 MPa) is significantly less than that predicted by theoretical models of open cell foams and is the first measure of the mechanical properties of these materials.

Proposed Work for FY06

In FY06, we plan to (1) measure the change in density with high spatial resolution in graded density materials using SRCT, (2) characterize the pore structure in mechanically deformed metal oxide foams using SAXS, (3) characterize the pore structure as a function of density for Ta_2O_5 and as a function of preparation condition in other metal oxide foams. These three steps will provide input for finite element modeling to provide the information needed for machining these materials. We also plan to conduct in-situ wetting studies.

Publications

Kucheyev, S. O. et al. (in press). “Monolithic, high surface area, three-dimensional GeO_2 nanostructures.” *Phys. Rev. B*. UCRL-JRNL-214093.

Nano-Barometers: An In Situ Diagnostic for High-Pressure Experiments

James S. Stolken 05-ERD-009

Abstract

We will develop an in situ diagnostic for high-pressure experiments capable of providing local peak pressure information at high resolution (<1 µm) and over a broad range of pressure (30–300 kbar). The diagnostic consists of nanosensors, embedded in the medium to be measured, that record local peak pressure and are read after the experiment using a variety of microspectroscopy techniques. We will explore the scale dependence, concentration limits, and loading sensitivity of the nanosensors. The sensors will be calibrated over a wide range of pressures and a broad range of deformation conditions in experiments with a diamond anvil cell, gas gun, and laser. The manufacture, deployment, and readout of the nanosensors will be thoroughly documented.

The nanoscale pressure sensor developed as part of this project would have applications in many defense and stockpile-stewardship-related experiments at static pressures ranging from 30 to 300 kbar. A natural follow-on of this project would be to extend the capability to dynamic loadings such as shocks and to design new materials to extend the useful pressure range of the proposed sensors to both lower and higher pressures. Beyond these immediate opportunities, the proposed research would be the nexus through which a variety of sensor materials could be developed for a wide range of nanotechnology applications.

Mission Relevance

The development of an in situ, nanometer-scale pressure sensor will provide both a valuable tool for many existing high-pressure applications and as an enabling technology for new uses and novel experiments. These complement many existing Laboratory efforts in multiscale modeling, material failure and fracture, and laser-driven experiments, furthering LLNL's national security mission related to stockpile stewardship and underlying technology for advanced materials and nanosensors.

FY05 Accomplishments and Results

A key concern regarding the existence and nature of the densification mechanism in silica nanoparticles was addressed. The permanent shift in the Raman spectra of the nanosensors that we measured is consistent with the published results in bulk silica glass. To the best of our knowledge, this was the first such measurement in nanoparticles as a function of particle size. Each of the experimental points that we measured to define the pressure dependence of the Raman shift represents a sequence of spectra taken after the loading and unloading of each sample. These results have been presented at a major international conference on high-pressure phenomena.

Proposed Work for FY06

We plan to complete the in situ nanoscale pressure-sensing technology and calibrate it over a wide range of static pressures and a range of deformation conditions using micro-Raman and transmission-electron-microscope-based spectroscopy. The sensors will be subjected to varying levels of deviatoric and hydrostatic stress by varying the pressure medium within the diamond anvil cell.

Characterization and Control of Laser-Induced Modifications in KDP and DKDP Crystals

Stavros G. Demos 05-ERD-016

Abstract

Potassium dihydrogen phosphate (KDP) and deuterated KDP (DKDP) crystals are the only materials available for use as frequency converters from fundamental (1ω) to second or third harmonics (2ω or 3ω) in large-aperture lasers. The interaction of laser light with defect structures in KDP and DKDP crystals can lead either to damage or to their modification into less absorbing species. The precise conditions controlling these behaviors are not currently known. The objective of this project is to reveal fundamental mechanisms involved in the laser-induced defect reactions leading to bulk damage and laser conditioning in KDP and DKDP crystals. Our ultimate goal is to learn how to control these reactions.

We will significantly enhance our fundamental understanding of the physics involved in laser-induced bulk damage and laser conditioning in KDP and DKDP crystals. This will be accomplished using new diagnostic tools and methods to investigate the microscopic response of a variety of well-characterized materials at varying relevant physical parameters—including pinpoint size and density versus fluence, wavelength, and pulse length at 2ω, 3ω, and combinations thereof, as well as conditioning effectiveness as a function of laser parameters. This work will also extend current knowledge regarding the interaction of high-power laser light with large-bandgap materials.

Mission Relevance

This project supports LLNL's national security mission by providing novel diagnostic tools to quantify and predict the damage performance of optical materials for large-aperture laser systems and offering basic knowledge to optimize conditioning protocols for frequency-doubling and -tripling crystals. In the long term, this project will lead to novel technologies for materials characterization at future large-scale lasers.

FY05 Accomplishments and Results

We investigated the evolution of damage morphology and threshold in the laser conditioning of DKDP crystals at 1ω, 2ω, and 3ω for a total of nine possible combinations. Furthermore, we investigated the results of conditioning at 3ω using a variety of conditioning methods (e.g., increasing laser fluence) to reveal the limits of conditioning and the fastest way to reach them. We also investigated the evolution of damage morphology and threshold under simultaneous 2ω and 3ω irradiation and began investigating the mechanisms of damage, which, unexpectedly, appeared to be due to electronic defect states in the crystal. In collaboration with California State University Northridge, we completed the modeling of the electronic structure of various oxygen-defect clusters in KDP.

Proposed Work for FY06

In FY06, we will (1) develop new approaches to explain damage initiation and conditioning; (2) design specific experiments to test various aspects of the models; (3) develop a method to rapidly characterize the performance of a material by combining analytical or empirical expressions with limited experimental measurements (which will expedite characterization of test materials and quantitative prediction of the damage behavior of crystals); and (4) quantify damage performance in materials exposed to various conditioning schemes, materials grown in different growth conditions, and crystals exposed to conditions that increase the number of damage initiators.

Publications

Carr, C. W. et al. (2005). “Radiation produced by femtosecond laser-plasma interaction during dielectric breakdown.” *Opt. Lett.* **30**, 661. UCRL-JRNL-155867.

Carr, C. W. et al. (2005). “The nature of emission from optical breakdown induced by pulses of fs and ns duration.” *Proc. SPIE* **5647**, 494. UCRL-PROC-207943.

DeMange, P. et al. (2005). “A multi-dimensional investigation of laser conditioning in KDP and DKDP crystals.” *Proc. SPIE* **5991**. UCRL-PRES-215388.

DeMange, P. et al. (2005). “A new damage testing system for detailed evaluation of damage behavior of bulk KDP and DKDP.” *Proc. SPIE* **5647**, 343. UCRL-CONF-208281.

DeMange, P. et al. (2005). “Different precursor populations revealed by microscopic studies of bulk damage in KDP and DKDP crystals.” *Proc. SPIE* **5991**. UCRL-POST-215389.

DeMange, P. et al. (2005). “Microscopic characterization of laser-induced damage performance of large-size KDP and DKDP nonlinear crystals.” *Proc. SPIE* **5337**, 47. UCRL-PRES-204451.

DeMange, P. et al. (2005). “Multi-wavelength investigation of laser-damage performance in KDP and DKDP following laser annealing.” *Opt. Lett.* **30**, 221. UCRL-JRNL-206405.

DeMange, P. et al. (2005). “Nonlinear behavior of laser-induced damage in KDP and DKDP under multi-wavelength irradiation.” *Proc. SPIE* **5710**. UCRL-ABS-205692.

DeMange, P. et al. (2005). "System for evaluation of laser-induced damage performance of optical materials for large aperture lasers." *Rev. Sci. Instrum.* **75**, 3298. UCRL-JRNL-155704.

Negres, R. A. et al. (2005). "Laser-induced damage in DKDP crystals under simultaneous exposure to laser harmonics." *Proc. SPIE* **5991**. UCRL-PRES-215265.

Negres, R. A. et al. (2005). "Stochiometric changes in KH_2PO_4 crystals during laser-induced breakdown." *Appl. Phys. Lett.* **86**, 171107. UCRL-JRNL-206391.

Negres, R. A. et al. (2005). "Stochiometric changes to KH_2PO_4 during laser-induced breakdown." *Proc. SPIE* **5647**, 306. UCRL-PROC-208139.

Negres, R. A., P. DeMange, and S. G. Demos. (2005). "Investigation of laser annealing parameters for optimal laser-damage performance in deuterated potassium dihydrogen phosphate." *Opt. Lett.* **30**, 2766. UCRL-JRNL-211718.

Ceramic Laser Materials

Thomas F. Soules 05-ERD-037

Abstract

Our objective is to evaluate ceramic neodymium-doped yttrium-aluminum-garnet (Nd:YAG) as a laser-gain medium. Ceramic materials offer the same optical and thermal properties as single crystals, but have advantages such as flexibility in design, larger aperture capability, ease and robustness of manufacturing, potential multifunctionality, higher and more uniform activator concentrations, and less brittleness. We plan to make a small piece of transparent Nd:YAG using a nanoparticle material and to evaluate larger transparent ceramic pieces in actual high-power laser configurations using the solid-state heat-capacity laser (SSHCL) and optical characterization capabilities at LLNL.

We expect to be able to apply what we have learned, both in making and evaluating transparent ceramics, to development of new materials and laser architectures that cannot be achieved with single crystals. A good understanding of transparent ceramics opens up many design possibilities. For example, if neodymium-doped strontium fluoride (Nd:SrF_2) can be made as a ceramic, it would offer a 4× increase in lifetime over Nd:YAG and hence 4x diode bar savings in an energy-storage laser. Incorporating samarium-doped YAG as an integral edge cladding would allow for edge pumping of the SSHCL laser and no debonding. Benefits of these and other ideas would include increased power, simplicity of design, and cost savings.

Mission Relevance

This project supports the national security mission by providing ceramic laser-gain slab technology for high-powered lasers for defense applications such as destroying mortar shells, rockets, and land mines.

Current 10 × 10 cm neodymium-doped gadolinium-gallium-garnet (Nd:GGG) single-crystal slabs used in the SSHCL are very difficult to grow and the source is unreliable. Larger single-crystal GGG slabs for still more powerful lasers may not be possible. Other lasers, including those for inertial fusion, can also benefit from the properties of transparent ceramic laser materials.

FY05 Accomplishments and Results

In FY05, we (1) demonstrated that ceramic Nd:YAG slabs can be used in place of the current Nd:GGG single crystals in the SSHCL—as a result, the SSHCL was completely retrofitted with these slabs; (2) demonstrated several novel methods for suppressing amplified spontaneous emission (ASE) by bonding suppressors to the slabs with high index epoxy; and (3) collaborated with researchers at Stanford University to show that we could fabricate our own small pieces of transparent ceramic YAG. Transparent ceramics were also proposed for several other lasers including the next-generation inertial-fusion lasers. A test performed on the damage resistance of transparent YAG ceramics showed a small absorption at 3ω, and the damaged spots became darker with subsequent laser shots.

Proposed Work for FY06

In FY06, we propose to improve the transparency of our ceramic YAG. Also, we will explore other materials and architectures. Again, we will focus on making small pieces to establish feasibility and use a commercial vendor to provide larger pieces for incorporation into lasers. We will (1) study integrally incorporating ASE-suppressing edge cladding into the slab; (2) making a ceramic Nd:SrF_2 and understanding ytterbium-doped YAG as potential inertial confinement laser materials; (3) building scalable and formable shapes with integral ASE suppression by molding; (4) making a ceramic Faraday rotator material; and (5) developing a Q-switched laser gain material using commingled chromium-doped YAG and Nd:YAG.

Determination of the High-Pressure Melting Curve of Iron

Jonathan C. Crowhurst 05-ERD-039

Abstract

The profound geophysical importance of iron demands an accurate determination of its phase boundaries—in particular that of melting—to ultrahigh pressures and temperatures. This project will develop a novel experimental approach for the definitive determination of the iron phase diagram. This method will be based on internal heating of a diamond anvil cell (DAC), combined with the latest synchrotron diagnostic techniques of x-ray diffraction and nuclear forward scattering.

The end result of the project will be an experimental method for determining the phase diagram of iron, and other metals, at high temperatures and pressures. This result will resolve a long-standing issue in geophysics and lead to the refinement of numerous geophysical models. Results will be submitted for publication in high-profile, peer-reviewed journals.

Mission Relevance

By offering a practical and accurate means to determine the phase boundaries of metals at ultrahigh pressures and temperatures and providing reliable experimental data to core computational efforts, this research is applicable to stockpile-relevant materials such as actinides and alloys. It also supports LLNL's commitment to breakthroughs in fundamental science.

FY05 Accomplishments and Results

In FY05, we sustained a temperature of 3500 K at 40 GPa with an internally heated DAC, a temperature of >1000 K at 100 GPa using a laser-heated DAC, and developed the first pressure scale that is good above 1000 K by making Raman measurements in the DAC at the highest temperature to date, which is useful for precise determination of melting pressures. We observed melting in copper at much higher pressures than previously found by measuring resistance changes during melting. We synthesized the second noble metal nitride and have determined its stoichiometry. The same technique will also be used on recovered iron samples to check for reactions. We successfully used the technique of nuclear forward scattering to pinpoint melting in iron at around 25 GPa. We will continue to use this technique.

Proposed Work for FY06

In FY06, measurements of resistance during phase transitions of metals under extreme conditions will continue. We will use our developed technology and experience to investigate the melting of diamond in the <20-GPa range. The melting curve of diamond is important for simulating performance of fusion-class laser targets. Synchrotron x-ray-diffraction measurements will be made on diamond at high temperature and pressure, and synchrotron nuclear forward scattering measurements will be made on iron.

Publications

Goncharov, A. F. and J. C. Crowhurst. (2005). "Pulsed laser Raman spectroscopy in the laser heated diamond anvil cell." *Rev. Sci. Instrum.* **76**, 063905. UCRL-JRNL-210863.

Goncharov, A. F. et al. (2005). "Raman spectroscopy of cubic boron nitride under extreme conditions of high pressure and temperature." *Phys. Rev. B* **72**, 100104. UCRL-JRNL-218266.

Mitigation of Optical Damage Sites on UV Optics

Christian Mailhiot 05-ERD-066

Abstract

The objective of this research is to explore and advance the observation that laser-induced damage on the surface of an optic can be mitigated to extend its lifetime in a high-fluence laser system. We will explore mitigation approaches that use laser, mechanical, and chemical techniques for material removal, crack healing, and surface smoothing; and evaluate the acceptability of the resulting divots sizes and shapes with respect to downstream beam intensification. We will explore both single- and multiple-step approaches in which each step addresses an aspect of the mitigation process in the material. The mitigation effort will focus on fused silica and potassium dihydrogen phosphate (KDP) and emphasize understanding the fundamental materials science and underlying physical processes.

For fused silica, we will explore mitigation methodologies for damage sites ranging from 0.02 to 1 mm in diameter. Our technical approach uses a CO_2 laser and includes (1) varying the optical penetration depth by adjusting the laser wavelength; (2) using a galvo-steered laser to mitigate large damage sites; and (3) modifying the material evaporation rate by using different atmospheres. For KDP, we will examine methodologies based on micromachining and chemical etching. If successfully demonstrated and implemented, these mitigation techniques will significantly extend the useful lifetime of optics in a high-fluence laser system and drastically reduce the associated operational costs. The proposed effort will also advance our fundamental understanding of laser–materials interactions.

Mission Relevance

Successful development and implementation of mitigation techniques that reduce and potentially eliminate damage growth in high-value optical components will significantly benefit worldwide efforts to field large-aperture, fusion-class laser systems by allowing those systems to operate efficiently, reliably, cost effectively, and perform at or above their design specifications. Large-aperture, fusion-class laser systems are essential tools for investigating fundamental physical phenomena relevant to science-based stockpile stewardship, high-energy density science, and inertial confinement fusion, in support of stockpile stewardship and LLNL's energy security mission.

FY05 Accomplishments and Results

In fused silica, we successfully mitigated damage sites approximately 0.3 mm in diameter using a galvo-steered 10.6-µm CO_2 laser. Because of the greater penetration depth at shorter wavelengths, a 4.6-µm CO_2 laser was used to mitigate damage sites exhibiting a deep crack morphology. Thermal field simulations were performed to predict the temperature profile and crater formation in fused silica when illuminated by a CO_2 laser. In KDP, we successfully demonstrated the use of a precision (0.1-mm-radius) diamond tool to micro-machine spherical and Gaussian-shaped divots; investigated the use of ink jets to water-etch KDP surfaces; and performed wave propagation simulations to evaluate downstream beam intensification as a function of the shape and size of the mitigated sites.

Proposed Work for FY06

We will perform sensitivity studies of divot sizes and shapes on the surfaces of key optical components to establish acceptable criteria for downstream intensification of the laser light. For fused silica optics, we will further explore and optimize mitigation approaches using galvo-steered and frequency-doubled CO_2 lasers. For KDP crystals, we will optimize parameters for micro-machining divots of pre-determined sizes and shapes. We will also explore water-etch mitigation approaches based on ink jet and dip-pen nanolithography techniques. Confocal microscopy will be used to provide high spatial and temporal resolution measurements of the optical emission of mitigated sites. Finally, nondestructive diagnostics will be evaluated to ensure the robustness of our mitigation approaches.

Publications

Bass, I., G. M. Guss, and R. P. Hackel. (2005). “Mitigation of laser damage growth in fused silica with a galvanometer-scanned CO_2 laser.” *Proc. SPIE* **5991**. UCRL-ABS-216553.

Carr, C. W. et al. (2005). “A summary of recent damage-initiation experiments on KDP crystals.” *Proc. SPIE* **5991**. UCRL-PROC-216929.

Feit, M. D., A. M. Rubenchik, and J. B. Trenholme. (2005). “Simple model of laser damage initiation and conditioning in frequency conversion crystals,” *Proc. SPIE* **5991**. UCRL-PROC-216930.

Trenholme, J. B., M. D. Feit, and A. M. Rubenchik. (2005). “Size-selection initiation model extended to include shape and random factor.” *Proc. SPIE* **5991**. UCRL-PROC-216931.

A Fracture Mechanics and Tribology Approach to Understanding Subsurface Damage on Fused Silica During Grinding and Polishing

Tayyab I. Suratwala 05-ERD-067

Abstract

The objective of this study is to understand the creation process and characteristics of subsurface fractures formed during the grinding and polishing of brittle materials, specifically glass. We will experimentally characterize the morphology, number density, and depth distribution of various surface cracks (e.g., radial, Hertzian, and lateral) as a function of blanchard, fixed-abrasive, and loose-abrasive grinding, as well as pitch and pad polishing. The effects of load, abrasive particle (size, distribution, geometry, and velocity), slurry properties (pH and Baume), and lap material (pitch and pad) will be

measured. Indentation fracture properties of fused silica will also be measured to develop a global working model to predict subsurface damage (SSD) as a function of a given process step.

The work will produce a body of materials research and enhance manufacturing fabrication processes of benefit to high-energy, high-power laser systems. It will allow them to operate efficiently, reliably, cost effectively, and at or above their design specifications. In addition, this effort will help advance LLNL's efforts in optical materials science and laser technology.

Mission Relevance

The major benefit of this work is a more science-based approach to the fabrication of optical components, a critical and enabling technology for high-energy, high-power, fusion-class laser systems. These laser systems are essential to the Laboratory's mission in stockpile stewardship because of their central role in understanding weapons physics and materials under extreme conditions of temperature, pressure, and strain rate.

FY05 Accomplishments and Results

In FY05, we characterized smoothing by spectral dispersion by measuring fracture morphology, number density, depth distribution, and type for fused silica that had been ground and polished using the wedge technique. We also measured the indentation properties of fused silica, including Hertzian, radial, and lateral indent growth constants and indentation toughness, and changes in the morphology of indentation defects upon hydrofluoric acid etching. Key findings include the following: (1) surface fractures can be characterized by a single type of fracture called a trailing indent fracture, which is analogous to the fracture expected from a loaded, sliding particle; (2) measured depth distribution, regardless of the grinding process, follows a single exponential dependence; (3) SSD depth scales with surface roughness of the resulting sample; and (4) size (both depth and length on the surface) of the individual trailing indent cracks scale with size of the particle being used during the grinding process.

Proposed Work for FY06

In FY06, we will prepare and measure SSD using the wedge technique coupled with standard microscopy for different processes—focusing on the effects of load; particle size distribution; matrix material, including pitch, lap, or pad material; and crack propagation for multiple-step processes. These measured distributions, along with the corresponding crack characteristics, will be correlated to a fracture mechanics model to predict both the distribution and driving factors that control distribution. The model will involve taking an ensemble approach to a single indentation fracture by adding load distribution as a function of particle size to calculate the measured SSD distribution.

Publications

Menapace, J. et al. (2005). *Measurement of process-dependent subsurface damage in optical materials using the MRF technique*. SPIE Boulder Damage Symp. XXXVII—Optical Materials for High Power Lasers, Boulder, CO, Sept. 19–21, 2005. UCRL-PRES-215618.

Miller, P. et al. (2005). *The distribution of subsurface damage in fused silica*. SPIE Boulder Damage Symp. XXXVII—Optical Materials for High Power Lasers, Boulder, CO, Sept. 19–21, 2005. UCRL-PRES-215542.

Suratwala, T. (2005). *Subsurface mechanical damage distributions during grinding of fused silica*. Third Intl. Workshop Flow and Fracture in Advanced Glasses, State College, PA, Oct. 2–5, 2005. UCRL-PRES-215670.

CHARACTERIZATION OF THE EFFECT OF SHORT-PULSE EXPOSURE ON LASER DAMAGE SIZE, MORPHOLOGY, AND CONDITIONING IN WIDE-BANDGAP MATERIALS

Christopher W. Carr 05-ERD-071

Abstract

The objective of this project is to develop a fundamental understanding of how short-pulse (ranging from 100 up to 1000 ps) laser parameters affect conditioning and damage initiation in potassium dihydrogen phosphate and mono-deuterated potassium dihydrogen phosphate (KDP and DKDP) crystals. We will also develop a short-pulse laser with the desired operating parameters (wavelength, pulse length and shape, output power, repetition rate, and beam contrast) for devising optimum protocols to reduce bulk damage in crystals and predict surface damage growth on silicon dioxide (SiO_2). Conditioning effectiveness, pinpoint damage, pinpoint size, and pinpoint morphology will be measured in crystals, and growth rate of surface sites will be measured on SiO_2. From our experimental results, we will develop empirical models to provide insight into the underlying physics of energy deposition and material response to exposure to short-pulse lasers.

After identifying and optimizing laser parameters responsible for the conditioning effect, we will develop offline, laser-based conditioning protocols for crystalline KDP and DKDP materials. These protocols will significantly increase these materials' damage resistance, enhance their performance, and extend their useful lifetimes when used in high-energy, high-power, fusion-class laser systems. We will also develop a predictive model that can describe damage growth on the surface of SiO_2 and damage initiation in the bulk of KDP crystals in a large-aperture laser system. If successful, this work will significantly reduce the operating costs of fusion-class laser systems.

Mission Relevance

Crystalline KDP and DKDP are used for frequency conversion in large, fusion-class lasers. Protocols that extend the useful lifetime of these critical components will have a significant impact on the operating costs of such systems. This work therefore benefits stockpile stewardship and inertial confinement fusion, in support of LLNL's missions in national and energy security.

FY05 Accomplishments and Results

We (1) integrated a variable-pulse-width laser operating with pulse durations of 150 ps to 1 ns at 1064 nm; (2) showed the system capable of delivering 100-mJ pulses at 300 Hz; (3) studied conditioning for variable pulse durations and producing a 2× improvement in damage resistance of KDP for 532-nm operation; (4) developed a superior technique for evaluating conditioning effectiveness; (5) measured the physical and optical properties of laser-induced damage, determining that the micro-cavities, which account for damage in KDP crystals, are similar to those produced by high-energy underground explosions; (6) measured the effect of multiple wavelengths on damage site size and density; (7) measured and modeled the effect of pulse shape on damage site size and density; and (8) filed a record of invention on techniques for measuring laser-induced damage and its effect on laser-beam propagation.

Proposed Work for FY06

We plan to complete a short-pulse laser and use it to establish optimal laser-conditioning parameters, such as the optimum fluence step size, for DKDP third-harmonic generators and KDP second-harmonic generators. We will also determine the optical properties of bulk damage sites in crystals and measure damage growth and morphology of SiO_2 sites initiated at a variety of pulse durations. The effect of residual 1ω light on 2ω and 3ω damage growth will be studied, and the dependence on pulse duration of the growth threshold fluence of SiO_2 and the damage density in KDP will be measured.

Publications

Adams, J. J. et al. (2005). *Wavelength and pulselength dependence of laser conditioning and bulk damage in doubler-cut KH_2PO_4*. Boulder Damage Symp. XXXVII, Boulder, CO, Sept. 19–21, 2005. UCRL-PRES-215422.

Carr, C. W. and J. M. Auerbach. (in press). "The effect of multiple wavelengths on laser-induced damage in DKDP crystals." *Opt. Lett.* UCRL-JRNL-214793.

Carr C. W. et al. (2005). *A summary of recent damage-initiation experiments on KDP crystals*. Boulder Damage Symp. XXXVII, Boulder, CO, Sept. 19–21, 2005. UCRL-PRES-215423.

Feit, M. D., A. M. Rubenchik, and J. B. Trenholme. (2005). *Simple model of laser damage initiation and conditioning in frequency conversion crystals*. Boulder Damage Symp. XXXVII, Boulder, CO, Sept. 19–21, 2005. UCRL-PROC-216930.

Molecular Transport in One-Dimensional Lipid Bilayers: A Biological "Smoke Alarm"

Aleksandr Noy 05-LW-040

Abstract

The purpose of this project is to synthesize novel one-dimensional (1-D) lipid bilayers, use these structures to study ion transport through a single protein pore, and construct a biosensor for detection of bacterial toxins. The key element of the final structure is a lipid bilayer wrapped around a single carbon nanotube. The sensor detects incorporation of the pore-forming toxins into the bilayer and ionic transport through the pore.

This project will not only create a new class of functional bio-compatible nanostructures, but also will use them to do fundamental studies of molecular transport in these systems. In addition to building an important detection capability, the project addresses a number of interesting scientific questions, such as synthesis of 1-D organic nanostructures and studies of mass transport in one-dimensional environments. These studies also have the potential to enable several important biophysical applications, such as creation of subcellular-size ion-selective biocompatible electrodes, or a versatile platform for studies of mass transport through individual protein pores.

Mission Relevance

This project has the full potential to bring a set of novel and important capabilities in biodetection and scientific capabilities in biophysical research, synthesis, and characterization of organic nanostructures. The project supports the Laboratory's mission in homeland security and bioterrorism prevention. Biophysical aspects of this research are relevant to the DOE Genomics:GtL program. In addition, the project contributes to LLNL's mission in breakthroughs in fundamental science.

FY05 Accomplishments and Results

In FY05 we (1) demonstrated layer-by-layer synthesis of polymer shells on suspended carbon nanotube templates; (2) used vesicle fusion to wrap lipid bilayers around these hydrophilic support structures; (3) used fluorescence recovery after photobleaching measurements to demonstrate that these bilayers maintain lateral mobility—a key property for a functional membrane; (4) performed the first demonstration of protein insertion into 1-D lipid bilayers; (5) fabricated single carbon nanotube transistors for our sensor platform and showed that these devices can achieve an on/off ratio of greater than 10,000; and (6) published two articles in peer-reviewed journals.

Proposed Work for FY06

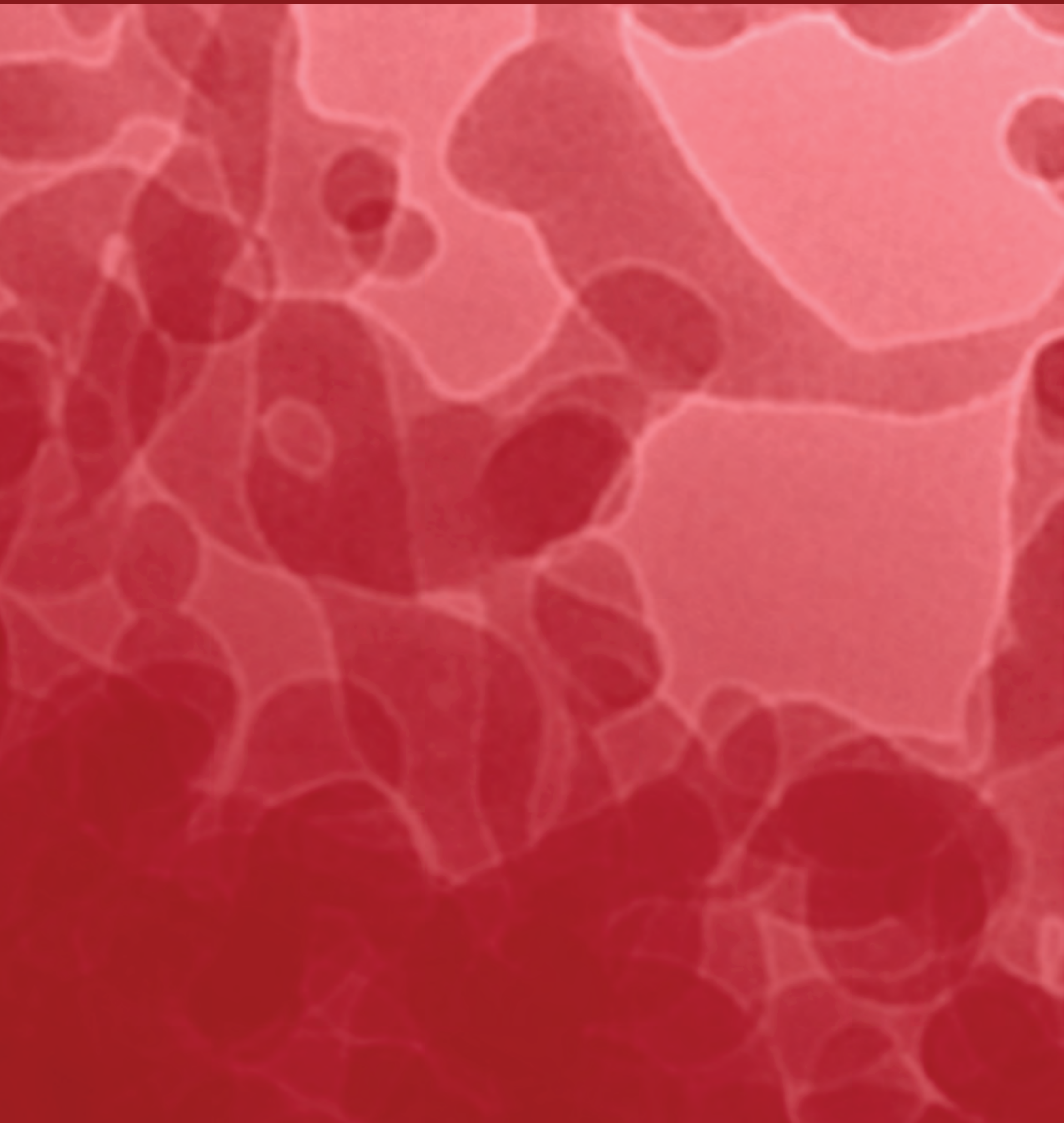
In FY06, we will (1) perform electrical characterization of our nanotube–lipid bilayer devices; (2) conduct protein pore insertion experiments; (3) demonstrate the operation of a novel device—a nanotube–ion channel biotransistor; and (4) evaluate the ability of this bionanodevice to detect pore-forming bacterial toxins.

Publications

Artyukhin, A. B. et al. (2005). "One-dimensional lipid bilayers on carbon nanotube templates." *J. Am. Chem. Soc.* **127**, 7538. UCRL-JRNL-205932.

Huang, S.-C. et al. (in press). "Persistence length control of the polyelectrolyte layer-by-layer self-assembly on carbon nanotubes." *J. Am. Chem. Soc.* UCRL-JRNL-214130.

MATHEMATICS AND COMPUTING SCIENCES



Biological and Synthetic Nanostructures Controlled at the Atomistic Level

Giulia A. Galli-Gygi 03-SI-001

Abstract

Nanotechnology holds great promise for many application fields, ranging from the semiconductor industry to medical research to national security. Novel, nanostructured materials are the fundamental building blocks upon which all these future nanotechnologies will be based. We are conducting combined theoretical and experimental investigations of the synthesis, characterization, and design techniques that are required to fabricate semiconducting and metallic nanostructures with enhanced properties. We have focused on developing capabilities that have broad applicability to a wide range of materials and can be applied both to nanomaterials that are currently being developed for nanotechnology applications and also to new, yet to be discovered, nanomaterials.

We are building expertise, computer simulation and modeling codes, chemical and vapor deposition synthesis techniques, and surface-sensitive spectroscopic characterization techniques for studying novel, nanostructured materials. In addition, we are identifying nanomaterials for designing biological sensing, detection, and separation devices. Our results have generated numerous high-profile scientific publications that have facilitated the recruitment of new postdoctoral researchers and staff to work at LLNL. In addition, we have set up and consolidated new laboratories for materials synthesis and characterization.

Mission Relevance

This proposal will help predict, develop, and design semiconductor materials with tailored properties for advanced biodetection needs in support of LLNL's national-security mission. This will provide a scientific foundation for the creation of next-generation technologies based on manipulating and controlling matter at the nanoscale, in support of the Laboratory's mission of breakthroughs in fundamental science and technology.

FY05 Accomplishments and Results

In FY05, we (1) developed the first techniques for chemically synthesizing crystalline silicon (Si) and germanium (Ge) nanoparticles, and grew the macroscopic superlattices from these nanoparticles; (2) refined our physical vapor deposition techniques to control the size and surface structure of nanoparticles; (3) developed a new collaboration with industry, which is extracting nanodiamonds from crude oil; (4) designed functionalized, fluorescent cadmium selenide (CdSe) nanoparticles to detect signaling pathways and to be detected non-invasively in an MRI; (5) collaborated with the University of California, Berkeley to predict the structure of CdSe nanoparticles and nanorods; (6) fabricated and simulated carbon nanotube molecular detection and separation devices; and (7) examined the properties of metallic nanoparticles for applications in bio-sensing and catalysis.

Proposed Work for FY06

In the final phase of this project, we will focus on completing each of the research tasks in this project and submitting the results for publication. We will (1) finalize our synthesis, characterization, and modeling of Si and Ge nanoparticles with controlled size and surfaces; (2) optically characterize our novel, nanoparticle superlattice structures; (3) complete studies examining how CdSe nanoparticles enter the nuclei of cells; and (4) demonstrate the chromatographic separation capabilities of microfluidic channels constructed from carbon nanotubes.

Publications

Chen, F. and D. Gerion (2004). "Fluorescent CdSe/ZnS nanocrystal-peptide conjugates for long-term imaging and nuclear targeting in living cells." *Nano Lett*. **4**, 1827. UCRL-JRNL-205304.

Drummond, N. et al. (2005). "Electron emission from diamondoids: A diffusion quantum Monte Carlo study." *Phys. Rev. Lett*. **95**, 096801. UCRL-JRNL-209819.

Galli, G. (2004). *Computational design of nanostructures and nanostructured materials*. UCRL-ABS-202352.

