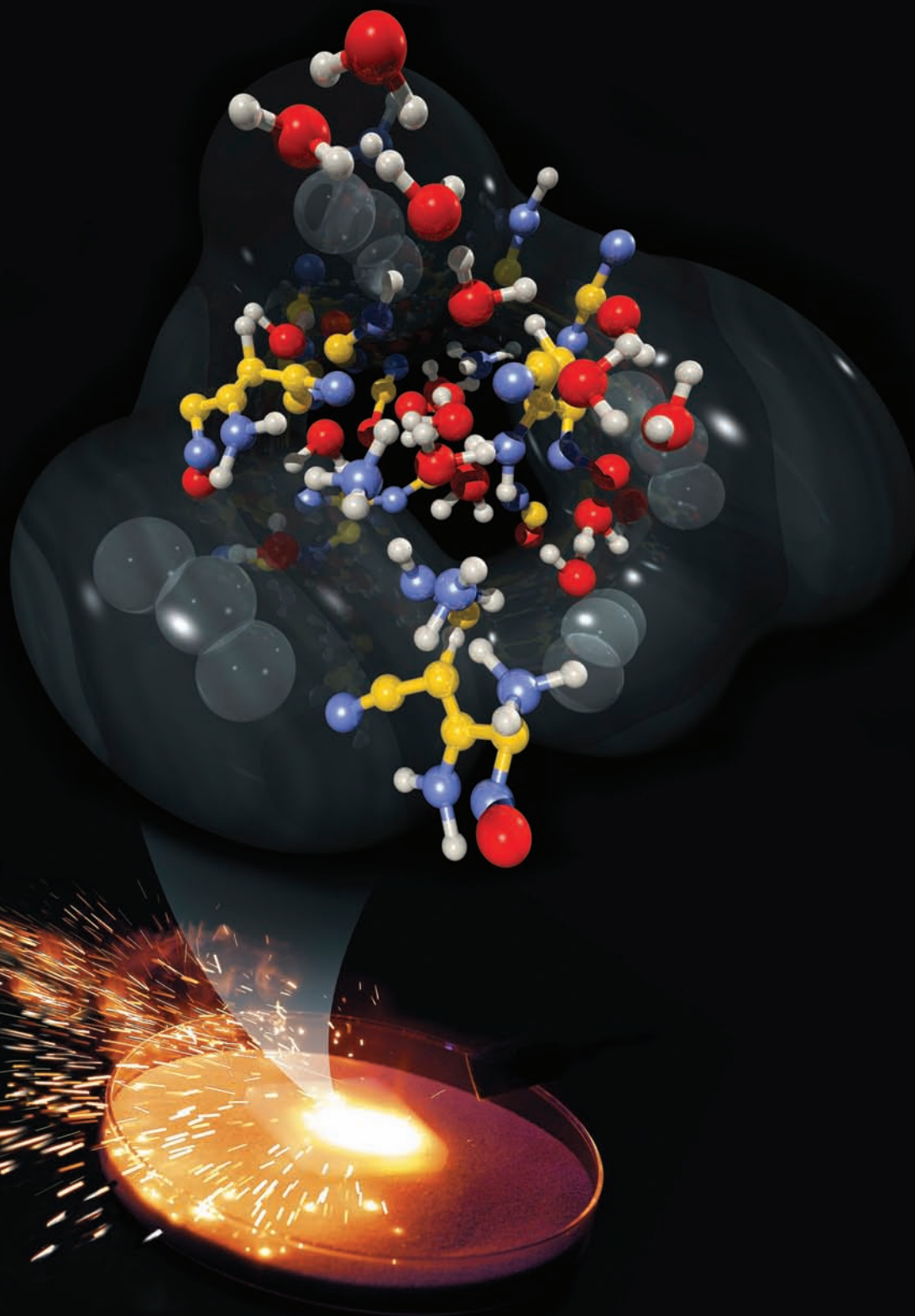


Laboratory Directed Research and Development
FY2007 **Annual Report**



Lawrence Livermore National Laboratory

About the Cover:

Sparks are emitted from rapid chemical reactions in a high-explosive material. This microscopic view shows a mixture of reaction intermediates observed during a computer simulation of nitromethane as it is detonated. Despite the extensive production and use of explosives for more than a century, their basic microscopic properties during detonation have not been unraveled. Researchers for LDRD project 06-SI-005, "Transformational Materials Initiative," have obtained an initial glimpse of explosives properties by performing the first quantum molecular dynamics simulation. They discovered that the explosive nitromethane undergoes a chemical decomposition and a transformation into a semimetallic state for a limited distance behind the detonation front. Their findings appeared in the December 9, 2007 online edition of the journal, *Nature Physics*. The project is devoted to providing the underlying science and technology for converting the U.S. nuclear weapons complex to one that is smaller, safer, and more agile.

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Acknowledgments

The LDRD Program extends its sincere appreciation to the principal investigators of the fiscal year 2007 projects for providing the content of the annual report and to the publications team. The LDRD Program also thanks the following members of the Laboratory Science and Technology Office for their many contributions to this publication: Dolores Lambert, administrator; Nancy Campos, database manager; Steve McNamara, computer specialist; and Cathleen Sayre, resource manager. We extend particular thanks to Rokaya Al-Ayat, who successfully directed the LDRD Program in 2007 and for many previous years.

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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



George Miller, Director

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 \$92.7 million for fiscal year 2007 sponsored 159 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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Laboratory Directed Research and Development

Overview



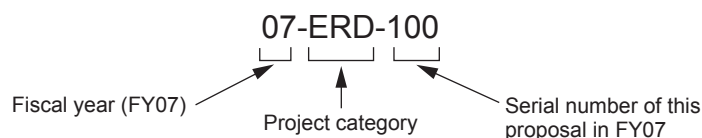
About the *FY2007 Laboratory Directed Research and Development Annual Report*

The Laboratory Directed Research and Development (LDRD) annual report for fiscal year 2007 (FY07) 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 Department of Energy (DOE)/National Nuclear Security Administration (NNSA) and Lawrence Livermore National Laboratory (LLNL) mission areas, the technical progress achieved in FY07, and a list of publications that resulted from the research in FY07.

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

About Lawrence Livermore National Laboratory

A premier applied-science laboratory, LLNL 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 DOE/NNSA laboratories, LLNL is managed by the Lawrence Livermore National Security, LLC. Since its inception in 1952, the Laboratory has fostered 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 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.

At LLNL, Laboratory Director George Miller and Deputy Director for Science and Technology Cherry Murray were responsible for the LDRD Program in FY07, and delegated 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, Rokaya Al-Ayat. The LDRD Program at LLNL is in compliance with *U.S. Department of Energy Order 413.2B* and other relevant DOE orders and guidelines.

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

The LDRD Portfolio Management Process

The FY07 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 FY07, the top-level LDRD strategic planning process was guided by the *2006 U.S. Department of Energy Strategic Plan*² and by the Laboratory's own long-range plan that will define the scientific and technical strategy for the coming decade. The DOE strategic plan articulates strategic themes for achieving the DOE mission of discovering solutions to power and secure America's future. In FY07, the Laboratory's LDRD Program strongly supported the DOE strategic themes:

1. **Energy Security**—Promoting America's energy security through reliable, clean, and affordable energy.
2. **Nuclear Security**—Ensuring America's nuclear security.
3. **Scientific Discovery and Innovation**—Strengthening U.S. scientific discovery, economic competitiveness, and improving quality of life through innovations in science and technology.
4. **Environmental Responsibility**—Protecting the environment by providing a responsible resolution to the environmental legacy of nuclear weapons production.

The Laboratory's long-range strategic science and technology (S&T) plan continues to guide 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).

The DOE/NNSA oversees the Laboratory'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. The value of LDRD to DOE as well as to the country is evidenced in a DOE 2007 report to

² 2006 U.S. Department of Energy Strategic Plan. <<http://www.energy.gov/about/strategicplan.htm>> (retrieved April 1, 2008).

Congress: “The DOE LDRD program offers a flexible mechanism by which the multi-program national laboratories maintain their vitality and, in the process, prepare themselves to help address the Nation’s future scientific and engineering challenges. . . .The flexibility inherent in the LDRD program is essential to maintaining the vitality of the laboratories that carry out the Department’s missions and national 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 LDRD 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 FY07, 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 FS projects throughout the year.

³ Department of Energy (DOE) FY 2007 Report to Congress—Laboratory Directed Research and Development at the DOE National Laboratories. <<http://www.cfo.doe.gov/CF1-2/Final%20FY%202007%20LDRD%20Report.pdf>> (retrieved February 27, 2008).

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 2007 Portfolio

Portfolio Overview

The FY07 LDRD portfolio was carefully structured to continue the LDRD Program's vigorous support for the strategic vision and long-term goals of DOE/NNSA and LLNL. The FY07 projects described in this annual report underwent a stringent selection process and received ongoing management oversight.