Puzder, A. et al. (2004). "The effect of organic ligand binding on the growth CdSe nanoparticles probed by ab-initio calculations." *Nano Lett*. **4**, 2361. UCRL-JRNL-207520.

Reboredo, F. A. and G. Galli (2005). "Theory of alkyl-terminated silicon quantum dots." *J. Phys. Chem*. **B109**, 1072. UCRL-JRNL-207526.

Reboredo, F. (2004). *Theory semiconductor quantum dots of group IV: Optical properties, surface control, and functionalization*. Particles 2004—Particle Synthesis, Characterization, and Particle-Based Advanced Materials, Orlando, FL, Mar. 6–9, 2004. UCRL-ABS-202130.

Trevor, M. et al. (2005). "Rapid degradation of alkanethiol-based self-assembled monolayers on gold in ambient laboratory conditions." *Surf. Sci*. **576**, 188. UCRL-JRNL-205694.

Williamson, A. J. et al. (2004). "Probing the electron density of states of germanium nanoparticles: A method for determining atomic structure." *Nano Lett*. **4**, 1041. UCRL-JRNL-203560.

Williamson, A. J. (2004). *Simulations of CdSe quantum dots*. Ann. American Physical Society March Mtg., Montreal, Canada, Mar. 22–26, 2004. UCRL-CONF-203007.

Williamson, A. J., F. A. Reboredo, and G. Galli (2004). "Chemisorption on semiconductor nanocomposites: A mechanism for hydrogen storage." *Appl. Phys. Lett*. **85**(14), 2917. UCRL-JRNL-203848.

Zaitsea, N. et al. (2005). “Optical properties of CdSe superlattices.” *J. Am. Chem. Soc.* **127**, 10221. UCRL-JRNL-209077.

Zaitseva, N. et al. (2005). “Faceted crystals precipitated from CdSe nanocrystal solutions: Superlattices or bulk selenium?” *Adv. Mater.* **17**, 1321. UCRL-JRNL-203438.

ICE: The Image Content Engine

James M. Brase 03-SI-003

Abstract

Advancements in imaging-sensor technologies (particularly for remotely sensed images) is resulting in volumes of data that overwhelm human analysts. The goal of this project is to develop a framework—the Image Content Engine (ICE)—that will allow analysts to interpret massive volumes of imagery in a timely fashion. The ICE approach is to focus attention of the analysts on a relatively few small areas with specified attributes. This will be achieved by capturing content extracted from images as nodes in semantic graphs whose links define relationships between pieces of image content and information obtained from non-image sources.

The ICE framework will allow content extracted from imagery—the most voluminous data source—to be seamlessly integrated with information from other sources. This will help break down the compartmentalized nature of analyst-based interpretation in the defense and intelligence communities. ICE will also greatly increase the productivity of image analysts by very efficiently focusing attention on potential objects of interest. The ICE approach also applies to other areas of experimental science (e.g., physics, biology, and environmental science), because mining massive archives of complex measurement data for new patterns and relationships is an emerging model for discovery in modern science.

Mission Relevance

The work supports LLNL’s mission in national security by developing technology to allow content extracted from images to be seamlessly integrated with information from other sources, enabling analysts to submit sophisticated queries relevant to defense, intelligence, counterterrorism, deployment strategies, determining the functions of enigma facilities, and detecting production of weapons of mass destruction.

FY05 Accomplishments and Results

In FY05, we (1) completed development and integration of tile-based feature extraction and query tools; (2) fully implemented the model-based, gradient direction-matching (GDM) image search technique and demonstrated its performance in application-specific problems; (3) completed a set of performance characterization experiments that quantify detection and false-alarm performance of the approach;

(4) designed a parallel version of the technique that allows us to scale performance of the method to arbitrarily large image databases; and (5) developed a set of relevance feedback techniques that allow users to enter performance information into the system to improve performance of subsequent searches.

Proposed Work for FY06

In FY06, our work will focus on two main areas. First, we will complete development of the parallel version of ICE and characterize its computational performance in large sets of images. We will characterize data movement through the computing cluster and optimize performance. Our second task is to improve performance of the GDM algorithm by incorporating region weighting, and by developing the ability to specify image queries by defining relations between detected image entities. The new query approach will allow us to decrease false-alarm levels in large image searches.

Publications

Abdulla, G. et al. (2004). *LSST data pipeline prototyping plans and strategy*. Large Synoptic Survey Telescope Data Management Kickoff Mtg., Tucson, AZ, May 10–11, 2004. UCRL-TR-204392.

Abdulla, G. et al. (2004). *Middleware for astronomical data analysis pipelines*. Astronomical Data Analysis Software and Systems XIV Conf., Pasadena, CA, Oct. 24–27, 2004. UCRL-CONF-209265.

Brase, J. M. et al. (2005). *ICE—A system for searching large image databases*. SPIE Defense and Security Symp., Orlando, FL, Mar. 29–Apr. 1, 2005. UCRL-PRES-210838.

Paglieroni, D. W., W .G. Eppler, and D. N. Poland (2005). P*hase sensitive cueing for 3D objects in overhead images*. SPIE Defense and Security Symp., Orlando, FL, Mar. 29–Apr. 1, 2005. UCRL-CONF-210371.

Paglieroni, D. W. (2005). *Phase sensitive cueing for 3D objects in overhead images*. UCRL-TR-210372.

Correction of Distributed Optical Aberrations

Kevin L. Baker 03-ERD-006

Abstract

The objective of this project is to demonstrate the use of multiple distributed deformable mirrors (DMs) to improve the performance of optical systems with distributed aberrations. This concept is expected to provide dramatic improvement in the optical performance of systems in applications where the aberrations are distributed along the optical path. Our approach uses multiple actuated DMs distributed to match the aberration distribution. The project will develop the algorithms necessary to determine the required corrections and simulate the performance of these multiple DM systems.

This project will develop the underlying theoretical and experimental capabilities for correcting distributed optical aberrations, both external and internal. Correcting distributed aberrations, such as in multi-conjugate adaptive optics systems, will allow the systems to achieve diffraction-limited resolution and a greatly enhanced field of view. Correcting internal aberrations, which are of relevance to the Large-Aperture Synoptic Survey Telescope (LSST), will be a prerequisite to enable the science performed on the telescope. This project will also develop a new technique for achieving field correction for short-pulse laser systems, which will increase both the delivered energy and intensity for the high-energy petawatt laser.

Mission Relevance

The development of advanced techniques for correcting distributed aberrations will support new programs in remote sensing and imaging for the Laboratory's national-security and homeland-security missions, and will also enable forefront astronomical science on the next generation of giant telescopes and on the LSST telescope.

FY05 Accomplishments and Results

The work in 2005 was focused primarily on developing the algorithms and code required to accurately correct systems with multiple deformable mirrors such as the LSST. A tomography code was developed to accurately determine the motion of the multiple deformable mirror surfaces to compensate aberrations induced in the system. This code allowed us to determine the number of wavefront sensors required to correct these systems and to optimize the placement of these wavefront sensors to minimize the impact on the focal plane array. New phase retrieval wavefront sensing codes were developed to handle the unique geometries encountered in fast, large field-of-view telescopes. A new method of field conjugation was developed and implemented in software.

Publications

Baker, K. L. (in press). "Tomographic reconstruction of high-energy-density plasmas with picosecond temporal resolution." *Opt. Lett.* UCRL-JRNL-215649.

Baker, K. L. (2005). "Least-squares wavefront reconstruction of Shack-Hartmann sensors and Shearing interferometers using multigrid techniques." *Rev. Sci. Instrum.* **76**, 053502. UCRL-JRNL-209572.

Baker, K. L. and E. A. Stappaerts. (in press). "Assessment of a single-shot, pixelated phase-shifting interferometer utilizing a liquid crystal spatial light modulator accepted" *Opt. Lett.* UCRL-JRNL-215836.

Baker, K. L. and J. Dunn. (2005). *Application of non-interferometric phase-sensitive visible/UV diagnostics in three and four dimensions and extension of non-interferometric phase-sensitive diagnostics into the soft x-ray regime.* 32nd IEEE Intl Conf. Plasma Science, Monterey, CA, June 18–23, 2005. UCRL-ABS-210149.

Adaptive Mesh-Refinement Algorithms for Parallel Unstructured Finite-Element Codes

Ian D. Parsons 03-ERD-027

Abstract

This project will produce algorithms and software for adaptive mesh-refinement (AMR) methods used to solve practical solid-mechanics and electromagnetics problems on multiprocessor, parallel computers using unstructured, finite-element meshes. The goal is to provide computational solutions that are accurate to a prescribed tolerance. The new tools will enable analysts to conduct more reliable simulations at reduced cost, both in terms of analyst and computer time. Research issues include effective error estimators for nonlinear structural mechanics and electromagnetics, local meshing at irregular geometric boundaries, and constructing efficient software for parallel-computing environments.

The technical approach taken by this project will produce AMR tools with applications in state-of-the-art engineering codes like Diablo for solid mechanics, and Eiger and EMSolve for electromagnetics. All of these codes are based on unstructured-mesh, finite-element methods, and will require similar algorithms for tasks such as mesh refinement and load balancing. Deliverables include the components of AMR techniques applied to unstructured finite-element discretizations, such as treatment of irregular geometries, constraint enforcement, intermesh transfer-of-state variables, and partitioning of adaptively refined domains. Model verification will be significantly easier and more transparent by combining error analysis and refinement.

Mission Relevance

Application of our robust, scalable AMR capabilities with simplified mesh generation and lower computational demands will provide a general capability to support LLNL's national-security mission by reducing the total costs of analyses that are performed in stockpile stewardship and other programs. Model verification will be significantly easier and more transparent by combining error analysis and refinement.

FY05 Accomplishments and Results

A parallel AMR capability has been added to Diablo. This includes data structures and algorithms to refine a user-defined mesh and error estimators based on patch recovery. Both isotropic and anisotropic refinement and de-refinement of a mesh are possible. A Gregory patch tracks evolving non-planar boundaries; this allows the mesh to capture the large geometric changes encountered in structural analysis. Software documentation has been produced using unified modeling language, forming the basis of abstraction of AMR schemes. For electromagnetics, both local patch and residual-based recovery have been developed and implemented within the EMSolve production code. Agreement between actual and estimated errors have been observed for several test problems.

Publications

Parsons, I. D. et al. (2005). "Parallel adaptive multimechanics simulations using Diablo." 8th U.S. National Congress on Computational Mechanics, Austin, TX, July 24–28, 2005. UCRL-ABS-210719.

Detection and Tracking in Video

Chandrika Kamath 03-ERD-031

Abstract

In this project, we are developing robust, accurate, and near-real-time techniques for detecting and tracking moving objects in video from a stationary camera. These techniques allow us to model interactions among the objects, thereby enabling us to identify normal patterns and detect unusual events. Our algorithms and software include techniques to separate the moving foreground from the background, extract features representing the foreground objects, track these objects from frame to frame, and post-process the tracks for display. The project focuses on video taken under less-than-ideal conditions, with objects of different sizes moving at different speeds, occlusions, changing illumination, poor weather, low resolution, and low frame rate.

This project is producing robust and accurate technology for video detection and tracking under less-than-ideal conditions. It is enhancing existing algorithms to address these situations, allowing us to better understand their limitations. This, in turn, determines the conditions under which successful surveillance is possible. The algorithms and software are also being applied to spatiotemporal data from computer simulations and can be used to mine text, audio, image, and video data simultaneously.

Mission Relevance

The capability to detect and track with video supports the national security mission of LLNL by enabling new monitoring and surveillance applications for counterterrorism and counterproliferation.

FY05 Accomplishments and Results

In FY05, we (1) put together a data pipeline to allow us to experiment with different algorithms in each step of the pipeline; (2) demonstrated our approach by successfully tracking in low-resolution, low-frame-rate video; (3) reduced the computation time for use of salient regions in tracking; and (4) obtained a better understanding of which methods work well under what circumstances, and the sensitivity of each method to the settings of various parameters. We also filed a patent on our new approach to video detection of moving objects.

Publications

Cheung, S. C. and C. Kamath. (2004). "Robust background subtraction with foreground validation for urban traffic video." *Eurasip. J. Appl. Signal Process.* **14**, 2330. UCRL-JRNL-201916.

Kamath, C. et al. (2005). *Salient points for tracking moving objects in video*. Electronic Imaging Symp. 2005, San Jose, CA, Jan. 16–20, 2005. UCRL-CONF-208738.

Scalable Discretization-Enhanced Solvers

Robert D. Falgout 03-ERD-033

Abstract

We propose to develop scalable, optimal linear-system solvers for discretizations of an important class of partial-differential equations (PDEs) that are handled poorly by existing linear-system solvers. These PDEs are used in accurate modeling of many phenomena studied at LLNL, particularly electromagnetics. The performance of codes for these applications can be adversely affected by poor performance of the linear-system solver. Development of scalable, optimal linear-system solvers for this class of PDEs will enable faster, more realistic simulations. To develop novel multigrid algorithms, we will exploit discrete representations of the large, oscillatory near-nullspaces that characterize these PDEs.

Our goal is to extend this linear solver capability to a whole new class of PDEs, which in turn will impact a large set of the Laboratory's physics codes. As a specific example, a scalable algorithm for implicit Maxwell's equations will enable the EMSolve application code to solve problems that are orders-of-magnitude larger (involving greater accuracy in the computed solution) than can currently be solved, significantly enhancing our capability in electromagnetics design and analysis.

Mission Relevance

An important class of PDEs are used in several LLNL applications. Helmholtz equations appear in light-propagation models for laser-plasma applications and in single-frequency source models for accelerator design. Maxwell's equations arise in broadband-frequency sources for accelerator design and optical scattering. Extending this linear-solver capability to linear systems that result from discretization of these equations will impact physics codes applicable to LLNL missions, especially in the area of stockpile stewardship.

FY05 Accomplishments and Results

We implemented a parallel definitive Maxwell equation solver in the hypre linear-solver library. It is currently used for structured-grid problems with smooth coefficients, but the software was engineered to make it easy to transition to a full unstructured-grid solver with strongly varying coefficients. We collaborated with the HYDRA code group at the Laboratory on this effort. We also developed and implemented a prototype solver for unstructured grid problems that works well for regularized magnetostatics problems. It is now being evaluated for use in Livermore's ALE3D code. Finally, we investigated a third approach for solving definitive Maxwell's equations on unstructured grids, and analyzed a new two-level preconditioning scheme for H(curl) bilinear forms.

Publications

Jones, J. E. and B. Lee. (in press). "A multigrid method for variable coefficient Maxwell's equations." *SIAM J. Sci. Comput.* UCRL-JC-204137.

Kolev, T. and P. Vassilevski. (2005). *Experiments with adaptive element agglomeration algebraic multigrid for H(div) and H(curl)*. 12th Copper Mountain Conf. Multigrid Methods, Copper Mountain, CO, Apr. 3–8, 2005. UCRL-PRES-210920.

Lee B. (2005). *Multigrid methods for equations of electromagnetics*. UCRL-PRES-215520.

Lee, B. and C. Tong. (2005). *An algebraic multigrid/multigraph-based approach for solving Maxwell's equations*. 8th European Multigrid Conf. Multigrid, Multilevel and Multiscale Methods, The Hague Scheveningen, Netherlands, Sept. 27–30, 2005. UCRL-PRES-215508.

Parallel Graph Algorithms for Complex Networks

Edmond T. Chow 03-ERD-061

Abstract

The intelligence community collects enormous amounts of data. To identify potential threats, analysts must sift through this information and understand the interactions among individual pieces or subsets of data. To facilitate this process, we are developing parallel and scalable algorithms for searching very large semantic graphs. This work will enable graph search on distributed parallel computers. We anticipate significant decreases in parallel communication time by using edge partitioning and reductions in search complexity by using heuristic search. The project has two aspects: (1) develop efficient parallel implementations using innovative data structures and algorithms and (2) develop algorithms that exploit the structure of semantic networks.

We expect to develop new algorithms and approaches for analyzing and searching semantic graphs of more than a billion nodes using many thousands of processors. This work will demonstrate that parallel computers can support very large semantic graphs. We anticipate that next-generation semantic graph architectures will be based on these parallel computing ideas.

Mission Relevance

Semantic graphs can be used to encode relationship data gathered for counterterrorism purposes. By exploiting the large aggregate memory of parallel computers, this research will enable the scaling to unprecedented sizes of complex network applications relevant to LLNL's national and homeland security missions. In addition, such parallel algorithms will also have application in complex networks in other disciplines, particularly the analysis of metabolic networks in support of LLNL's mission in bioscience.

FY05 Accomplishments and Results

We report two main accomplishments in FY05. First, we developed a new innovative parallel graph search algorithm for finding paths in massive semantic graphs. Running on 32,000 nodes of the BlueGene/L supercomputer, the code required only a few seconds to search a graph with 3.2 billion vertices and 32 billion edges, which is the largest ever explored by any graph search method. This work was nominated for a Gordon Bell prize at the 2005 Supercomputing Conference. Second, we created new graph metrics and probabilistic models based on the semantic graph ontology. Our methods allow us to infer the existence of links and subgraphs in semantic graphs, which reduces the cost of point-to-point searches and enables analysts to detect patterns in the semantic graph.

Publications

Barthelemy, M., E. Chow, and T. Eliassi-Rad. (2005). *Knowledge representation issues in semantic graphs for relationship detection.* 2005 AAAI Spring Symp. AI Technologies for Homeland Security, Stanford, CA, Mar. 21–23, 2005. UCRL-CONF-209845.

Chow, E., K. Henderson, and A. Yoo. (2005). *Distributed breadth-first search with 2-D partitioning*. 19th ACM Intl. Conf. Supercomputing, Cambridge, MA, June 20–22, 2005. UCRL-CONF-210829.

Yoo, A., et al. (2005). *A scalable distributed parallel breadth-first search algorithm on BlueGene/L*. Supercomputing Conf. 2005, Seattle, WA, Nov. 12–18, 2005. UCRL-CONF-213752.

A Computational Design Tool for Microdevices and Components Used in Pathogen Detection Systems

David P. Trebotich 03-ERI-003

Abstract

We propose to develop new computational models to simulate complex biological flows in integrated microsensors, and to validate these models experimentally. The proposed work is important because microfluidic designs have been chosen for the next-generation of pathogen-detection devices for chemical and biological security efforts. Our computational tool will (1) provide critical understanding of the fluid dynamics involved in microdevices; (2) shorten design and fabrication process to reduce costs; (3) optimize prototypes; and (4) provide a predictive capability for new, more advanced designs. Computational work will be performed in collaboration with University of California (UC) at Davis, and experimental work in collaboration with UC Berkeley.

We expect to produce computational tools for simulating complex biological flows in microdevices. With this capability, the design and fabrication cycle of microelectromechanical system devices, such as the “biobriefcase,” can be shortened by months per cycle by eliminating trial-and-error design. This tool will help to optimize current prototypes and will lead to new, more advanced designs.

Mission Relevance

The new computational models to simulate complex biological flows developed in this project will further LLNL’s mission in countering the proliferation and use of weapons of mass destruction.

FY05 Accomplishments and Results

We (1) pursued more physics and chemistry in our algorithm development; (2) did a proof-of-concept calculation for flow of genomic DNA in an extraction device for the biobriefcase polymerase-chain-reaction module; (3) devised two approaches to particle interactions for bead-rod polymer dynamics: soft potentials and rigid constraints; and (4) demonstrated the fundamental issue of elastic-wave propagation involved in the high Weissenberg number problem for viscoelastic flow used to model particle-laden DNA solutions. Simulations compare well with experimental results. Our approach for treating complex geometry has been shown to be successful and robust by the ability to simulate biological flows in critical anatomies such as a stenotic carotid artery and a diseased trachea.

Publications

Nonaka, A. et al. (2005). "A computational model with experimental validation for DNA flow in microchannels." *Technical Proc. 2005 NSTI Nanotechnology Conf. and Trade Show*, vol. 3, 712–715. UCRL-POST-211217.

Trebotich, D. et al. (2005). "A tightly coupled particle-fluid model for DNA-laden flows in complex microscale geometries." *Computational Fluid and Solid Mechanics 2005*, 1018–1022. New York: Elsevier. UCRL-CONF-208132.

Trebotich, D., P. Colella, and G. H. Miller. (2005). "A stable and convergent scheme for viscoelastic flow in contraction channels." *J. Comput. Phys.* **205**, 315. UCRL-JC-153079.

Trebotich, D. and G. H. Miller. (2005). "Modeling and simulation of DNA flow in a microfluidic-based pathogen detection system." *Proc. 3rd Ann. Intl. IEEE-EMBS Special Topic Conf. Microtechnologies in Medicine and Biology*, 353–355. UCRL-CONF-209334.

Trebotich, D. and G. H. Miller. (2004). *Computational model of bio-fluids in multi-scale systems*. Mtg. American Physical Society Division of Fluid Mechanics, Seattle, WA, Nov. 21–23, 2004. UCRL-PRES-208219.

THREE-DIMENSIONAL VECTORIAL TIME-DOMAIN COMPUTATIONAL PHOTONICS

Jeffrey S. Kallman 04-ERD-004

Abstract

Three-dimensional (3-D) time-domain (TD) design tools are fundamental to developing new technologies to create nonlinear optical devices for data generation, transmission, manipulation, and detection. This project proposes to develop state-of-the-art simulation tools for the design and analysis of next-generation 3-D photonic integrated circuit devices. The simulation codes developed in this

project will include models for optical gain and nonlinearities, and microscopic, non-uniform, inhomogeneous structures. We expect to develop a new suite of 3-D tools general enough to be adapted to specialized problems in several areas, and flexible enough to be applicable to the design of future mixed-signal systems and stand-alone systems throughout the electromagnetic spectrum.

This project is expected to result in a set of sophisticated 3-D TD tools that designers can use to model new, complex photonic devices, and to advance scientific understanding. By developing this unique modeling capability, LLNL will maintain a leadership role in the worldwide scientific and computational community, and be better positioned to carry out our core missions. We expect to publish, patent, and license (where possible) the results of this work.

Mission Relevance

New capabilities developed in this project will benefit LLNL's national-security missions that rely heavily on advanced photonics. Applications include encryption circuits for secure communication and remote sensing for counter- and nonproliferation missions; high-bandwidth optics for advanced laser science and high-density optical interconnects for high-performance computing, both in support of stockpile stewardship; and microsensors for weapons miniaturization.

FY05 Accomplishments and Results

FY05 accomplishments include (1) writing a nonlinear vector finite-element beam propagation solver; (2) writing a significantly faster matrix-less solver for EMSolve (which will speed simulation of materials with time-varying constitutive parameters); (3) incorporating new finite-element operators (wedge products) into EMSolve; (4) incorporating a drift diffusion model for carriers (electron-hole pairs) into EMSolve; (4) writing software to extract constitutive parameters for quantum well materials as a function of carrier density; and (5) using these results to examine designs for Auston switch terahertz sources, examine the effects of phased arrays of Auston switches, and simulate the time course of electron-hole carrier diffusion in bulk semiconductors.

Proposed Work for FY06

Work in FY06 will include developing and incorporating gain and spontaneous emission algorithms into the EMSolve code and replacing Quench3D's scalar beam-propagation solver with a wide-angle vector finite-element beam-propagation solver. The upgrade to EMSolve will allow modeling of optically driven terahertz sources and vertical cavity surface emitting lasers. The upgrade to Quench3D will allow examination of the polarization dependence of light emitted from semiconductor lasers, as well as the polarization dependence of gain-quenched laser logic. In addition, work will continue on modeling devices for a variety of applications related to nonproliferation, defense technologies, and advanced lasers.

Locally Adaptive Mesh Refinement for Linearly Scaling Electronic Structure Calculations

Jean-Luc Fattebert 04-ERD-012

Abstract

In this project, we propose to investigate a new scalable algorithm for efficient electronic structure calculations, the most computationally expensive part of first-principles molecular dynamics simulations. In current application codes, the algorithm scales as the cube of the number of electrons and thus limits the size of tractable simulations. We hope to develop a new algorithm that scales linearly with the number of electrons while preserving the accuracy of standard methods, and investigate a new way of representing the electronic structure using localized orbitals. Fine grids will be used to resolve the "center" of localized wave functions, and coarser grids will represent the tails of the functions. The new approach will use the SAMRAI parallel adaptive mesh refinement framework.

The final product of this research will be a fast and efficient algorithm for electronic structure calculations that may be used as the core of a first-principles molecular dynamics code. This algorithm could potentially reduce by more than one order of magnitude the cost of simulations of physical systems of the order of 1000 atoms, thus allowing larger simulations and longer molecular dynamics, and opening the possibility of investigating new physical phenomena.

Mission Relevance

Achieving an efficient, accurate, and scalable algorithm for computing the electronic structure of a system with many atoms would have a large impact in many research fields that have applications to stockpile stewardship, such as molecular dynamics simulation of materials or fluids at high temperature and pressure, and to determine the equation of state of various materials.

FY05 Accomplishments and Results

In FY05, we developed and tested an iterative algorithm to solve efficiently the Kohn-Sham equation discretized by quadratic finite elements on locally refined grids. In particular, we (1) developed an efficient multigrid preconditioner to speed up convergence; (2) implemented functionalities for full density-functional theory calculations with pseudopotentials in this framework; (3) implemented new functionalities into the SAMRAI library to allow function-dependent mesh refinement; and (4) tested the scaling of these new functionalities on target size problems, demonstrating very low parallelism overhead.

Proposed Work for FY06

In FY06, we will use the functionalities developed so far to implement an algorithm for computing the electronic structure of a system of N electrons in operations of the order of $N \log N$. We will evaluate the efficiency, practical scaling and accuracy of this approach on real problems with thousands of electrons. Timing and accuracy will be compared to state-of-the-art plane waves and finite difference calculations. We will also evaluate how this approach will allow us to treat metals and carry out energy-conserving molecular dynamics.

Publications

Fattebert, J.-L. et al. (2005). "Electronic structure code based on existing parallel AMR infrastructure." *Bull. APS* **50**, 126. UCRL-ABS-208361.

Exfiltration Interdiction Algorithm Development

David A. Knapp 04-ERD-047

Abstract

We will conduct basic research on generalized methods for detecting and interdicting data exfiltrated from classified to unclassified networks. Current approaches to the problem are limited to very application-specific techniques. Our goal is to invent and develop general techniques that are new and innovative.

Success in this project will lay the foundation for a large number of applications in information security. Protecting classified and sensitive information from exfiltration is a goal of all government institutions. This work will establish a cornerstone with application not only in this area, but in many other areas of information security.

Mission Relevance

This work will create new concepts that can be applied to the problem of data exfiltration, in support of LLNL's mission in national security.

FY05 Accomplishments and Results

We have completed an evaluation of the software framework developed for three approaches to interdiction. The algorithms created have been incorporated into the software framework. The results for the algorithms were presented in a national forum with other researchers working in this field, and further work on these algorithms and techniques has now been funded by external sources.

Protein Classification Based on Analysis of Local Sequence-Structure Correspondence

Adam T. Zemla 04-ERD-068

Abstract

We propose to develop a protein-structure comparison system to generate information about sequence correspondence between related protein structures. The ability to verify sequence-based alignments by comparison to the correct structural alignments will significantly improve the quality of protein structure modeling, protein classification, and protein function recognition. There are no fully automated libraries of protein folds that provide credible information about sequence correspondence (alignments) between protein structures. We will deliver a structure alignment program (STRAL) that will generate detailed information about the regions of global and local similarities between protein structures, and will establish a sequence-structure database of structural alignments.

The software and database resulting from this project will demonstrate how the problem of automation of the protein structure classification can be solved. The ability to verify sequence-based alignments by comparison to the correctly calculated structural alignments significantly improves the quality of protein modeling, function recognition, and identification of regions on protein surfaces as candidates for ligand binding sites. Because accurate structural analysis is requisite to computational protein-based detection schemes, this work will improve the success rate and reduce the cost for choosing regions in proteins for antibody or high-affinity ligand recognition, and will improve our ability to identify possible cross-reactive proteins related to the protein targeted for detection.

Mission Relevance

The proposed protein structure comparison system will have immediate applicability in research related to the Laboratory's homeland-security mission. Our proposed STRAL database of protein sequence-structure motifs used for protein structure classification will provide a dramatic advance in protein modeling capability that will enable us to predict more high-quality protein signature targets for pathogens of interest in homeland security.

FY05 Accomplishments and Results

We developed a protein structure comparison algorithm to generate information about sequence–structure correspondence between related proteins. Our STRAL system was shown to be capable of evaluating the level of overall structure similarity, and also of generating detailed information about the regions of local similarities between compared structures. We also designed a set of numerical criteria that use detected structurally conserved regions for automated protein structure classification. The prototype protein structure database we developed clusters proteins based on their similarity in identified structural motifs. Our automated clustering method detects relationships between proteins on the level of structural families that are in very good agreement with manual classifications using the Structural Classification of Proteins database.

Publications

Beuning, P. J. et al. (in press). "Non-cleavable UmuD variant that acts as a UmuD' mimic." *J. Biol.Chem*. UCRL-JRNL-216587.

Geisbrecht, B. V. (2005). "Crystal structures of EAP domains from *Staphylococcus aureus* reveal an unexpected homology to bacterial superantigens." *J. Biol. Chem*. **280**, 17243. UCRL-JRNL-202639.

Zemla, A. et al. (2005). "AS2TS system for protein structure modeling and analysis." *Nucleic Acids Res*. **33**, 111. UCRL-JRNL-209684.

Zemla, A. et al. (2005). *A novel structure-driven approach for protein classification*. UCRL-POST-216262.

Zhou, C. E. et al. (2005). "Computational approaches for identification of conserved/unique binding pockets in the A chain of ricin." *Bioinformatics* **21**, 3089. UCRL-JRNL-209388.

Zhou, C. E. et al. (2005). *Computational approaches for identification of signatures for protein-based diagnostics*. Gordon Research Conf. on Chemical and Biological Terrorism Defense, Buellton, CA, Jan. 31–Feb. 4, 2005. UCRL-POST-209138.

Zhou, C. et al. (2005). *Computational approaches for identification of targets for protein-based diagnostics*. UCRL-POST-211564.

Zhou, C. et al. (2005). *Computational approaches for identification of targets for protein-based diagnostics*. UCRL-POST-211564.

Zhou, C. et al. (2004). *High-throughput selection of protein-based signature targets for detection of bio-threat agents*. ASM Conf. Functional Genomics and Bioinformatics, Portland, OR, Oct. 6–9, 2004. UCRL-POST-207543.

Electro-Thermal-Mechanical Simulation Capability

Daniel A. White 04-ERD-086

Abstract

This project will research and develop numerical algorithms for three-dimensional (3-D) electro-thermal-mechanical (ETM) simulations and incorporate them into LLNL's computational mechanics codes ALE3D and Diablo. A coupled 3-D ETM simulation solves, in a self-consistent manner, the equations of electromagnetics, heat transfer, and nonlinear mechanics. Research will include advection of electromagnetic quantities in an ALE3D setting, algorithms for electrical contact and slide surfaces, and electromagnetic boundary conditions. Extensive algorithm analysis and code verification will be performed to ensure the model equations are solved correctly. To validate the simulation capability, we will compare simulation results to available measured data from magnetic flux compression experiments.

A robust, accurate ETM simulation capability developed in this project will enable LLNL physicists and engineers to better support applications such as explosively driven magnetic flux compressors, electromagnetic launchers, inductive heating and mixing of metals, and microelectromechanical systems. We expect to publish our results in leading peer-reviewed journals in the field.

Mission Relevance

Large-scale computer simulation is a core competency of LLNL. The ETM simulation tools developed in this project will find application in weapons systems for the stockpile stewardship mission. In addition, ETM simulation tools are needed for designing the electromagnetic launchers that have been proposed for DOE equation-of-state research and DoD missile defense and artillery systems.

FY05 Accomplishments and Results

We investigated several different formulations for the electromagnetics: the electric-field formulation, the magnetic-field formulation, and the magnetic vector potential–electric scalar potential formulations. (The key difference between the three formulations is the natural and essential boundary conditions.) We developed a consistent method for computing the magnetic flux density, B, and the induced eddy current density, J, for each of the three formulations, and verified the convergence. These results were accepted for publication in the *IEEE Transactions on Magnetics*. We began to incorporate the electromagnetics into the ALE3D and Diablo codes. In each code the electromagnetics is update in an operator-split manner: the $J \times B$ force is added to the momentum equation and the $\mathrm{J}\cdot\mathrm{E}$ heating to the heat equation.

Proposed Work for FY06

In FY06 we will (1) investigate constrained transport and constrained interpolation in the advection of electromagnetic quantities to create, in ALE3D, the ability to advect electromagnetic fields while still maintaining the divergence-free character of the fields; (2) develop new algorithms for contact and slide surfaces because the continuity conditions of electromagnetic fields and currents across material interfaces is somewhat different than for mechanical stress; and (3) investigate meshless Green's function methods and symmetrically coupled overlaid mesh methods for dealing with zero conductivity regions.

Publications

Solberg, J., et al. (2005). *Coupled electromagnetics/thermal mechanics/solid mechanics simulations using DIABLO*. 8th U.S. National Congress on Computational Mechanics, Austin, TX, July 25–27 2005. UCRL-ABS-209901.

White, D. A., K. Koning, and R. Rieben. (in press). "Development and application of compatible discretizations of Maxwell's equations." *Compatible Spatial Discretizations*. New York: Springer. UCRL-BOOK-212729.

White, D. A. and Rieben R. (in press). "Verification of higher order finite-element solution of transient magnetic diffusion problems." *IEEE Trans. Magn*. UCRL-JRNL-212411.

A New "Natural Neighbor" Meshless Method for Modeling Extreme Deformations and Failure

Michael A. Puso 04-ERD-088

Abstract

The objective of this project is to develop a fully Lagrangian meshless-particle approach to model extreme deformation and failure for analyses such as earth penetration and dam failure. The method should overcome the numerical problems of other meshless approaches by applying a special nodal integration technique that ensures rank stability and by using a "natural neighbor" interpolation scheme that should eliminate tensile instabilities. Material quantities will be stored at node points and so will not need to be re-mapped or advected. The method will be effectively meshless (i.e., nodes will define the model) and will be incorporated into the DYNA3D code.

If successful, the new approach will provide an improved method for modeling extreme events. For example, because the method is fully Lagrangian, better descriptions of anisotropic material damage of concrete can be applied; even water spilling over a damaged dam could be modeled with this approach. Being meshless, the approach could be used for applications where nondestructive evaluation is required, from weapons analysis to biomechanics. The method is more stable than smooth-particle hydrodynamics methods, faster than element-free Galerkin meshless methods, and circumvents the advection required by arbitrary Lagrangian-Eulerian techniques, making it applicable to a much larger class of problems.

Mission Relevance

The proposed approach treats both solid and fluid mechanics and is applicable to several important LLNL national security and homeland security missions. Analyses of earth penetration and terrorist vulnerability evaluation of infrastructure elements (e.g., dams) are target applications for this new approach. As-built x-ray tomography of laser targets and in-vivo MRI imaging for biomechanics create "point clouds" and are good examples where some form of meshless method is needed for expediting stress analyses.