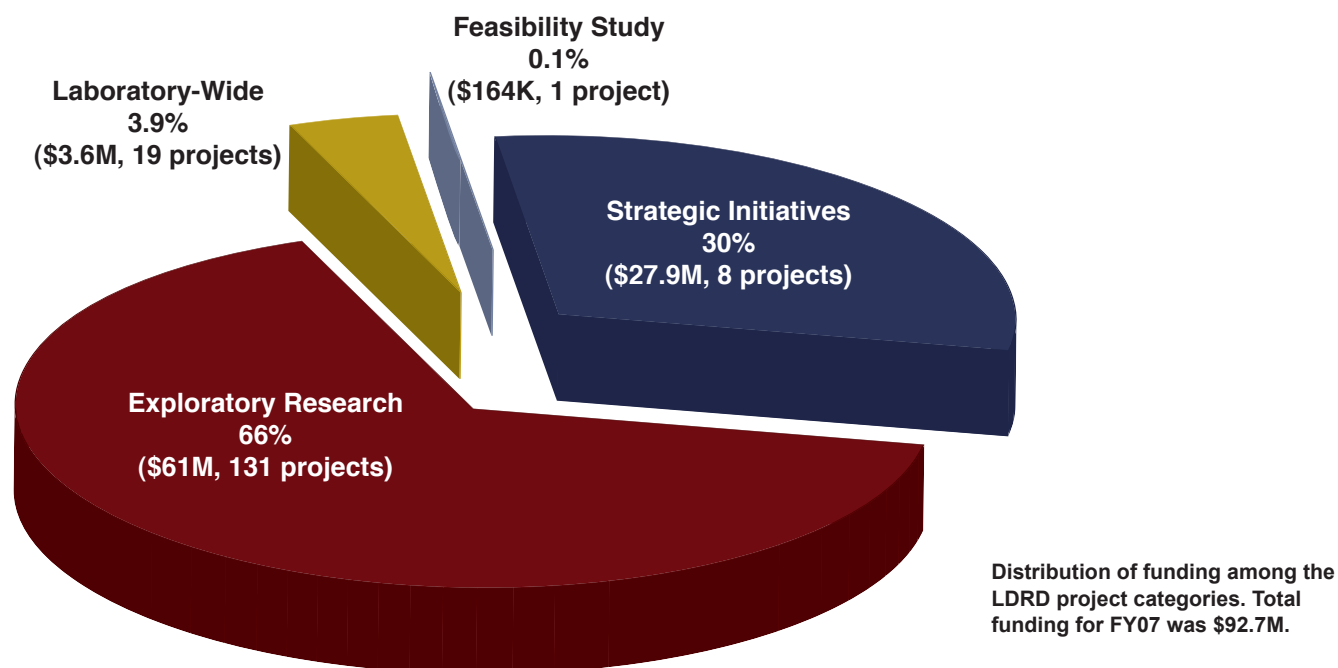
In FY07 the LDRD Program funded 159 projects with a total budget of \$92.7 million. The distribution of funding among the LDRD project categories is shown in the pie chart on page 7.

Strategic Initiative

In FY07, the LDRD Program funded eight SI projects. Although the SI category represented only 5% of the total number of LDRD projects for FY07, it accounted for 30% of the budget. SI projects ranged in funding from \$1.8 to \$7.5M.

Exploratory Research

The LDRD Program funded 131 ER projects for FY07. The largest project category, ERs accounted for 82% of the number of LDRD projects and 66% of the budget for the fiscal year. Projects in this year's ER category ranged in budget from \$24K to \$4.8M.



Laboratory-Wide Competition

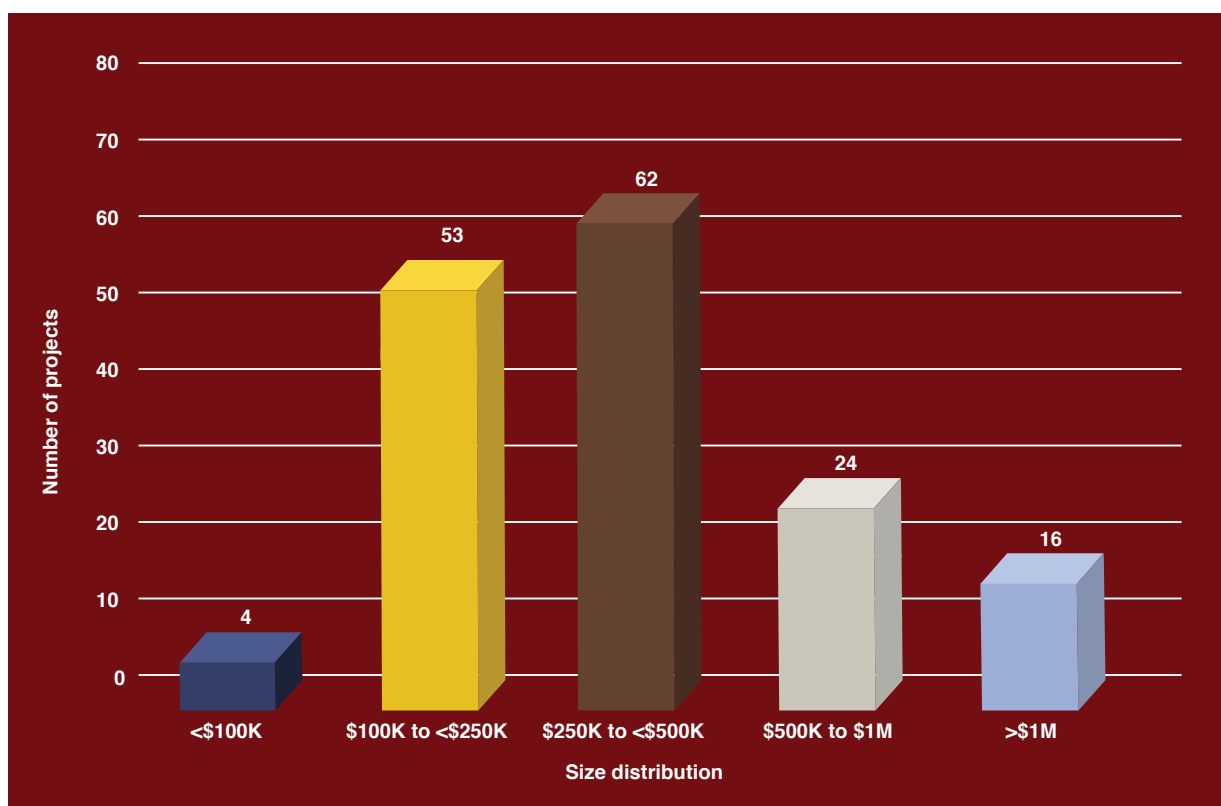
In FY07, 19 LW projects were funded, which represent 12% of the LDRD projects for the year and 3.9% of the budget. LW projects are limited to \$225K/year funding, with a few exceptions. The LW projects in FY07 ranged in funding level from \$122 to \$227K.

Feasibility Study

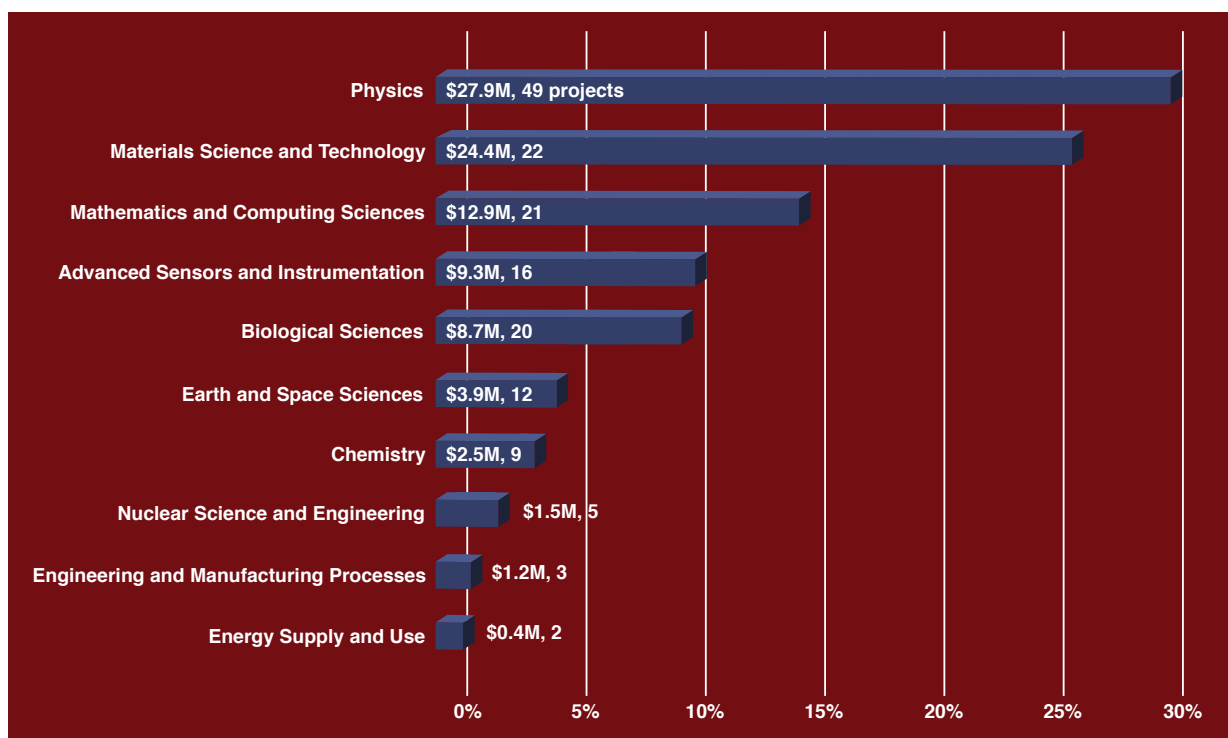
The LDRD Program funded just one FS project for \$64K in FY07. FS projects are limited to \$125K and a 12-month duration.