FY05 Accomplishments and Results

In FY05, we found that our original FY04 nodal integration technique was not completely stable and that low-energy modes exist. To rectify this, we added an additional stabilization term. In one conference paper and one journal article, we demonstrated analytically and numerically that this new method is completely stable and consistent. We also developed a novel technique to analytically compute a tight upper bound for the explicit time step to assure robustness. To our knowledge, no one else has succeeded in doing this with a particle meshless method. We analyzed numerous benchmark problems and began analyzing very-large-deformation problems in which connectivity changes drastically.

Proposed Work for FY06

In FY06 we will (1) complete the large-deformation implementation, primarily through "tuning" based on benchmark results; (2) apply new approaches to penetration-type problems (e.g., modifying current material models for required damage capabilities); (3) look at an adaptive point insertion framework based on error indicators as opposed to error estimators; (4) implement domain decomposition that can use different times steps; and (5) implement special integration to other interpolation functions (e.g., the moving least squares interpolation), which can be used for strain gradient methods.

Publications

Puso, M. A. (2005). “An improved linear tetrahedral element for plasticity.” *Proc. 2005 Joint ASCE/ASME/SES Conf. Mechanics and Materials*. UCRL-CONF-211697.

Puso, M. A. et al. (2005). *A new stabilized nodal integration approach*. Third Intl. Workshop Meshfree Methods, Bonn, Germany, Sept. 12–15, 2005. UCRL-PRES-214433.

Internet Ballistics: Identifying Internet Adversaries Despite Falsified Source Addressing

Anthony Bartoletti 04-ERD-095

Abstract

This project will determine the degree to which an Internet adversary can be identified through packet traffic features imposed unwittingly by the attacker’s choice of platform, software, algorithm settings, and network locations, with focus on hostile scan activity. Most network security efforts require adversary identification and characterization, but rely primarily upon easily spoofed or illicit Internet Protocol (IP) addresses. This research seeks to supplant IP address with other evidence that is characteristic of the adversary. We will measure the influence on packet arrival patterns from varied attack software, platform performance, and intervening network conditions by employing controlled experiments with reference platforms. Mathematical characterization will employ wavelets.

If successful, the project will be able to separate adversary-specific traffic signatures from network-specific traffic signatures and will provide a testable foundation for a system of attacker attribution that can be employed in augmenting and disambiguating adversary profiles. In the cybersecurity realm, added confidence in source attribution can lead to improved discovery of related intrusions that might have been overlooked because of deliberately obscured source-IP addresses. A precise regimen of measures best suited to feature separation and detection will be detailed. Directions for further research in improving the underlying metrics will be indicated.

Mission Relevance

This work will support Laboratory and DOE/NNSA’s national security missions in cybersecurity and will benefit counterintelligence efforts through increasing analysts’ ability to relate what might otherwise appear as unrelated network activities.

FY05 Accomplishments and Results

An automated throughput system is in place to collect, partition, and characterize scan traffic upon which a range of data transforms and wavelet signatures may be applied. Controlled tests conducted in a closed network produced baseline behaviors for the tcpreplay packet-injection tool, Nmap, Strobe, and Hping2 for scan rates ranging from 100 to 4000 probes per second. We have successfully used the methodology to distinguish Nmap scans from others, and further partitioning among Nmaps scans in accord with (hidden) attacker-controlled options. Selected wavelet measures have proven effective in producing distinct clusters. A preliminary University of California (UC) Davis tool (ScanVis) has served well to refine our measures. A joint paper was accepted for presentation to the 2005 VizSEC conference.

Proposed Work for FY06

The set of controlled scans will be expanded to test the effectiveness of our methods to separate features imposed by attack hardware and Internet location. Each test will be performed multiple times and at a fixed set of rates. The work of Tadayoshi Kohno at UC San Diego in "clock-skew" identification will be leveraged to refine measures on the network component for these tests. The ScanVis tool will assist in isolating those combinations of measures best able to discriminate these varied sources of signal present in the traffic signatures. A formal paper indicating the most effective measures for isolating adversary drivers will be submitted for publication in a peer-reviewed journal.

Publications

C. W. Muelder, K. Ma, and A. Bartoletti. (2005). *A visualization methodology for characterization of network scans*. VizSEC 2005, Minneapolis, MN, Oct. 26, 2005. UCRL-CONF-217880.

Petascale Simulation Initiative

David R. Jefferson 04-ERD-102

Abstract

This project will dramatically increase LLNL's capability in scalable multiphysics and multiscale simulations for stockpile stewardship and other applications. The focus will be development of algorithms and software for efficient coupling of multiphysics calculations in a Multiple Program, Multiple Data (MPMD) environment on massively parallel computers, and on adaptive algorithms for multiscale modeling. The architecture developed will facilitate coupling existing, independently written codes as components, leading to new multiphysics and analysis capabilities. Advances to the programming environment will provide more flexible, and potentially more efficient, use of massively parallel computers at LLNL. Adaptive sampling will enable more efficient use of multiscale models.

A scalable, simulation-component ("symponent") architecture (SA) will be developed and implemented for multiphysics codes, and the core capabilities will be demonstrated with a full-scale simulation on a high-performance computing platform. Adaptive sampling algorithms will be created and tested,

permitting significantly more efficient use of subscale physical models in full-scale simulations. The code coupling and adaptive sampling software will be portable, usable with other LLNL codes, and applicable to a broad class of material models. The software will take advantage of MPMD parallelism and dynamic launching of component codes. All of the above will be demonstrated in a large-scale simulation using a detailed subscale polycrystalline material model.

Mission Relevance

This project supports stockpile stewardship, homeland security, and other mission areas that require the multiphysics, multiscale synthesis of existing models. The SA will facilitate coupling existing codes, shortening development time for complex applications. MPMD capabilities exercised in the demonstration problem will help achieve greater machine efficiency. Adaptive sampling applied to multiscale material modeling will be useful for many applications requiring subscale physics.

FY05 Accomplishments and Results

In FY05 we (1) completed the first version of the SA interface specifications and ran the initial implementation on the development platform; (2) launched the finite-element code ALE3D and a polycrystal plasticity code as symponents; (3) ran the first version of the multiscale coupling model in parallel but discovered efficiency issues; (4) implemented and tested the first generation of sampling and interpolation algorithms; and (5) routinely ran test problems to exercise the core SA and sampling algorithms; (6) assessed BlueGene/L availability for symponent research, concluding that targeting Blue Gene/L in the scope of this project would not be feasible; (7) ceased porting the code to Blue Gene/L and began porting to an AIX platform instead.

Proposed Work for FY06

In FY06 we will (1) produce Version 2 of the symponent runtime system with high reliability, cleaner interfaces, and better services, scaling, and instrumentation; (2) symponentize ALE3D to enable launching of independent processes for computing fine-scale model results; (3) develop a distributed, high-dimensional database to facilitate parallel adaptive sampling algorithms; (4) create next-generation sampling and coupling algorithms to improve efficiency and accuracy; (5) create a regression suite for the symponent architecture and coupled simulations; (6) conduct performance studies to identify and mitigate bottlenecks; and (7) continue porting the code to run on AIX and other platforms if appropriate.

Publications

Arsenlis, A. et al. (in press). "Generalized in situ adaptive tabulation for constitutive model evaluation in plasticity." *Comp. Meth. Appl. Mech. Eng.* UCRL-JRNL-211883.

Catalyzing the Adoption of Software Components

Thomas G. Epperly 05-ERD-012

Abstract

We are developing technology to automate the componentization of existing source code, which will in turn improve the productivity of code developers. Software component technology has redefined state-of-the-art software engineering practice in industry, enabling codes to be larger, more modular, scalable, robust, and amenable to change. Component technology can also benefit LLNL. Our in-house expertise and tools uniquely position us to conduct research in automated componentization with more sophisticated techniques than those in the literature. By providing a lower-cost path towards componentization, we will help LLNL maintain the lead in delivering large, modular, scalable, robust, and flexible simulation codes in the pursuit of science.

This project will provide (1) technology to automate the construction of components by automatically partitioning existing functions into components and generating Babel interfaces; (2) multiple alternative componentizations; (3) the ability to visualize large-scale applications and their decompositions into components; (4) the means to manually tailor componentization; (5) metrics to evaluate interface suitability; (6) static analysis to determine correct usage of components; (7) techniques to optimize performance of applications using components; and (8) a system to build the generated components.

Mission Relevance

Many Laboratory activities depend on scientific computation to further science in the national interest—particularly science relevant to national security. This project supports various LLNL missions by providing a concrete approach to the development of components within existing LLNL applications. This work will make the development and maintenance of large-scale applications cheaper, more robust, and more flexible.

FY05 Accomplishments and Results

In FY05 we (1) developed a prototype for automatically generating Scientific Interface Definition Language code for a C/C++ source; (2) began developing the initial program visualization capabilities in collaboration with Vaxjo University in Sweden; (3) wrote prototype algorithms for allocating functions to components by experimenting with clustering algorithms applied to the Implicit Radiation Solver package; and (4) drafted a component performance optimization strategy.

Proposed Work for FY06

In FY06 we will (1) perform initial case studies for migrating small to medium-sized libraries from source code to component-based systems, which will involve an initial integration of the different research areas; (2) implement prototype performance optimization algorithms to reduce the runtime overhead of using component interfaces; (3) present multiple alternative componentizations for software packages to allow users to consider multiple alternative designs; and (4) hire a postdoctoral researcher to assist in this work.

Publications

Quinlan, D. J. et al. (2005). *Toward the automated generation of components from existing source code*. 2nd Workshop Productivity in High-End Computing, San Francisco, CA, Feb. 13, 2005. UCRL-CONF-208403.

Quinlan, D. J., S. Ur, and R. Vuduc. (in press). “An extensible open-source compiler infrastructure for testing.” *Lect. Notes Comp. Sci.* UCRL-PROC-217691.

LOCAL: Locality-Optimizing Caching Algorithms and Layouts

Peter G. Lindstrom 05-ERD-018

Abstract

This project is investigating layout and compression techniques for large, unstructured simulation data to reduce bandwidth requirements and latency for visualization and related tasks. Our goal is to eliminate the data-transfer bottleneck (e.g., from disk to memory and from central processing unit to graphics-processing unit) through cache-coherent data access and by trading underutilized computing power for bandwidth and storage. We are approaching this by (1) designing algorithms that both enforce and exploit compactness and locality in unstructured data and (2) adapting offline computations to a novel stream-processing framework that supports pipelining and low-latency sequential access to compressed data. The scalable algorithms developed in this project run on both end-user desktops and dedicated visualization clusters.

We expect to achieve dramatic improvements in disk and memory usage, effective bandwidth, data-access latency, and cache reuse, which will result in more efficient random and sequential access to unstructured grid data. These improvements will lead to the ability to manage larger data sets and store more complete simulation data dumps for post-analysis and visualization. They will additionally provide new capabilities and order-of-magnitude performance improvements in simulation setup, offline mesh processing, interactive data queries, and real-time paging and rendering. These bandwidth- and latency-efficient techniques will become increasingly valuable on next-generation supercomputing architectures such as BlueGene/L with limited local disk capacity.

Mission Relevance

Our research will serve as an important aid in managing and visualizing large data sets from scientific and engineering simulations by supporting analysis and interactive exploration of terascale data sets for the stockpile stewardship mission.

FY05 Accomplishments and Results

After a midyear start, we (1) implemented a prototype of streaming triangle mesh compression that achieves a 50× speedup, uses 75× less memory compared to the state of the art, and reduces data by

20×; (2) designed an algorithm for cache-oblivious triangle mesh layouts that support random access, and demonstrated a 2–10× speedup in various visualization applications; (3) designed a streaming simplifier for triangle and tetrahedral meshes with a 15× speedup over the state of the art using only 20 MB of random access memory; (4) devised representations and defined metrics for measuring the compactness of unstructured streaming meshes; and (5) designed and implemented an algorithm for out-of-core optimal stream layout.

Proposed Work for FY06

In FY06, we plan to (1) design algorithms for the streaming compression of tetrahedral and hexahedral meshes (with 10× to 100× reduction); (2) investigate progressive coding of graphics primitives for interactive visualization; (3) study application-, operating system-, and hardware-managed caching schemes and their performance differences; (4) design compact scalar field representations that exhibit locality in field value (e.g., for fast isocontouring); (5) generalize and apply lossless floating-point compression to simulation data (with up to 10× reduction); and (6) design cache-aware layouts and compare with cache-oblivious ones.

Publications

Isenburg, M. and P. Lindstrom. (2005). *Streaming meshes*. IEEE Visualization, Minneapolis, MN, Oct. 23–28, 2005. UCRL-CONF-211608.

Isenburg, M. et al. (2005). *Streaming formats for geometric data sets*. Workshop Massive Geometric Data Sets, Pisa, Italy, June 2005. UCRL-ABS-212587.

Isenburg, M., P. Lindstrom, and J. Snoeyink. (2005). "Lossless compression of predicted floating-point geometry." *Comput. Aided Des.* **37**, 869. UCRL-JRNL-208490.

Isenburg, M., P. Lindstrom, and J. Snoeyink. (2005). "Streaming compression of triangle meshes." *Proc. Eurographics Symp. Geometry Processing*, Vienna, Austria, July 4–6, 2005. UCRL-CONF-210481.

Yoon, S.-E. et al. (2005). "Cache-oblivious mesh layouts." *ACM Transactions on Graphics* **24**, 886. UCRL-JRNL-211774.

Risk Analysis of Secure Knowledge Discovery

Deborah W. May 05-ERD-041

Abstract

This project seeks to quantify the risks associated with applying knowledge-discovery (KD) rule sets within a mixed-classification knowledge-management system. Many DoD and Department of Homeland Security communities rely on people to manually classify information based on non-quantitative methods. This research will formalize and automate the process of making sound classification judgments based on a quantitative risk analysis. It will also enable automated classification of algorithmically derived information to be made on a massive scale—a much needed capability for LLNL's next-generation knowledge fusion systems. The project will quantify the risks of both too much security, by measuring knowledge fragmentation in graph-modeled information, and too little security, by measuring the potential for information leakage.

Our main goal is to develop a mathematical basis for quantifying risks of automating security in systems. The results from this research will provide a valuable tool for engineers, scientists, and decision makers to understand the risks associated with classification policies. Using this tool, they will be able to make classification decisions based on rigorous and mathematical analyses of the knowledge-sharing loss resulting from overly stringent security policies, and the risk of information leakage resulting from overly lenient security. The tool also offers the user technologies that will maximize both security and knowledge sharing. In addition, this project will enable automated classification of information discovered or extracted by computer algorithms.

Mission Relevance

This project will provide the science base for LLNL to develop sound information security policies, secure KD rule sets, and robust information-to-insight applications that support information systems for a wide range of national security and homeland security missions.

FY05 Accomplishments and Results

In FY05, we developed graph metrics that account for semantic links and ontology-restricted topologies, and developed a software infrastructure for experimenting with metrics by applying them to synthetically generated ontologies and graphs. Having experimented extensively with random, scale-free, and small-world graphs of varying sizes and densities, we validated the new metrics, which we are applying to three real intelligence community graphs. With the software framework and metrics in place, we have begun to simulate security policies by removing nodes/links. A comparison of the metric results before and after node/link removal provide us with the measure of robustness to fragmentation possessed by each graph type.

Proposed Work for FY06

The project's focus areas for FY06 include quantifying the risks of lax security policies by analyzing the potential for inadvertent information leaks. To determine what formal security properties are possessed by a rule set, we will draw on an analogy between KD rule sets and data compression algorithms, and we will apply data compression analysis techniques to rule sets to determine to what extent input data can be deduced from output data. Thus, we will determine what features of the rule sets may be predictive of their security properties. Combined with our FY05 work, this finding will enable us to demonstrate a quantitative risk analysis approach for choosing security labeling policies, KD rule sets, and information models that achieve a desired balance of security and sharing.

A New Method for Wave Propagation in Elastic Media

Anders Petersson 05-ERD-079

Abstract

Simulation of elastic-wave-propagation phenomena is essential for many LLNL applications, including monitoring possible underground nuclear explosions or other seismic events, and nondestructive evaluation of complex parts. We propose to develop significant improvements for the traditional finite-difference technique that (1) allow a fully second-order accurate treatment of boundary conditions in complex domains to handle topography and internal layers and (2) use local mesh refinement to avoid partial oversampling of the solution because of varying wave speeds.

This project will result in a verified, accurate, and efficient elastic-wave-propagation code for numerical simulation in complex two- and three-dimensional (2-D and 3-D) media. This open-source code will be used for many applications at LLNL and in the scientific community. The computer software will support applications ranging from 2-D simulations in nondestructive testing to 3-D earthquake modeling using compute power ranging from desktop workstations (e.g. SUN, LINUX, and OSX platforms) to massively parallel high-performance machines (e.g., MCR, Thunder, and Frost). We plan to validate the code against benchmark problems relevant to LLNL program applications and to publish our research in journals and conference proceedings.

Mission Relevance

Simulation of seismic-wave-propagation phenomena is essential for the success of many applications in support of LLNL national security missions. This includes strong ground-motion prediction for the Enhanced Test Site Readiness Program, the Yucca Mountain Program, nuclear explosion monitoring and underground facilities characterization, and nondestructive testing for locating imperfections in critical components—for example, in optics for large fusion-class lasers.

FY05 Accomplishments and Results

We have developed a finite-difference algorithm for one uniform Cartesian grid, with variable wave speeds and density throughout the domain. A free-surface boundary condition has been implemented on the top surface, which currently is assumed flat, and far-field boundary conditions are of the first-order Clayton–Engquist type. We have also implemented general point-force and moment-source terms. Parallel input/output interfaces have been implemented for reading complex 3-D geophysical material data as well as outputting 2-D slices of the solution and synthetic seismograms. The code has been verified using the method of manufactured solutions and by comparing with previous computational results for 1-D layered seismic models. The code runs both on desktop workstations and in parallel.

Proposed Work for FY06

Effects of surface topography will be included by implementing free-surface boundary conditions on curved 3-D surfaces. The current 2-D methodology requires a linear system to be solved for the boundary points at each time step. While this approach can be generalized to three dimensions, we will also investigate alternative discretizations that decouple the boundary points. Work will begin for satisfying jump conditions across internal material interfaces, which will reduce numerical dispersion errors. This technique will combine the above method for free-surface boundary conditions with a previously developed approach for jump conditions for the scalar wave equation. Verification will continue using manufactured solutions and the code will be validated against relevant measurements.

Publications

Kreiss, H. O. and N. A. Petersson. (in press). “An embedded boundary method for the wave equation with discontinuous coefficients.” *SIAM J. Sci. Comput*. UCRL-JRNL-215702.

Nilsson, S. (2005). *Elastic wave propagation on Cartesian grids with embedded boundaries*. Third MIT Conf. Computational Fluid and Solid Mechanics, June 17, 2005, Cambridge, MA. UCRL-PRES-212820.

Petersson, N. A. et al. (2005). *Simulating wave propagation in solids*. Southern California Earthquake Center (SCEC) Ann. Mtg., Sept. 11–14, 2005, Palm Springs, CA. UCRL-PRES-215201.

Efficient and Reliable Data Exploration via Multiscale Morse Analysis and Combinatorial Information Visualization

Valerio Pascucci 05-ERI-002

Abstract

We are developing a new visualization framework based on general-purpose data-analysis tools coupled with information visualization techniques. The framework will allow fast computation and effective display of metadata roadmaps guiding the exploration of terabytes of raw data. We are using Morse

analysis to build multiscale models of fundamental structures that are ubiquitous in scientific data. The large size and complexity of the topology graphs obtained will require new and general multiscale graph models that we will apply to the exploration of other combinatorial models. The environment will use progressive rendering of multiple linked views and present the graphs with context information that improves the overall data exploration and understanding process.

The success of this project will yield new data-exploration modalities for smooth and discrete data. At the scientific level, this will contribute new basic research both in information visualization and in topology-based data analysis. In these areas we will develop new multi-resolution representation models and external memory algorithms and data structures. On the practical level, our technology will allow us to develop tools for data analysis and presentation that will improve the effective speed of accessing the information stored in terascale scientific data sets and in large semantic graphs. This will be accomplished both by increasing performance of the display methods and by integrating multiple presentation modalities for improved data understanding.

Mission Relevance

This research will enable new techniques for analysis and visualization of large scale scientific data of the type generated by LLNL's stockpile stewardship program as well as more general discrete data representing complex combinatorial relations. We target the needs of scientists and decision makers who need to explore such datasets with the confidence that important features are not overlooked.

FY05 Accomplishments and Results

In FY05 we developed some of the core software tools by (1) constructing a simplified topology with tight error bounds; (2) constructed robust Jacobi sets for two-dimensional (2-D) scalar fields; (3) computing integrals on the Jacobi sets to evaluate the correlation between 2-D fields; (4) demonstrating the robust computation of critical points in the 3-D sampled data; (5) implementing a framework with linked views coordinating multiple presentation modalities; (6) scaling the interface for the external memory visualization of large graphs; and (7) completing the definition of data layout and file formats for storing large graphs that cannot fit in main memory. In addition, we published our results in refereed journals and conference proceedings.

Proposed Work for FY06

In FY06 we will extend the 2-D techniques to the volumetric case and work on a complete and robust computation of the 3-D Morse complex with the main goal of testing time simplification of Jacobi sets over time. Specifically we will (1) use topological analysis to segment bubble structures in mixing fluid dynamics simulations; (2) use time tracking to provide detailed information and global summaries of turbulent mixing behavior; (3) start integrating the topology components with the visualization framework based on linked views; and (4) start extending the tree visualization tools to the case of general graphs first introducing a moderate number of cycles and then scaling to unconstrained connectivity.

Publications

Bremer, P.-T. and V. Pascucci. (in press). “A practical approach to two-dimensional scalar topology.” *Topology-Based Methods In Visualization*. UCRL-BOOK-216201.

Bremer, P.-T., V. Pascucci, and B. Hamann. (in press). “Adaptive refinements in hierarchical topological model.” *Mathematical Foundations of Scientific Visualization, Computer Graphics, and Massive Data*. UCRL-BOOK-218178.

Bremer, P.-T., V. Pascucci, and B. Hamann. (2005). “Maximizing adaptivity in hierarchical topological models using extrema trees.” *Proc. Intl. Conf. Shape Modeling and Applications* **2005**, 298. UCRL-PROC-216200.

Edelsbrunner, H. et al. (2005). “Local and global comparison of continuous functions.” *Proc. IEEE Conf. Visualization* **275**, 280. UCRL-PROC-208677.

Guerra, C. and V. Pascucci. (2005). “Line-based object recognition using Hausdorff distance: from range images to molecular secondary structure.” *Image Vision Comp.* **23**, 405. UCRL-JRNL-208551.

Gyulassy A. et al. (2005). “Topology-based simplification for feature extraction from 3-D scalar fields.” *Proc. IEEE Conf. Visualization* **2005**, 535. UCRL-PROC-216195.

Gyulassy, A. et al. (2005). “A topological approach to simplification of three-dimensional scalar functions.” *IEEE Trans. Visual. Comp. Graph*. UCRL-JRNL-208700.

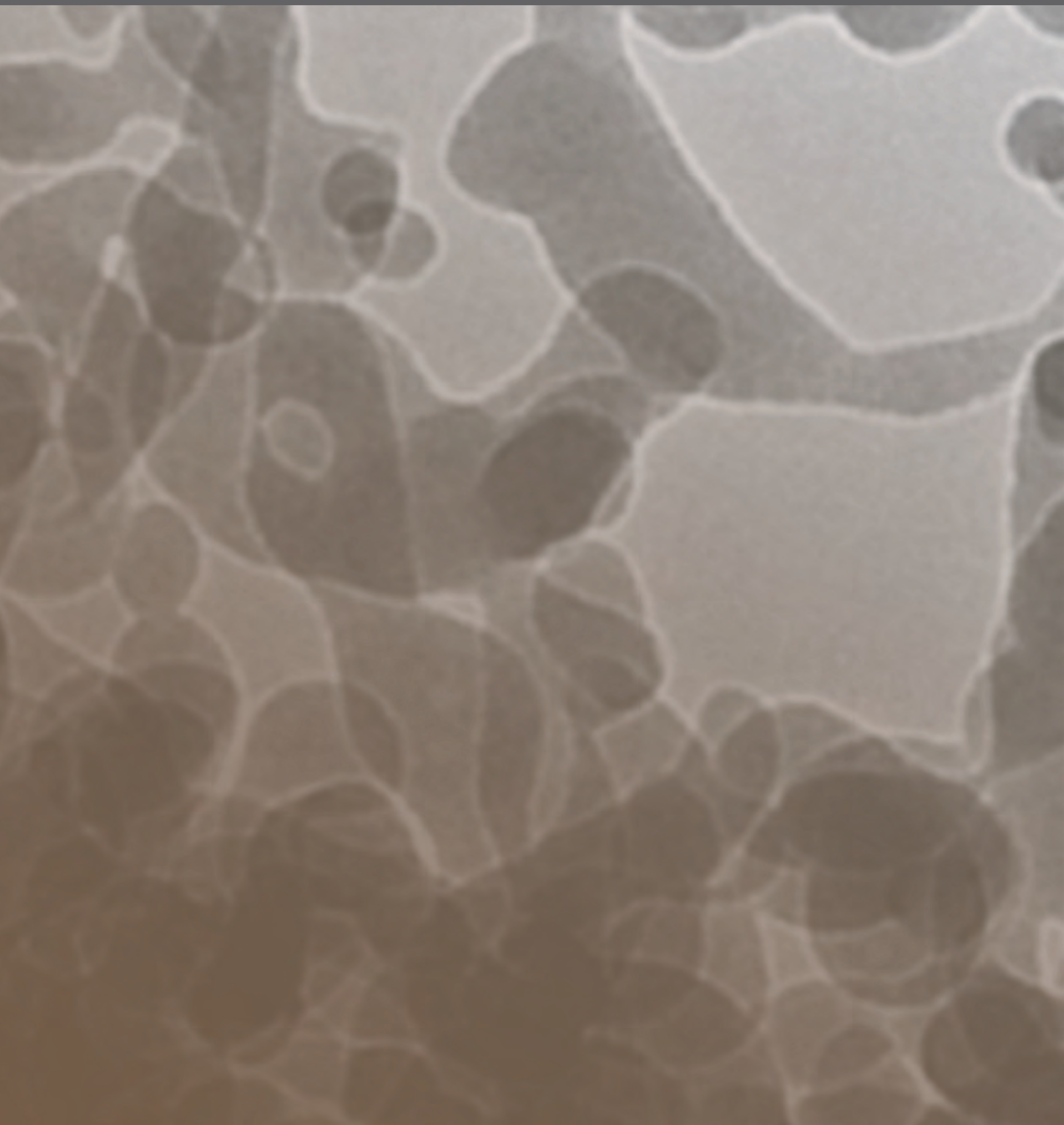
Natarajan, V. and V. Pascucci. (2005). “Volumetric data analysis using Morse-Smale complexes.” *Proc. Intl. Conf. Shape Modeling and Applications* **2005**, 320. UCRL-JRNL-211774.

Pascucci V. (in press). “Multi-resolution streaming techniques for visualization and analysis of scientific data: from structured to unstructured meshes.” *Proc. SIAM Conf. on Parallel Processing for Scientific Computing*. UCRL-PROC-217952.

Pascucci, V., K. Cole-McLaughlin, and G. Scorzelli. (in press). “Multi-resolution computation and presentation of contour trees.” *Mathematical Foundations of Scientific Visualization, Computer Graphics, and Massive Data*. UCRL-BOOK-216196.

Yoon S. A. et al. (in press). “Cache-oblivious mesh layouts.” *ACM Trans. Graph*. UCRL-JRNL-211774.

Nuclear Science and Engineering



Mononitride Fuel Development for SSTAR and Space Applications

Jor-Shan Choi 03-ERD-019

Abstract

We have manufactured modified, nitride-based natural-uranium and mono-uranium fuel pellets for the small, secure, transportable autonomous reactor (SSTAR) currently promoted as the lead fast reactor (LFR) for DOE's Generation IV Initiative. Using nitride-based fuel would make the reactor compact, long-lived, and hence, reduce refueling frequency. These are key features in proliferation-resistant reactor designs. In addition, computational methods were used to analyze performance of the nitride-based fuels in a space-reactor environment. The spent nitride-based fuels were also studied to verify the models and demonstrate feasibility of reprocessing and final disposal in a geologic repository.

Modified uranium nitride-based fuel pellets with content as high as 75 wt% of hafnium nitride were manufactured, but because of the temperature limit of the sintering furnace, lower than expected density of the modified uranium nitride fuel pellets was achieved. Hafnium is a neutron absorber, and its presence in the uranium nitride fuel pellets enabled inclusion of a higher content of fissile material (i.e., high enrichment), and hence, a potentially higher burn-up for the fuel. The successful demonstration of fuel-performance analysis with the SPACEPIN computer code for 6 at.% burn-up of SP-100 nitride-based fuel enhanced our computational capability. However, elaborated effort with computational material science, such as multiscale modeling and phase-field methodology for higher burn-up, are needed for the SSTAR design.

Mission Relevance

This project supported LLNL's energy-security and national-security missions by developing and manufacturing advanced nuclear fuel for SSTAR designs. Such reactors, fueled with the advanced nitride-based fuel, would be fabricated and sealed by nonproliferation-trusted nations, shipped to other nations, and operated for about 30 years—after which, the fuel in the sealed reactor core would be returned to the trusted nation for disposition. The advanced nitride-based fuel manufactured in this project contributed also to the DOE's Advanced Fuel Cycle and Generation IV initiatives, which call for the development of a compact liquid-metal-cooled nuclear reactor for deployment in an expanding nuclear energy future.

FY05 Accomplishments and Results

In FY05, we produced modified uranium nitride fuel pellets with high content of hafnium nitride, though in a lower-than-expected density because of the temperature limit of the sintering furnace. Computational capability of predicted performance of irradiated nitride-based fuel was demonstrated with the computer codes SPACEPIN, LIFE4Rev1, and SIEX3. These codes, when coupled with other computational material science methodologies such as the multiscale and phase-field models, could enhance our modeling capability for material development. Also, we collaborated with the University of California, Berkeley in developing a reprocessing and waste-disposal model for nitride-based fuel. These accomplishments met our goals of fuel manufacturing, modeling and computing, and university collaborations.

Publications

Ebbinghaus, B., J.-S. Choi, and T. Meier. (2003). “A modified nitride-based fuel for long core life and proliferation resistance.” *Proc. Global 2003—Atoms for Prosperity: Updating Eisenhower’s Global Vision for Nuclear Energy*, New Orleans, LA, Nov. 16–23, 2003. UCRL-CONF-200563.

Meier, T., B. Ebbinghaus, and J.-S. Choi. (2003). “Facilities for development of modified nitride-based fuel pellets.” *Proc. Global 2003—Atoms for Prosperity: Updating Eisenhower’s Global Vision for Nuclear Energy*, New Orleans, LA, Nov. 16–23, 2003. UCRL-CONF-200564.

Broadband Radiation and Scattering

Robert M. Sharpe 04-ERD-017

Abstract

This project will enhance our computational electromagnetics capability in broadband radiation and scattering analysis, including electromagnetic interference and electromagnetic compatibility, noise analysis, broadband radar, and accelerator wakefield calculations. Our codes are limited by the accuracy of radiation boundary conditions (RBCs), which truncate space. We will develop improved RBCs by (1) extending the perfectly matched layer (PML) approach to non-Cartesian meshes and (2) developing discrete-time-domain, boundary-integral techniques, which are compatible with high-accuracy, finite-element methods and capable of arbitrary accuracy. The accuracy and efficiency of the two approaches will be compared for a variety of radiation and scattering problems.

The ultimate deliverable is an enhanced computational electromagnetics (CEM) capability that can provide accurate and efficient computational solutions to broadband radiation and scattering problems. The algorithms for improved RBCs will be incorporated into LLNL’s existing EMSolve code. The result will be a 10- to 1000-fold improvement in simulation accuracy. Problems that heretofore were not solvable will be addressed. Improved algorithms and our existing high-performance computer hardware will place the Laboratory’s CEM activity among the top capabilities in the world, thus attracting new talent to LLNL.

Mission Relevance

Our work supports the national-security mission by reducing the time and money spent in building and testing existing programs, and will enable computer simulations for new devices and systems, performance analysis of systems critical to nonproliferation efforts, and the design of micropower impulse radar and other microwave systems.

FY05 Accomplishments and Results

We have completed initial development for the full parallel hybrid, finite element–boundary element code. We have validated the basic parallel algorithms and made comparisons with less-accurate absorbing boundary conditions, and have collaborated with a professor at the University of Washington, an expert on time-domain integral equations. Several stability issues have been addressed and better quadrature techniques have been developed and implemented. Several research papers have been submitted for publication, and results have been presented at national conferences.

Proposed Work for FY06

The main effort will be to improve computational efficiency of the hybrid code by developing and implementing fast algorithms for the boundary element solutions. We are collaborating with a professor at the University of Washington, an expert on time-domain integral equations and fast algorithms for electromagnetics—filtered Green's functions, fast Fourier transforms, and fast QR algorithms (an algorithm to calculate eigenvalues and eigenvectors of a matrix) are under consideration. We will also compare the new hybrid code results with those from our previously developed, high-order PML boundary conditions.

Publications

Fasenfest, B. et al. (2005). *Differential forms basis functions for better conditioned integral equations*. IEEE Intl. Symp. Antennas and Propagation, Washington, DC, July 3–8, 2005. UCRL-CONF-208620.

Pingenot, J. et al. (2005). *Surface based differential forms*. IEEE/ACES Intl. Conf. Wireless Communications and Applied Computational Electromagnetics, Honolulu, HI, Apr. 3–7, 2005. UCRL-CONF-208620.

Decontamination of Terrorist-Dispersed Radionuclides from Surfaces in Urban Environments

Robert P. Fischer 05-ERD-029

Abstract

Cleanup and demolition costs following a terrorist attack with a radiological dispersal device (RDD) could be very high—estimated in excess of several billion dollars. Identification of a new generation of decontamination agents that are highly specific to key radionuclides would represent a significant breakthrough in facilitating post-attack cleanup. This project proposes to identify such agents, to characterize their performance on urban surfaces likely to require remediation, and optimize means for deployment of a comprehensive decontamination system. If successful, the project will identify and optimize a new generation of radionuclide-specific decontamination agents and design a comprehensive decontamination system for responding to an attack with an RDD. This system will be a substantial improvement over existing response capability. Results of this study will be published in peer-reviewed journals.

We will publish the results of the various milestones throughout the expected three-year duration of the project. The first year of research will yield specific results regarding radionuclide substrate interactions that could be utilized immediately by decontamination personnel in the event of an attack with an RDD. During the second year, we expect to publish results concerning optimizing specific agents for use in decontamination operations. During the concluding year of research, we will publish specific results on the design of a comprehensive decontamination system for response to RDD attacks.

Mission Relevance

This project enhances U.S. capability to respond effectively to a radiological dispersal device. It supports national security, homeland security, and environmental risk missions by developing effective decontamination agent(s) for use in urban settings, and advancing the basic scientific knowledge of radionuclide-substrate interactions in the urban environment.