The top chart on page 8 shows the funding distribution by dollar amount for the 159 FY07 projects—72% of the projects were in the \$100 to \$500K range, with 2.5% falling below \$100K. Projects in the \$500K to \$1M funding range accounted for 15% of the total, and another 10% of the projects received more than \$1M. The average funding level for the 159 projects was \$583K.

The percentage of LDRD funding and number of projects in each research category for FY07 is shown in the bottom chart on page 8.



Number of projects and levels of funding. The average funding level for an LDRD project in FY07 was \$583K.



Percentage of LDRD funding and number of projects in each research category in FY07.

Highlights of 2007 LDRD Accomplishments

In FY07, 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 FY07 highlights that exemplify LDRD's noteworthy research results, timely support for the Laboratory's S&T plan as well as critical national needs, and external recognition of Laboratory personnel.

Stockpile Science and Technology

"Transformational Materials Initiative" (06-SI-005)

Randall L. Simpson, Principal Investigator

This project is developing science and technology to help make the U.S. nuclear weapons complex smaller, safer, and more agile. It will also help ensure the continued success of the Stockpile Stewardship Program—one of the Laboratory's key missions. To this end, the project team is creating new materials, processes, and diagnostics to reduce cost and time required to produce and maintain the stockpile, enhance weapon safety, ensure future stockpile longevity, and optimize stockpile performance. The multidisciplinary team combines capabilities in materials synthesis, characterization, theory, and modeling to deliver cutting-edge advances in high explosives, multifunctional materials, metals, and sensing.

Achievements made in FY07 include preparing a new explosive composite with improved quality, completing new simulations of shocked high explosives, preparing test specimens of advanced metals, developing three sensing technologies, and conducting preliminary testing of a capability to monitor criteria of interest. High-explosive loading results showed novel and promising results in material ductility.

The team's ambitious plans for FY08 include scaling up the manufacture of the new high explosives to the kilogram level to test detonation and low-temperature mechanical properties, extending first-principles studies on the detonation of nitromethane to triamino-trinitro-benzene, demonstrating



Livermore's Transformational Materials Initiative team combines expertise in materials synthesis, characterization, theory, and modeling to advance the technology of high explosives, multifunctional materials, metals, and sensing. Their goal is to ensure the continued success of the Stockpile Stewardship Program by enabling a smaller, safer, and more agile U.S. nuclear weapons complex.

communication to a microelectromechanical systems-based load sensor for realistic materials, refining simulations and understanding of the effects of processing parameters on high-explosives loading response, and synthesizing a unified, catalytically active polymer nanocomposite.

High-Energy-Density Science and Technology

“Nonequilibrium Phase Transitions” (04-ERD-108)

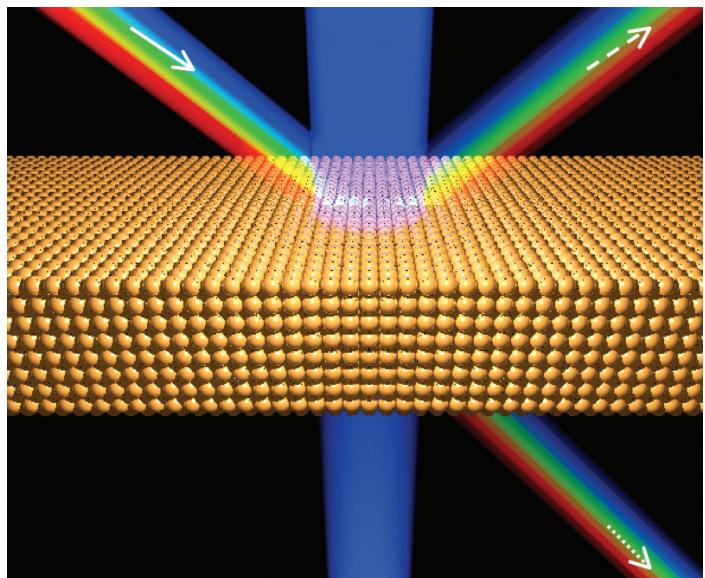
Andrew Ng, Principal Investigator

The study of phase transitions in an extreme, nonequilibrium regime is a scientific frontier that holds promise for discovering new phases, metastable states, chemical reaction pathways, and biological functioning processes. The team is examining lattice disordering and melting, quantifying the role of electronic excitation on phase-transition kinetics, and developing approaches in finite-temperature condensed matter for constructing an equation of state. In collaboration with the University of Toronto, the project is using measurements to correlate optical and structural properties under ultrafast laser excitation to help develop density functional theory approaches.

Deliverables include time-correlated data on materials’ optical and structural properties. The data will be generated by tracking solid–liquid to liquid–plasma transitions under ultrafast excitation conditions and will be used to benchmark quantum simulations. This will lead to a new understanding of the connection between electronic (optical) and atomistic (structural) behavior, opening up possibilities of manipulating phase stability and boundary. Success in this area also will help elucidate the convergence of condensed matter and plasma physics, a critically missing link in basic scientific understanding. This work supports the Laboratory’s mission in stockpile stewardship.

In FY07 the team completed a 600-femtosecond, 30-kiloelectronvolt electron gun and demonstrated the viability of single-shot, ultrafast electron diffraction measurement with gold nanofoils heated by a 400-nm laser beam. In addition, new findings elucidated ionization dynamics and nonequilibrium effects on phonon modes. Overall, the project’s most notable achievement is the discovery of quasi-steady states of superheated lattice with highly excited nonequilibrium electrons. This fundamentally changes the existing understanding of warm dense matter produced by ultrafast excitation of a solid using laser

For the first systematic study of phase transitions in an extreme, nonequilibrium regime, researchers have demonstrated a 30-nm-thick freestanding gold nanofoil that is heated with a femtosecond, 400-nm laser beam (blue) to produce a nonequilibrium, high-energy-density state. It is then probed by a frequency-chirped, broadband (450- to 800-nm) laser beam (blue to red) in reflection and transmission measurements.



x-ray-charged particles. The team also discovered the persistence of band structure in athermally melted metals, which was prominently featured in journals such as *Nature of Materials News*. These achievements played a large part in the project's principal investigator receiving an LLNL Science and Technology Award.

Nuclear, Radiative, and Astrophysical Science and Technology

“Finding and Characterizing Rare Events in Two Next-Generation Particle Astrophysics Experiments” (07-ERD-056)

Adam Bernstein, Principal Investigator

This project team is collaborating on a larger effort to resolve one of the most important scientific issues of the 21st century—direct detection of dark matter. Another goal is measurement of the neutrino-oscillation parameter θ_{13} , which has nearly the same level of scientific significance. To this end, the team is building detectors with unprecedented levels of sensitivity and with record low levels of systematic error. The skills being developed in this project have direct relevance to Laboratory global nuclear security missions. In particular, the advanced simulation capabilities, event-selection algorithms, and advanced detection concepts being pursued have immediate relevance to ongoing reactor monitoring and special nuclear material detection efforts by the DOE and the Department of Homeland Security. In addition, this project increases LLNL's already significant competency in the detection of antineutrinos—which are a potentially valuable signature for reactor monitoring and treaty verification—and will be concurrently applied to nonproliferation efforts.