FY05 Accomplishments and Results

Project accomplishments for FY05 include a major sampling campaign of concrete, metal, and grime surfaces performed throughout the Bay Area Rapid Transit system and the CalTrans Caldecott tunnel. Using a unique sampling technique developed during this project, we collected samples without altering the surface deposition layer. Thin sections of material were analyzed by optical and electron microscopy. Results show a high iron, zinc, lead, and carbon content in a 100-µm grime layer.

We also conducted an extensive analysis of the collected samples. Thermodynamic modeling was used to screen over 500 possible decontamination agents, examining effectiveness, selectivity, and potential matrix interference. A short list of candidate agents was generated, and the role of pH on binding efficiency was investigated. Sorption experiments were begun to investigate the binding of americium and cesium on urban grime.

Proposed Work for FY06

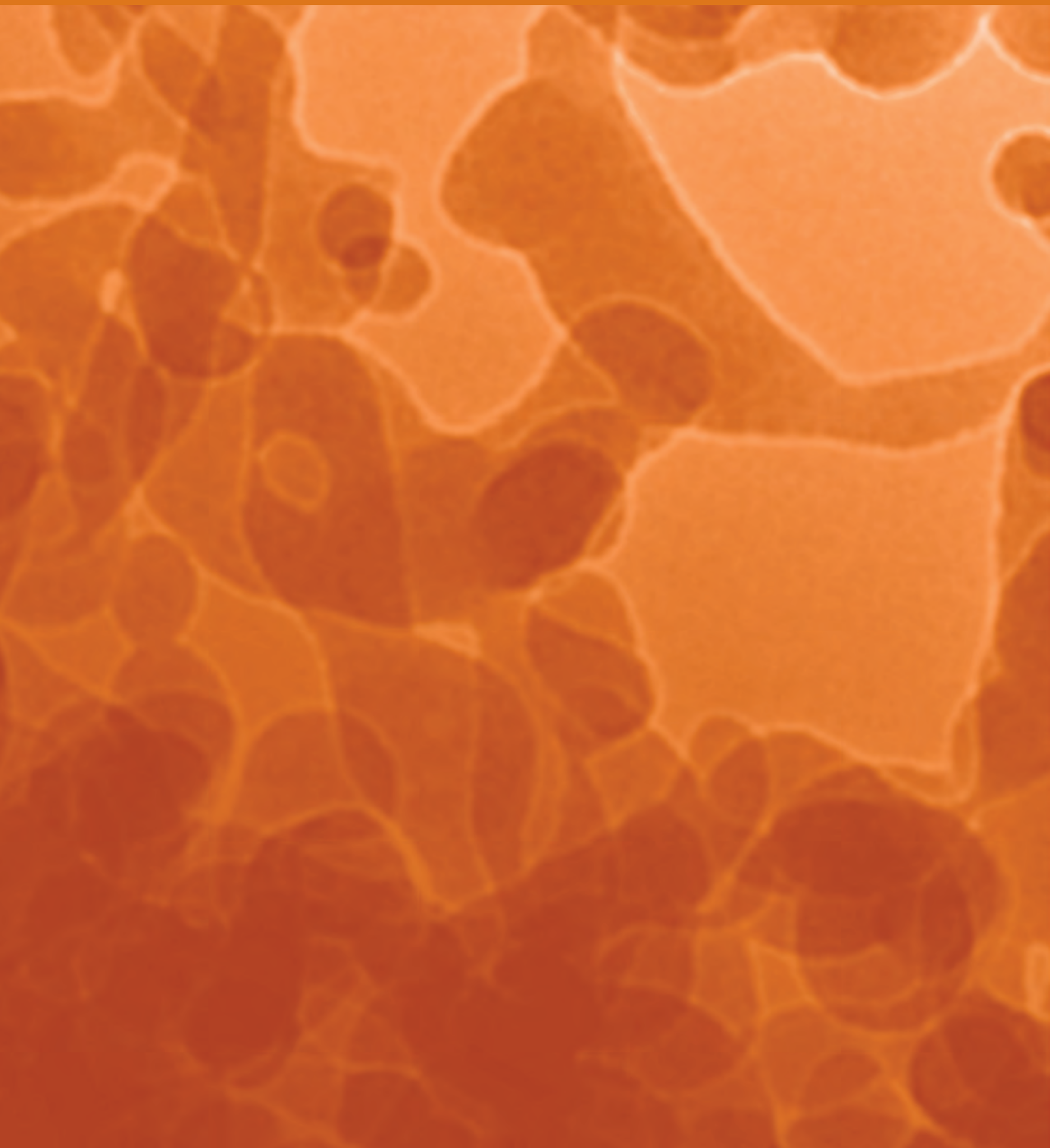
In FY06 we will: (1) synthesize the most promising chelators to perform laboratory and model validation experiments; (2) develop an urban surface testing capability to study radionuclide interaction and agent effectiveness on more simplistic yet empirical surfaces (such as calcite, hematite, and silica) for comparison with real urban surfaces and grime; (3) continue experimental and model evaluation of radionuclide interaction with urban surfaces and grime layers so that we can understand the fundamental science of surface chemistry and interactions; (4) investigate how experimental and model data can be incorporated into fate and transport models for incident response; and (5) answer the basic question about what effect grime has on the fate and transport of radionuclides.

Publications

Fischer, R. and M. Sutton. (2005). *LLNL decontamination science project*. UCRL-PRES-209724.

Sutton, M. and R. Fischer. (2005). *Research in decontamination of terrorist-dispersed radionuclides from surfaces in urban environments*. UCRL-TR-213942.

PHYSICS



A Revolution in Biological Imaging

Henry N. Chapman 02-ERD-047

Abstract

This project develops the science for ultrahigh-resolution imaging of biological materials with x-ray free-electron lasers (XFELs), which are in development. We solve several critical problems in XFEL imaging, including modeling the atomic motion of sample and image reconstruction. Our calculations are used to determine limits to single-molecule imaging as a function of fluence and pulse length. We assess the importance of XFELs and existing light sources to life-science programs, and experimentally test x-ray imaging techniques on a three-dimensional (3-D), micron-sized test object.

The results from our models and experiments indicate that atomic-resolution imaging on XFELs should be feasible. The pioneering work that was performed in this project, such as the hydrodynamic model of the Coulomb explosion and the first lensless 3-D x-ray imaging, will establish LLNL as a premier center for ultrahigh-resolution imaging. Indeed, the 3-D x-ray images achieved here are the world's highest resolution 3-D x-ray images of a non-periodic object. Our work provides the basis that enables imaging at XFELs, and will be used to determine a plan for developing the required technologies. Atomic-resolution imaging of macromolecules, the ultimate result of this work, will have an enormous impact on structural biology and medicine.

Mission Relevance

Improved tomography algorithms will benefit stockpile stewardship. As a specific example, diffraction-imaging techniques can be applied to the study of warm dense matter, a critical regime of weapons physics. Single-molecule imaging will allow researchers to determine the structure of virtually any macromolecule, protein, or virus, which furthers LLNL's missions in both biodefense and bioscience to improve human health. Our research also enhances the capabilities of the Linac Coherent Light Source, a high-priority project of the DOE Office of Science, in support of LLNL's mission in breakthrough science and technology.

FY05 Accomplishments and Results

Using our models, we determined the effectiveness of strategies to allow longer pulses. In particular, we found that a water tamper around the molecule can increase the allowable pulse duration to 50 fs. We extended our hydrodynamic model to longer wavelengths, by including inverse Bremsstrahlung effects, to design validation experiments at the Hamburg Vacuum-Ultraviolet FEL. We collaborated on experiments at the Sub-Picosecond Pulse Source to synchronize an infrared laser with the x-ray pulses, a prerequisite for future experiments. The relative jitter could be measured to a standard deviation of 200 fs. We implemented our 3-D image reconstruction code on a computer cluster, which enabled us to reconstruct 3-D images at 10-nm resolution of our test object and of tantalum oxide foam, a component of fusion-class laser target capsules.

Publications

Cavalieri, A. L. et al. (2005). "Clocking femtosecond x-rays." *Phys. Rev. Lett.* **94**, 114801. UCRL-JRNL-207122.

Chapman, H. N. (2005). *Estimates of signals in LCLS diffraction imaging experiments*. UCRL-TR-210230.

Chapman, H. N. (2005). *Prospects for high-resolution imaging of macromolecules with x-ray lasers*. DOE Single Molecule Research in the New Millenium, Rockville, MD, Apr. 10–12, 2005. UCRL-ABS-211605.

Chapman, H. N. (2004). *Strawman design of the LCLS imaging experiment*. Ultrafast Science and LCLS Experiments Workshop, Stanford, CA, Oct. 25–26, 2004. UCRL-PRES-207913.

Hau-Riege, S. P. (2004). *Pulse requirements for electron diffraction imaging of single biological molecules*. UCRL-TR-207533.

Hau-Riege, S. P. (2004). *The role of damage in x-ray imaging of biological molecules*. Ultrafast Science and LCLS Experiments Workshop, Stanford, CA, Oct. 25–26 2004. UCRL-PRES-207385.

Howells, M. R. et al. (in press). "An assessment of the resolution limitation due to radiation-damage in x-ray diffraction microscopy." *J. Electron Spectrosc.* UCRL-JRNL-208398.

Lindenberg, A. M. et al. (2005). "Atomic-scale visualization of inertial dynamics." *Science* **308**, 392. UCRL-JRNL-210233.

Nugent, K. A. et al. (2005). "Diffraction with wavefront curvature: a path to unique phase recovery." *Acta Crystallogr. A* **61**, 373. UCRL-JRNL-211569.

Chemical Dynamics of High-Pressure Interfaces

Eric R. Schwegler 03-ERD-001

Abstract

The presence of an interface is central to many areas of science ranging from the study of physical properties such as melting and structural phase transitions, to chemical properties such as catalysis and corrosion resistance. In this project, we have worked to develop and apply ab initio simulation methods to the study of complex interfacial systems at the microscopic level. We have built on our existing expertise in first-principles molecular dynamics by incorporating two-phase simulation techniques. These simulation methods have been applied to a set of low-atomic-number materials that are relevant to potential high-power laser experiments.

The primary benefit of this project has been in the development and application of a new capability for investigating interfacial systems under extreme conditions. The success of this project depended on utilizing the unique computational capabilities of the Laboratory. In particular, application of first-principles molecular dynamics to interfacial systems would not be feasible without access to a parallel computing facility and state-of-the-art simulations codes. The techniques on which we have focused are general and can be applied to a wide range of systems. Our research has already led to several high-profile publications and has stimulated new experiments.

Mission Relevance

This research supports LLNL missions in national and energy security by providing a detailed understanding of materials that undergo changes under pressure, melting, and phase transitions. This understanding will particularly benefit the stockpile stewardship program.

FY05 Accomplishments and Results

During FY05, we (1) determined the B2–liquid and the B1–B2 phase boundaries in lithium hydride (LiH); (2) developed an approach for combining quantum Monte Carlo with melting temperatures based on density functional theory and demonstrated it for LiH; (3) examined the mechanism behind the liquid–liquid phase transition in hydrogen in more detail; (4) determined the first-order influence of quantum effects on the hydrogen melt curve; (5) completed two-phase simulations of the melting of ice-VII at 10, 30, and 50 GPa (a manuscript summarizing this work is currently being prepared); and (6) completed and submitted for publication the phase diagram of carbon at 10 Mbar.

Publications

Correa, A. A. et al. (2005). *Falcone Phase diagram of carbon from ab initio simulations*. American Physical Society Mtg., Los Angeles, CA, Mar. 21–25, 2005. UCRL-PRES-210690.

Correa, A. A., S. Bonev, and G. Galli. (in press). “Carbon under extreme conditions: Phase boundaries and electronic properties from first principles.” *Proc. Natl. Acad. Sci. U.S.A.* UCRL-JRNL-216286.

Galli, G. and E. Schwegler. (2005). *First-principles simulations of water: Recent progress and open challenges*. American Chemical Society National Mtg. and Exposition, San Diego, CA, Mar. 13–17, 2005. UCRL-ABS-207530.

Lin, J. F., E. Schwegler, and C. S. Yoo. (in press). “Phase diagram and physical properties of H_2O at high pressure and temperatures: Applications to planetary interiors.” *American Geophysical Union Monograph: Earth’s Deep Water Cycle*. UCRL-JRNL-215773.

Ogitsu, T. et al. (2005). *Ab-initio study of the B1-B2 phase transition in LiH and its impact on melting*. Study of Matter at Extreme Conditions Mtg., Miami, FL, April 17–21, 2005. UCRL-ABS-211462.

Ogitsu, T. et al. (2005). *First-principles simulations of LiH: Towards the B2 phase*. 20th Congress Intl. Union of Crystallography, Florence, Italy, August 23–31, 2005. UCRL-ABS-210644.

Schwegler, E. (2005). *Structural and dynamical properties of water under ambient and high-pressure conditions*. American Physical Society Mtg., Los Angeles, CA, Mar. 21–25, 2005. UCRL-PRES-210674.

Schwegler, E. et al. (2004). "Towards an assessment of the accuracy of density functional theory for first principles simulations of water II." *J. Chem. Phys.* **121**, 5400. UCRL-JRNL-204466.

Trave, A. et al. (2005). *Beryllium at high pressure and temperature: A first principles molecular dynamics study*. American Physical Society Mtg., Los Angeles, CA, Mar. 21–25, 2005. UCRL-ABS-209040.

Quantum Electrodynamics and Electron Collisions in the Superstrong Fields of K-Shell Actinide Ions

Peter Beiersdorfer 03-ERD-004

Abstract

This project studies the rich physics of actinide ions and develops new capabilities for doing so, including a high-energy microcalorimeter sensitive to 100-keV ions. The ultrastrong nuclear field found only in the heaviest ions offers rich new physics that will advance atomic modeling and enhance our understanding of nuclear physics. We will determine electron-impact excitation cross-sections, dielectronic recombination, atomic energy levels, the Lamb shift, and the energy levels of isomeric nuclear states. The project combines several LLNL capabilities: the world's only source of stationary high-Z ions [the Super Electron Beam Ion Trap (SuperEBIT)]; access to the relevant actinides; high-resolution, hard x-ray detectors developed in collaboration with NASA; and unique theory capabilities.

This project will provide the most definitive test of high-field, bound-state quantum electrodynamics ever undertaken, identify the role of new forces active in high-Z collisions, and quantify their significance for spectral modeling of high-temperature plasmas. The project will determine the energy splitting of the thorium-229 (^{229}Th) ground state from the lowest-lying isomeric state, and will create a new class of hard x-ray (up to 100 keV) detector (the microcalorimeter) and the highest-resolution extreme ultraviolet (EUV) spectrometer ever developed.

Mission Relevance

This world-class science effort will test the range of validity of collisional theory important for LLNL's mission in stockpile stewardship. This world-class science effort also extends the Laboratory's core competencies in atomic and nuclear physics, and will attract talented young scientists to the Laboratory.

FY05 Accomplishments and Results

In FY05 we performed our final high-resolution x-ray measurement of ^{229}Th using the hard x-ray microcalorimeter developed for this purpose. This measurement provided a value within 0.5 eV of the energy of its isomeric ground level, dramatically revising the value and accuracy of what was previously known about this level. Measurement of the 2s Lamb shift of uranium was completed, resulting in the first determination of the two-loop Lamb shift term and providing the most stringent test of bound-state quantum electrodynamics in strong nuclear fields. Finally, a 2-J laser system was completed and successfully achieved isotope injection into SuperEBIT.

Publications

Beiersdorfer, P. et al. (in press). “Measurement of the two-loop Lamb shift in lithiumlike U^{89+}.” *Phys. Rev. Lett.* UCRL-JRNL-211310.

Brown, G. et al. (in press). “Laboratory astrophysics and atomic physics using the NASA/GSFC microcalorimeter spectrometers at the LLNL electron beam ion trap and radiation properties facility.” *Nucl. Instrum. Methods Phys. Res. A.* UCRL-JRNL-214698.

Brown, G. V. et al. (in press). “High-resolution gamma-ray spectroscopy with a NASA/GSFC microcalorimeter at the Lawrence Livermore National Laboratory.” *Bull. APS.* UCRL-ABS-215796.

Quinet, P. et al. (2005). *Multiconfiguration Dirac-Fock wavelengths and transition rates in the x-ray spectra of highly charged Ga-like ions from Yb^{39+} to U^{61+}*. UCRL-JRNL-217878.

Trabert, E, et al. (2005). “Soft-x-ray spectra of highly charged Os, Bi, Th, and U ions in an electron beam ion trap.” *Cana. J. Phys.* **83**, 829. UCRL-JRNL-208760.

Vilkas, M. J., Y. Ishikawa, and E. Trabert. (2005). *Relativistic multireference many-body perturbation theory calculations on F-, Ne-, Na-, Mg-, Al-, Si-, and P-like xenon ions*. UCRL-JRNL-217882.

Exploring Properties of Quantum Chromodynamics with Proton–Nucleus and Deuteron–Nucleus Collisions

Michael D. Heffner 03-ERD-005

Abstract

The standard model of strong interactions predicts the presence at high temperatures of a phase transition in which nucleons dissolve into a plasma of quarks and gluons. Relativistic heavy-ion collisions provide the only laboratory-controlled method available for producing and studying this phase of matter, and is the primary goal of the Relativistic Heavy Ion Collider (RHIC) at Brookhaven National Laboratory. Experimental proof of production of this phase relies on a set of observations that are currently inconclusive. The key to their resolution lies in studying baseline proton–nucleus collisions. We will upgrade the Pioneering High-Energy Nuclear Interaction Experiment (PHENIX) at RHIC to characterize the fragmentation of the nucleus in proton–nucleus collisions to help resolve if the phase has indeed been observed.

A new detector installed in PHENIX to characterize proton–nucleus collisions will aid in the systematic study of bulk nuclear matter at high energy densities and help resolve a longstanding question: can we

create a new phase of matter, the quark–gluon plasma, in the laboratory and study its properties? We expect to gain a much improved understanding of physics in this regime.

Mission Relevance

This work furthers LLNL's national-security mission by advancing cutting-edge techniques in nuclear radiation detection and state-of-the-art computer simulation of complex events. This project will also help recruit top scientists in these fields through leading research in nuclear and particle physics.

FY05 Accomplishments and Results

The LLNL upgrade to PHENIX, funded by this LDRD, was successful in making measurements of the baseline proton–nucleus interactions at RHIC. New data collection and analysis in the last few years at RHIC have generated comparisons of baseline measurements to nuclear collision measurements in the gold—gold system, that have shown that the matter generated in the relativistic heavy ion collision is qualitatively different from ordinary nuclear matter, and is consistent with a plasma of quarks and gluons. One PHENIX paper that uses measurements from the LLNL upgrade has been submitted to *Physical Review Letters*, and we expect to submit another in the next year. The upgrade will also continue to be used in future runs at RHIC.

Electron Production and Collective Field Generation in Intense Particle Beams

Arthur W. Molvik 03-ERD-008

Abstract

The future of heavy-ion fusion could depend on predicting and controlling electron cloud effects (ECEs), which limit the performance of present positron and ion storage rings. To better understand the phenomena and develop mitigating mechanisms, this project is conducting theoretical and experimental studies of electron-cloud generation, trapping, transport, and effects on ion beams. Experiments are carried out at the Heavy-Ion Fusion Virtual National Laboratory. Theoretical studies are coordinated with a Lawrence Berkeley National Laboratory LDRD project that combines a code that models electron clouds with the particle-in-cell code, WARP, for modeling self-consistent beams in accelerators. The project will validate the models and tools through comparison with experimental data.

This project will lead to development of definitive theoretical and experimental tools for studying ECEs in advanced accelerators. If successful, the project will further the fundamental understanding of gas desorption.

Mission Relevance

By addressing ECEs in ion beams, the models and diagnostic techniques developed in this project will be applicable to high-intensity accelerator technology, in support of LLNL's stockpile stewardship and fusion energy missions.

FY05 Accomplishments and Results

WARP simulations met the objective of replicating experimental results: agreement ranges from semi-quantitative agreement to close quantitative agreement. This code was developed, in collaboration with Lawrence Berkeley National Laboratory (LBNL), by adding self-consistent electron and gas populations to the beam-dynamics particle-in-cell code, WARP, and combining it with the electron-cloud code from LBNL, POSINST. A breakthrough electron mover speeds computations by one to two orders of magnitude. Measurements were developed of each source of electrons and their accumulation during a pulse. WARP modeled electron oscillations and a "Z" shaped distortion of ion velocity space when electrons from an end wall are permitted to flood the beam. These tools are being used by the accelerator community.

Publications

Cohen, R. H. et al. (2005). "Simulating electron clouds in heavy ion accelerators." *Phys. Plasmas* **12**, 56708. UCRL-JRNL-209229.

Cohen, R. H. et al. (2005). "Simulating electron cloud effects in heavy-ion beams." *Nucl. Instrum. Methods Phys. Res. A* **544**, 210. UCRL-CONF-204532.

Cohen, R. H. et al. (2004). "Electron-cloud simulation and theory for high-current heavy-ion beams." *Phys. Rev. ST Accel. Beams* **7**, 124201. UCRL-JRNL-205555.

Molvik, A. W. et al. (2005). *Experiments studying desorbed gas and electron clouds in ion accelerators*. Particle Accelerator Conf., Knoxville, TN, May 16–20, 2005. UCRL-CONF-212340.

Molvik, A. et al. (2005). "Experimental studies of electrons in a heavy-ion seam." *Nucl. Instrum. Methods Phys. Res. A* **544**, 194. UCRL-JRNL-204975.

Stoltz, P. H. et al. (2005). "Electron loss, range and electron yield comparisons of the CRANGE ion-material interaction code." *Nucl. Instrum. Methods Phys. Res. A* **544**, 502. UCRL-JRNL-216226.

Stoltz, P. H. et al. (2004). "Simulation of heavy-ion induced electron yield at grazing incidence." *Phys. Rev. ST Accel. Beams* **7**, 103201. UCRL-JRNL-207387.

Vay, J-L. et al. (2005). *Filling in the roadmap for self-consistent electron cloud and gas modeling*. Particle Accelerator Conf., Knoxville, TN, May 16–20, 2005. UCRL-PROC-216104.

Vay, J-L. et al. (2005). *Initial self-consistent 3D electron cloud simulations of the LHC beam with the code WARP+POSINST*. Particle Accelerator Conf., Knoxville, TN, May 16–20, 2005. UCRL-PROC-216105.

A Coupled Turbulence-Transport Model for Edge Plasmas

Thomas D. Rognlien 03-ERD-009

Abstract

We are developing the first predictive turbulence-based model of plasma and neutral characteristics in tokamak fusion devices. Experiments show that edge-plasma properties are key to high fusion-energy gain, and simulations are needed to reliably design large devices. The project uses parallel computer codes to solve magnetized-fluid equations for both the fast-timescale turbulence in three dimensions (3D) and the much slower toroidally averaged transport (profile evolution) in 2D. Multi-scale coupling of these processes is essential because the turbulence is driven by plasma gradients and flows, and in turn, the profile-determining transport is the result of turbulent plasma fluxes and plasma/surface interactions that produce neutral particles via recycling and sputtering.

The project produces a new edge-plasma model based on self-consistent turbulence and profile evolution, a major step beyond previous models. Consequently, temperature and density at the boundary of the hot fusion plasma can be modeled, as well as wall heat loads from escaping plasma and line radiation. The model can be used to help design and guide operation of future devices such as the International Thermonuclear Experimental Reactor. The coupling algorithms developed for turbulence/transport interactions have application to other important complex fluid systems of national interest such as climate and combustion, where there is a need to describe the long-time consequences of fast turbulence and slow system evolution.

Mission Relevance

This project advances fusion-energy research in support of LLNL's energy-security mission and directly advances the Laboratory's recognized excellence in developing comprehensive models of the complex edge-plasma region. Such models require numerical algorithms utilizing large, coupled physics simulations on LLNL's high-performance computers, which are also used to support the Stockpile Stewardship Program. The project also furthers the Laboratory's mission to develop breakthroughs in fundamental science.

FY05 Accomplishments and Results

Coupling has been demonstrated for the two major types of tokamak edge geometries: magnetically diverted and material limiters. We have advanced the coupling procedure to allow many of turbulence/transport iterations between codes to be performed automatically, and also developed key physics diagnostics. The coupling demonstrates statistical steady-state edge plasmas for moderately unstable plasmas, and large intermittent transport for strongly unstable plasmas—the latter require development of time-dependent simulations. Inclusion of a radial electric field model demonstrates substantial suppression of the strong intermittent transport, thereby allowing the edge plasma temperature to increase, and leading to the expectation of improved energy confinement and fusion gain.

Publications

Rognlien, T. D. (2005). *Consistent turbulence/transport coupling for pedestal/SOL regions*. Intl. Tokamak Physics Activities Pedestal Mtg., St. Petersburg, Russia, Oct. 3–5, 2005. UCRL-PRES-207627.

Rognlien, T. D. (2005). "Understanding of edge plasmas in magnetic fusion energy devices." *Plasma Phys. Control. Fusion* **47**, A283. UCRL-JNRL-207633.

Rognlien, T. D. and M. V. Umansky. (2005). *Tokamak edge transport simulations with an Er model for varying collisionality regimes*. 47th American Physical Society—Division of Plasma Physics Mtg., Denver, CO, Oct. 24–28, 2005. UCRL-ABS-213822.

Rognlien, T. D. et al. (2005). *Results and issues for pedestal turbulence simulation from drift waves and MHD modes*. Intl. Tokamak Physics Activities Pedestal Mtg., Kyoto, Japan, Apr. 18–21, 2005. UCRL-PRES-211552.

Rognlien, T. D. et al. (2005). "Simulation of plasma fluxes to material surfaces with self-consistent edge turbulence and transport for tokamaks." *J. Nucl. Mater.* **337**, 327. UCRL-JC-204447.

Rognlien, T. D., M. V. Umansky, and X. Q. Xu. (2005). *Edge radial electric field generation and dynamics in tokamaks*. 10th Intl. Atomic Energy Agency H-Mode Technical Mtg., St. Petersburg, Russia, Sept. 28–30, 2005. UCRL-PRES-215660.

Umansky, M. V. et al. (2005). *Edge-plasma transport utilizing 3D fluid turbulence simulations of cross-field fluxes*. 2005 Transport Task Force Mtg., Napa, CA, Apr. 6–9, 2005. UCRL-ABS-210464.

Umansky, M. V. et al. (2005). *Self-consistent calculation of turbulence and transport in the tokamak edge*. American Physical Society Annual Mtg. of Division of Plasma Physics, Denver, CO, Oct. 24–28, 2005. UCRL-ABS-216496.

Umansky, M. V., T. D. Rognlien, and X. Q. Xu. (2005). "Simulation of turbulence in the divertor region of tokamak edge plasmas." *J. Nucl. Mater.* **337**, 266. UCRL-JC-207046.

Strain-Rate Scaling of Deformation Mechanisms

James M. McNaney 03-ERD-015

Abstract

Our objective is to determine material-deformation mechanisms in copper at ultrahigh pressures and strain rates. Further, we intend to assess the scalability of the results across a wide range of strain rates to determine if what we learn about deformation mechanisms at very high strain rates is applicable to

other regimes. To accomplish our objectives, we will use controlled, shockless loading along with soft-capture recovery on three experimental facilities: the Omega laser, the LLNL gas gun, and a high-explosives facility. Post-shot characterization will allow us to determine deformation mechanisms. Modeling simulations will guide experimental planning and mechanistic interpretation.

We expect to demonstrate the methodology for determining deformation mechanisms over a wide range of strain rates and ultrahigh pressures. The results from this project will provide basic scientific information in unexplored regions of pressure and strain rates, as well as providing a demonstrable scientific underpinning for the high-pressure strength measurements critical to the Stockpile Stewardship Program. Additional benefits include initiating an effort to investigate solid-state material behavior under extreme conditions, and recruiting young scientists.

Mission Relevance

Deformation mechanisms provide the scientific underpinning of strength measurements at ultrahigh pressures. This project supports LLNL's national-security mission by developing a methodology for both assessing these deformation mechanisms and determining the scalability of high-strain-rate results, which will be critical input for long-range, science-based stockpile stewardship.

FY05 Accomplishments and Results

The project was successfully completed in FY05. We concluded a strain-rate scaling study of deformation in copper using three platforms: laser, high explosive, and gas gun. For each, we conducted isentropic loading experiments, including recovery and post-mortem examination to determine the operative high-pressure, high-strain-rate deformation mechanism. Large scale molecular dynamics of single crystal and polycrystalline material identified operative mechanisms of the deformation. We found major differences in behavior that appear to be caused by the large strain-rate differences among the various platforms. Interestingly, the shock response among the platforms is uniform. The work led us to develop a strain-rate and pressure-dependent model for determining the operative deformation mechanism.

Publications

Bringa, E. M et al. (2005). "Ultrahigh strength in nanocrystalline materials under shock loading." *Science* **309**, 1838. UCRL-JRNL-211450.

Jarmakani, H. N. et al. (2005). *Quasi isentropic compression of copper*. 14th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 5, 2005. UCRL-POST-213507.

McNaney, J. M. et al. (2005). *Near-isentropic, high pressure laser-driven deformation response of copper*. 14th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 5, 2005. UCRL-PRES-213835.

McNaney, J. M. et al. (2004). "High pressure, laser driven deformation of an aluminum alloy." *Met. Trans. A*. **35**, 2625. UCRL-JC-152750.

Determining Phonon Dispersion Curves for Delta-Phase Plutonium–Gallium Alloys

Joe Wong 03-ERD-017

Abstract

The goal of this project is to measure the phonon-dispersion curves (PDCs) of face-centered-cubic (fcc) plutonium–gallium (Pu–Ga) alloys, as well as their temperature and pressure dependencies. Pure Pu exhibits six solid-state phase transitions, with large volume changes, along the path to melting: alpha, beta, gamma, delta, delta´, epsilon, and liquid. The delta phase has desirable mechanical properties. Alloying with Ga expands the delta-phase field from high to below room temperature. To measure PDCs of Pu–Ga alloys, we will use x-ray scattering methods employing high-brightness synchrotron sources to eliminate both the neutron absorption problem of Pu and single-crystal requirements for conventional PDC measurements with neutron inelastic scattering. We will obtain full dispersion curves along the three principal crystallographic directions in the fcc structure of a Pu–Ga alloy, and their dependence on Ga concentration and temperature to elucidate the softening of the T(111) branch. The dispersion curves will provide, for the first time, a bona-fide lattice dynamic data for any Pu-bearing material, and serve as experimental inputs to realistic modeling and theory of correlated 5f-electron systems.

Mission Relevance

This project will provide much-needed basic lattice dynamical data for phase stability and property simulations of Pu materials relevant to LLNL science-based stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we (1) reproduced the T(111) branch bending of the phonon dispersion curve at room temperature, and obtained temperature-dependence data of the L(100) branch, hence C11; (2) obtained both (111) and (110) orientation images from room temperature down to 200 K using a beam tightly focused to 20-µm spot; (3) used grain-size mapping with micro-scanning diffraction to show that optical microscopy overestimates the Pu–Ga grain size by about 30%; and (4) designed, fabricated, and successfully tested online a cryo-chamber for Ga-dependence TDS measurements at the Advanced Photon Source, and obtained both (111) and (110) orientation images from room temperature down to 200 K with 0.1 K stability over 30+ hours.

Publications

Wong, J, et al. (2005). "Crystal dynamics of delta fcc Pu–Ga alloy by high resolution inelastic x-ray scattering." *Phys. Rev. B* **72**, 064115. UCRL-JRNL-207578.

Wong, J, et al. (2004). "Imaging phonons in a fcc Pu–Ga alloy by thermal diffuse x-ray scattering." *Appl. Phys. Lett.* **84**, 3747. UCRL-JC-151455.

Colliding Nanometer Beams

Jeffrey B. Gronberg 03-ERD-044

Abstract

The goal of this project is to develop and demonstrate an alignment and metrology frame that will allow beam position monitors (BPMs) to reach nanometer resolution in the measurement of the transverse position of an electron beam. Future linear colliders will achieve a high event rate by focusing electron beams down to spot sizes of a few nanometers. Stabilization systems to hold the final magnetic optics steady at that level are under development, but definitive tests would require construction of a linear collider. Stabilization system failure would be a showstopper. A stabilization system test using a single beam is possible with nanometer-resolution BPMs. A metrology and alignment frame for a set of BPMs will be designed, built, and operated to demonstrate nanometer resolution.

The project will determine whether existing cavity BPMs can achieve nanometer resolution. This would be a more than order-of-magnitude improvement in current beam diagnostics and would provide sufficient signal sensitivity to monitor bunch tilt, which is expected to have a significant effect on the luminosity of future accelerators. A working nanometer-resolution BPM would allow the stabilization scheme required for colliding beams at a linear collider to be tested, greatly improving our confidence in the chosen linear collider design.

Mission Relevance

The DOE advisory panel on high-energy physics has identified a teraelectronvolt-scale electron-positron linear collider as the next big project in high-energy physics, and LLNL is a charter member of the U.S. research and development consortium for the design of such a machine. This project will allow a potential showstopper to be tested before detailed design and construction and to advance our capability in nanometer metrology, which will support DOE and LLNL's mission in fundamental science and help recruit young talent.

FY05 Accomplishments and Results

Detailed data analysis of the system has shown a system resolution of 20 nanometers. Resolution has been determined to be limited by electronic noise in the cavity BPM readout. The vibrational stability of the alignment frame is below this level. The metrology frame was completed. A paper describing our results has been submitted to the Nanobeams conference.

Publications

Chung, C. et al. (in press). "Performance of a nanometer-resolution BPM system." *Proc. NanoBPM Conference*. UCRL-CONF-215334.

Laser–Matter Interactions with a 527-Nanometer Drive

Siegfried Glenzer 03-ERD-070

Abstract

The primary goal of this research is to develop an understanding of laser-matter interactions with 527-nm light (2ω) for studies—including material strength, radiation transport, and hydrodynamics—of interest to many Laboratory programs. The potential of significantly greater energy delivered onto target at 2ω enables a wide variety of experiments that scale strongly with energy and allow us to probe increasingly higher regimes of pressure, temperature, and strain rate in matter. As part of this project, we are also developing the enabling technology and prototype instrumentation to diagnose a high-fluence laser beam for energy, power, near-field intensity profile, and full-aperture backscatter at 2ω.

As a result of this work, we will resolve important scientific questions relating to laser-plasma interactions, laser-target coupling, preheat and hohlraum filling, and gain a better understanding of the advantages and limitations of 2ω illumination relative to 3ω for high-energy-density (HED) and inertial-confinement fusion (ICF) applications. We will also pave the way for critical stockpile-stewardship-related experiments to be performed in matter in regimes of pressure, temperature, and strain rate much higher than previously accessible.

Mission Relevance

Five key areas within the Stockpile Stewardship Program would significantly benefit from the ability to conduct experiments with a 2ω drive: ignition, material dynamics, experiments using special nuclear materials, nuclear weapons effects testing, and hydrodynamics. This project will also enable cutting-edge research in other LLNL mission areas such as ICF and HED physics.

FY05 Accomplishments and Results

We demonstrated efficient 2ω laser beam propagation and small laser backscatter consistent with ignition with 2ω beams. The newly activated transmitted beam diagnostics have measured up to 80% transmission through a 2-mm-long high-temperature plasma. This value indicates that most of the laser energy will be deposited into the ignition hohlraums during an ignition experiment satisfying the main requirement for 2ω ignition. This value is also significantly larger than the transmission through gasbags, which is 25%. The main interpretation of this finding is the presence of high temperatures in the new hohlraum target so that the stimulated Raman scattering instability is below threshold and laser light is not backscattered.

Proposed Work for FY06

We propose to complete the Omega 2ω laser propagation and scattering experiments by directly measuring electron temperature using 4ω Thomson scattering. The Thomson scattering diagnostics will allow us to infer temperatures with high accuracy and with high spectral and temporal resolution. This measurement will benchmark 2-D radiation-hydrodynamics modeling of this target and will allow scaling the present findings to ignition experiments and verifying that 2ω ignition hohlraums will be successful. We will also carry out a campaign on the Precision Diagnostics Station to assess the performance of optical and beam diagnostic components for producing high-quality 2ω beams.

Publications

Glenzer, S. H. et al. (2005). "Progress in large-scale-length laser–plasma interactions." *Nucl. Fusion* **44**, 185. UCRL-155301.

Mackinnon, A. J. et al. "Implementation of high-energy 4ω Thomson-scattering diagnostic beam on the Omega laser." *Rev. Sci. Instrum.* **75**, 3906. UCRL-203615.

Niemann, C. et al. "Intensity limits for propagation of 0.527 5m laser beams through large-scale-length plasmas for inertial confinement fusion." *Phys. Rev. Lett.* **94**, 08505. UCRL-202697.

Niemann, C. et al. "Transmitted laser beam diagnostic at the Omega laser facility." *Rev. Sci. Instrum.* **75**, 4171. UCRL-203442.

Suter, L. J. et al. "A summary of explorations into the use of green light for high-gain, high-yield experiments on the National Ignition Facility." *Nucl. Fusion* **44**, 140. UCRL-201547.

Characterization and Optimization of High-Energy K-Alpha X-Ray Sources

Richard A. Snavely 03-ERD-072

Abstract

This project will explore the generation, characterization, and optimization of high-energy (20- to 100-keV) K-alpha x-ray backlight sources for application to high-energy petawatt (HEPW) radiography experiments at future large lasers for fusion research. Our goal is to develop the capability to predict and optimize source parameters in HEPW experiments. To this end, we will characterize key K-alpha source parameters as a function of laser and target parameters in experiments at LLNL's Janus-pumped, ultrashort-pulse laser facility and (if possible) at higher-energy, petawatt-scale laser facilities.

By careful characterization of source parameters, we expect to illuminate the fundamental physics responsible for optimization of x-ray yields with respect to these parameters and, by learning more about the physics, to be able to optimize backlight targets for specific applications. In particular, we will measure x-ray source sizes and yields from high-atomic-number targets at up to 100 keV, determine whether and how laser pre-pulses affect source characteristics, and use shifted K-alpha line signatures to determine local heating that might affect electron-beam distributions and therefore source size. We will also investigate physically restricted sources, which could enhance source brightness.

Mission Relevance

The use of x-ray sources as backlights for radiography experiments is an established technique for studying hydrodynamics, equations of state, and other properties of materials under extreme conditions of high temperature and density. Our project will investigate how to most effectively perform experiments at future large lasers for fusion research by using high-energy x-ray backlights to study large-volume, high-atomic-number (Z) materials. Success in this effort is directly relevant to the stockpile-assessment activities in support of LLNL's national security mission.

FY05 Accomplishments and Results

In FY05, we took additional measurements of K-alpha and bremsstrahlung source sizes and K-alpha yields from solid-foil targets as functions of laser intensity, pulse duration, and laser spot size. Importantly, we identified critical physics issues concerning electron transport in the target volume that affect the efficiency of K-alpha emissions. We found that enhanced electron–target coupling from refluxing electrons implies extreme energy densities and high plasma temperatures. The effects of these on K-alpha production is now understood. We also developed new diagnostic capabilities for this regime that will carry on to higher-energy x-ray sources.

Proposed Work for FY06

We will (1) measure the spectrally resolved high-Z K-alpha emissions using high-resolution crystal spectroscopy and determine how laser pre-pulses and angle of incidence affect source characteristics, including yield and source size; (2) investigate the effects physically restricted sources, such as small foils, have on yield and source size; (3) explore fundamental issues regarding laser–electron interactions, electron–target coupling, and photon versus electron ionization cross-sections at high energy densities with a view to x-ray yield optimization in high-Z fluors; (4) continue investigating source size reduction physics with issues regarding target tamping and yields; and (5) develop new high-energy diagnostics.

Publications

Gregori, G. et al. (2005). "Experimental characterization of a strongly coupled solid density plasma generated in a short-pulse laser–target interaction." *Contrib. Plasma Phys.* **45**, 284. UCRL-ABS-210496.

Park, H. S. et al. (2005). "Characteristics of high-energy Ka and bremsstrahlung sources generated by short-pulse petawatt lasers." *Rev. Sci. Inst.* **95**, 4048. UCRL-CONF-203568.

Snavely, R. A. et al. *Advanced plasma diagnostics in the petawatt–fast ignition region*. 8th Intl. Workshop Fast Ignition of Fusion, Tarragona, Spain, June 29, 2005. UCRL-ABS-209876.

Thoebald, W. et al. (in press). "Hot surface ionic line emission and cold K-inner shell emission from petawatt-laser irradiated Cu foil targets." *Phys. Plasma*. UCRL-JRNL-217895.

A Compact Accelerator for Proton Therapy

George J. Caporaso 03-ERD-073

Abstract

High-gradient insulator technology developed at LLNL, coupled with the dielectric wall accelerator (DWA) concept, has produced the potential for a compact, high-voltage, short-pulse accelerator. This accelerator is being developed as a flash x-ray radiography source for stockpile stewardship. Successfully exploiting this technology to build small proton accelerators could revolutionize cancer radiation therapy and make this effective treatment widely available. Proton therapy is superior to x-ray therapy, yet is only practiced in a few locations in the U.S. because of the large size and high cost of these facilities. In addition, this work may lead to compact accelerators for other applications such as high-energy physics, Compton scattering, and free-electron laser (FEL) drivers.

We have three major objectives. The first is to examine the feasibility of a DWA architecture that meets the requirements for proton therapy. This architecture will be verified by simulation, and construction and testing of scalable prototypes. Second, we will develop an understanding of the beam parameters needed for standard and novel proton radiotherapies. Third, we will evaluate the use of this short-pulse accelerator to produce Compton sources and advanced light sources. At the conclusion of this effort, we will know whether the DWA can be applied to proton therapy.

Mission Relevance

The project focuses on using DWA in a regime that has the potential to produce the highest accelerating gradient of any non-plasma-based acceleration scheme. These accelerators could be used for medical treatment, Compton scattering sources, FEL drivers, and high-energy physics, in support of LLNL missions in national security, biotechnology to improve human health, and breakthroughs in fundamental science. Furthermore, it will enhance capabilities in induction linac technology for stockpile stewardship.

FY05 Accomplishments and Results

We met the major project objective to determine feasibility of a compact proton accelerator using DWA technology, and have filed several records of invention related to this research. Specific FY05 accomplishments include: (1) validation of several potential accelerator architectures by first principles, three-dimensional, time-domain electromagnetic simulations; (2) development of a concept for a long-lifetime, compact proton source capable of supplying the required beam current; (3) calculations from source-to-patient showing straightforward beam dynamics; (4) demonstration of silicon carbide photoconductive switching up to an average gradient of 27.5 MV/m; and (5) demonstration of an alternate method of generating high gradient using Mylar Blumleins and an oil gap switch at 28 MV/m in a matched load.

Publications

Nunnally, W. et al. (2005). *High electric field, high current packaging of SiC photo-switches*. IEEE Intl. Pulsed Power Conf., Monterey, CA, June 13–17, 2005. UCRL-CONF-212630.

Rhodes, M. (2005). *Ferrite-free stacked Blumlein pulse generator for compact induction linacs*. IEEE Intl. Pulsed Power Conf., Monterey, CA, June 13–17, 2005. UCRL-CONF-212632.

Rhodes, M. (2005). *UV pre-ionized rail-gap switch for stacked Blumlein pulse generators*. IEEE Intl. Pulsed Power Conf., Monterey, CA, June 13–17, 2005. UCRL-CONF-212639.

Sampayan, S. et al. (2005). *Development of a compact radiography accelerator using dielectric wall accelerator technology*. 2005 Particle Accelerator Conf., Knoxville, TN, May 16–20, 2005. UCRL-CONF-212251.

A High-Efficiency, Grazing-Incidence Pumped X-Ray Laser

James Dunn 03-LW-001

Abstract

Our primary objective is to demonstrate a new x-ray laser pumping geometry where short-wavelength (<20 nm) sources can be operated at high 10-Hz rates, and potentially scaled to 1 kHz. This generates high-average-power tabletop x-ray lasers using small-scale laser drives that complement current third- and future fourth-generation light sources, such as the Linac Coherent Light Source. We will improve laser-pumping efficiency by 10 to 30 times in some cases, and demonstrate a grazing-incidence pumped 10-Hz x-ray laser. Plasma characterization, simulations of laser coupling, x-ray laser production, and propagation are required. The work will be conducted on the existing LLNL Jupiter Facility high-power, short-pulse Callisto and COMET lasers.

The grazing-incidence pumping geometry improves the laser-drive plasma coupling and therefore x-ray laser efficiency. We expect to achieve a high-output, 10-Hz x-ray laser operating at 13 to 20 nm with about 1-ps pulse duration, equivalent to near-synchrotron-level average brightness, but on a tabletop and at low cost. The goal is to perform a demonstration with useable output for applications relevant to LLNL programs, requiring high repetition rate and high output capability of the source. This technique can be extended to many other x-ray laser applications requiring a single-shot, high-output, short wavelength (<4.5 nm) laboratory x-ray laser operating in the water window. The new Titan petawatt-class laser at LLNL would have the capability as a short-wavelength x-ray laser driver.

Mission Relevance

The x-ray probing of ultrafast processes in materials such as actinides is relevant to stockpile stewardship. A tabletop source would be ideal for 13-nm wavelength metrology of extreme ultraviolet lithography optics for microchip development, in support of LLNL's mission in fundamental science and technology. This x-ray laser could operate below 4.5 nm, giving a single-shot capability lasting 100 fs for microscopy and holography of biological cells, in support of bioscience both for our missions in homeland security and human health.

FY05 Accomplishments and Results

A better understanding of x-ray laser output on spatial dependence of the nickel-like ion fraction in the plasma was achieved through study of x-ray laser parameters and simulations. This helps in design of future shorter-wavelength x-ray lasers. Several papers are in preparation. In addition, we collaborated with the Engineering Research Center for Extreme Ultraviolet Science and Technology funded by the National Science Foundation, and co-wrote two papers on high-repetition-rate source development. Further work is ongoing: the design and layout of future experiments using petawatt-class lasers—Titan at LLNL or PHELIX at GSI in Germany—to drive water-window x-ray lasers were studied, with the objective of performing an experiment later in 2006.

Publications

Dunn, J. (2005). U.S. *Activities in the development of plasma-based x-ray lasers*. Plasma-Based X-Ray Lasers Workshop, Prague, Czech Republic, Sept. 1, 2005. UCRL-CONF-216149.

Dunn, J. et al. (2005). *Characterization and applications of laser-driven x-ray lasers*. Atomic Spectroscopy in High Fields Workshop, Piaski, Poland Sept. 6–11 2005. UCRL-CONF-216064.

Dunn, J. et al. (2005). *X-ray laser source development and applications at Lawrence Livermore National Laboratory*. UCRL-PRES-216144.

Dunn J., R. Keenan and V. N. Shlyaptsev. (2005). "Grazing incidence pumping for high efficiency x-ray lasers." *Proc. SPIE* **5919**, 591905-1. UCRL-PROC-216002.

Filevich, J. et al. (2005). "Observation of a multiply ionized plasma with index of refraction greater than one." *Phys. Rev. Lett.* **94**, 035005. UCRL-JRNL-207242.

Keenan, R. et al. (2005). "High-repetition-rate grazing-incidence pumped x-ray laser operating at 18.9 nm." *Phys. Rev. Lett.* **94**, 103901. UCRL-JRNL-204477.

Keenan, R. et al. (2004). *Grazing incidence pumped x-ray lasers at LLNL*. UCRL-BOOK-208545.

Larotonda, M. A. et al. (2004). "Characteristics of a saturated 18.9 nm tabletop laser operating at 5 Hz repetition rate." *IEEE J. Sel. Top. Quantum Electron.* **10**, 1363. UCRL-JRNL-209395.

Luther, B. M. et al. (in press). "High repetition rate collisional soft x-ray lasers based on grazing incidence pumping." *IEEE J. Sel. Top. Quantum Electron.* UCRL-JRNL-217566.

Smith, R. F. et al. (2005). "Plasma conditions for improved energy coupling into the gain region of the Ni-like Pd transient collisional x-ray laser." *Phys. Rev. E* **72**, 036404. UCRL-JRNL-207182.

KINETIC SIMULATION OF BOUNDARY-PLASMA TURBULENT TRANSPORT

Bruce I. Cohen 04-SI-003

Abstract

This project will develop a kinetic code to model the tokamak plasma boundary region, allowing first-principles predictions of the edge transport barrier, the greatest source of uncertainty in projecting the fusion power output of next-generation tokamak reactors like the International Thermonuclear Experimental Reactor (ITER). The code will include a nonlinear kinetic description of the edge plasma in realistic three-dimensional (3-D) magnetic geometry. Molecular dynamics simulations will provide improved models of the plasma hydrocarbon contamination source. The code will use efficient numerical methods to solve the edge-plasma coupled equations in a 5-D phase space on massively parallel computers, and provide the software structure to support continuing physics development by multi-institution collaborations.

This project will deliver the world's first ab initio integrated model of the boundary region of fusion plasmas, which recent work suggests may provide an accurate method of predicting and optimizing the fusion power of ITER. A full-fidelity ITER calculation will be within the capabilities of our emerging petaclass computers, such as BlueGene. A code architecture compliant with the goals of the DOE Fusion Simulation Project (FSP) will facilitate continued development of the physics by multi-institution collaborations, allowing the models developed under this project to be extended into a simulation of the full tokamak as part of the FSP.

Mission Relevance

This project supports the energy-security mission of the Laboratory by advancing the physics required for magnetic fusion energy through the use of high-performance computing, advanced software technology, and fundamental simulation of the properties of materials. Fusion has been a core mission for LLNL since its founding. This project is timely because the U.S. has rejoined ITER negotiations, and a comprehensive modeling capability is viewed as necessary for effective use of ITER.

FY05 Accomplishments and Results

In FY05 we (1) developed and tested an efficient and scalable multigrid solver for field equations in four dimensions; (2) met the key FY05 milestone by demonstrating a 4-D kinetic code with field solver; (3) completed and tested a tokamak magnetic geometry module; (4) merged code development into a framework, added radial drifts and parallel acceleration, put the collision operator into the framework, tested it, and added a calculation of velocity moments needed by other modules; (5) began an initial physics calculation of neoclassical transport in a tokamak; and (6) developed realistic models for carbon and hydrogen targets, began to compute a table of sputtering versus hydrogen energy, began testing chemical-rate methods (ChemKin code), and did the initial sensitivity study of the edge plasma to carbon sputtering using the UEDGE code.

Proposed Work for FY06

We will extend the code to five dimensions (3-D space and 2-D velocity) and enable kinetic edge plasma simulation as well as couple self-consistent plasma-to-plasma-wall interactions. An initial study will be conducted of performance and scaling of 4-D and 5-D kinetic code on parallel computers. We will compare OOPID carbon models with iterative coupling between OOPID and UEDGE codes for impurity density, provide a fluid impurity model for kinetic code and benchmark it with experiments, and assess the need for kinetic extension. A 3-D kinetic Monte Carlo code will be developed for a detailed model of the graphite divertor surface, and used with ChemKin and OOPID to model impurity reflux and study surface roughness and sputtering interaction. We will calculate the edge bootstrap current and validate against experiments and perform the first self-consistent simulations of edge pedestal formation.

Publications

Marian, J. et al. (2005). "Simulation of carbon production from material surfaces in fusion devices." *Bull. Am. Phys. Soc.* **50**(86) CP1, 56. UCRL-PRES-217014.

Nevins, W. M. et al. (2004)."Kinetic simulation of boundary plasma transport." *Bull. Am. Phys. Soc.* **49**, 166. UCRL-PRES-211784.

Qin, H. et al. (2005). "General gyrokinetic equations for edge plasmas." *Proc. 10th Intl. Workshop Plasma Edge Theory in Fusion Devices*. UCRL-PROC-215824.

Xiong, Z. et al. (2005). "Simulation of plasma transport in a toroidal annulus with TEMPEST." *Bull. Am. Phys. Soc.* **50**(86) CP1, 55. UCRL-ABS-322586.

Xiong, Z. et al. (2005). *Towards an efficient field solve for edge-plasma simulations*. 2005 Intl. Sherwood Theory Conf., Apr. 11–13, 2005, Stateline, NV. UCRL-ABS-210030.

Xu, X. (2005). *Continuum gyro-kinetic simulation*. 4th Workshop Nonlinear Plasma Sciences and Intl. School on Plasma Turbulence and Transport, Hangzhou, China, Aug. 30–Sept. 3, 2005. UCRL-PRES-215222.

Xu, X. Q. et al. (2005). "Four-dimensional continuum gyrokinetic code: Neoclassical simulation of fusion edge plasmas." *Bull. Am. Phys. Soc.* **50**(85) CP1, 54. UCRL-ABS-322858.

Xu, X. Q. et al. (2005). *Status of continuum edge gyrokinetic code physics development*. 2005 Intl. Sherwood Theory Conf., Apr. 11–13, 2005, Stateline, NV. UCRL-PROC-212647.

Xu, X. Q. et al. (2004). "Algorithm development for gyrokinetic simulations of boundary plasmas." *Bull. Am. Phys. Soc.* **49**, 166. UCRL-PRES-211815.

Xu, X. Q. and W. M. Nevins. (2004). *Development of a gyro-kinetic Vlasov simulation code for boundary plasmas*. 2004 Intl. Sherwood Theory Conf., Apr. 26–28, 2004, Missoula, MT. UCRL-PRES-211817.

Zepeda-Ruiz, L. A. et al. (2005). *Simulations of carbon sputtering and plasma-sheath chemistry interactions at fusion reactor divertor plates.* 2005 Intl. Sherwood Theory Conf., Apr. 11–13, 2005, Stateline, NV. UCRL-ABS-215980.

Development of Absolute Spectroscopic Diagnostics for Nonlocal-Thermodynamic-Equilibrium Plasmas

Christina A. Back 04-ERD-019

Abstract

The goal of this project is to develop a quantitative understanding of nonlocal-thermodynamic-equilibrium (NLTE) plasmas. We are conducting experiments using low-density aerogel foams to measure the temporal evolution of the K- and L-shell emission of highly ionized species. The data will be sufficiently accurate to create benchmarks that can be used to refine and potentially validate the Laboratory's NLTE codes, such as CRETIN. In particular, we will use experiments to address the recombination processes in the density regime of 1019 to 1022 cm^{-3} of mid-atomic-number elements. The experiments will provide temporally resolved, absolute measurements of the emission over a spectral range of 200 to 1000 eV. Data will be compared with calculations at each stage of the project.

The data obtained will resolve long-standing discrepancies in the study of laser-produced plasmas, and enable better modeling of physical processes relevant to stockpile stewardship. These experiments investigate the NLTE phenomena that impact high-power laser experiments. Deliverables will include (1) absolute spectra from an NLTE plasma; (2) temporal correlation of K- and L-shell spectra; and (3) a new capability to perform absolute emission measurements in the soft x-ray regime, which provide an important new constraint for simulations.

Mission Relevance

Accurate understanding of the mechanisms of x-ray production is an essential component of energy balance and x-ray transport in stockpile stewardship applications, and for design and production of high-energy-laser x-ray sources. The physical conditions present in many important stockpile stewardship applications and virtually all radiation-dominated laboratory experiments are also found in NLTE plasmas, which is relevant to weapons effects.

FY05 Accomplishments and Results

Comparisons with calculations showed that the absolute measurements are within a factor of three of the calculations. This excellent success for the first attempt in the validation of L-shell kinetics gives us confidence that the atomic energy levels and rates are well represented. Relative features do not change significantly during the last nanosecond of the 4-ns laser heating pulse, showing that the experiments successfully produce a steady-state plasma. After the end of the laser pulse, the plasma recombines and the ionization slowly shifts to a longer wavelength, and thus a lower ionization state. The next set of experiments have been designed to produce much more uniform plasmas and provide much better spectral resolution and Thomson-scattering temperature measurements.

Proposed Work for FY06

One additional set of experiments is planned for FY06 to experimentally determine the plasma temperature, correlated with the spectrum, and investigate plasma electron-density gradients through interferometry. Analysis of the experimental results will include radiation hydrodynamics simulations to investigate effects from nonuniform laser heating and hydrodynamic motion, NLTE atomic kinetics simulations of the ionization (and energy) balance, and spectral modeling to simulate the observed time-dependent spectral features. Simulation results will be compared to data in each of these areas, with the goal of identifying discrepancies between the simulations and data, and understanding sensitivities to key aspects of the simulations.

Publications

Back, C. A. et al. (in press). "Absolute, time-resolved emission of non-LTE L-shell spectra from Ti-doped aerogels." *J. Quant. Spectrosc. Radiat. Transf.* UCRL-JRNL-211261.

Chung, H.-K. et al. (in press). "FLYCHK: simple but generalized population kinetics and spectral model." *High Energ. Densit. Phys.* UCRL-JRNL-213347.

Constantin, C. et al. (2005). "Supersonic heat-wave propagation in laser-produced underdense plasmas." *Phys. Plasmas* **12**, 063104. UCRL-JRNL-203465.

Scott, H. A. (in press). "Non-LTE radiation transport in high radiation plasmas." *High Energ. Densit. Phys.* UCRL-JRNL-209013.

Electronic Transitions and Phonons in f-Band Metals at High Pressures

Choong-Shik Yoo 04-ERD-020

Abstract

Unusual phase transitions driven by electron correlation effects occur in many f-band metals. The exact nature of these transitions has not been well understood, including the short-range correlation effects themselves, their relation to long-range crystalline order, the role of magnetic moments and order, and the critical behavior, among other issues. Many of these questions represent forefront physics challenges central to stockpile materials. In this study, we propose to investigate electronic phase transitions occurring in solid and liquid f-band metals at high pressures and temperatures using the nation's brightest third-generation synchrotron x-ray source, the Advanced Photon Source. This work will be complemented by new theoretical activities.

The success of the proposed study will result in significant advances of our current understanding in two forefront scientific areas of condensed matter physics: electron correlations and phonons of f-band metals. The study will deliver fundamental data to challenge and validate theoretical understanding of f-electron calculation and interatomic forces and will also develop and apply cutting-edge experimental and theoretical tools.

Mission Relevance

This LDRD should result in better understanding of forefront physics challenges central to stockpile actinides such as electron-correlation effects, phonons, and liquid equations of state for stockpile stewardship. These studies on strongly correlated systems additionally provide opportunities to develop collaborations with leading scientists in the field and attract talented young physicists to the Laboratory.

FY05 Accomplishments and Results

In FY05 we (1) determined volume collapse transitions for the rare-earth metals praseodynium (Pr), neodynium (Nd), europium (Eu), and gadolinium (Gd) and for the transition metal compounds europium nitride (EuN) and manganese oxide (MnO), all to 100 GPa and above, by using in situ angle-resolved x-ray diffraction; (2) obtained, for the first time, low-energy f-electronic states of Gd up to 110 GPa using resonant inelastic x-ray scattering (RIXS); (3) determined magnetic moments in MnO up to 110 GPa using high-resolution x-ray emission spectroscopy (these results were published in *Phys. Rev. Lett.*); and (4) studied the feasibility of determining the phonon density of state of dysprosium (Dy) at 8 and 13 GPa using nuclear RIXS (NRIXS) in a diamond anvil cell.

Proposed Work for FY06

In FY06 we propose to (1) determine the local magnetic moments of f-electrons in light rare-earth metals (Pr, Nd, Eu) across the volume-collapse transitions using high-resolution x-ray emission spectroscopy; (2) exploit the mixed-valence electronic states of Pr up to 100 GPa using RIXS; and (3) determine the phonon consequences in the electronic-volume collapse transition in Dy at 70 GPa using NRIXS.

Publications

Iota, V. et al. (2005). *Electronic structure and magnetism in highly compressed 3D-transition metals*. UCRL-JRNL-215558.

Kim, E. et al. (in press). “Martensitic fcc-to-hcp transformations in solid xenon under pressure: a first-principles study.” *Phys. Rev. Lett.* UCRL-JRNL-216056.

Maddox, B. et al. (2005). *Second-order rutile-to-$CaCl_2$ phase transition in half-metallic CrO_2 at high pressures*. UCRL-JRNL-215314.

Maddox, B., et. al. (2005). “Resonance inelastic x-ray scattering across the volume collapse transition in Gd at high pressure.” 2005 Materials Research Society Fall Mtg., Boston, MA, Nov. 28–Dec. 2, 2005. UCRL-ABS-213159.

Maddox, B. R. et al. (2005). *Kondo-like 4f delocalization in Gd at high pressures*. UCRL-JRNL-217382.

Yoo, C. S. et. al. (2005). "First-order isostructural Mott transition in highly compressed MnO." *Phys. Rev. Lett.* **94**, 115502. UCRL-JRNL-204811.

Short-Pulse Laser Absorption and Energy Partition at Relativistic Laser Intensities

Ronnie Shepherd 04-ERD-023

Abstract

We are conducting detailed and integrated experiments for the first comprehensive measurements of laser absorption and energy partitioning in solid targets heated with an ultrashort laser pulse focused to relativistic laser intensities (>10^{17} W/cm^2). The measurements will include determining the density scale length, a critical parameter in understanding the contribution of absorption processes dominant at low laser intensities. In parallel, we propose to benchmark LSP, a new collisional particle-in-cell (PIC) model that is planned as the primary modeling tool for future petawatt and high-energy-laser experiments. The resulting information will be used to produce high-brightness, short-pulse-laser-produced broadband x-ray sources for these experiments.

We expect to (1) measure scale length versus laser intensity, scale length versus absorption, relative hot electron and thermal electron production, magnetic field strength versus laser intensity, and particle energy distribution versus laser intensity; (2) perform energy-scaling absorption experiments; and (3) interface the measurements to codes to improve the fundamental physics used in high-intensity laser–matter interaction models. If successful, we will develop a predictive capability usable for experimental design and data analysis on petawatt lasers.

Mission Relevance

This work will provide insight in an area relevant to weapons physics (e.g., proton generation and x-ray back illuminators) and experiments at future large fusion-class lasers (e.g., hot electrons for fast-ignitor schemes) by acquiring high-quality physics data. This research will also help recruit scientists in high-energy-density physics, in support of stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we succeeded in measuring the absorption from 10^{17} to 10^{20} W/cm^2, a major accomplishment in the high-intensity laser–matter interaction field. During this experiment, we managed to field several additional diagnostics to provide information about the temperature and direction of the nonthermal electrons and the source of energy loss because of instabilities driven at the critical surface. We also measured the energy partition between thermal and non-thermal electrons from 10^{17} to 10^{19} W/cm^2. We made the first-ever time-dependent measurement of energy transfer from non-thermal to thermal electrons in a high-intensity-laser heated solid.

Proposed Work for FY06

For FY06, we plan to extend our energy partition measurement from 10^{19} to 10^{21} W/cm^2. Having observed an unusual increase in the coupling to thermal electrons at 10^{19} W/cm2, we must determine if this trend continues at higher energies. We will also extend our laser absorption measurement to s-polarized laser light, which is important for understanding the physics of the high absorption observed in p-polarized light at high laser intensities. If time permits, we will perform experiments to determine the magnetic field strength and the energy and time dependence of the protons generated in the interaction. The magnetic field and proton energy and time measurements could have a significant impact on transport and short-pulse-laser-generated proton applications.

Publications

Chen, H. et al. (2005). *Observation of energy partition shift between thermal and non-thermal electrons at relativistic laser intensities*. 32nd IEEE Intl. Conf. Plasma Science, Monterey, CA, June 18–23, 2005. UCRL-ABS-211312.

Shepherd, R. et al. (2005). *Observation of indirect, time-dependent heating of solid targets from ultra-intense, ultrashort pulse laser light*. 32nd IEEE Intl. Conf. Plasma Science, Monterey, CA, June 18–23, 2005. UCRL-ABS-210902.

Shepherd, R. et al. (2005). *Short pulse laser absorption and solid target heating at relativistic laser intensities*. LLNL Science Day 2005, Livermore, CA, May 23, 2005. UCRL-POST-211632.

The Creation of a Neutron Star Atmosphere

Richard I. Klein 04-ERD-028

Abstract

Extreme conditions of density and temperature that are relevant to stockpile stewardship are similar to those of low-altitude atmospheres of magnetized neutron stars and accretion disks around black holes. This project will assess the feasibility for experiments on future petawatt lasers to reproduce aspects of these astrophysical environments. It will, furthermore, assess the possibility of multidimensional codes to successfully guide experiments to produce and detect photon-bubble instabilities observed by x-ray satellite and predicted by LLNL. We will determine the scaling from the astrophysical to the laboratory environment and test key components of the design on existing lasers; perform simulations to estimate temperatures, radiation fields, and magnetic fields; and perform experiments on existing ultra-intense lasers.

The experimental design developed in this project will advance the study of extreme physics generated by strongly radiative flows and laser–plasma interactions for the laboratory study of both distinct astrophysical phenomena and the physics of extreme conditions relevant to stockpile stewardship. Establishing a laboratory capability to probe the physics of accreting, magnetized compact objects will enhance LLNL’s international reputation in high-energy-density (HED) physics.

Mission Relevance

This project has direct relevance to understanding the extreme conditions of HED, ultrashort-pulse-laser–matter interactions that are relevant to LLNL's stockpile stewardship mission and will be a driver for advanced scientific applications at future petawatt lasers where Laboratory personnel will carry out their work. The world-class science that this project represents will improve recruiting and forge links with the U.S. astrophysical community.

FY05 Accomplishments and Result

In FY05 we (1) designed a set of experiments to conduct at the Rutherford Appleton Laboratory Vulcan petawatt laser to assess the hottest thermal and radiation temperatures possible in petawatt laser–solid density interactions; (2) conducted an extensive set of 10 shots on Vulcan using the long-pulse (10-ps) and short-pulse (0.4-ps) mode; (3) obtained high-quality data with seven diagnostic instruments; (4) showed the thermal temperature to be several hundred electronvolts; (5) established a unique computational capability with radiation-hydrodynamic, particle-in-cell (PIC), and the LSP atomic physics codes to interpret results and optimize designs for maximum temperatures; (6) began extreme ultraviolet polarimetry to measure high magnetic field strengths at the JanUSP laser; and (7) began scaling studies.

Proposed Work for FY06

In FY06 we will (1) complete the PIC–LSP atomic physics analysis of our data to establish the extreme temperatures possible in petawatt laser–solid interactions; (2) conduct experiments with proton deflectometry and optical Faraday rotation using JanUSP to measure the B fields of such interactions; (3) finish our analytical analysis to establish the scaling parameters from the astrophysical to the laboratory petawatt environment and the phase space of those parameters that give rise to the astrophysical phenomena; (4) assess whether existing laboratory platforms can achieve the scaled conditions required to duplicate the astrophysical phenomena; and (5) assess whether actual petawatt lasers can achieve such conditions.

Publications

Gregori, G. et al. (2005). "Experimental characterization of a strongly coupled solid density plasma generated in a short-pulse laser target interaction." *Contrib. Plasma Phys.* **45**, 284. UCRL-JRNL-211077.

Moon, S. J. et al. (2005). "A neutron star atmosphere in the laboratory with petawatt lasers." *Astrophys. Space Sci.* **298**, 293. UCRL-JRNL-211863.

New Generation X-Ray Optics: Focusing Hard X Rays

Regina Soufli 04-ERD-032

Abstract

We propose to design, build, and deploy a 30- to 70-keV reflective system for dynamic characterization of shocked materials at the single-grain scale. The first system we are building will collect the emergent beam from the Picosecond Laser Electron Interaction for Dynamic Evaluation of Structures (PLEIADES) x-ray source, then monochromatize and refocus it on the target to increase the power by two orders of magnitude. Using hard x rays, the focusing system will significantly increase the flux on target and will provide a smaller spot size than conventional methods. The optic will be constructed using a combination of technologies that will be integrated into a single system for the first time, thus providing a new x-ray diagnostic capability.

Studying material properties at the single-grain scale will require a new x-ray diagnostic capability. The homogeneity provided by such a small sample will lead to a dramatic increase in understanding dynamical processes in matter. Beam monochromatization by our reflective element will eliminate background and thus significantly improve the qualitative and quantitative content of dynamic diffraction results. By increasing the power and resolution of the currently available sources and techniques by orders of magnitude, this new diagnostic will enable dynamical studies of homogeneous material samples and propagating shocks, and detailed studies of physical properties of micrometer-scale objects.

Mission Relevance

The demonstration of this novel technological capability will have immediate impact on x-ray diagnostics for target and plasma characterization at future large fusion-class lasers, in support of the stockpile stewardship mission. The new capability will also find application for basic science missions at other next-generation lasers and light sources (e.g., the Linac Coherent Light Source), in major NASA programs [Nuclear Spectroscopic Telescope Array (NuSTAR) and Constellation-X], and for medical applications to improve human health.

FY05 Accomplishments and Results

In FY05 we designed and constructed a 29.2-keV reflective optic composed of segmented glass substrates coated with multilayers of graded-period thickness and developed, fabricated, and tested prototype optics from novel plastic substrate technologies pioneered at LLNL. With these new structures, we designed a focusing optic and fielded it on the CERN Axion Solar Telescope (CAST), a particle astrophysics experiment that is searching for a leading candidate for dark matter. As a result of our work, we joined the international CAST collaboration as full members. We also designed and fabricated multilayer dispersive elements to be used in tandem with reflective optics to further enhance the resolving power of our optical systems for future experiments on shocked materials.

Proposed Work for FY06

In FY06 we plan to construct and install additional optics for the CAST dark-matter experiment. We will also develop a systems-level model that combines the astrophysics of deep surveys of active galactic nuclei with predictions of the assembly error budget and optics performance to predict the scientific performance of NuSTAR. This will allow the development of deconvolution techniques to improve the science and will guide possible enhancements of the assembly process, with the goal of markedly increasing the science return of NuSTAR. We will also continue the design and fabrication of multilayer dispersive structures as resolution-enhancing elements in high-energy experiments. The performance of these elements will be demonstrated through testing at synchrotron facilities.

Publications

Graessle, D. E. et al. (2004). "Iridium optical constants from synchrotron reflectance measurements over 0.05 to 12 keV x-ray energies." *Proc. SPIE* **5538**, 72. UCRL-PROC-206831.

Pivovaroff, M. J. et al. (2005). "Progress of focusing x-ray and gamma-ray optics for small animal imaging." *Proc. SPIE* **5923**, 65. UCRL-PROC-214412.