At the San Onofre Nuclear Generating Station, the team is working to measure an as-yet undetected antineutrino interaction process, known as coherent neutrino–nucleus scattering. To this end, the team has deployed a radiation detector with unprecedented sensitivity to low-energy nuclear recoils. The detector contains a noble liquid target and is being used to measure the faint but high-probability coherent scatter that induces these recoils. The high rate per unit volume of this weak signal allows



Louisiana State University collaborator William Coleman, LLNL principal investigator Adam Bernstein, and LLNL co-investigator Steve Dazeley (left to right) with a prototype gadolinium-doped water Cerenkov antineutrino detector developed to explore rare events in next-generation particle astrophysics. The device detected antineutrinos emitted from the San Onofre Nuclear Generating Station. Antineutrinos interacting in the water generate a unique pattern of faint blue flashes of Cerenkov light, which the detector records.

a ten- to hundred-fold reduction in the size of the detector, making tabletop antineutrino detection a realizable goal in the coming years.

In FY07, the team achieved a world-record limit on the detection of supersymmetric dark matter in a liquid-xenon detector, beating the previous record by nearly an order of magnitude. The limit was set with a 10-kg dual-phase xenon detector, and the results published in *Physical Review Letters*. In collaborating with other organizations, the team also began planning a 300-kg xenon detector for the Homestake Deep Underground Science and Engineering Laboratory in South Dakota. This includes designing the electronics and simulating the performance of the 300-kg detector. A custom detector Monte Carlo capability was also developed for detailed calibration studies with the Double Chooz detector in France.

In FY08, the team will simulate, assemble, and deploy a prototype of the 300-kg dark-matter detector and perform simulation and design work for a 2-m-diameter gadolinium-doped water shield to screen backgrounds from this highly sensitive detector. Simulation capability for the Double Chooz antineutrino detector will also be expanded.

Science and Technology at the Intersection of Chemistry, Biology, and Materials

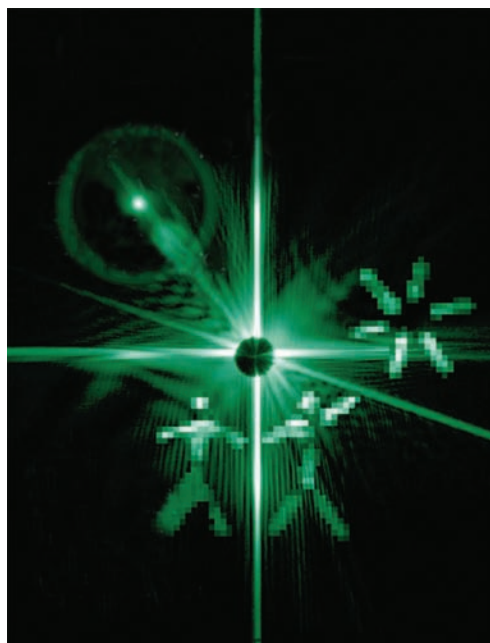
“Biological Imaging with Fourth-Generation Light Sources” (05-SI-003)

Henry N. Chapman, Principal Investigator

Groundbreaking experiments have been proposed to test key concepts of single-molecule x-ray free-electron laser (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 manipulation and orientation of single particles in space and time to interact with XFEL pulses. The experimental results will be compared with high-fidelity modeling to understand how to use XFEL to determine the atomic structure of biological macromolecules, protein complexes, viruses, and spores. Specific goals are to determine the duration and fluence of XFEL pulses required for single-molecule imaging, demonstrate image-reconstruction methods, and perform ultrahigh-resolution three-dimensional imaging of container-free particles.

Single-molecule imaging will further LLNL's missions in both biodefense and bioscience to improve human health. In addition, improved tomography algorithms developed will benefit stockpile stewardship, such as diffraction imaging techniques applicable to the study of warm dense matter. This research will also enhance the capabilities of the Linac Coherent Light Source (LCLS) beyond the baseline design, a high-priority project of the DOE Office of Science, in support of LLNL's mission in breakthrough science and technology.

In FY07, the project team achieved a major milestone—the first-ever demonstration of the ultrafast diffractive x-ray imaging of free injected particles. They also characterized their injection system at the FLASH free-electron laser facility in Germany and found it capable of carrying out experiments at higher resolution at the upcoming LCLS. Images of injected living hydrated biological cells and test particles were reconstructed. The method of time-delay holography was also improved; these results were featured on the cover of *Nature Physics*.



A technique was demonstrated for reconstructing the image of an experimental sample using scattered x-rays at the beginning of a single 25-femtosecond soft x-ray pulse generated by a free-electron laser, as featured on the December 2006 cover of *Nature Physics*. The 1- μm stick figures were patterned onto a silicon nitride film, and their images were captured just moments before evaporating at a temperature of 60,000 K. The biological imaging project is working to determine the atomic structure of biological macromolecules, protein complexes, viruses, and spores.

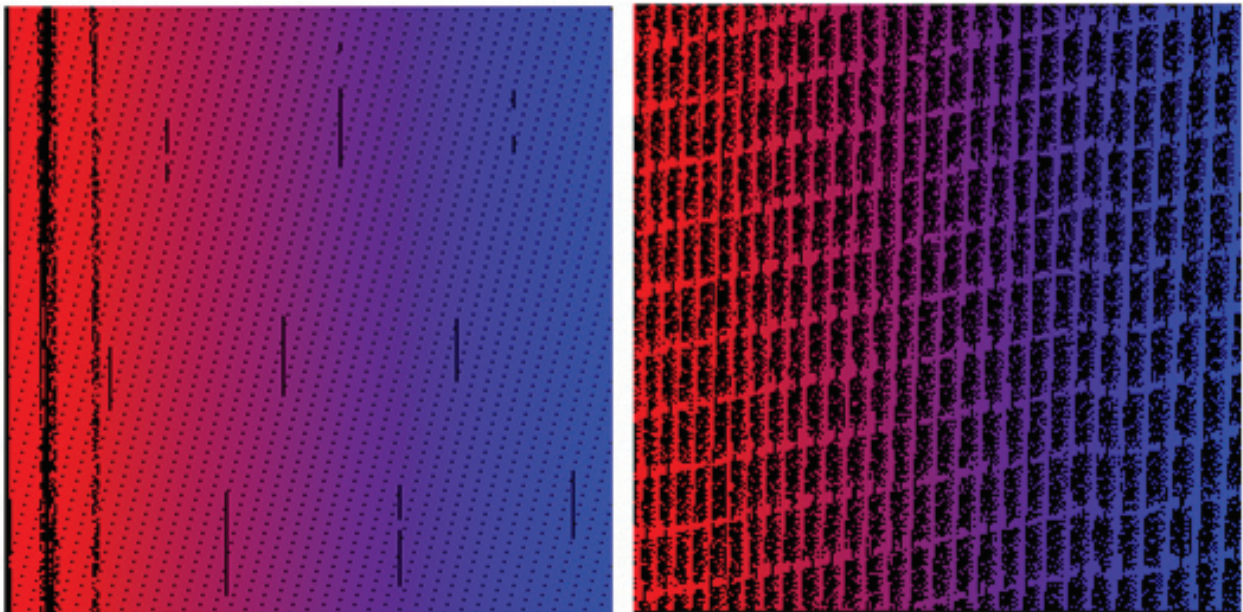
In FY08, the team will take advantage of the increased x-ray penetration of FLASH to image biological cells beyond radiation-damage limits. They will also increase the efficiency of the particle injection system and diagnostics for use in the three-dimensional imaging of reproducible structures, which will be done with gold rod nanoparticles and asymmetric virus particles; further develop time-resolved imaging methods (including the tomographic time-resolved imaging of ablation); measure the effect of a tamper on XFEL-induced particle explosion; and develop a plan for the first experiments at the LCLS based on FLASH experiments and modeling.

Information, Simulations, and Systems Science and Technology

“Internet Ballistics: Identifying Internet Adversaries Despite Falsified Source Addressing” (04-ERD-095)

Anthony Bartoletti, Principal Investigator

Cybersecurity is an important part of the Laboratory’s national security mission. Most network security efforts require adversary identification and characterization but rely primarily upon Internet protocol (IP) addresses, which are easily falsified. This project is studying how accurately an Internet adversary can be identified through unwittingly transmitted packet-timing-related information. Controlled experiments will determine how packet arrival patterns are affected by the attacker’s platform, attack software, algorithm settings, and intervening network locations and conditions, with a focus on hostile scan activity. The aim is to separate adversary-specific traffic signatures from network-specific traffic signatures and provide a testable foundation for an enhanced system of attacker attribution that should improve the discovery of intrusions that might otherwise be overlooked because of deliberately obscured source IP addresses. This work also supports counterintelligence efforts by increasing analysts’ ability to relate what might otherwise appear as unrelated network activities.



Internet ballistics research to identify Internet adversaries employs various wavelet signatures to capture multiscale features in the detailed time and target patterns of hostile scan activity. Individual hostile scans reveal both regular and irregular time and target pattern aberrations, evidence of the attack tool and algorithm settings (left), or of Internet routing conditions and packet delivery interference (right).