Soufli, R. et al. (2005). *Optical profilometry and atomic force microscopy characterization of novel EUV and x-ray optical substrates*. SPIE Optics and Photonics Mtg., San Diego, CA, Jul. 31–Aug. 4, 2005. UCRL-ABS-205091.

Soufli, R. et al. (2005). *Polymer smoothing processes for EUV and x-ray optical substrates*. 2nd International Symp. Technologies and Applications of Photoelectron, Tsukuba, Japan, Feb. 1–3, 2005. UCRL-ABS-208399.

Soufli, R. et al. (2005). "Substrate smoothing for high-temperature condenser operation in EUVL source environments." *Proc. SPIE* **5751**, 140. UCRL-PROC-212377.

Nanomechanics: Strength and Structure for Nanotechnology

Robert E. Rudd 04-ERD-043

Abstract

In this computational study, we are developing the fundamental principles of nanomechanics, which concerns the mechanical behavior of nanoscale structures, by advancing the theoretical understanding of novel mechanical phenomena at the nanoscale, and by developing and validating computational methodologies suitable for nanomechanical systems. Nanomechanical theory and the computational tools we are developing are important to design nanoscale systems. We focus on mechanical nanoscale processes driven by surface effects and how they lead to novel physical laws. We are investigating two

kinds of systems: (1) nano-electromechanical systems (NEMS) to study size-dependent stiffness and strength effects and (2) epitaxial nanostructures, which exemplify nanomechanical self-assembly.

We expect to develop a new theory of stiffness and strength of nanostructures and a deeper understanding of how structures are produced in nanomechanically driven self-assembly. This work has extensive implications for the mechanical response of nanoscale structures in NEMS, foams, and nanotechnology and biotechnology. In particular, calculation of the stiffness of NEMS using classical and ab initio quantum techniques will enable quantification of nanoscale changes to the laws of mechanics for the oscillating beams found in NEMS. Development of thermodynamic and atomistic kinetic models of epitaxial island formation will provide a more powerful description of this self-assembly process.

Mission Relevance

The scientific understanding and computational tools developed have potential application to the control of interface features in fusion-class laser capsules, development of next-generation biothreat detectors for homeland-security applications, and mechanical characterization of protein–ligand binding. Development of high-quality NEMS structures is of interest for defense applications as well as for advanced scientific devices. Applications associated with dislocation nucleation are relevant to the Advanced Simulation and Computing Program, which supports stockpile stewardship. In addition, the fundamental principles of nanomechanics discovered in this project will contribute to the Laboratory's basic science mission.

FY05 Accomplishments and Results

In FY05, we invented and used new atomistic (empirical and ab initio) techniques to calculate elastic moduli of nanostructures at finite temperature, used the new dislocation dynamics code to calculate the behavior of dislocations in nanoplates, and implemented new algorithms in the kinetic Monte Carlo (KMC) code. Milestones in each of these areas were met, including significant advances in the theory of elasticity of nanoscale beams and in the computer code for nanoscale plasticity. In addition, we made advances in the nanomechanical theories of force spectroscopy and cantilever-based sensing. We submitted several articles to peer-reviewed journals, presented research findings at invited talks at conferences and universities, and contributed to a patent application.

Proposed Work for FY06

In FY06, we will (1) simulate novel dynamical behavior of NEMS through molecular dynamics (MD) and concurrent finite-element method and MD studies of dissipation in NEMS, and conduct an MD study of plasticity of metallic ligaments; (2) conduct the first discrete dislocation-dynamics simulation of metallic nanorods to demonstrate a new capability for robust treatment of surface image stresses in complex geometries, including the first dynamic calculation of geometry-dependent size effects in nanosystems; and (3) and if funding permits, implement a new MD coupling in the KMC code for general strain relaxation to simulate the growth of semiconductor epitaxial quantum dots. We expect to submit several more journal articles based on this research.

Publications

Ratto, T. V. et al. (in press). “Nonlinearly additive forces in multivalent ligand binding to a single protein revealed with force spectroscopy.” *Langmuir*. UCRL-JRNL-214218.

Rudd, R. E. (2005). *Connecting atomistic and continuum level modeling*. Workshop Multiscale Modeling in Solids, Montreal, Canada, Apr. 28–30, 2005. UCRL-PRES-211807.

Rudd, R. E. (2005). *Lanczos and recursion techniques for multiscale modeling*. Bonding and Structure—Theory and Applications, Oxford, UK, July 7–9, 2005. UCRL-PRES-213992.

Rudd, R. E. (2005). *Multiscale elastic relaxation in kinetic Monte Carlo via a Lanczos technique: Nanostructure evolution*. Nanotech 2005, Anaheim, CA, May 8–12, 2005. UCRL-PRES-212171.

Rudd, R. E. (2004). *Multiscale modeling of elasticity and dissipation in NEMS resonators*. 2nd Intl. Conf. Multiscale Materials Modeling, Los Angeles, CA, Oct. 11–15, 2004. UCRL-PRES-207281.

Rudd, R. E. and J. Q. Broughton. (2005). “Coarse-grained molecular dynamics: Nonlinear finite elements and finite temperature.” *Phys. Rev. B* **72**, 144104. UCRL-JRNL-212644.

Tang, M. (2005). *Direct dislocation dynamics simulation of annihilation radius in thin films*. Materials Research Society Spring Mtg., San Francisco, CA, Mar. 28–Apr. 1, 2005. UCRL-PRES-203523.

High-Average-Power, High-Energy, Short-Pulse Fiber Laser System

Jay W. Dawson 04-ERD-048

Abstract

Our objective is to develop the technology for high-energy (>10 mJ), high-average-power (>300 W) fiber laser systems. The primary limits to scaling the output energy in a fiber laser are nonlinear interactions and beam quality for larger core sizes. To increase the output energy of a fiber amplifier, we are developing a novel waveguide design that distributes the optical intensity of the light more evenly across the core. Consequently, smaller cores can produce pulses with higher output peak powers.

We will develop the key technology components for a high-energy, high-average-power, short-pulse fiber laser system to scale the pulse energy and average output power of short-pulse fiber laser systems by an order of magnitude beyond previously reported results. We will also demonstrate a prototype system. This should solve many longstanding research hurdles to make the technology suitable for applications in materials processing, high-resolution x-ray imaging, and short-pulse front ends for large-scale, high-energy laser systems. We expect this research to lead to a number of spin-offs.

Mission Relevance

High-average-power, high-energy, short-pulse fiber laser systems that are easy to use, reliable, and efficient are relevant to (1) high-energy petawatt laser front ends for future large laser systems; (2) cutting, drilling, and other materials-processing applications; and (3) small-spot-size, high-flux x-ray sources. All three of these areas support DOE and LLNL's national-security mission, specifically in stockpile stewardship.

FY05 Accomplishments and Results

In FY05 we (1) completed construction of a 10-W version of the laser system, including the final amplifier; (2) obtained encouraging system measurements showing good stability and excellent recompression of a chirped pulse, including pulse widths of less than 1 ps with pulse energies of 0.14 mJ; (3) had our linear frequency modulation (LFM) fiber results independently verified by a commercial company; (4) finished the final version of the LFM fiber and began testing; (5) began work on a compressor based on a high-average-power grating; and (6) acquired additional large-mode-area fibers for testing as final amplifiers in the system.

Proposed Work for FY06

In FY06, we plan to complete a final assessment of the maximum pulse energy achievable from our laser design. First, we will complete the 100-W version of the laser system. Then, we will use the system to generate high-brightness x rays for imaging applications. We will also collaborate with the University of Southampton to improve the stature of overall fiber lasers.

Development and Application of a Predictive Computational Tool for Short-Pulse, High-Intensity Target Interactions

Max Tabak 04-ERD-054

Abstract

The goal of this theory-and-computation effort is to produce and validate a computational tool that can model a high-density plasma driven by high-intensity laser light. Integral components of this model will be coupling between laser light and a dense plasma, production and transport of relativistic electrons, self-consistent production and transport of large-scale electric and magnetic fields, realistic equations of state (EOS), electrical and thermal transport properties of thermal plasmas, and coupling to a hydrodynamic (burn) code. This tool will be used to model past and ongoing experiments driven by high-intensity lasers to provide optimized designs for applications such as fast ignition.

If the project is successful, we will be able to model experiments driven with ultra-intense lasers, from laser–plasma interaction to the heating of dense plasmas with intense beams of particles. This will enable the design and optimization of various applications in high-energy-density physics and fusion energy. We will also design experiments for future large laser systems to obtain EOS and opacity data of relevance to stockpile stewardship; improving the efficiency of radiography techniques; and improve fast-ignition target designs, reducing the short-pulse laser energy necessary for ignition.

Mission Relevance

Powerful short-pulse, high-intensity lasers are expected to enable a number of applications that are important to defense and fusion energy but difficult, if not impossible, to accomplish by other means. By providing theory and computational support for an integrated high-intensity, short-pulse laser effort, including laser construction and experiment, this project support LLNL's missions in national and energy security.

FY05 Accomplishments and Results

In FY05 we (1) linked the output of the explicit particle-in-cell code Z3 to the electromagnetic code LSP, producing beam spreading in qualitative agreement with experiment; (2) began a theoretical effort to calculate theoretical growth rates for the coupled Weibel filamentation–two-stream instabilities (our models of heating of cone-wire systems agreed with experiment quantitatively); (3) modeled a fast-ignition implosion for the Omega Extended Performance (EP) laser in a hydrocode, coupled the data to LSP, and calculated the fuel heating; (4) designed targets that couple more than 50% of incident electron energy to a proton beam; (5) calculated relativistic electron stopping power and scattering in a dense plasma whose dielectric function is given in the random phase approximation; and (6) began comparison of LSP simulations with the two-stream instability.

Proposed Work for FY06

We will (1) use the hydrocode-linked LSP to optimize fast ignition experiments at Omega EP; (2) create a catalog of laser–plasma interactions, varying plasma scale height, laser intensity, and angle of incidence for realistic beams and improve the linkage between Z3 and LSP; (3) compare general models of filament growth in collisional and collisionless plasmas with LSP simulations and define the saturated state of these instabilities; and (4) install the transport models into LSP.

Publications

Atzeni, S. and M. Tabak. (in press). "Overview of ignition conditions and gain curves for fast ignition." *Plasma Phys. Control. Fusion*. UCRL-JRNL-213654.

Cottrill, L. A. et al. (2005). *Electron beam-plasma interaction physics*. 47th Ann. Mtg. American Physical Society Division of Plasma Physics, Denver, CO, Oct. 25, 2005. UCRL-PRES-214745.

Cottrill, L. A. et al. (2005). *Optimization of Ka emission yields for short-pulse high-intensity laser–solid interactions*. 8th Intl. Workshop Fast Ignition of Fusion Targets, June 27–July 1, 2005, Tarragona, Spain. UCRL-ABS-205335.

Cottrill, L. A. et al. (2005). *Simulations of relativistic electron beam instabilities for applications to fast ignition*. 47th Ann. Mtg. American Physical Society Division of Plasma Physics, Denver, CO, Oct. 25, 2005. UCRL-POST-216238.

Hatchett, S. et al. (2005). *Hydrodynamics of conically-guided fast ignition targets*. 8th Intl. Workshop Fast Ignition of Fusion Targets, June 27–July 1, 2005, Tarragona, Spain. UCRL-ABS-209939.

King, J. A. et al. (2005). “Ti K alpha radiography of Cu-doped plastic microshell implosions via spherically bent crystal imaging. *Appl. Phys. Lett.* **86**, 191501. UCRL-JRNL-210564.

King, J. A. et al. (2005). “Characterization of a picosecond-laser-generated 4.5-keV Ti K-alpha source for pulsed radiography.” *Rev. Sci. Instrum.* **76**, 76102. UCRL-JRNL-210865.

Logan, B. G., D. A. Callahan, and M. Tabak. (in press). “Assessment of potential for ion driven fast ignition.” *Fusion Sci. Tech.* UCRL-JRNL-208443.

Mackinnon, A. J. et al. (2005). *Integrated fast ignition programs*. 8th Intl. Workshop Fast Ignition of Fusion Targets, June 27–July 1, 2005, Tarragona, Spain. UCRL-ABS-210079.

Tabak, M. (2005). *Capsule optimization techniques for fast ignition*. 47th Ann. Mtg. American Physical Society Division of Plasma Physics, Denver, CO, Oct. 25, 2005. UCRL-ABS-210652.

Tabak, M. (2005). *Capsule optimization techniques for fast ignition*. 4th Intl. Conf. Intl. Fusion Sciences and Applications, Biarritz, France, Sept. 5–9, 2005. UCRL-POST-214864.

Tabak, M. (2005). *Introduction to fast ignition*. UCRL-PRES-213389.

Tabak, M. (2005). *On the path to inertial fusion energy*. 4th Intl. Conf. Intl. Fusion Sciences and Applications, Biarritz, France, Sept. 5–9, 2005. UCRL-PRES-214860.

Tabak, M. and D. A. Callahan. (2005). *Fast ignition driven by heavy ion beams*. UCRL-PRES-214669.

Tabak, M. and D. A. Callahan. (2005). “Models of gain curves for fast ignition.” *Nucl. Instrum. Methods Phys. Res.* **A544**, 48. UCRL-CONF-204393.

Tabak, M. et al. (in press). “Fast ignition: overview and background.” *Fusion Sci. Tech.* UCRL-JRNL-213396.

Tabak, M. et al. (2005). *Campaign: understand and control high-energy-density plasmas*. UCRL-PROP-206335.

Tabak, M., R. Stephens, and K. A. Tanaka. (2004). "Review of progress in fast ignition." *Phys. Plasmas* **12**, 57305. UCRL-CONF-207839.

Tabak, M., R. Stephens, and K. A. Tanaka. (2004). *Review of progress in fast ignition*. 46th Ann. Mtg. American Physical Society Division of Plasma Physics, Savannah, GA, Nov. 2, 2004. UCRL-ABS-205284.

Town, R. P. J. et al. (2005). *Integrated modeling of short-pulse laser-plasma experiments*. 47th Ann. Mtg. APS Division of Plasma Physics, Denver, CO, Oct. 24–28, 2005. UCRL-ABS-210615.

Town, R. P. J. et al. (2005). *LSP calculations of cone-wire experiments*. 8th Intl. Workshop Fast Ignition of Fusion Targets, June 27–July 1, 2005, Tarragona, Spain. UCRL-ABS-209626.

Town, R. P. J. et al. (2005). "Simulations of electron transport for fast ignition using LSP." *Nucl. Instrum. Methods Phys. Res.* **A544**, 61. UCRL-PROC-204414.

Surrogate Nuclear Reactions and the Origin of Heavy Elements

Jutta E. Escher 04-ERD-057

Abstract

This work explores an innovative method for indirectly determining reaction cross sections by "surrogate" nuclear reactions. This is important because a large number of nuclear reactions relevant to astrophysics and Laboratory programs cannot be measured with currently available techniques. We will provide the theoretical and experimental framework for planning and analyzing experiments that will allow us to obtain cross sections of interest. We intend to design, carry out, and analyze experiments that provide benchmarks for the surrogate nuclear reaction method. Our applications will focus particularly on reactions involving unstable nuclei that play a key role in the production of elements between iron and uranium.

The long-term goal of this research is establishment of a new method for indirectly determining cross sections for a large class of nuclear reactions. Indirect approaches to reactions are becoming increasingly important because nuclear physics applications, particularly in the area of astrophysics, require information on reactions involving unstable isotopes. While currently available indirect methods focus on direct reactions, the surrogate nuclear reaction approach allows us to address the need for compound nuclear-reaction cross sections. Such cross sections are extremely important for achieving an improved understanding of the astrophysical s-process (slow neutron capture), and for obtaining significant new insights into synthesis of heavy elements.

Mission Relevance

Establishing a novel technique for determining reaction cross sections for unstable nuclei will satisfy critical needs for science-based stockpile stewardship and homeland-security applications that require reliable information about reactions involving unstable nuclei. The basic science research in this project also supports the Laboratory's mission in breakthrough science and technology.

FY05 Accomplishments and Results

In FY05, we (1) developed models and corresponding new codes that allow us to describe formation of a spherical compound nucleus in a surrogate reaction; (2) studied the decay of a compound nucleus and its dependence on spin-parity distribution of the decaying nucleus; (3) improved the previously used experimental setup and moved it to nearby Lawrence Berkeley National Laboratory; (4) performed and begun to analyze surrogate reaction experiments that will serve as benchmarks for the method, and provide information on hitherto unknown reaction cross sections; (5) begun to expand our theoretical studies to include deformed nuclei; and (6) presented a number of invited and contributed conference talks, and published several short articles and conference proceedings.

Proposed Work for FY06

In FY06, we will (1) implement further improvements and generalizations of our theoretical approach—in particular, include a treatment of pickup reactions and expand our models to describe surrogate reactions on deformed nuclei; (2) design, carry out, and analyze relevant experiments to test our models and establish benchmarks; and (3) examine under which circumstances, simplifications or approximations to the surrogate method can be utilized to determine relevant cross sections, and study the limitations of the surrogate approach.

Publications

Ahle, L. et al. (2004). *The surrogate method for reaction cross sections*. UCRL-POST-208192.

Church, J. A. et al. (2005). "Surrogate reactions for nucleosynthesis: ^{102,104}Ru(a,a') as surrogate reactions for ^{101,103}Ru(n,g)." *Bull. Am. Phys. Soc.* **50**, 161. UCRL-ABS-212950.

Escher, J. (2005). *Indirect methods for nuclear reaction data*. Perspectives on Nuclear Data in the Next Decade Workshop, Bruyere-le-Chatel, France, Sept. 26–28, 2005. UCRL-ABS-211693.

Escher, J. (2005). *Nuclear reactions with unstable nuclei and the surrogate reaction technique*. 21st Winter Workshop Nuclear Dynamics, Breckenridge, CO, Feb. 5–12, 2005. UCRL-ABS-209125.

Escher J. et al. (2005). "Surrogate nuclear reactions—An indirect method for determining reaction cross sections." *J. Phys. G: Nucl. Part. Phys.* **31**, S1687. UCRL-JRNL-210888.

Escher, J. et al. (2005). *The surrogate method—An indirect approach to compound-nucleus reactions*. 21st Winter Workshop Nuclear Dynamics, Breckenridge, CO, Feb. 5–12, 2005. UCRL-PROC-211557.

Escher, J. and F. S. Dietrich (2005). *Determining cross sections for reactions on unstable nuclei: A consideration of indirect approaches*. 2nd Argonne/MSU/JINA/INT RIA Workshop: Reaction Mechanisms for Rare Isotope Beams, East Lansing, MI, Mar. 9–12, 2005. UCRL-PROC-212586.

Escher, J. et al. (2004). *Cosmic questions and microscopic answers: Understanding the origins of the heavy elements*. 2004 Laboratory Women's Forum, Berkeley, CA, Oct. 26–27, 2004. UCRL-POST-207398.

Escher, J. et al. (2004). *Nuclear reactions with unstable nuclei and the surrogate reaction technique*. 2004 Fall Mtg. Division of Nuclear Physics, American Physical Society, Chicago, IL, Oct. 27–30, 2004. UCRL-PRES-207521.

Stellar Astrophysics and a Fundamental Description of Thermonuclear Reactions

William E. Ormand 04-ERD-058

Abstract

This project is investigating new methods to provide a comprehensive understanding of reactions between light nuclei in hot, dense environments, such as stellar interiors and implosion capsules for future large fusion-class lasers. The project will develop a new theoretical framework that describes the dynamics of nuclear collisions based on the fundamental interactions of nature. In addition, the project will undertake a theoretical study of the quantum corrections to electron screening in thermal plasmas to resolve a discrepancy exhibited in previous theoretical approaches and investigate the viability of using fusion-laser facilities to observe a wide range of stellar thermonuclear reactions.

Achieving an accurate and predictive theoretical view of thermonuclear reactions in thermal plasmas based on fundamental physical properties will have far-reaching applications for astrophysics. Benefits include improved stellar modeling capabilities that could improve our knowledge of neutrino oscillations, better databases important to stockpile stewardship, and higher confidence in understanding the dynamics of fusion-class laser target capsules. The ability to perform stellar astrophysics experiments at such facilities could lead to an exciting new experimental program.

Mission Relevance

A more comprehensive understanding of nuclear reactions in hot, dense systems based on fundamental physical properties will be achieved. In particular, this project will support the nuclear data effort for the Laboratory's stockpile stewardship mission and provide insight for experiments to be performed at future fusion-class laser facilities. In addition, by advancing the understanding of stellar evolution, this project supports LLNL's basic science mission.

FY05 Accomplishments and Results

We (1) developed and published the formalism detailing radial-cluster overlaps and an approximation to the full formalism to compute astrophysical s-factors and scattering phase shifts; (2) applied the approximation to the ^{7}Be(p,γ) and ^{11}Be(n, γ) reactions and performed preliminary calculations for ^{3}He(^{4}He,γ)^{7}Be, achieving excellent agreement with measured ^{7}Be(p, γ) s-factors and good agreement with the ^{4}He + n phase shift; (3) set up electron screening problem in Keldysh formalism; (4) uniquely determined the form of the three-nucleon interaction as derived from effective-field theories (EFT); and (5) carried out large-basis calculations for light nuclei. Our calculations indicate that EFT potentials give a quantitative description of the low-lying structure of ^{10}B.

Proposed Work for FY06

In FY06, for the reaction component, we plan to develop the formalism to derive folding potential for the scattering solution in asymptotic region, develop the formalism to compute the renormalized radial-cluster form factors, integrate three-nucleon interactions to reaction calculations and determine their sensitivity on s-factors, and start developing codes to include four-body effective interactions. We plan to hire a postdoctoral researcher to investigate the physics of two-temperature plasmas and nonequilibrium effects on nuclear reactions in a plasma. Plasma effects to be examined include relaxation, conductivities, and ion loss. Depending on the progress for reactions and screening, we will re-examine capsule simulations.

Publications

Barrett, B. R. et al. (2005). "No-core shell model calculations in light nuclei with three-body interactions." *Nucl. Phys. A* **746**, 579C. UCRL-PROC-201751.

Forssen, C. et al. (2005). "Ab initio study of the level ordering anomaly in ^{11}Be." *Bull. Am. Phys. Soc.* **50**, 48. UCRL-ABS-209120.

Forssen, C. et al. (2005). "Large basis ab initio shell model investigation of the ^{9,11}Be isotopes." *Phys. Rev. C* **71**, 044312. UCRL-JRNL-208555.

Forssen, C. et al. (2005). "Study of the ^{10}Be(n, γ)^{11}Be reaction and strong E1 transitions in ^{11}Be using ab initio wave functions." *Bull. Am. Phys. Soc.* **50**, 71. UCRL-PRES-215619.

Gueorguiev, V. G., P. Navratil, and W. E. Ormand. (2005). "Chiral three-nucleon interactions and the structure of light nuclei." *Bull. Am. Phys. Soc.* **50**, 148. UCRL-PRES-215539.

Navratil, P. (2005). "Ab initio nuclear structure and nuclear reactions in light nuclei." *Int. J. Mod. Phys. E* **14**, 85. UCRL-PROC-206321.

Navratil, P. (2005). "Cluster form factor and spectroscopic factor calculation in the ab initio no-core shell model." *Bull. Am. Phys. Soc.* **49**, 103. UCRL-PRES-207490.

Navratil, P. (2005). "Cluster form factor calculation in the it ab initio no-core shell model." *Phys. Rev. C* **70**, 054324. UCRL-JRNL-205655.

Navratil, P., C. Bertulani, and E. Caurier. (2005). "^{7}Be(p, γ)^{8}B S-factor from ab initio wave functions: I. overlap integral calculation." *Bull. Am. Phys. Soc.* **50**, 92. UCRL-PRES-211568.

Navratil, P. et al. (2005). "Ab initio no-core shell model calculations using realistic two- and three-body interactions." *Eur. Phys. J. A* **25**, 481. UCRL-PROC-208310.

Navratil, P. et al. (2005). "No-core shell model and reactions." *AIP Conf. Proc.* **791**. UCRL-PROC-211912.

Navratil, P. et al. (2005). "S-factors of capture reactions important for astrophysics from ab initio wave functions." *Bull. Am. Phys. Soc.* **50**, 70. UCRL-PRES-215417.

Ormand, W. E. et al. (2005). "The structure of light nuclei with interactions derived from chiral effective-field theory." *Bull. Am. Phys. Soc.* **50**, 149. UCRL-PRES-215625.

Stetcu, I., et al. (2005). "Effective operators with the ab initio no-core shell model." *Phys. Rev. C* **71**, 044325. UCRL-JRNL-208306.

Stetcu, I. et al. (2005). "Electromagnetic transitions with effective operators." *Int. J. Mod. Phys. E* **14**, 95. UCRL-PROC-205656.

Vary, J. P. et al. (2005). "Shell model in a first principles approach." *Nucl. Phys. A* **746**, 123C. UCRL-PROC-201765.

High-Energy-Density Galaxy Jets

Willem Van Breugel 04-ERD-059

Abstract

Jets, ionizing radiation, and high-energy particles from active supermassive black holes and exploding stars profoundly affect the formation of new stars in their parent galaxies. At LLNL, we are in a unique position to investigate this by combining our expertise in complex, multidimensional numerical simulations and high-energy experiments, with our access to world-class observatories. We propose to continue our multidisciplinary studies of the interaction of jets with interstellar medium clouds, and the energetic processing of astrophysically relevant materials using an ion beam accelerator.

We will (1) analyze astronomical observations at optical, x-ray, and radio wavelengths and determine input parameters for numerical jet–cloud collisions; (2) improve our numerical simulations code COSMOS to perform more realistic, three-dimensional simulations of jet–cloud collisions; (3) conduct further irradiation experiments of astrophysically relevant materials using the ion accelerator, and analyze the results; and (4) obtain astronomical observations of galaxies to investigate the effects of active black holes on interstellar dust grains.

Mission Relevance

Our research will help validate computer codes used to simulate high-energy-physics phenomena critical to nuclear physics research and to experiments at future large lasers for fusion studies. Our laboratory experiments, combined with astronomical observations, will provide new information about the effects of high-energy particles on materials under extreme conditions, which will further the national security mission.

FY05 Accomplishments and Results

In FY05 we (1) improved the COSMOS code by including the effects of magnetic fields—directions and strengths of the fields dramatically affected the cooling, size, and distribution of cloud fragments; (2) obtained new radio and optical observations that provide strong evidence, predicted by the COSMOS simulations, that radio jets can trigger radiative cooling of warm clumpy gas, and subsequent star formation; (3) presented these results at several meetings; (4) completed analysis of our LLNL irradiation experiments and submitted this for publication; and (5) performed numerical simulations using a thermal spike model, confirming that electronic excitations in astrophysical dust by "low" (0.1 to 1 GeV) cosmic rays can vitrify interstellar dust, as observed.

Proposed Work for FY06

In FY06 we will (1) perform radiation experiments at higher energies than before, which will more accurately represent conditions in the interstellar medium; (2) perform surface chemistry experiments to determine if molecule formation is affected by vitrifying interstellar dust; and (3) perform more detailed numerical simulations of the effects of cosmic rays on dust grains.

Publications

De Breuck, C. et al. (2005). "Detection of two massive CO systems in 4C 41.17 at z = 3.8." *Astron. Astrophys.* **430L**, 1. UCRL-JRNL-207484.

Van Breugel, et al. (2005). "Star formation in high pressure, high energy density environments: Laboratory experiments of ISM dust analogs." *Proc. Symp.: The Dusty and Molecular Universe—A Prelude to Herschel and ALMA*, 91–92. UCRL-JRNL-209106.

Venemans, B. P. et al. (2005). "Properties of Lyα emitters around the radio galaxy MRC 0316-257." *Astron. Astrophys.* **431**, 793. UCRL-JRNL-206071.

Venemans, B. P. et al. (2004). "Discovery of six Lyα emitters near a radio galaxy at z ~ 5.2" *Astron. Astrophys.* **424L**, 17. UCRL-JRNL-206783.

High-Brightness, Laser-Driven, X-Ray Source for Nanoscale Metrology and Femtosecond Dynamics

John K. Crane 04-ERD-064

Abstract

This project will develop a new, bright, ultrafast x-ray source based on laser-driven K-alpha generation that will produce an x-ray flux 10 to 100 times greater than current microfocus x-ray tubes. The short-pulse (~100 fs) duration of this x-ray source also makes it ideal for observing time-resolved dynamics of atomic motion in solids and thin films. We are currently developing this K-alpha x-ray source using the existing Falcon terawatt laser system, measuring x-ray yield, emitter size, and temporal pulse length as a function of laser pulse and target characteristics. The measurements are compared with a model. The results will lead to a high-average-power fiber-laser-driven x-ray source.

We expect to develop a high-average-brightness x-ray source with subpicosecond temporal and submicron spatial resolution. This source could be used for the in situ radiography of beryllium shells and cryotargets, dynamic diffraction experiments with actinides, and microscopic imaging of chemical and biological systems, nanostructures, and advanced semiconductor devices. Careful measurements of K-alpha x-ray generation versus laser and target parameters will help verify codes being developed for high-energy-density physics and backlighters. When combined with high-average-power, ultrafast fiber-laser technology, this x-ray source could serve as a multiple-application facility.

Mission Relevance

An improved, high-average-power x-ray source will be useful to many applications that support the national security mission, such as the in situ characterization of cryotargets for fusion ignition, three-dimensional imaging of actinides under dynamic stress (when used with ultrafast electron diffraction technology), advanced streak cameras, x-ray detectors, and x-ray optics.

FY05 Accomplishments and Results

In FY05 we performed experiments to characterize the x-ray yield and emitter spot size for thin copper foil targets. The object was to optimize the K-alpha x-ray yield for the similar laser characteristics expected for the fiber-laser driver. In these experiments, we measured a conversion efficiency from laser energy to K-alpha x-ray energy of 1.5×10^{-4}. We also developed an automated Mylar debris shield to protect the focusing parabola from debris when operating for long periods; made a target positioner for measuring target location and target motion during continuous operation; measured an x-ray spot size of 57 µm and a target motion of 10 µm; and wrote an interface for the Tiger code that models electron transport in the material.

Proposed Work for FY06

In FY06 we will complete the K-alpha source measurements on the Falcon laser and compare the data with our models, and complete the 100-W fiber laser and use it to produce x-rays and characterize the results. X-ray emission experiments with the fiber laser will be performed at a reduced repetition rate, but we will begin designing a source that accommodates the laser's full repetition rate. A final report will describe the path toward a high-brightness, multi-user x-ray facility.

Ultrafast, In Situ Probing of Shocked Solids at the Mesoscale and Beyond: A New Paradigm for Materials Dynamics

Hector E. Lorenzana 04-ERD-071

Abstract

This project will conduct the first unambiguous in situ measurements of the microstructural evolution of metals under high strain rates using ultrafast x-ray and electron probes to produce diffraction, scattering, and imaging measurements. By directly probing lattice response in real time down to nanometer and nanosecond resolution (a new paradigm), we will provide critical insight about the fundamental mechanisms governing macroscopic behavior. We will develop these novel dynamic measurements using electron microscopes, laser-produced x-ray sources, and accelerator x-ray sources. We will shock samples with high-power lasers and electric mini-flyers.

We expect that measurements from this research will produce new understanding of solid–solid phase transformations and damage evolution. Specific physical observables will include kinetics of crystalline restructuring, elastic-to-plastic transition, dislocation and defect concentrations, and void growth, all in real time. Measurements will directly connect lattice response with continuum behavior, important to materials science and practical applications of metals under shock loading.

Mission Relevance

Success in stockpile stewardship, energy research, and other mission areas demands a fundamental understanding of dynamic materials behavior. The techniques demonstrated in this project will be applicable to current and future Laboratory experiments in support of stockpile stewardship. Capabilities demonstrated will drive development of computational tools for predicting performance, safety, and reliability of nuclear weapons, as well as complex issues important to high-power laser research.

FY05 Accomplishments and Results

We produced the first dynamic x-ray diffraction observations of a pressure-driven phase transformation in iron. The measurements were also able to identify the atomic pathway of the transition. High signal-to-noise-ratio single-pulse (100 ps) static x-ray scattering measurements of void distributions in incipiently spalled vanadium show that dynamic experiments are possible. Time-resolved static imaging of void-like features at the Janus laser identified the necessary conditions for dynamic experiments. Dynamic electron microscopy has measured a solid-state phase transition in titanium and a solid–liquid interface under transient melting conditions.

Proposed Work for FY06

During FY06, we will perform a series of quantitative measurements in situ during shock loading. First, we will begin our study of lattice kinetics using diffraction. Our work in iron will continue with the investigation of the effect of crystal orientation on the observed phase transformation. Additional diffraction experiments will assess kinetics of the phase transformations. We will also measure, for the first time, the damaged state during dynamic fracture using time-resolved, small-angle x-ray scattering. We will demonstrate scattering from geometrically representative and shock recovered samples. Then, we will synchronize the synchrotron x-ray pulse with the shock drive to enable scattering studies at the Advanced Photon Source.

Publications

Campbell, G. et al. (2005). *The HCP to BCC phase transformation in Ti characterized by nanosecond electron microscopy*. Solid–Solid Phase Transformations in Inorganic Materials 2005, Phoenix, AZ, May 29–June 3, 2005. UCRL-PROC-213217.

Kalantar, D. H. et al. (2005). "Direct observation of the alpha–epsilon transition in shock-compressed iron via nanosecond X-ray diffraction." *Phys. Rev. Lett.* **95**, 075502/1. UCRL-JRNL-211109.

Kleinschmidt, H., A. Ziegler, and G. Campbell (in press). "Phase transformation analysis in titanium at nanosecond time-resolution." *J. Appl. Phys.* UCRL-JRNL-209974.

Remington, B. et al. (in press). *Materials dynamics at extreme pressures and strain rates*. Intl. Symp. Plasticity, Kaui, HI, Jan. 3–8, 2005. UCRL-PROC-206341.

Remington, B. et al. (2005). "Accessing high pressure states relevant to core conditions in the Giant planets." *Astrophys. Space Sci.* **298**, 235. UCRL-PROC-204070.

Swift, D. C. et al. (2005). "Dynamic response of materials on subnanosecond time scales, and beryllium properties for inertial confinement fusion," *Phys. Plasmas* **12**, 056308. UCRL-JRNL-208779.

Non-Equilibrium Phase Transitions

Andrew Ng 04-ERD-108

Abstract

Exploring non-equilibrium phase transitions is a scientific frontier that holds promises for discovering new phases, metastable states, chemical reaction pathways and biological functioning processes. This project will conduct the first systematic study of phase transitions in an extreme, non-equilibrium regime to correlate electronic excitation, lattice disordering, and structural changes, and to advance finite-temperature condensed matter theory for understanding dynamic material behavior at the first-

principles level. The project will use experimental measurements of optical and structural properties under ultrafast laser excitation as the basis for developing density functional theory approaches. Work in ultrafast electron diffraction is being conducted in collaboration with University of Toronto.

We expect to obtain time-correlated data on optical and structural properties by tracking solid–liquid to liquid–plasma transitions under ultrafast excitation conditions. These data will benchmark quantum simulations based on the density functional theory approach. If successful, the project will achieve new understanding of the connection between electronic (optical) and atomistic (structural) behavior, opening up possibilities of manipulating phase stability and boundary while validating new developments in theory to improve predictive power. Success in this area will also help describe the convergence of condensed matter and plasma physics, a critically missing link in basic scientific understanding.