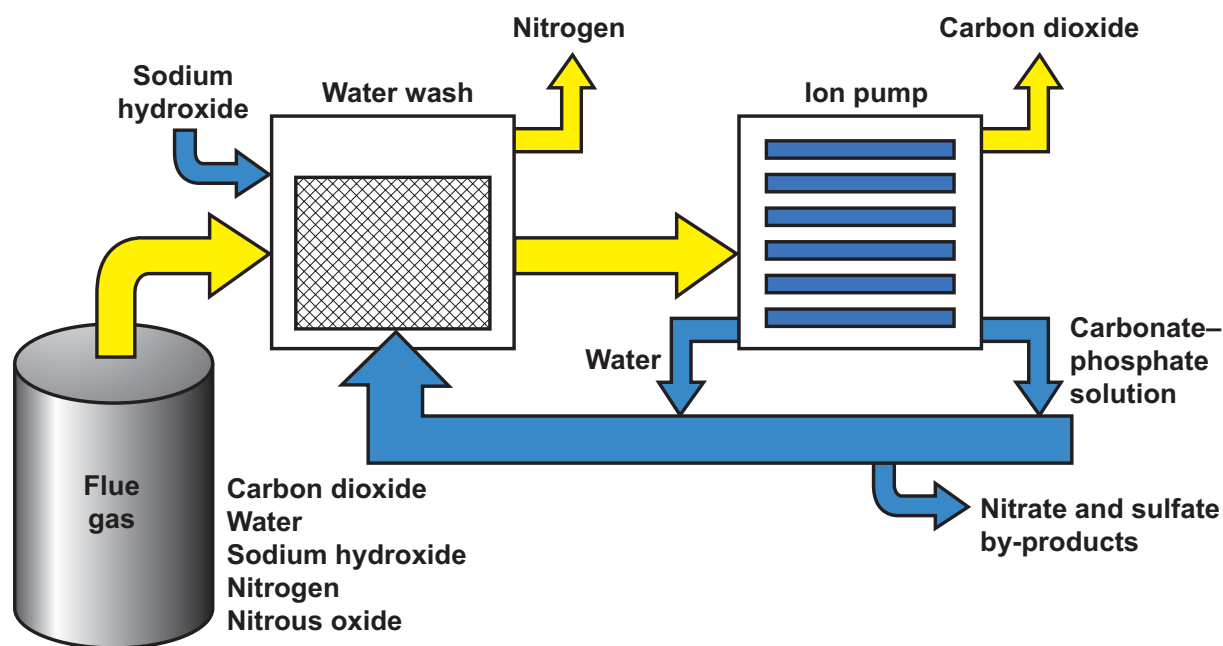
In FY07, the team conducted and characterized 700 class-B network scans, employing three contemporary scan tools with parameter variation, involving both nonrouted and Internet-routed pathways. Each scan was analyzed using 110 separate datamode-wavelet signature combinations and ranked for effectiveness in separability of the known tool, parameter, and location groupings. Overall, this project created a robust capability to distinguish tools and parameters in the presence of packet-routing unknowns and to distinguish scan routing given accurate tool and parameter identification. In collaboration with the University of California at Davis, the team also produced an analytic tool employing these techniques to enhance hostile scan discrimination, providing a foundation for the behavior-based detection of adversary activity and the identification of adversaries.

Energy and Environmental Science and Technology

“Separation of Carbon Dioxide from Flue Gas Using Ion Pumping” (06-ERD-014)

Roger Aines, Principal Investigator

Reducing carbon dioxide emissions is a crucial, high-profile aspect of the Laboratory’s mission in environmental management. The main barrier to lowering carbon emissions generated during energy production is the lack of a cost-effective means of separating carbon dioxide from combustion sources. This project is investigating the possibilities of separating carbon dioxide from flue gas by ionic pumping of carbonate ions dissolved in water. The ion pump dramatically increases dissolved carbonate ion in solution and hence the overlying vapor pressure of carbon dioxide gas, allowing its removal as a pure



A proposed method for lowering carbon emissions generated during energy production. Flue gas is first dissolved in slightly alkaline water, which passes into the ion pump to produce a concentrate from which carbon dioxide is released. Phosphate is added to buffer pH and increase carbon dioxide carrying capacity. The concentrate is recycled to the water wash, and nitrate and sulfate will be concentrated with the bicarbonate and either removed separately as solids or evolved in gas form.

gas. This novel approach to increasing the concentration of extracted gas permits new approaches to treating flue gas, because the slightly basic water used as the extraction medium is impervious to the trace acid gases that destroy existing solvents used in conventional methods, and no pre-separation is necessary. Modeling and laboratory experiments will demonstrate the chemistry of the method, which is expected to offer a dramatically improved solution for removing carbon from hydrocarbon combustion at power plants.

In FY07 the team successfully demonstrated that their method works at flue-gas concentrations—a significant milestone. They also addressed another significant hurdle for large-scale implementation by demonstrating that the method can be implemented with any of the three most widely used water desalinization techniques—reverse osmosis, electrodialysis, or thermal distillation. These two accomplishments represent a major step forward in validating this method's economic utility.

Work in FY08 will focus on using the ion pump as a carbon dioxide concentrating mechanism. Gas and fluid flow will be tested in multiple-plate ion pumps to determine whether the plate materials can be optimized for the carbon separation method. A major goal is to determine the parameters necessary for a pilot-scale test of the technology. The team will also investigate the feasibility of producing drinking water during the carbon-separation process, which would further enhance the method's economic viability.

Patents resulting from LDRD-funded research as a percentage of all LLNL patents from 1999 to 2007.

	1999	2000	2001	2002	2003	2004	2005	2006	2007
All LLNL patents	84	93	89	97	71	95	93	63	62
LDRD patents	45	35	42	27	29	55	51	29	32
LDRD patents as percentage of total	54%	38%	47%	28%	41%	58%	55%	46%	52%

Patents and Publications

Projects sponsored by LDRD consistently account for a large percentage of the patents issued for LLNL research, especially considering that beginning in FY07 the LDRD Program funding represents 8% of the Laboratory's budget (i.e., 8% burdened, or no less than 6% in available funding). Before that, LDRD accounted for 6% of the total LLNL budget, with the exception of FY2000, when the LDRD Program received only 4% of the budget. The number of patents resulting from LDRD-funded research since 1999 is shown in the table above. 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.

Scientific journals further the progress of science by reporting new research—sometimes marking fundamental breakthroughs—and serving as part of the permanent scientific record. Each year, Laboratory scientists and engineers publish more than 1,000 papers in a wide range of peer-reviewed journals. In FY07 there were 1,162 such articles, of which 237 (20.4%) were related to LDRD projects. In addition, several LDRD-supported projects were featured on journal covers or named as significant articles of the year, as detailed in the "Awards and Recognition" section starting on page 17.

Collaborations

Collaborations are absolutely essential to the conduct of research and development in LDRD. By collaborating formally and informally with other national laboratories, academia, and industry, LDRD principal investigators (PIs) are able to access world-leading facilities and knowledge—both in the U.S. and abroad—and serve as active and prominent members of the broad scientific and technical community. This, in turn, allows LDRD PIs to leverage LLNL's achievements and capabilities. In addition, LDRD collaborations often result in outside scientists and engineers being recruited to LLNL.

In FY07, the PIs of 83 LDRD projects (52% of the total projects funded) engaged in 140 formal collaborations. Collaborating institutions included the University of California (38% of total collaborations), other academic institutions (33%), DOE sites (9%), and other collaborators (e.g., other government agencies and industry, 20%). These statistics do not include the numerous informal collaborations that PIs pursue in the course of their LDRD projects.

Awards and Recognition

Alameda County Women's Hall of Fame. Hope Ishii was inducted into the Alameda County Women's Hall of Fame as the 2007 Outstanding Woman in Science during a ceremony at the Fremont Marriott Hotel. Ishii was recognized for her research as a member of the Laboratory team that performed the first-ever study of cometary material collected by the Stardust mission, whose work was supported by LDRD (03-ERI-007).

American Physical Society Fellow. Peter Amendt was honored in 2007 as an American Physical Society Fellow for 2006 through their Division of Plasma Physics. He was honored for "seminal contributions to the development of indirectly driven single- and double-shell inertial confinement fusion physics necessary for the demonstration of laboratory-scale ignition." Amendt's related LDRD projects include research on double-shell laser target design (01-ERD-003) and target fabrication science and technology (05-SI-005).



Hope Ishii (second from right) is pictured with Laboratory Science and Technology Office Director Rokaya Al-Ayat, Physics and Advanced Technologies Associate Director Bill Goldstein, and Institute of Geophysics and Planetary Physics Director John Bradley.

Best Application Paper. A team supported in part by LDRD was awarded Best Application Paper award at the IEEE Visualization 2006 meeting held in Baltimore, Maryland. Daniel Laney, Peer-Timo Bremer, Ajith Mascarenhas, Paul Miller, and Valerio Pascucci won for their paper, "Understanding the Structure of the Turbulent Mixing Layer in Hydrodynamic Instabilities," which was enabled by Pascucci's LDRD project on data exploration with multiscale analysis and information visualization (05-ERI-002).

Chemistry Highlights 2006. *Chemical & Engineering News* cited Peter Weber's LDRD-supported research using the Lab's nanoscale secondary ion mass spectrometry system to image cell membranes with spatial resolution better than 100 nanometers for the first time (04-ERD-039). The journal's rundown of chemistry highlights in 2006 noted that the new method, in which samples are bombarded by tightly focused beams of cesium ions, could lead to a better understanding of biological membranes.



Peter Amendt

Edward Teller Fellows. Henry Chapman and Dmitri Ryutov, both from the Physics and Advanced Technologies Directorate, were recognized for their outstanding scientific contributions and technical leadership. The 2007 fellowship award allows the recipients to do a year's worth of self-directed work that will benefit the Laboratory. Chapman's award will allow him to explore new strategic research to create the techniques and technologies to build the coherent imaging end station at the Linac Coherent Light Source at the Stanford Linear Accelerator Center and perform single-particle imaging. Chapman's research in biological imaging with advanced light sources has been supported by LDRD (02-ERD-047 and 05-SI-003). Ryutov's award will enable him to explore new strategic direction in the area of plasma physics, an area he has explored with

LDRD support of a project on interaction of magnetized plasma with structured surfaces (98-LW-023).

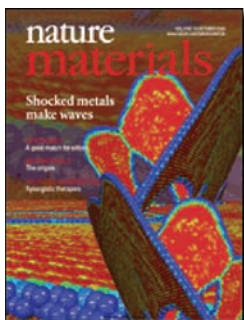
Hitchcon Award. The International Association of GeoChemistry named the 2006 article “Sources of Groundwater Nitrate Revealed Using Residence Time and Isotope Methods” by researchers at Livermore and the University of Arizona as winner of the Hitchcon Award, marking the year’s most significant paper published in the journal *Applied Geochemistry*. This research was partially supported by Bradley Esser’s LDRD project on nitrate biogeochemistry and reactive transport in California groundwater (03-ERD-067).



Henry Chapman (middle) and Dmitri Ryutov (right) with LLNL Director George Miller.

Houtermans Award. Jimi Badro, an LDRD collaborator at the Laboratory, was awarded the 2006 Houtermans Award given annually by the European Association of Geochemistry. It is awarded in recognition of an outstanding publication or series of publications by a young scientist under the age of 35, within the fields of geochemistry or cosmochemistry. Specific LDRD projects include mapping phonons at high pressure for phase transformations and stability (04-ERD-106) and exploration of core formation chemistry (06-ERI-002).

Journal Covers. In FY07, several LDRD-supported projects were featured on the covers of peer-reviewed journals. The February 2007 issue of *Genome Research* featured a method developed by Gabriela Loots for predicting biological tissue-specific enhancers as part of the LDRD project on developing transgenic technologies to study genome regulation and architecture (07-ERD-046). Femtosecond diffractive imaging with a soft x-ray free-electron laser was featured on the December 2006 cover of *Nature Physics*, based on Henry Chapman’s LDRD project on biological imaging with fourth-generation light sources (05-SI-003). The October 2006 issue of *Nature Materials* featured a cover depicting large-scale simulations that successfully model the different stages of shocked metals, part of LDRD research conducted by Eduardo Bringa on high-strain-rate deformation of nanocrystalline metals (04-ERD-021). Also in October 2006, *Angewandte Chemie* featured a cover article on metallic nanowires as immunoassay platforms for pathogen detection, work supported by an LDRD project on a microfluidic system for solution array-based bioassays, with PI George Dougherty (03-ERD-024).



Journal covers of LDRD projects on modeling the different stages of shocked metals (left), metallic nanowires as immunoassay platforms (middle), and predicting biological tissue-specific enhancers (right).

LLNL Science and Technology Awards. In 2007, the Laboratory awarded two Science and Technology Awards to teams of Laboratory scientists and engineers to celebrate significant recent contributions that, in many cases, were widely acknowledged both internally and externally to the Laboratory. Both teams were involved with LDRD-supported research. Andrew Ng led a team that was recognized for seminal contributions to the study of warm dense matter under nonequilibrium extreme conditions (04-ERD-108). Dennis Slaughter was the PI of a team developing an innovative new technique to detect special nuclear materials in fully loaded containers (02-ERD-064 and 07-ERD-019).

Nano 50 Awards. The top 50 technologies, products, and innovators that significantly impact nanotechnology are awarded by the publishers of *Nanotech Briefs*. All three of the Livermore projects honored in 2007, the first from the Laboratory to win this award, were supported in part by LDRD. Greg Nyce was recognized for the development of new nanoporous, low-density materials while working on the LDRD project for laser target fabrication science and technology (05-SI-005). Olgica Bakajin received her award in the innovation category for the discovery and experimental demonstration of ultrafast transport in carbon nanotubes (03-ERD-050). Jeff Tok and George Dougherty were recognized for their work on the development of pathogen-sensing nanosensors based on multistripped metallic nanowires during their LDRD project of a microfluidic system for solution array-based bioassays (03-ERD-024).

NNSA/Defense Programs' Weapons Awards of Excellence. 2007 awards were given for work performed in 2005 for outstanding contributions to the nation's nuclear weapons program. Among them was a team cited for executing a significant scientific and engineering program leading to the determination of pit lifetimes for U.S. warheads, partially enabled by LDRD support of a Choong-Shik Yoo project on electronic transitions and phonons in metals at high pressures (04-ERD-020).

Optical Society of America Fellow. Henry N. Chapman was recognized as a fellow for his "contributions to x-ray microscopy, coherent x-ray imaging, x-ray optics, and EUV lithography."



Bruce Goodwin, LLNL associate director for Defense and Nuclear Technologies; David Crandall, NNSA deputy administrator for Research Development and Simulation; and W. Steven Goorum, NNSA assistant deputy administrator for Military Application and Stockpile Operations, with the pit lifetime team.

A member of the Optical Society of America for more than 10 years, Chapman attributes his success to the research opportunities at the Laboratory, especially as provided by the LDRD Program. Chapman's LDRD-supported research includes biological imaging with advanced light sources (02-ERD-047 and 05-SI-003).

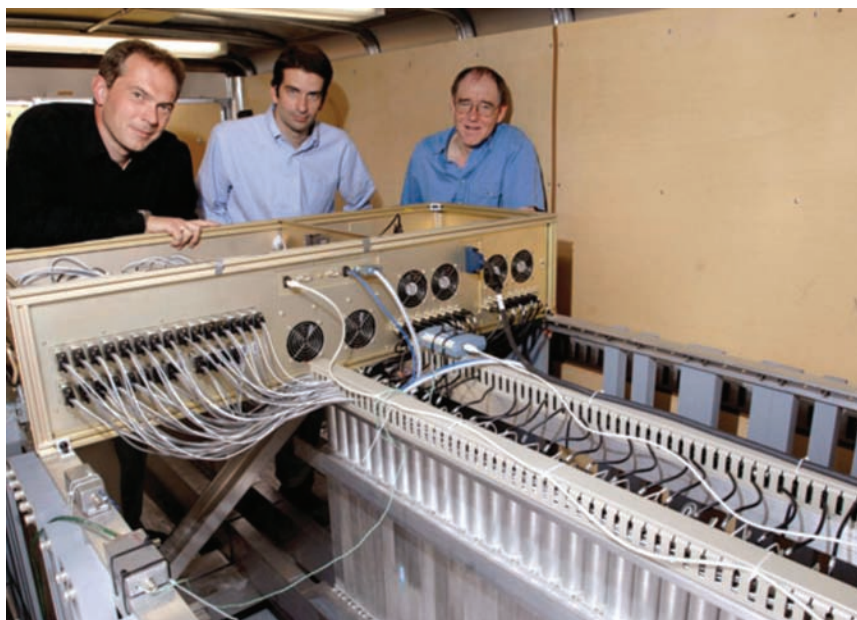


Henry Chapman

Outstanding Technology Development. George Caporaso, Steve Sampayan, and Genaro Mempo were honored for their technology transfer efforts of the dielectric wall accelerator for proton therapy in the 2007 Far West Regional competition sponsored by the Federal Laboratory Consortium for Technology Transfer. The consortium is a nationwide network of federal laboratories that provides a forum to develop strategies and opportunities for linking the laboratory mission technologies and expertise with the marketplace. The technology was enabled with support from LDRD for research on the ultrahigh gradient dielectric wall accelerator (93-DI-044) and a compact accelerator for proton therapy (03-ERD-073).