Mission Relevance

Ultrafast optical and atomistic diagnostics for nanoscale experiments, coupled with theory development using high-performance computing, will provide increased understanding of phase transitions and kinetics for equation-of-state data development in support of the stockpile stewardship mission.

FY05 Accomplishments and Results

In FY05 we (1) measured optical properties linking disassembly of a laser-heated solid to lattice energy density as a critical theory benchmark; (2) measured spectral dielectric function to show changes in intraband and interband transitions, opening the possibility of probing band structure collapse; (3) in collaboration with the University of Texas, accessed ultrafast electron diffraction (UED) data to correlate lattice disordering and disassembly; (4) began construction of our electron gun; (5) implemented QBOX code to compare effects of excited electrons in silicon and aluminum, revealing a rapid onset of non-thermal melting only in silicon; (6) started to develop transport and equation-of-state calculations; and (7) performed fluorescence measurements, which showed a clear correlation between fluorescence and dielectric function.

Proposed Work for FY06

In FY06 we will (1) perform a systematic comparison of spectral dielectric function and fluorescence spectrum over a wide range of laser excitation energy densities; (2) complete construction of our femtosecond electron gun and perform UED measurements to correlate structural properties with optical properties; and (3) use quantum molecular dynamics simulations to calculate dielectric function and conductivity for comparison with experimental data, and to calculate the nonequilibrium melt line of gold for comparison with experimental observations.

Publications

Ao, T. et al. (in press). "Optical properties in non-equilibrium phase transitions." *Phys. Rev. Lett.* UCRL-JRNL-218075.

Ping, Y. et al. (2005). *The color of warm dense gold.* IEEE Intl. Conf. Plasma Science, Monterey, CA, June 20–23, 2005. UCRL-ABS-212528.

Widmann, K. et al. (2005). *Measurements of single-state electrical conductivities of warm dense gold.* Intl. Workshop Warm Dense Matter, Vancouver, Canada, Oct. 6–8, 2005. UCRL-ABS-215918.

Widmann, K. et al. (2004). "Single-state measurement of electrical conductivity of warm dense gold." *Phys. Rev. Lett.* **92**, 125002. UCRL-ABS-204709.

Development of Insulating Liquids for Detecting and Imaging of Low-Energy Particles

Adam Bernstein 04-LW-017

Abstract

Noncryogenic detectors capable of providing high-efficiency, high-resolution spatial imaging and good energy resolution could help solve a variety of long-standing questions in the detection of low-energy (keV to MeV) particles. Ion drift within insulating liquids could allow development of new imaging detectors with wide application to outstanding problems in fundamental physics, medical imaging, and fissile-material detection. We propose a staged research program aimed at developing and exploiting this largely unexplored detection technique.

If successful, this work will enable development of a wide class of imaging, energy-resolving detectors capable of measuring the properties of 10-keV to 10-MeV particles with high efficiency at room temperatures. This will help solve problems for detection in low-rate applications such as the detection of weakly radioactive materials and rare particle decays, as well as medical imaging applications in which low doses are imperative.

Mission Relevance

This work is broadly applicable to NNSA and LLNL's national-security mission, through the possibility of improved detection of fissile materials. It could make a significant contribution toward development of a new technological capability for biomedical imaging, and may also considerably improve sensitivity for a rare process known as neutrinoless double-beta decay, of central interest to DOE and LLNL's mission in breakthroughs in fundamental physics in the 21st century.

FY05 Accomplishments and Results

In FY05, we (1) confirmed the existence of electron and ion signals using x-ray irradiation; (2) established that the mobility of the liberated electrons and ions were consistent with expectations; (3) discovered that recombination effects were small because ion and electron signals persisted even at low sweep voltages, an important consideration for practical reasons; and (4) developed a clear path forward for using insulating liquids as a simpler means of performing spectroscopy of MeV-scale gamma rays and neutrons for use in a variety of radiation detection applications. We did not see individual particle interactions as we had hoped. We believe this was because of the low level of purity in the liquid and cell components we used, but did not have the time or resources needed to resolve this issue.

Relativistic Antimatter Plasmas Created by Ultraintense Lasers

Scott C. Wilks 04-LW-020

Abstract

The goal of this project is to generate and characterize, in the laboratory, a relativistic electron–positron (e^+e^-) plasma that is theorized to be the origin of the physical mechanisms of gamma ray bursts observed near the boundaries of black holes. We propose to study the formation of this peculiar state of matter with a combination of experiment and modeling. We will build the first-ever e^+e^- spectrometer for laser-plasma physics and use it to characterize the e^+e^- jet in ultraintense laser–solid interactions. Scaled laboratory astrophysics experiments will then be designed and conducted on existing and planned laser facilities. We plan to model the experimental results and test novel target configurations with three-dimensional computer simulation codes.

If successful, we expect to achieve the highest density relativistic e^+e^- plasma ever generated in a controlled fashion on earth. This will open up an entirely new window to understanding this peculiar state of matter thought to exist only in energetic astrophysical objects. We expect to build and patent the first e^+e^- spectrometer. In addition, we will produce a point design for one of the first laboratory astrophysics experiments to be fielded on future large laser facilities with petawatt capability. Developing a laboratory astrophysics capability is an exciting new tool that will attract talented young researchers to LLNL.

Mission Relevance

The knowledge concerning the generation of hot dense matter using ultrashort, ultra-intense lasers that was gained from this project will be important in employing these lasers to support the Laboratory’s stockpile stewardship mission.

FY05 Accomplishments and Results

In FY05, we finished building a novel e^+e^- spectrometer and filed a record of invention for the device. Upon fielding our device on a petawatt laser in England that is currently the world's most powerful short-pulse laser, we measured simultaneous electron and positron energy spectra of a dense e^+e^- jet formed using a gold target. By comparing these experimental results to simulation results using particle-in-cell code, we concluded that important laser–plasma interactions in the preformed plasma acted to limit the maximum electron energies that can be achieved. This information resulted in important modifications to our point design for generating dense e^+e^- jets using future petawatt laser facilities. In this project, development of a spectrometer that quantitatively characterizes pair plasmas produced by such lasers has resulted in a diagnostic that will, for several years to come, play a role in virtually all LLNL efforts that require short-pulse laser–solid interactions.

Publications

Chen, H. and S. C. Wilks. (2005). "Hot electron and positron energy distributions from ultra-intense laser–solid interactions." 4th Intl. Conf. Inertial Fusion Science and Applications, Biarritz, France, Sept. 4–9, 2005. UCRL-ABS-212922.

Chen, H. and S. C. Wilks. (2004). "Evidence of enhanced effective hot electron temperatures in ultra-intense laser–solid interactions due to reflexing." Short-Pulse Laser–Matter Computational Workshop, Pleasanton, CA, Aug. 25, 2004. UCRL-PRES-206188.

Chen, H. et. al. (2005). "Hot electron energy distributions from ultra-intense laser–solid interactions." 47th Ann. Mtg. American Physical Society Division of Plasma Physics, Denver, CO, Oct. 24–28, 2005. UCRL-ABS-214200.

Fractality of Fracture Surfaces in Polycrystalline Materials

Bryan W. Reed 04-LW-036

Abstract

We propose to combine statistical mechanics models of fracture in random media with detailed experimental data that characterize certain polycrystalline materials and their fracture behavior. The computer model we have developed provides a fast way to find the weakest path through a random medium as an estimate for the fracture path typically showing a self-affine fractal behavior. To date this model has been applied only to simplified, toy model systems. In this project, electron backscatter diffraction data and molecular dynamics simulations of various grain boundaries will provide input for the model, including the topology of the grain boundary network and the strength properties of the boundaries. The results will then be compared with the experimental fracture of the system.

By linking theoretical models and methods with actual experimental data of fracture surfaces, this project will provide many interesting basic science results and insight into the behavior of polycrystalline materials and their strength properties. In addition, the project will produce a database of strength values of various grain boundaries and the effect of bismuth in them, calculated using molecular dynamics.

Mission Relevance

The statistical mechanics model for fracture in random media and the analysis of polycrystals developed in this project enhance the Laboratory's core competence in dynamic fracture of metals, which supports the Laboratory's national security mission, and specifically stockpile stewardship.

FY05 Accomplishments and Results

This project broke new ground in understanding the roles of microstructural length scales in intergranular failure. Grain boundary network correlations were shown to create a supragrain length scale that appears to be a key factor distinguishing grain-boundary-engineered material from normal material. We developed a coherent suite of techniques (including theoretical analysis), from a new group-theoretical perspective, computer simulations that produce remarkably realistic polycrystalline structures with very few arbitrary parameters, data analysis methods for electron back-scatter diffraction, and fractal analysis of fracture surfaces measured with scanning confocal microscopy. These techniques and insights have strong potential for a broad range of applications.

Publications

Gertsman, V. Y. and B. W. Reed. (2005). "On the three-dimensional twin-limited structure." *Z. Metallk.* **96**, 1106. UCRL-JRNL-210611.

Kumar, M. and B. W. Reed. (2005). *Influence of grain boundary engineering processing on fracture processes*. Materials Science and Technology 2005, Pittsburgh, PA, Sept. 25–28, 2005. UCRL-ABS-210680.

Reed, B. W. and M. Kumar. (in press). "Mathematical methods for analyzing highly-twinned grain boundary networks." *Scr. Mater.* UCRL-JRNL-212206.

Reed, B. W. et al. (2005). *On the role of multi-length-scale microstructural correlations in the fracture of grain boundary engineered materials*. 2005 MRS Spring Mtg., San Francisco, CA, Mar. 28–Apr. 1, 2005. UCRL-ABS-207552.

Laser Pulse Compression by Stimulated Raman Scattering in a Plasma

Robert Kirkwood 04-FS-011

Abstract

We have developed a new technique to increase the power of a laser by using three-wave stimulated mixing and pulse compression in a plasma. The nonlinear wave response needed to compress a laser pulse of approximately 1 ns into a 1-ps pulse was studied, and a 1-ps seed pulse was amplified by as much as 37 times, acquiring its power from the pump beam. Our results show the intensity and density dependence of the amplification process and indicate the plasma conditions needed for compressing a 1-ns pulse at higher beam intensity. When successful, this plasma-based approach will be an advance over present pulse-compression techniques, because the power-handling limits of a plasma (>10^{14} W/cm^2) are much higher than those of conventional solid-state optics (<10^{11} W/cm^2).

We have demonstrated for the first time the amplification of a 1-ps beam by intersection with a 1-ns beam in a plasma. The amplification was measured at two different plasma densities and temperatures providing data needed to benchmark laser plasma interaction simulations and design a compression scheme for an approximately 1-ns pump pulse with the best achievable efficiency. These results are expected to lead to a low-risk plan for attaining 10 to 100 kJ of energy in a short pulse at future fusion-class lasers. This work also has measured stimulated Raman scattering (SRS) gain rates on a time scale short enough to identify saturation mechanisms. The data is being prepared for publication in a peer-reviewed journal, and we will complete a second longer paper.

Mission Relevance

These experiments will provide great leverage to existing and planned fusion-class lasers, and will have broad application in inertial fusion, radiation sources, and particle accelerators. All these topics are of primary importance to NNSA and Laboratory missions in national security, particularly stockpile stewardship.

FY05 Accomplishments and Results

We have used the improved target and beam transport system to perform experiments demonstrating amplification of a 1-ps pulse with a high laser intensity 1-ns pulse in a low-density plasma. The experiments demonstrate for the first time that a net increase of power in the short-pulse beam can be produced by SRS in a plasma that can coexist with a high intensity 1-ns beam, and is an essential step toward plasma compression of a 1-ns pulse. Measurements at different beam intensities and densities will allow the plasma and beam conditions to be optimized for a pulse-compression experiment carried out with a higher intensity short-pulse beam.

Publications

Kirkwood, R. K. et al. (2005). *Amplification of 1 ps pulse length beam by stimulated Raman scattering of a 1 ns beam in a low density plasma*. IEEE Intl. Conf. Plasma Science, Monterey, CA, June 20–23, 2005. UCRL-CONF-213152.

Kirkwood, R. K. et al. (2005). "Observation of amplification of a 1 ps pulse by SRS of a 1 ns pulse in a plasma with conditions relevant to pulse compression." *Bull. Am. Phys. Soc.* **50**, 26. UCRL-ABS-213638.

Two-Phase Noble Liquid–Gas Detectors for Detection of Coherent Elastic Neutrino Scattering

Adam Bernstein 04-FS-032

Abstract

This project aims to establish the feasibility of a new class of very low-noise and -background particle detectors, known as two-phase noble gas ionization detectors, which may prove capable of detecting the faint but high-rate coherent neutrino-nucleus scattering signal. Coherent scattering is predicted by and calculable within the Standard Model, but has never been measured. At low momenta, the neutrino can elastically scatter off all the nucleons in the nucleus coherently, leading to a recoil of the whole nucleus in its ground state, for which the elastic scattering cross section is enhanced by a factor of Z^2, where Z is the atomic number. This feasibility study will allow us to model, build, and test a gas phase detector, from which data will be analyzed to establish or reject feasibility of a larger-scale experiment to pursue the detection of coherent elastic neutrino-nucleus scattering.

If successful, we expect to (1) build a low-cost platform for direct experimentation with noise-reduction strategies for coherent scattering detectors; (2) determine the likelihood of being able to detect 1 to 10 electrons produced at rest in a noble gas (xenon) through the use of their luminescent signal under an accelerating voltage; and (3) establish the feasibility of a larger-scale experiment. This work is significant because it allows us to begin actual experimental determination of the prospects for detection of coherent scattering of neutrinos, a long predicted yet unmeasured feature of weak interactions.

Mission Relevance

This world-class measurement would have immediate impact on the Standard Model of particle physics, and would influence cosmology and astrophysics by confirming or correcting widely used star and supernovae opacity calculations that are of interest to stockpile stewardship. Practical national-security applications of this technology include a compact device for monitoring nuclear reactors by detection of antineutrinos.

FY05 Accomplishments and Results

In FY05, we (1) tested the luminescence, drift, and noise properties of our system, thereby making essential progress in establishing the feasibility of detection of few electron signals in the gas phase; (2) reached an impressive noise floor (i.e., detection limit) of about 25 primary electrons, compared to the long-term goal of 10 or fewer primary electrons; (3) developed a clearer understanding of the remaining hurdles to achieve a lower electron detection goal, learning enough to conclude that the remaining problems should be readily solvable in future work. In short, this project achieved its goal of establishing the feasibility of dual-phase detection.

Biological Imaging with Fourth-Generation Light Sources

Henry N. Chapman 05-SI-003

Abstract

We propose to develop capabilities to carry out single-molecule atomic-resolution imaging at future x-ray free-electron laser (XFEL) facilities. Our goal is to perform groundbreaking experiments at new and existing sources to test the key concepts of single-molecule XFEL imaging, including measurement of the Coulomb explosion of particles in intense ultrashort x-ray beams, lensless x-ray imaging beyond the radiation-damage limit, and the manipulation and orientation of single particles in space and time to interact with XFEL pulses. We will crosscheck experiments with substantial modeling efforts to understand the new abilities that the XFEL will bring to biological imaging.

Each of our experiments, which will be a world first and a major new result in x-ray science in every case, will determine (1) the duration and fluence of XFEL pulses required for single-molecule imaging; (2) demonstrate reconstruction methods; and (3) perform ultrahigh-resolution three-dimensional (3-D) imaging of container-free particles, for which new technologies in biological sample preparation will be developed. Together, these experiments will demonstrate the extraordinary science achievable with XFELs and the impact they will have on structure determination of biological macromolecules, protein complexes, viruses, and spores.

Mission Relevance

Improved tomography algorithms will benefit stockpile stewardship. As a specific example, diffraction-imaging techniques can be applied to the study of warm dense matter, a critical regime of weapons physics. Single-molecule imaging will allow the structure of virtually any macromolecule, protein, or virus to be determined, which furthers LLNL's missions in both biodefense and bioscience to improve human health. Our research also enhances the capabilities of the Linac Coherent Light Source (LCLS), a high-priority project of the DOE Office of Science, in support of LLNL's mission in breakthrough science and technology.

FY05 Accomplishments and Results

We carried out a rigorous characterization of our images of test objects and aerogel, establishing a resolution of 10 nm, the world's highest resolution 3-D x-ray images of nonperiodic material. An apparatus for experiments on the Vacuum-Ultraviolet Free-Electron Laser (VUV-FEL) in Hamburg, Germany has been designed (utilizing our hydrodynamic code for particle/x-ray interaction) and built. The experiments will yield the first validation of models of XFEL molecular imaging. The apparatus utilizes a novel VUV bandpass mirror, and the modular design allows pump-probe experiments with a time resolution of 30 fs. We have built a test chamber for the development of particle injection systems for future VUV-FEL experiments and for the LCLS, and we have determined a method to "repair" diffraction patterns of molecules to overcome the effects of ionization.

Proposed Work for FY06

In FY06, we will perform experiments at the world's brightest soft x-ray source, VUV-FEL in Hamburg. This will be the first experimental verification of models of the damage done to biological materials by XFEL pulses. Initial measurements will be made on multilayered and lithographically fabricated test structures; then measurements will be made on the explosion of organic particles. Explosions will be measured from the diffraction pattern of the XFEL pulse itself scattered from the particles. These will be compared with calculations of Mie scattering to obtain the particle size as a function of pulse fluence. We will test the feasibility of pump-probe experiments with inherently high time resolution. We will develop an efficient particle injector, and simulate the injection of molecules, using the LCLS located at the Stanford Linear Accelerator Center. Those results will be compared to molecular dynamics codes run on a Thunder-class computer. Additionally, our 3-D imaging experiments will be extended to 5-nm resolution.

Publications

Barty, A. et al. (2005). *High-resolution ab initio three-dimensional coherence x-ray diffraction microscopy*. Intl. Workshop Phase Retrieval and Coherent Scattering, June 15–17, 2005, Porquerolles, Provence, France. UCRL-CONF-213044.

Chapman, H. N. (2005). *Imaging molecules with x-ray free-electron lasers*. National Academy of Science German–American Frontiers of Science, June 2–5, 2005, Irvine CA. UCRL-PRES-213584.

Chapman, H. N. (2005). *Spherical focusing mirror for the VUV–FEL*. UCRL-TR-215700.

Gaffney, K. J. et al. (2005). "Observation of structural anisotropy and the onset of liquidlike motion during the nonthermal melting of InSb." *Phys. Rev. Lett.* **95**, 125701. UCRL-JRNL-215427.

Hau-Riege, S. P. (in press). "Extending the size-parameter range for plane-wave light scattering from infinite homogeneous circular cylinder." *Appl. Opt.* UCRL-JRNL-211307.

Hau-Riege, S. P. et al. (2005). "Pulse requirements for x-ray diffraction imaging of single biological molecules." *Phys. Rev. E* **71**, 061919. UCRL-JRNL-211307.

Jacobsen, C. J. et al. (2005). *Towards community software for diffraction microscopy*. Intl. Workshop Phase Retrieval and Coherent Scattering, June 15–17, 2005, Porquerolles, Provence, France. UCRL-CONF-213196.

Peele, A. G. et al. (2005). *Coherent diffractive imaging using curved-beam illumination*. Intl. Workshop Phase Retrieval and Coherent Scattering, June 15–17, 2005, Porquerolles, Provence, France. UCRL-CONF-213203.

Starodub, D. et al. (in press). "Damped and thermal motion of laser-aligned hydrated macromolecules for diffraction." *J. Chem Phys*. UCRL-JRNL-215872.

Physics from the MIPP Experiment

Peter D. Barnes 05-ERD-007

Abstract

This research project will deliver the essential physics data needed to accurately model particles produced by high-energy protons interacting in thick objects. The data are needed to model the NuMI/MINOS neutrino beam at Fermilab, enabling high-accuracy neutrino measurements. The data are also needed for modeling proton radiography experiments. The project will support acquisition and analysis of the required physics data using the Fermilab E907 MIPP experiment—which can measure particle production and total cross sections from proton, pion, and kaon beams from 5–120 GeV/c momentum on nuclear targets from hydrogen to uranium—and also on the Fermilab NuMI Beam Target for the MINOS experiment.

The data obtained from the MIPP experiment will allow determination of the cross sections with 2% relative and 5% absolute error. The charged-particle cross sections obtained for radiography will allow high-accuracy modeling and analysis of past experiments at Brookhaven National Laboratory, and future experiments at facilities such as the Advanced Hydrotest Facility. The data obtained for NuMI/MINOS will allow a reduction of the neutrino spectrum systematic error (the dominant systematic error) from 10% to approximately 2%. The cross sections will be published and entered into LLNL databases and made available to simulation codes, and will enhance strong collaborations with DOE Office of Science national laboratory and university groups, including support for a Purdue graduate student.

Mission Relevance

The project will deliver total cross sections, particle production cross sections, and scattering distributions from proton, pion, and kaon interactions in the momentum ranges and on targets relevant to the Laboratory's stockpile stewardship mission. Simultaneously, these precision measurements will also enhance the physics yield from a DOE Office of Science priority, the MINOS experiment.

FY05 Accomplishments and Results

The FY05 work consisted primarily of operations and data taking. By the end of FY05, we had acquired a total of 13 million events from pion, kaon, and proton beams on the full set of experimental targets (liquid hydrogen, beryllium, carbon, bismuth, and the NuMI target). The data run will continue until approximately March 2006. At that time, we will have approximately 18 million events. The primary deliverable for FY05 was the priority 1 data set, which was accomplished in June 2005. We have also made major progress on the data analysis software. Data reconstruction software exists for all detector systems. In June we completed our first end-to-end reconstruction through the entire data sample in hand at that time. This was a test pass to identify deficiencies in the software.

Proposed Work for FY06

We will implement improved pedestal and gain corrections for data from the Cerenkov and time-projection chamber detectors. We will also approach completion on particle identification capability for every track in every event in these detectors. With the collaboration we will complete the data reconstruction software for the entire experiment. Accomplishing these goals will allow us to focus in the final year on acceptance and efficiency studies, the final cross sections, and table insertion.

Neutron-Capture Cross-Section Measurements at DANCE

Winifred E. Parker 05-ERD-011

Abstract

Important nuclear cross sections (n,gamma) are not well known and are very difficult to model. Isotopes important to the astrophysical s-process have no experimental measurements at all. Also, some isotopes of interest to stockpile stewardship have measurements that disagree with each other. We propose to perform neutron-capture cross-section measurements using radioactive targets, a novel and high-risk technique, using the Detector for Advanced Neutron Capture Experiments (DANCE) array, and the white neutron source at Los Alamos National Laboratory. Each measurement will be performed in two stages: target preparation and cross-section measurement. In addition, we will develop the ability to make radioactive targets.

We will increase knowledge of the properties of unstable nuclei, including level-density information and statistical theory of gamma-ray decay. In astrophysics, the impact of this work will be significant because current models give results that differ from global isotopic abundances by an order of magnitude. No measurements have been performed on the radionuclides that we propose to study. Furthermore, modern approaches involving quantification of margins and uncertainties (QMU) require improved physics input. If this new technique involving unstable nuclei as targets is successful, the problems of europium (Eu) and gadolinium (Gd) cross sections for stockpile stewardship calculations may also be solved.

Mission Relevance

This project supports the national security mission. If successful, the unstable target approach will directly impact stockpile science, as well as nuclear and astrophysical science. It will open the window to experimental data that apply directly to stockpile certification. Many detectors were added to nuclear devices to determine details of the nuclear detonation, and for many of the product radionuclides, cross sections for destruction are unknown. Therefore, our measurements will ultimately benefit QMU.

FY05 Accomplishments and Results

In FY05 we performed our first measurements using the array of barium fluoride crystals at DANCE and ^{151,153}Eu (stable) targets at a neutron energy range of 1 eV to 100 keV. We began analysis of these measurements, and the data gave information about the gamma cascade following neutron capture. Our first radioactive target to be made at LLNL will be americium-242m (^{242m}Am), and the material for this was purified in FY05. We also made (stable) natural Gd and ^{152,154}Gd targets. The ^{242m}Am target completion will be timed to match the experiment start, because it is a radioactive target. The ^{153}Gd radioactive target, intended to have been made in FY05, will be made in FY06 because the isotope production facility was closed for several months in FY05. Data collection for the natural Gd and ^{152,154}Gd (n,gamma) cross sections began in FY05.

Proposed Work for FY06

In FY06, we (1) expect to finish the data analysis for ^{151,153}Eu (n,gamma) measurements and publish the cross-section results; (2) perform ^{152,154}Gd and ^{242m}Am capture cross-section measurements at DANCE, and analyze the data from those experiments; (3) fabricate and run the next set of targets, including radioactive ^{153}Gd; and (4) investigate making a krypton-86 gas target. Results from the Eu cross-section data will resolve a 20-year discrepancy in existing data sets, and allow results from previous mix experiments to be correctly interpreted. The Eu data, together with the ^{152,154}Gd data will be used to describe the statistical theory of the gamma-ray decay of an excited nucleus. We intend to determine accurate cross sections for use in astrophysics and stockpile stewardship.

Publications

Agvaanluvsan, U. et al. (2005). *Investigation of the statistical properties of stable Eu nuclei using neutron-capture reactions*. Intl. School on Contemporary Physics-III, Aug. 8–12, 2005, Ulaanbaatar, Mongolia. UCRL-PROC-215969.

Agvaanluvsan, U. et al. (2005). *The energy and multiplicity correlation of resonances in ^{151,153}Eu(n,g) Reaction*. 2nd Joint Mtg. Nuclear Physics Divisions of the American Physical Society and the Physical Society of Japan, Maui, HI, Sept. 18–22, 2005. UCRL-ABS-212883.

Agvaanluvsan, U. et al. (2005). *The radiative strength function using the neutron-capture reaction on $^{151,153}Eu$*. 12th Intl. Conf. Capture Gamma-Ray Spectroscopy and Related Topics, Notre Dame, IN, Sept. 4–9, 2005. UCRL-PROC-215984.

Wilk, P. et al. (2005). *Gadolinium neutron-capture measurements with DANCE*. Interlaboratory Working Group 39, Los Alamos, NM, May 23–25, 2005. UCRL-POST-212490.

Hysteresis and Kinetic Effects during Liquid–Solid Transitions

Frederick H. Streitz 05-ERD-014

Abstract

Our goals for this project are threefold: (1) gain insight into the kinetics of solid–solid and liquid–solid transition; (2) determine the shape of the high-pressure and -temperature melt line for metals in general; and (3) understand how hysteresis during multiple crossings of the melt line affect the liquid–solid transition. Acquiring this information is the first step towards developing a dynamic equation-of-state (EOS) model. These goals will be accomplished by investigating the high-pressure resolidification of prototypical materials, such as iron (Fe), bismuth (Bi), tin (Sn), and water, through combinations of shock and isentropic compression in gas guns using graded-density impactors. In addition to these experiments, companion calculations will be performed at the hydrodynamic and atomic scales.

The success of this project would represent the first experimental measurement of the time scale for phase transitions under high pressure and temperature. Specifically, it will produce the first-ever look at the kinetics of pressure-induced transitions, including resolidification, in molten metals. The insights gained into the kinetics of solid–solid transitions and resolidification represent vital input for the validation of several simulation codes for stockpile stewardship, including the incorporation of a next-generation multiphase, multi-table EOS in hydrodynamic codes. This project will also produce the first-ever description of hysteresis effects during multiple crossings of the melt line, possibly introducing new physics.

Mission Relevance

This project supports the national security mission through increased understanding of the dynamics of materials under extreme conditions, particularly rapid resolidification in metals under conditions of high pressure and temperature, which is specifically applicable to stockpile science.

FY05 Accomplishments and Results

For the initial phase of this project, we began a study of the kinetics in a simple shock experiment with Bi solid–solid transitions, exploiting large changes in resistivity between the multiple low-pressure solid phases. We redesigned our target holder to minimize the signal-to-noise ratio using the results of test shots fired with a copper sample, which represents the worst case, as it has a much higher conductivity. With this newly designed target, we acquired very clean resistivity data in real time during the I-III transition in Bi. Experiments at different shock strengths showed the effects of kinetics during the transitions. By acquiring experimental data on a pressure-induced phase transition in Bi, we successfully achieved our goal for this first year.

Proposed Work for FY06

We will continue our study of solid–solid transitions in Bi and other materials using both conductivity and planar Doppler velocimetry, marking the first time such a study (with concurrent diagnostics) has been undertaken. By correlating particle velocity information with conductivity data (and the results of hydrodode simulations), we aim to achieve our goal of reproducibly identifying phase kinetics during a transition. Using multiple diagnostics as appropriate, we will begin exploring the kinetics of resolidification by shock melting and then compressing samples along an isentrope, using a combination of designer bullets and more conventional target design at the gas gun. Although we will focus on Bi, we aim to investigate solid–solid and liquid–solid phase transitions in Fe, if time permits.

Fully Atomistic Simulations of Hydrodynamic Instabilities and Mixing

Alison Kubota 05-ERD-020

Abstract

This project will use LLNL's large-scale computational capabilities to investigate the fundamental underlying connection between hydrodynamic instabilities and local, collective, and time-correlated behaviors and fluctuations at the atomic scales. Using Purgatory, a massively parallel classical molecular dynamics code that has been tested and validated for MCR and Thunder, and classical atomistic methods, the project will model hydrodynamic instabilities and mixing processes from the initial linear phase, through the nonlinear, to the final turbulent phase. We will perform simulations of the Rayleigh-Taylor, Richtmyer-Meshkov, and Kelvin-Helmoltz instabilities in three dimensions using up to several billions of atoms to understand the atomic-scale picture of these phenomena.

If successful, this study will provide novel insights on how to address the weaknesses and uncertainties in continuum-type fluid codes used at LLNL and elsewhere. We expect that this work will lead to a number of high-profile publications. This work demonstrates the enormous impact of large-scale computation on important cutting-edge scientific discovery and innovation.

Mission Relevance

By providing improved understanding for addressing applications such as the effect of plutonium aging on performance, fusion-class laser target design, and tera- and petascale code and visualization development, this work supports missions in stockpile stewardship, energy security, and breakthroughs in fundamental science and technology.

FY05 Accomplishments and Results

During FY05, we developed techniques that are essential to understanding large molecular dynamics simulations and connecting their results with experimental results. In addition, we developed methods to extract dynamic strength and elucidate the structure of shockwaves as they pass through materials and interfaces, which is relevant to the Richtmyer-Meshkov instability. This project has provided the basis for a close collaboration between LLNL and Washington State University to continue this work and further develop this capability to help understand and interpret the results of other experiments.

Publications

Kubota, A., D. Reisman, and W. G. Wolfer. (2005). *ASC supercomputers predict effects of aging on materials*. UCRL-TR-214867.

Kubota, A., D. Reisman, and W. G. Wolfer. (2005). *Effects of aging on shock and isentropic compression response in face-centered-cubic metals*. 14th Am. Phys. Soc. Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 31, 2005. UCRL-ABS-213504.

Optical Properties as a Real-Time In Situ Materials Diagnostic at Extreme Conditions

Jeffrey H. Nguyen 05-ERD-030

Abstract

Although characterizing the state of materials at subnanosecond time resolution is critical in experiments studying the time evolution of phase transitions, realistic diagnostics are scarce. This project will develop the use of optical properties as a diagnostic tool to obtain materials information, including inferred crystal structure, in real time from experiments at LLNL's gas gun. We propose to measure accurate optical properties and to leverage calculations of these same quantities that are funded by Advanced Simulations and Computing (ASC). Direct comparison between theory and experiment will enable the association of the observed optical properties with other materials properties by establishing spectroscopic fingerprints for each crystalline phase as well as the liquid.

The primary deliverable will be a diagnostic capability for analyzing and measuring in situ the structure of a material in real time as it is subjected to extreme temperatures and pressures in dynamics experiments on gas guns (e.g., at LLNL and JASPER), lasers, and Sandia's Z-machine. This diagnostic

capability will thus enable exciting new science. Moreover, the real-time nature of the diagnostic will enable us to infer structural quantities as they are changing on a subnanosecond time scale. This information is not only essential for understanding experiments conducted on very disparate time scales, from subnanosecond laser shocks to minutes or hours in a diamond anvil cell, but is crucial to developing a theory of dynamic phase transitions for incorporation into existing hydrodynamic simulation codes.

Mission Relevance

This project will develop a real-time, in situ diagnostic to characterize crystal structures with subnanosecond resolution for actinide research, in support of the stockpile stewardship mission. This diagnostic will be portable to experiments at other gas guns, high-power lasers, and the Z-machine to support basic science research. The project also leverages and supports ASC-funded theoretical effort on optical properties for stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we (1) measured optical properties with monochromatic light and detected various phase transitions in shock-compression experiments: iron (Fe) (alpha–epsilon), bismuth (Bi), and tin (Sn) (solid–liquid); (2) began analyzing results from water solidification in preparation for quasi-isentropic compression experiments; (3) developed software needed for the more complicated analysis of broadband experiments; (4) began the first broadband experiment; (5) completed calculations on some phases of Fe, Bi, and aluminum (Al); and (6) published a paper in *Physical Review B* that describes the optical properties of Al across the solid-to-liquid transition and demonstrates the efficacy of optical properties as a melt diagnostic.

Proposed Work for FY06

To identify crystal structures unambiguously, we will make broadband measurements of the optical properties. Building on work in FY05, in which we demonstrated the optical technique with monochromatic light, in FY06 we propose to fully develop the measurement and complex analysis of broadband optical properties. Phase transitions in Fe (solid–solid), Bi, and Sn (solid–liquid) are some of the candidates for these experiments. On the theory side, we propose to calculate the corresponding optical properties as a function of temperature and pressure. If successful, we should be able to identify crystal structures under extreme pressure and temperature conditions in real-time in gas-gun experiments.

Publications

Nguyen, J. H. (2005). *Recent advances in dynamic compression: tailored thermodynamic paths and high pressure-temperature optical properties*. UCRL-PRES-215633.

Nguyen, J. H. et al. (2005). *Crystal structure characterization using high-pressure, temperature-optical properties*. American Physical Society March Mtg. 2005, Los Angeles, CA, Mar. 21–25, 2005. UCRL-ABS-208363.

Nguyen, J. H. et al. (2005). *Crystal structure characterization using optical properties*. AIRAPT Intl. Conf. High-Pressure Science and Technology, Karlsruhe, Germany, June 5, 2005. UCRL-ABS-208816.

Nguyen, J. H. et al. (2005). *High-temperature, high-pressure emissivities*. 14th APS Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 5, 2005. UCRL-ABS-214205.

Patterson, J. R. et al. (2005). *Optical properties of shock-compressed materials*. American Physical Society March Mtg. 2005, Los Angeles, CA, March 21–25, 2005. UCRL-PRES-214217.