R&D 100 Awards. In 2007, Laboratory technologies won five R&D 100 Awards from *R&D Magazine*. Of these, LDRD support contributed to four awards:

- **Fast Detection of a Punctured Lung.** A new handheld diagnostic device can quickly detect pneumothorax, a medical condition caused by having air trapped in the space between the wall of the chest cavity and the lung. John Chang was the PI on two different LDRD projects that led to the diagnostic device: impulse radar application for cardiac monitoring (01-ERD-088) and concealed threat detection (02-ERD-061).
- **Detecting Nuclear Materials: Large Area Imager.** A collaborative research effort focusing on radiation-sensing expertise has produced an innovative technology that allows investigators to pinpoint illicit nuclear material from a moving truck with total insensitivity to variations in an area's radiation field. This work was partially supported by a Lorenzo Fabris LDRD project on



Lorenzo Fabris, Thomas Karnowski, and Klaus Zioc (left to right) and the large-area imager.

the long-range passive detection of fissile material (03-ERD-048).

- **Retinal Camera to Capture Early Stages of Eye Disease.**

A new instrument could revolutionize retinal imaging, providing eye doctors with the capability to detect, diagnose, and treat blinding retinal diseases more successfully. Several LDRD projects with Scott Olivier as the PI led to this instrument, including advanced wavefront control (98-ERD-061), diffraction-limited adaptive optics and human visual acuity (01-LW-036), and correction of distributed optical aberrations (03-ERD-006).



The *Hydre* development team: (left to right) Van Emden Henson, Rob Falgout, Ulrike Meier Yang, Tzanio Kolev, Charles Tong, Allison Baker, Barry Lee, Jeffrey Painter, and Panayot Vassilevski.

- ***Hydre* Library of Linear Solvers.** *Hydre* is an innovative software package that offers the most comprehensive suite of scalable linear solvers available for large-scale scientific simulations, with some algorithms appearing for the first time in a linear solver library. Robert Falgout was the PI for an LDRD project on scalable discretization-enhanced solvers (03-ERD-033), which helped enable the *Hydre* software.

SPIE Fellow. Scot Olivier is one of 56 new fellows of the International Society for Optical Engineering (SPIE) for 2007. Fellows are members of distinction who have made significant scientific and technical contributions in the fields of optics, photonics and imaging. Olivier was selected for his achievements in adaptive optics. He leads one of the premier optics groups in the world and has made significant contributions in the application of adaptive optics to astronomy, human vision science, high-power lasers, and remote sensing with support from LDRD projects on advanced wavefront control (98-ERD-061), diffraction-limited adaptive optics and human visual acuity (01-LW-036), and correction of distributed optical aberrations (03-ERD-006).



Scot Olivier

Laboratory Directed Research and Development

Advanced Sensors and Instrumentation

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.

FY07 Accomplishments and Results

In FY07 we (1) constructed an aneurysm temperature measurement system, (2) collaborated with the University of California at Berkeley to conduct detailed temperature and flow studies using a cardiac flow system, (3) used our temperature measurements to assess the thermal effects of SMP heating on arterial tissue, (4) performed a series of CFD simulations on anatomically correct aneurysms, and (5) demonstrated the fabrication and delivery of an SMP dialysis needle adapter (modified from the originally proposed bifurcated stent). This project resulted in significant follow-on work.

Publications

Baer, G., et al., in press. "In vitro deployment of laser-activated shape memory polymer vascular stent," *Biomed. Eng. Online*. UCRL-JRNL-226900.

Baer, G., et al., 2007. "Shape-memory behavior of thermally stimulated polyurethane for medical applications," *J. Appl. Pol. Sci.* **103**(6), 3882. UCRL-JRNL-224906.

Maitland, D. J., et al., 2007. "Prototype laser-activated shape memory polymer foam device for embolic treatment of aneurysms," *J. Biomed. Opt. Lett.* **12**, 030504. UCRL-JRNL-227411.

Wilson, T. S., et al., 2007. "Shape memory polymers based on uniform aliphatic urethane networks," *J. Appl. Poly. Sci.* **106**(19), 540. UCRL-JRNL-227443.

Developing Advanced Radiography Capabilities at Future Large Fusion-Class Lasers

Hye-Sook Park 05-ERD-006

Abstract

We propose to develop bright, high-energy, high-resolution radiography solutions for high-energy-density (HED) experiments at future high-power laser facilities. These experiments require probing imploding targets made of bigger, thicker, and more dense materials than those used currently. We will develop backlighters with an energy of greater than 15 keV and spatial resolution of less than 10 μm to properly diagnose experimental conditions. We will study high-intensity laser interaction and radiation transport in small and confined volumes and utilize these physics properties to generate bright high-energy K-alpha sources from microfoil and microwire targets. We intend to demonstrate their brightness and resolution using various short-pulse laser facilities.

If successful, this project will lead to a petawatt radiography diagnostic for use on HED experiments at future fusion-class lasers. We will have a suite of hard x-ray sources, optics, and detectors that will be applicable to high-resolution, high-energy x-ray radiography. We will also significantly enhance our knowledge of high-intensity laser-coupling efficiencies and electron transport for short-pulse lasers. This will contribute to the development of reliable simulation models for numerous applications such as fast ignition, proton heating, laboratory astrophysics, and high-temperature, solid-density physics.

Mission Relevance

This project supports the Laboratory's national security mission by providing optimal radiography techniques for stockpile stewardship experiments such as material strength, HED with mid- to high-Z capsules, and double-shell ignition experiments.

FY07 Accomplishments and Results

We have achieved all milestones set for this year. Specifically, we (1) tested the greater than 15-keV, one- and two-dimensional radiography sources; (2) used the Titan short-pulse laser at Livermore to measure the K-alpha brightness as a function of target materials from molybdenum at 17.5 keV to lead at 75.0 keV and compared the results with simulation; (3) measured the brightness and spatial resolution of microwire targets for two-dimensional radiography; (4) estimated, from our measurements, the signal-to-noise ratio for fusion-class laser experiments—our calculations show that the Advanced Radiographic Capability laser at the National Ignition Facility will deliver enough brightness for HED experiments; and

(5) utilized various hard x-ray detectors including cameras and curved crystal spectrometers to demonstrate their performance.

Publications

Park, H. S., et al., 2006. *High-resolution 15–100 keV K-alpha radiography for high-energy density experiments*. American Physical Society 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-PRES-225579.

Park, H. S., et al., 2007. *Quasi-isentropic material property studies at extreme pressures: from Omega to NIF*. Intl. Conf. Inertial Fusion Sciences and Applications, Kobe, Japan, Sept. 9–14, 2007. UCRL-PRES-234443.

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

John F. Regan 05-ERD-049

Abstract

The goal of this project is to develop a practical, 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 chain reaction (PCR) assay panel for simultaneous detection of multiple respiratory pathogens, (2) optimize assay performance in complex sample matrices, (3) validate the multiplexed PCR assays and diagnostic platform using clinical samples, 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, point-of-care 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.

FY07 Accomplishments and Results

In FY07, we completed development of an instrument capable of performing multiplexed assays that are specific for respiratory pathogens, biothreat agents, and even agricultural threats. We also conducted a clinical study at the University of California Davis Medical Center to compare the accuracy and sensitivity of the instrument to that of viral cultures. We filed a record of invention describing the respiratory multiplex assay and wrote two manuscripts: one detailing the accomplishments of this project and another describing the processing and screening of samples for any pathogen of choice using our automated instrument. In summary, although this project focused on detecting respiratory pathogens, the instrument developed can also be used for biothreat assay panels developed at LLNL. The ability of this point-of-care pathogen detection instrument to utilize different assays suggests numerous potential biodefense applications.

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

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 screen 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 in human effluents. 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. 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. It will also improve capabilities for emergency response to bioterrorist attacks and triage. This project also provides new avenues for strong collaborations with researchers at two University of California campuses: Davis and Berkeley.

Mission Relevance

If successful, 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 support the Laboratory's mission in biotechnology.

FY07 Accomplishments and Results

In FY07, we focused on respiratory disease, specifically efforts to show that unique mass-spectrometry signatures can be obtained with BAMS respiratory pathogens (including live, nonvirulent strains of *Mycobacterium tuberculosis*). We demonstrated that some mass-spectrometry signatures are preserved when intact organisms are analyzed by BAMS and that the signatures are detectable on a single-cell level. We also demonstrated the feasibility of detecting surrogates for respiratory pathogens in lung surfactant and in exhaled breath condensate with BAMS, addressing the challenge of detecting characteristic marker molecules from pathogens embedded in such a complex human matrix.

Publications

Bogan, M. J., et al., "Online aerosol mass spectrometry of single micrometer-sized particles containing poly(ethylene glycol)." *Rapid Comm. Mass Spectrom.* **21**, 1214. UCRL-JRNL-227120.

Frank, M., et al., 2007. "Modular sampling and analysis techniques for the real-time analysis of human breath." *Proc. IEEE Sensors 2007 Conference*. UCRL-PROC-232624.