Advanced Studies of Hydrogen at High Pressures and Temperatures

William J. Evans 05-ERD-036

Abstract

The goal of this project is to study hydrogen at high pressures (megabars) and temperatures (kilo-Kelvin). Properties of hydrogen in this regime are important to a range of sciences, including condensed-matter theory and modeling, planetary science, and energy storage. High-pressure hydrogen has been the subject of intensive theoretical and experimental studies, both static and dynamic. Our high-pressure and high-temperature experiments bridge the unexplored gap between static and dynamic experiments. We are applying our recent advances in state-of-the-art high-pressure, x-ray, laser, and spectroscopic capabilities. These studies target important issues such as the equation of state at high pressures and temperatures, metallization, phase lines, and novel phase transitions.

The rich physics in the regime between dynamic and static studies are targeted by this proposal. Shock-wave studies (150 GPa, 3000 K) find a liquid metallic phase, while static work (300 GPa, 77 K) has identified only solid insulating states. The goal is to find the pressure and temperature states bridging these regimes that contain transitions in the properties of hydrogen that reconcile these disparate results—that is, the phase lines for melting, metallization, and dissociation. Further, we seek to measure the melt line, liquid–liquid (molecular–nonmolecular) transition, and the metallic fluid state predicted by theory. Such discoveries in hydrogen would impact our understanding for defense applications, Jovian planets, and hydrogen energy storage.

Mission Relevance

The deliverables of equation of state and phase transitions of high pressure and temperature hydrogen directly address needs in stockpile stewardship (specifically, the extreme dynamics of materials). These data will provide valuable data benchmarks for validating predictive theory efforts and code developments. Further, this work provides us with valuable experience and infrastructure for investigations of hydrogen energy storage, in support of LLNL's national and energy security missions.

FY05 Accomplishments and Results

After a midyear start, we (1) conducted measurements of high pressure and temperature solid phases and the melt line of hydrogen; (2) used our cryogenic loading capability to capture hydrogen in a high-pressure diamond anvil cell; and (3) measured the coherent anti-Stokes Raman spectra of deuterium up to 70 GPa, discovering sidebands on the vibron, which we tentatively identified as Anderson localized vibrons. This work positioned us to begin measurements of the melt line in FY06.

Proposed Work for FY06

In FY06 we will measure the pressure dependence of the hydrogen melt line, molecular bond energy, and structure of the liquid phase. For these measurements, we will use heating techniques, coherent anti-stokes Raman spectroscopy, and x-ray scattering. Building on our work on the solid phase in FY05, we will extend and further understand the liquid-phase properties.

Hydrodynamic, Atomic Kinetic, and Monte Carlo Radiation Transfer Models of the X-Ray Spectra of Compact Binaries

Christopher W. Mauche 05-ERD-044

Abstract

Our project models the x-ray spectra of compact (white dwarf, neutron star, and black hole) binaries by constructing highly detailed, three-dimensional (3-D) hydrodynamic models of the plasma flow in these binaries, calculating the x-ray spectra of every point in the flow, and transporting this radiation through the flow to the observer. These activities are being accomplished with the FLASH hydrodynamic package, LLNL atomic models, and our Monte Carlo radiation transfer code, using the massively parallel computing resources of LLNL. These models will be used to interpret the existing Chandra and XMM-Newton and future Constellation-X and XEUS high-resolution x-ray spectra of compact binaries.

If successful, the project will produce highly detailed, 3-D, time-dependent hydrodynamic models of the flow of plasma in compact binaries. These models will be used to produce the first realistic x-ray spectral models of compact binaries with sufficient detail to predict relative and absolute line strengths and line shapes as a function of binary phase. Although focused initially on high-mass x-ray binaries, the capabilities we will develop will be applicable to all types of x-ray sources dominated by photo-ionized plasmas. Therefore, the results will have far-reaching and long-term importance in x-ray astrophysics.

Mission Relevance

By providing hydrodynamic, atomic kinetic, and Monte Carlo radiation transfer models of the x-ray spectra of compact binaries, this project contributes to computer codes that simulate nuclear explosives for the stockpile stewardship mission.

FY05 Accomplishments and Results

In FY05, we (1) implemented the FLASH hydrodynamics package, tested it on various benchmark problems, and began doing simulations of high-mass x-ray binaries (HMXBs) using a 2-D Cartesian adaptive-mesh grid including the gravity of the neutron star, a phenomenological expression for the radiative driving of O-star wind, and Sutherland and Dopita radiative cooling; (2) used the photoionization code XSTAR to calculate, as a function of ionization parameter and temperature, the ionization balance and heating and cooling of wind photoionized by the neutron star; (3) began calculating the radiative driving of the wind; (4) began developing modules to incorporate these features into FLASH; and (5) began performing realistic 2-D simulations of HMXBs.

Proposed Work for FY06

During FY06, we will (1) extend our calculations to three dimensions; (2) implement radiation transfer in FLASH; (3) conduct a series of 3-D radiation hydrodynamic simulations of HMXBs; (4) use the resulting structures to perform Monte Carlo radiation transfer calculations of the observed x-ray spectra; (5) compare these theoretical spectra to Chandra and XMM grating spectra of various HMXBs; (6) investigate ways to adapt our code, and what we learn about radiation hydrodynamics, to other national-security-relevant activities; (7) hire a postdoctoral researcher, who will greatly expand our ability to utilize our code; and (8) begin to write papers on our results.

The Opacity of the Solar Interior

Pravesh K. Patel 05-ERD-045

Abstract

For this project we will make experimental measurements of the opacity of materials in the high-density, high-temperature regime of stellar physics. Opacity, which governs radiation transport through a material, is of fundamental importance in plasmas at very high energy densities and radiation-dominant regimes. However, opacity calculations are extremely complex, and little or no data exist for benchmarking models at high energy densities. We will obtain such data for the first time by utilizing a new set of ultrashort-pulse heating and x-ray backlighting techniques on next-generation petawatt-class laser facilities. Measurements will be made at solid density (2.3 to 8.9 g/cm^3), and at temperatures ranging from 20 to 100 eV.

At the conclusion of the project we expect to have obtained frequency-resolved opacity data for a set of heavy elements (carbon, nickel, and iron) abundant in the solar interior. For accurate comparison with modeling, we will also independently characterize the density, temperature, and uniformity of the heated samples. The results will access plasma densities at least two orders of magnitude higher than any previous measurements, and will provide the first experimental data able to validate LLNL opacity codes in this high-temperature, high-density regime.

Mission Relevance

This project will greatly enhance our understanding of the physics of high-energy-density plasmas. Improvements to opacity codes in this radiation-dominant regime are of central importance to LLNL's mission in stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we made progress in several key areas. A six-week experimental slot in November and December of 2004 on the 400-J petawatt laser at the Rutherford Appleton Laboratory in England enabled us to test four of the new techniques: (1) proton heating at high temperature; (2) proton beam collimation; (3) x-ray thermal backlighting; and (4) x-ray scattering. (Some of these research areas are being pursued in partnership with other LDRD projects.) The experiment itself involved 21 scientists from LLNL and 6 collaborating institutions. Good results were obtained in each of the four areas and we are now working intensively on data analysis and modeling using a combination of hydro, particle-in-cell, and radiation transport codes. This project has succeeded in attracting three young scientists to the Laboratory.

Proposed Work for FY06

In FY06, we will transition these tools to the new petawatt Titan laser at LLNL and continue development of individual techniques necessary for a fully integrated, frequency-resolved opacity experiment: construction of additional diagnostics, including a high-efficiency, extreme ultraviolet flat-field spectrometer and a time-resolved x-ray spectrometer. We also expect to advance our modeling capabilities as we analyze data from the development of the individual techniques, which in themselves should be highly publishable.

Split-Beam, Short-Pulse Final Optics and Characterization for High-Energy Short Pulses

Igor Jovanovic 05-ERD-060

Abstract

High-energy short pulses can be generated by large-scale neodymium (Nd):glass lasers in a chirped-pulse amplification scheme that utilizes a compact, folded compressor in a split-beam geometry. Recompression and focusing of high-energy, short pulses can be accomplished with novel multilayer dielectric (MLD) diffraction gratings and off-axis parabolic mirrors, respectively. The goals of this project are (1) the demonstration of MLD gratings and off-axis parabolic mirrors in split-beam, kilojoule-energy, short-pulse systems and (2) development of precision metrology for determining the alignment, in both space and time, of a focused split-beam pulse. This metrology is necessary for integrated tests under vacuum to determine damage thresholds of MLD diffraction gratings and mirrors.

We will develop several key technology components for the back end of a high-energy, short-pulse, split-beam laser system, and also perform prototype testing of those components. This project should solve most of the outstanding technology challenges associated with focusing and characterizing such laser systems. The split-beam concept represents a new paradigm for achieving very high-power, short-pulse laser operation. This work is expected to result in numerous publications and new intellectual property.

Mission Relevance

New back-end components and advanced metrological methods are relevant to the compression of multi-kilojoule pulses in chirped-pulse amplification systems for use in x-ray radiography, the scaling of high-energy, short-pulse systems to ~100 kJ using split-beam configurations with application to fast ignition, and the development of a novel pulse metrology capability for high-energy, short-pulse, split-beam systems, all in support of the Laboratory's national security (stockpile stewardship) and energy security (fusion energy) missions.

FY05 Accomplishments and Results

In FY05 we (1) developed a conceptual design for a diagnostics system table for a high-energy, split-beam short-pulse (HESBSP) system; (2) designed the beam path for delivery of the HESBSP system to the diagnostics system; (3) procured major commercially available components of the diagnostics system; (4) evaluated several commercially available components for use with the diagnostics system; (5) made significant experimental progress on compressor alignment diagnostics; (6) developed a concept for temporal diagnostics and commenced with construction of custom instruments required for HESBSP temporal metrology; and (7) made significant progress in commissioning of the diagnostics test laser.

Proposed Work for FY06

In FY06 we will (1) complete commissioning of the diagnostics test laser for HESBSP surrogate generation; (2) design and construct beam transport technology for transporting the diagnostics test laser to the diagnostics table; (3) complete design of the temporal diagnostics system; (4) construct a majority of the custom temporal diagnostics instrumentation; (5) complete purchases of commercially available instruments; (6) evaluate commercial diagnostics instruments performance; and (7) complete designs for integration of the diagnostics table in the HESBSP system.

Publications

Rushford, M. C. et al. (in press). "Diffraction grating eigenvector for translational and rotational motion." *Opt. Lett*. UCRL-JRNL-214177.

Rushford, M. C. et al. (2004). *Coherent addition of grating apertures: Out of plane metrology & issues*. 2004 Intl. Conf. Ultrahigh Intensity Lasers, Tahoe City, CA, Oct. 3–7, 2004. UCRL-ABS-206990.

Rushford, M. C. et al. (2004). *Coherent grating tiling via "lambda mount" all-optical encoder*. 2004 Intl. Conf. Ultrahigh Intensity Lasers, Tahoe City, CA, Oct. 3–7, 2004. UCRL-PRES-206992.

Precision Split-Beam, Chirped-Pulse, Seed Laser Technology

Christopher P. Barty 05-ERD-061

Abstract

High-energy-density (HED) science and inertial fusion are of strategic importance to national security. A significant enhancement of national HED capability would result by adding one or more high-power, short-pulse laser beams to existing and planned HED facilities. This proposal will provide critical technology that would enable such additions. Existing low-energy, seed laser technologies for high-energy, short-pulse lasers are unreliable and ill-suited for use with large-scale (e.g., 10 kJ per beam line) laser systems. This project will study, by constructing integrated subsystems, all issues required for the production of multiple-aperture, separately timed and dispersed seed pulses suitable for amplification in large-scale neodymium (Nd):glass laser systems.

We will develop seed laser technology for scaling high-energy, short-pulse laser beams on 40-cm-aperture Nd:glass laser systems across the DOE complex. This technology will enable the spatial multiplexing of amplified short pulses in the same beam line, a new paradigm for achieving very high power in short-pulse lasers. In addition to increasing the overall short-pulse energy extractable from a single beam line, this technology may significantly increase the utility of a short-pulse system by allowing multiple-time-frame x-ray imaging and other applications. This research is likely to result in numerous patents and publications.

Mission Relevance

The DOE complex has a large, nationwide investment in high-energy lasers, particularly high-energy, short-pulse lasers for research in high-field laser–matter interactions. This project, which to our knowledge is the only one in the DOE complex pursuing critical technology on the use of fiber lasers for short-pulse front ends, supports the Laboratory's national security mission, specifically stockpile stewardship.

FY05 Accomplishments and Results

In FY05, we (1) drafted a detailed design for a prototype seed laser system that was reviewed by an extensive team of HED experts to validate its compatibility with existing and proposed systems; (2) constructed an all-fiber mode-locked laser suitable for use in a split-beam seed laser system; (3) amplified chirped pulses to greater than 100 μJ; and (4) demonstrated that a chirped fiber Bragg grating can be used as a pulse stretcher with good final pulse compression.

Proposed Work for FY06

In FY06 we will integrate the components constructed in FY05 to create a complete prototype system. We will (1) design, build, and test appropriate diagnostics for this system; (2) fully test and characterize this system for use as a split-beam seed laser; and (3) test the utility of this system in generating coherent pulses that can be interferometrically summed. This latter task is of particular scientific significance because it is a pathway that would allow multiple beamlines on existing and planned DOE HED physics facilities to be summed to permit the exploration of new laser power regimes—for example, an exawatt laser system.

Publications

Dawson, J. W. et al. (2005). "Fiber laser front ends for high-energy short pulse lasers." *Proc. SPIE* **5709**, 37. UCRL-CONF-209779.

Dark Matter and Dark Energy Science

Leslie J. Rosenberg 05-ERD-063

Abstract

Three decadal surveys have shown the importance of large-aperture synoptic survey telescopes for time-domain and cosmological studies of distant objects. We propose to develop the basic tools and techniques for investigating "dark sector" cosmological science with the next generation of large-aperture, real-time telescopes. The critical research involves determining if extremely small distortions (shears) in images of faint background galaxies can be measured to reveal the distribution of dark matter and dark energy in the universe. If successful, this project would significantly enhance the prospects for building such a telescope and greatly enhance its scientific potential.

The limiting systematic uncertainties in cosmological observations are because of uncorrectable effects in the telescope point spread function (PSF) caused by the atmosphere. One anticipated result of this project will be the predicted star-corrected PSF components expected at a representative site, given the expected distribution of atmospheric parameter. The second result will be a program to determine if atmospheric models can be extrapolated to the huge aperture and long exposure time such a novel telescope would offer. This research will yield scientific publications and play a key role in determining the feasibility of building these synoptic telescopes (such as the proposed Large-Aperture Synoptic Survey Telescope and Panoramic Survey Telescope and Rapid Response System) for weak-lensing science.

Mission Relevance

The technologies required for a weak-lensing program on the scale of the next-generation instruments are directly relevant to LLNL's national security mission. In particular, large telescopes and gigapixel focal planes are required for the next generation of surveillance measurements. The algorithms and techniques used for weak-lensing analyses would also enhance capabilities that support nonproliferation.

FY05 Accomplishments and Results

The optical simulation team delivered two packages to model the sky, atmosphere, and instrument. One package features a high-fidelity model of the atmosphere; the other, an end-to-end capability to model sky through charge-coupled devices. The data we took at the Southern Astrophysical Research (SOAR) and Gemini telescopes in Chile are being analyzed, and two papers are in preparation: one on the Gemini images and the other on SOAR wavefront reconstruction. Both these analyses add to the confidence that coming instruments will achieve the weak-lensing scientific goals. The associated atmosphere turbulence and wind data have been reduced and cataloged. It is likely these data will be the focus of another round of papers and will integrate our data and modeling. Overall, we have established a basis of confidence in the capability of large, wide-field imagers for weak-lensing science.

Proposed Work for FY06

The main activity for FY06 will combine simulation components into a generally usable package. This package will allow other simulation teams to use this software for their design activity. In particular, these simulations are in demand by the data pipeline, data filter, and analysis groups from all the next-generation experiments. We will begin to generate and archive these simulated data, and also will apply the simulation tools to the feasibility of measuring small second-order PSF shifts (shear) across the image plan. We will also apply our SOAR and Gemini datasets to the question of how well our models simulate the atmosphere, a major activity in the coming year.

Development of Hot, LTE-Tunable Radiation Sources for Material Science Studies and Simulating Radiation Transport in Dense Astrophysical Plasmas

Marilyn B. Schneider 05-ERD-068

Abstract

This project seeks to develop a hot radiation source (radiation temperature ~350 eV) and to characterize this source using x-ray spectroscopy. A key feature of this source will be that design parameters can be changed to vary from local thermal equilibrium (LTE) conditions to highly non-LTE conditions. (In this case, LTE implies that the radiation and electron temperatures are equal.) This source can be used for materials studies under extreme conditions or can probe plasmas in astrophysical regimes, thus extending the range of plasma conditions that can be probed in laboratory experiments. The source will be designed by taking previously studied half-hohlraum (known as “halfraum”) cans heated by intense lasers, making them into pipes (or hohlraums), and putting a target in the center. This research will address several issues, including successfully shielding the target from hot electrons, holding back plasma filling, and developing the x-ray probes that can diagnose the plasma conditions (radiation temperature, electron temperature, electron density, mass density).

If successful, the project will result in a well-characterized radiation source with tunable “LTE-ness,” meaning that, with a radiation temperature of ~350 eV, the electron temperature can be tuned from 350 eV to ~10 keV. This source will provide the spectroscopic tools to diagnose plasma conditions such as those that occur in accretion disks around black holes, to accurately measure radiation brightness of materials under extreme conditions, and to probe how the material properties change as plasma conditions deviate from LTE.

Mission Relevance

This basic science research in high-energy-density physics will find applications in x-ray spectroscopy, astrophysics, opacity, and experiments with high-power lasers that support the stockpile stewardship mission. In addition, the project contributes to the Laboratory’s mission in basic science.

FY05 Accomplishments and Results

After a midyear start, we (1) developed a conceptual design to use the thin back wall of a hot hohlraum to drive a physics package; (2) tested the design at the Omega laser, measuring the radiation drive from the back wall for two hohlraum sizes; and (3) designed and fielded a hard-x-ray L-band spectrometer during these shots. The shots returned preliminary data showing it is feasible to spectrally resolve gold L-shell lines to obtain the charge state of gold and deduce the electron temperature. (Hot hohlraums produce very hot electrons from laser–plasma interactions, and these electrons can preheat a physics package). We also obtained the time history of suprathermal electrons for the first time.

Proposed Work for FY06

In FY06, we will (1) design a low-atomic-number (Z) witness plate whose thermal expansion and soft x-ray spectrum is sensitive to the radiation drive at the location of the physics package; (2) field the witness plate, driving it first by the single back wall of a hot hohlraum whose radiation drive is measured; (3) field the symmetrical configuration (i.e., witness plate between thin back walls of two hohlraums); (4) use the L-band spectrometer to measure Z and use the resulting data in atomic physics codes to deduce electron temperature; and (5) design, build, calibrate, and test a spectrometer to measure the thermal emission of the witness plate, and field the spectrometer at Omega.

Publications

Constantin, C. G. et al. (2005). “Laser-plasma interactions in high-energy-density plasmas.” *Proc. 4th Intl. Conf. Inertial Fusion*, Biarritz, France, Sept. 4–9, 2005. UCRL-CONF-214910.

Constantin, C. G. et al. (2005). *Stimulated Raman scattering from hot, underdense plasmas*. 32nd Intl. Conf. Plasma Science, Monterey, CA , June 19–23, 2005. UCRL-ABS-210353.

Schneider, M. B. et al. (2005). *Coupling of high power lasers to small, hot targets at the National Ignition Facility*. 32nd Intl. Conf. Plasma Science, Monterey, CA , June 19–23, 2005. UCRL-ABS-210351.

Schneider, M. B. et al. (2005). "X-ray flux and x-ray burnthrough experiments on reduced-scale targets at the NIF and Omega lasers." *Proc. 4th Intl. Conf. Inertial Fusion*, Biarritz, France, Sept. 4–9, 2005. UCRL-CONF-214849.

Widmann, K. et al. (2005). *Determination of the electron temperature of a hot gold plasma utilizing high-resolution x-ray spectroscopy of the L-band emission*. 32nd Intl. Conf. Plasma Science, Monterey, CA , June 19–23, 2005. UCRL-ABS-210248.

Technology Basis for Fluorescence Imaging in the Nuclear Domain

Christopher P. Barty 05-ERD-083

Abstract

We propose to develop the technical basis for a promising class of active interrogation techniques that would use new laser-based, gamma-ray sources to image the isotopic composition of well-shielded objects, and thus enable a novel and effective method of detecting concealed, highly enriched uranium (HEU). Isotopic selectivity can be accomplished by high-energy, x-ray/gamma-ray excitation and monitoring nuclear resonance fluorescence (NRF) from target nuclei. Research will include development of NRF experiments to determine useful fluorescent transitions in uranium-235 (^{235}U) and plutonium-239 (^{239}Pu) and modeling high-risk issues related to generating high-brightness, laser-based gamma rays and NRF-based nuclear material detection schemes.

Mission Relevance

Detection of HEU is the preeminent grand challenge in the area of domestic nuclear security. This work aims to provide a viable solution that can be fielded to address this challenge. In addition, the new gamma-ray sources required would be of use for a number of NNSA missions, including precision radiography of stockpile components, time-resolved study of energetic material dynamics, and precision metrology of advanced targets for large, fusion-class lasers.

FY05 Accomplishments and Results

In FY05 we (1) evaluated experimental techniques that would use existing LLNL hardware and allow the measurement of NRF transition locations, strengths, and widths in ^{235}U and ^{239}Pu; (2) numerically modeled various material detection schemes that use NRF based detectors; (3) numerically simulated broad-area NRF and Compton scattered light and possible large solid-angle energy discrimination detectors; and (4) designed laser and accelerator components, which will allow generation of 680-keV photons of sufficient brightness for NRF demonstration experiments using ^{238}U as the target material. For the latter, frequency conversion schemes for generation of precision ultraviolet pulses were investigated.

DEVELOPING THE PHYSICS BASIS OF FAST-IGNITION EXPERIMENTS AT FUTURE LARGE FUSION-CLASS LASERS

Michael H. Key 05-ERI-001

Abstract

This project will establish the physics basis, measurement techniques, and numerical designs for future integrated fast ignition (FI) experiments. The FI is an innovative approach to achieving fusion, and this research will be conducted in U.S. and foreign laser facilities, in collaboration with U.S. universities and other organizations. The team is comprised of experts in fast ignition, numerical modeling, and experiments. Advanced modeling of the short-pulse ignition process with particle-in-cell (PIC) and hybrid PIC codes will be benchmarked against experiments and used to assess short-pulse heating in the design of experiments at future, fusion-class lasers. Experiments will develop new diagnostics of electron isochoric heating and will study and optimize heating by proton beams.

The deliverables include optimized hydrodynamic designs for FI targets specific to large, fusion-class lasers, with near-term scaled designs based on both electron and proton ignition tested experimentally at Omega. This near-term work will provide a significant contribution to the forefront of FI research and experiments worldwide.

Mission Relevance

The work directly supports the national- and energy-security mission areas of stockpile stewardship and fusion energy by advancing work on short-pulse and high-energy petawatt lasers.

FY05 Accomplishments and Results

The project team (1) successfully carried out three experiments on the Rutherford Appleton Laboratory petawatt laser; (2) implemented new spectroscopic diagnostics for target heating in the 300- to 1000-eV range; (3) implemented simultaneous dual-wavelength, extreme ultraviolet (XUV) emission imaging and streaked 68-eV imaging of electron and proton isochorically heated plasmas, and are currently modeling these data using the Large-Scale Plasma (LSP) code; and (4) developed a hydro design for direct drive on the Omega extended performance (EP) laser. In FY05, 14 papers on FI were either submitted or have already been published in peer-reviewed journals, including *Nature* and *Physical Review Letters*.

Proposed Work for FY06

In FY06, (1) a zirconium–potassium alpha fluorescence diagnostic will be tested on the Callisto and Titan lasers over a range of FI-relevant temperatures; (2) hydro design of the proton FI capsule implosion at the Omega EP laser will be developed; (3) proton conversion efficiency and optimized proton focusing will be studied; (4) proton and electron designs will be scaled for Omega, using PIC and hybrid PIC techniques to study and optimize the electron source and electron transport in cone geometry; (5) diagnostic development will include dual-wavelength, streaked XUV emission imaging, implementation of spatially resolved extreme ultraviolet spectroscopy, and development of streaked fluorescence imaging at 4.5 or 8 keV; and (6) the PSC code will be applied to proton-focusing studies and the LSP code adapted to model proton focusing and energy deposition.

Publications

Allen, M. (2004). “Direct experimental evidence of back-surface ion acceleration from laser-irradiated gold foils.” *Phys. Rev. Lett.* **93**, 0265004. UCRL-JRNL-204174.

Gregori, G. (2005). “Experimental characterization of a strongly coupled solid density plasma generated in a short-pulse laser target interaction.” *Contrib. Plasma Phys.* **45**, 285. UCRL-JRNL-211077.

Habara, H (2004). “Ion acceleration from the shock front induced by hole boring in ultraintense laser–plasma interactions.” *Phys. Rev. E.* **70**, 046414. UCRl-JRNL-155688.

King, J. A. et al. (2005) “Characterization of a picosecond laser generated 4.5 keV Ti K-alpha source for pulsed radiography.” *Rev. Sci. Instrum.* **76**, 076102. UCRL-JRNL-210865.

Mackinnon, A. J. (2004). “Proton radiography as an electromagnetic field and density perturbation diagnostic.” *Rev. Sci. Instrum.* **75**, 4183–4186. UCRL-CONF-203614.

Parks, H. S. et al. (2004) “Characteristics of high-energy Kα and bremsstrahlung sources generated by short-pulse petawatt lasers.” *Rev. Sci. Instrum.* **75**, 4048–4050. UCRL-CONF-203568.

Stephens, R. (2005). “Implosion hydrodynamics of fast ignition targets.” *Phys. Rev. E.* **69**, 066414. UCRL-JRNL-208003.

Tabak, M. (2005). “Review of progress in fast ignition.” *Phys. Plasmas* **12**, 057305. UCRL-CONF-207839.

Measuring Plasmon Density of States in Warm Dense Matter

Siegfried Glenzer 05-ERI-003

Abstract

This research conducts novel x-ray scattering experiments that demonstrate the measurement of optical properties of dense matter. Almost any type of dense material will be accessible by this novel technique, including hot, solid-density plasmas; compressed matter; and cold matter under high pressure such as occurs in inertial confinement fusion, laboratory astrophysics, materials science, and high-energy density experiments. The experiments will be conducted on multiple laser and synchrotron facilities, in collaboration with research teams at the University of California at Berkeley, Los Angeles, and San Diego.

The project is expected to accomplish five milestones: (1) measure plasmon states in cold dense matter; (2) demonstrate improved data quality and accuracy using the narrow-band scattering technique in the noncollective regime; (3) measure the plasmon spectra in hot isochorically heated plasmas and in shock-compressed matter; (4) test theories of dense matter with plasmon spectra and independent measurements of temperature and density using noncollective scattering; and (5) achieve the complete determination of the plasmon density of states in cold matter and in dense plasmas. The results will be used to test theoretical models in regimes where no experimental data have been obtained previously.

Mission Relevance

Measuring the optical properties of dense matter will provide experimental data in high-energy-density regimes that are important for the stockpile stewardship mission. This project supports the Laboratory's mission in breakthrough science, nurtures collaborations with the high-energy-density physics research community, and attracts talented scientists to the Laboratory.

FY05 Accomplishments and Results

In FY05, we began experiments at the Omega laser in Rochester, New York, where we have demonstrated suitable conversion efficiencies for laser to chlorine Lyman alpha radiation and fielded a high-efficiency spectrometer for measuring forward x-ray scattering. The first experiments demonstrated accurate data of the spectral bandwidth and the dielectronic satellite contribution. The results show that this line is suitable to measure plasmon spectra that will uniquely test the theory of dense matter. We have submitted three papers to peer-reviewed journals and presented three invited talks. Also, Siegfried Glenzer received a Humboldt Research Award from the Alexander von Humboldt Foundation in Bonn, Germany.

Proposed Work for FY06

In FY06, the first experiment we are planning will demonstrate benchmark data of plasmon spectra at the Omega laser. In addition, the first experiments on cryogenic deuterium and on lithium hydride will be performed to measure the compressibility directly with x-ray scattering. Experiments on other laser facilities will include Rutherford Appleton Laboratory and free-electron lasers at the Deutsche

Elektronen Synchrotron Facility in Hamburg, Germany. These measurements will be the first to demonstrate the density measurement from the shift of the plasmon peak in compressed matter. We expect a publication in *Nature* or *Physical Review Letters* at the end of FY05.

Publications

Gregori, G. et al. (2005). “Experimental characterization of a strongly coupled solid density plasma generated in a short-pulse laser target interaction.” *Contrib. Plasma Phys.* **45**, 284. UCRL-JRNL-211077.

Pak, A. et al. (2005). “X-ray line measurements with high-efficiency Bragg crystals.” *Rev. Sci. Instrum.* **75**, 3747. UCRL-CONF-203748.

Understanding the Nuclear Magnetic Fields

Elmar C. Trabert 05-LW-006

Abstract

There is reasonable doubt whether recently developed models of magnetic field generation in nuclei are correct, since a spectral line predicted by these models could not be found in a laser-excitation experiment. Correct models are needed for the interpretation of parity non-conservation (PNC) experiments, as well as for nuclear materials and energy research. We propose to conduct high-precision measurements of the hyperfine structure in praseodymium (Pr), thallium (Tl), and bismuth (Bi) to provide data needed to develop the correct nuclear models and to distinguish between current models for n=2 electron energy splittings in lithium- (Li-) like ions. The experiment, carried out at the LLNL SuperEBIT electron beam ion trap, utilizes passive emission spectroscopy using a high-resolution soft-x-ray spectrometer.

Standard Model uncertainties stem largely from the uncertainties with which various nuclear parameters are known. An improved knowledge of these parameters will be essential for further progress with PNC experiments. We expect to achieve data that are precise enough to distinguish between recent predictions, and thus meaningful for tests of the Standard Model. The results will guide improved descriptions of the nuclear-atomic interaction. Due to the level of worldwide scientific interest in PNC, we expect to publish our results in high-profile, peer-reviewed journals.

Mission Relevance

The proposed work, at the interface of atomic and nuclear physics, supports the Laboratory’s stockpile stewardship mission and reinforces LLNL’s mission in breakthrough science and technology. The project encourages collaborative efforts for the precision study of nuclear-atomic interactions and will attract talented scientists to the Laboratory.

FY05 Accomplishments and Results

In FY05 we (1) successfully upgraded our instrumentation—extending the upper cutoff of the spectrometer to 65 Å, achieving optimal focusing, and improving resolving power from 1200 to 3000, which was well beyond our target; (2) successfully injected boron into SuperEBIT; (3) demonstrated spectral calibration of our Tl and Bi measurements with boron and nitrogen lines; (4) began measurement of Bi and Tl; (5) successfully identified hyperfine splitting in Bi; and (6) began the analysis and writeup of the Bi data.

Proposed Work for FY06

In FY06, our instrumentation will be redesigned to allow measurements from about 50 to 140 Å; the current design allows measurements only between 10 and 65 Å. The new design will feature a slanted exit plane on the grating housing. We will demonstrate our new capability with a measurement of the 0.20-eV splitting of ^{141}Pr (atomic number = 59). This measurement of the hyperfine splitting far from the doubly magic ^{208}Pb (atomic number = 82) nucleus will be a very stringent test of nuclear field models, as they need to deal with a complicated open nuclear shell. We aim to publish our results in the peer-reviewed literature, because understanding field generation in the nucleus is fundamental to nuclear materials and nuclear energy research in particular and to the building blocks of nature in general.

High-Pressure, Multi-Mbar Conductivity Experiments on Hydrogen: The Quest for Solid Metallic Hydrogen

Damon D. Jackson 05-LW-027

Abstract

Ultradense hydrogen has long been the subject of intense experimental and theoretical research because of, in part, the fascinating physics that arise from this seemingly simple system. Metallic hydrogen has long been considered to be the prototypical system for the study of insulator-to-metal (I-M) transitions, and has been predicted to have a high-pressure, high-temperature superconducting transition near 100 K. We plan the first direct measurement of the expected I-M transition above 3 Mbar using designer diamond anvils, membrane diamond anvil cells, and the cryogenic loading of hydrogen. These three techniques were developed and are in routine use at LLNL, and will be combined for the first time to investigate the possibility of solid metallic hydrogen.

Mission Relevance

A detailed understanding of the properties of hydrogen at high pressure is important to stockpile stewardship and will also benefit work in planetary science and hydrogen energy storage in support of LLNL missions in breakthrough science and energy security.

FY05 Accomplishments and Results

We integrated LLNL technologies to both pressurize hydrogen and measure its electrical resistivity. This included adapting membrane diamond anvil cells, cryogenic loading of samples, and designer diamond anvils so that they all worked together as one unit. We also developed new techniques to measure the photoconductivity of samples under pressure using designer diamonds. All of this has laid the groundwork for pressurizing materials and investigating their I-M transitions.

Proposed Work for FY06

We will focus our efforts on pressurizing hydrogen up to the 3-Mbar region required for detecting its I-M transition. This will involve modifying the diamonds so that they have the optimum geometry required to reach these pressures. We expect that several loadings will be required before we successfully reach these pressures—therefore, we will require the full year to obtain the best possible results.

Fiber Laser Replacement for Short Pulse Titanium: Sapphire Oscillators—Scalable Mode Locking to Record Pulse Energies

Jay W. Dawson 05-LW-062

Abstract

Fiber lasers are enabling a new generation of higher average power, more robust solid-state lasers. Although titanium (Ti):sapphire lasers are the de facto standard in ultrafast solid-state lasers, fiber lasers may offer performance (100 fs, 15 nJ, 1 W) rivaling that of Ti:sapphire lasers. To date, a physical demonstration of this was limited in pulse energy to 5 nJ due to the mode-locking mechanism. We propose to demonstrate a new and improved mode-locking mechanism based on waveguide bend-induced loss by building a fiber laser with an order of magnitude more energy per pulse than any other fiber laser in existence.

We expect to demonstrate a novel nonlinear optical switch and use it as a mode-locking mechanism in constructing the highest pulse energy fiber laser ever built (~500 nJ). This new laser would be less expensive, easier to use, more compact, and have higher output energies and powers than Ti:sapphire short-pulse lasers. Further, the resulting laser will provide a scalable pathway to oscillators with much higher pulse energies that will eliminate the need for the initial amplification stages that typically follow oscillators in high-energy laser systems. We expect this work to lead to several patents and publications.

Mission Relevance

Fiber-laser, short-pulse lasers with record pulse energies would find applications for the stockpile stewardship mission, including direct micromachining of parts with an oscillator, waveguide writing, multiphoton microscopy, and petawatt systems with reduced amplified spontaneous emission.

FY05 Accomplishments and Results

We were able to increase the energy of pulses emitted by a conventional, nonlinear polarization evolution oscillator to 25 nJ. The higher pulse energy exceeded our previous best by an order of magnitude and all previous reports by nearly a factor of two. We believe that we succeeded in creating the "self-similar" pulses recently reported by another group. In nonlinear bend-loss measurement, we used an amplifier to create 1-μJ pulses and measured bend loss as a function of pulse energy to verify that nonlinear bend loss can be used as a pulse discriminator. However, these measurements suffered repeatability issues.