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 infer the existence of more than 200 planets orbiting nearby stars. These discoveries were 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, high-contrast "extreme" AO systems (ExAO) is needed to probe the environments of other stars on scales comparable to the size of our solar system. This project will use existing AO to probe 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 will also develop advanced AO algorithms and sensors for very high-order precision AO, allowing systems an order of magnitude more powerful than current technology to be controlled efficiently, optimally, and accurately. Technologies developed for this project will be key to a wide range of future AO work in remote sensing, laser beam control, biomedical applications, and the next-generation Gemini Planet Imager.

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 counterproliferation and nonproliferation missions. This project also supports LLNL's mission in breakthroughs in science and technology.

FY07 Accomplishments and Results

During FY07 we (1) carried out a series of observations of young stars using new image-processing techniques developed to extract moving signals from slowly varying noise backgrounds, (2) collaborated with the Laboratory for Adaptive Optics at the University of California at Santa Cruz to experimentally verify fundamental concepts and algorithms developed at LLNL, and (3) developed an innovative new algorithm that uses Fourier domain control to predict changes in the atmospheric distortions of light as multiple turbulent layers across a telescope aperture. Thus, we have potentially paved the way to improvements in the performance of AO systems for many applications by a factor of two.

Publications

Konopacky, Q. M., et al., 2007. "Measuring the mass of a pre-main-sequence binary star through the orbit of TWA 5A." *Astron. J.* **133**, 2008. UCRL-JRNL-227824.

Marois, C. M., B. Macintosh, and T. Barman., 2007. "GQ Lup B visible and near-infrared photometric analysis." *Astroph. J. Lett.* **654**, L151. UCRL-JRNL-223503.

Poyneer, L., B. Macintosh, and J.-P. Veran, 2007. "Fourier transform wavefront control with adaptive prediction of the atmosphere." *J. Opt. Soc. Am.* **24**, 2645. UCRL-JRNL-225647.

Siegler, N., et al., 2007. "Discovery of a 66-mas ultracool binary with laser guide star adaptive optics." *Astron. J.* **133**, 2320. UCRL-JRNL-227799.

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

Raymond J. Beach 05-ERD-062

Abstract

A high-power, short-pulse laser would significantly enhance existing and planned research in high-energy-density science as well as the capabilities of inertial fusion facilities. To help develop the technology needed to realize such enhancements, this project will investigate the scientific and technical issues related to creating an integrated system for the injection, setup, compression, and monitoring of split-beam and high-energy, short-pulse generation from a single aperture of a neodymium:glass laser system. We aim to develop new amplifier and pulse compressor technologies that meet the stringent operating requirements and

architectural constraints of large-scale amplification systems capable of producing kilojoule-class pulses.

If successful, we will develop new techniques for (1) multiple sub-aperture pulse generation and pulse-width control; (2) precision pointing and characterization of split-beam pulses; (3) precision alignment of a folded, mixed-grating pulse compressor; (4) precision alignment and characterization of split-beam pulse compressors; and (5) multipulsing of large optical switches to block back-propagating beams on large-scale, kilojoule-class inertial fusion facilities. The ability to manipulate and combine multiple independent beams to achieve very-high-power, short-pulse operation represents a new paradigm in high-energy-density science and will enable multiple time-frame, K-alpha radiographic diagnostic capability.

Mission Relevance

This project furthers LLNL's missions in stockpile stewardship by developing powerful new radiographic diagnostics, and in energy security by enhancing fast ignition, which would lead to potential fusion energy applications. In addition, the research will support Laboratory efforts in breakthrough science and technology by enabling studies of matter under extreme conditions of temperature and pressure, with applications in astrophysics, planetary physics, stellar interiors, and basic material dynamics.

FY07 Accomplishments and Results

In FY07, we successfully (1) assembled a prototype split-beam injection system, which demonstrated the precision metrology required for beam-pointing and centering, as well as the level of prepulse suppression needed to ensure that chirped beams will not impact the utility of standard nanosecond-duration beam lines; (2) performed a partial characterization of the split-beam injection concept using prototype hardware; (3) assessed the isolation level possible with our optical isolator scheme; and (4) identified an engineering approach that will allow our developed split-beam concept to be installed with minimal interruption and impact to the ongoing operation of a working facility.

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 use the SQUID response to read out the signal from 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.

The most important result will be the demonstration of the rf-SET readout by a microstrip SQUID amplifier that is sensitive enough to detect a single spin in a solid-state system. This will be a breakthrough for quantum computations and quantum information (QC/QI) applications. We expect to demonstrate the quantum limit for charge amplification and position measurement in nanomechanical resonators. Our results will be published in peer-reviewed journals, and university collaborations will attract top-echelon postdoctoral researchers to the Laboratory.

Mission Relevance

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

FY07 Accomplishments and Results

In FY07, we improved the match between the microstrip amplifier and the rf-SET, which was our major goal. We also created a prototype of a novel, compact quadrature hybrid for constructing a balanced amplifier consisting of two microstrip SQUIDs. This balanced amplifier should solve problems caused by impedance mismatches between the SET and the amplifier. We also completed noise tests of the amplifiers.

Proposed Work for FY08

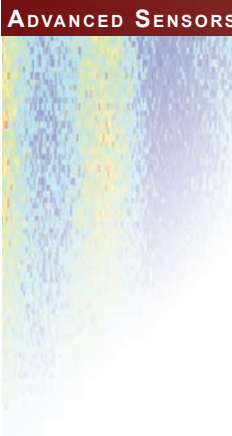
In FY08, we plan to complete experimental work by (1) finalizing results on the charge noise sensitivity for the SET, (2) measuring flux noise sensitivity for the SQUID, and (3) completing simulations for the microstrip amplifier. In addition, we plan on publishing the results of our findings in high-profile scientific journals. We are hopeful that new, ultrasensitive detectors using the same principle of resonant circuit readout employing microstrip SQUID amplifiers can be developed for use by various government agencies.

Terascope: Terahertz Spectroscopic Imaging for Standoff Detection of High Explosives

Robert Deri 05-ERD-076

Abstract

We are developing a standoff detection and identification method for locating concealed high-explosives. Our approach uses emerging spectroscopic and imaging technologies in the terahertz frequency regime to develop a systems concept and multispectral detection algorithms, as well as simulate the behavior of such a system in the presence of atmospheric absorption, obscurant losses, and system noise through a concealing material. The algorithms are a two-channel approach that compares the return signal from two points on



a target to remove the effects of the intervening atmosphere, and then compares the return signals to known spectral signatures.

If successful, this project will lead to increased safety of U.S. citizens at home and abroad and will stimulate improvements in terahertz technology. This will be the first demonstration of concealed high-explosives detection at a 50-m standoff. We anticipate that this result will provide the impetus for development of usable standoff high-explosives detectors based on this technology.

Mission Relevance

By developing a terahertz spectroscopy capability for the standoff detection and identification of covert high explosives, this research supports Laboratory missions in national and homeland security.

FY07 Accomplishments and Results

In FY07, we developed a systems concept/link budget and algorithms for standoff terahertz spectral imaging. Specifically, we (1) collected terahertz data on common high-explosives materials, (2) developed a systems concept/link budget for a terahertz spectral imager, (3) developed algorithms for terahertz spectral imaging and, (4) executed Monte Carlo simulations of system performance. Algorithms developed deconvolved the spectral modifications induced by atmospheric propagation. Results showed the proposed system can achieve detection of bulk high explosives at safe standoff distances (between 30 and 50 m) even when the explosive is concealed by layers of fabric. Simulated receiver-operating characteristic curves showing probabilities of detection and false alarm demonstrated excellent discrimination of C-4 explosive against innocuous background material such as skin and lactose.

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 isolate the pathogen quickly from biological samples for analysis. Persistent technology issues in the process of identifying and quantifying the presence of pathogenic agents have been sample handling and preparation that must precede any assay, along with the need for improved multiplex assays for rapidly-mutating RNA viruses. 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 dielectrophoretic techniques to separate and purify viruses—the most transmissible and infectious agents—from biological and environmental samples. We will also create less-costly, but more general multiplex assays for viruses, using multiplex, ligation-dependent probe amplification.

We expect the new capabilities developed in this project to reduce the time required to identify a new pathogen (e.g., SARS) by up to an order of magnitude, in part, by better matching methods for sample preparation with the capabilities of emerging assay technologies. 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, detect, and identify engineered and naturally emerging viruses, 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.

FY07 Accomplishments and Results

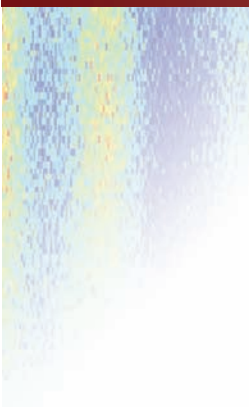
In FY07, we (1) created validated three-dimensional computer simulation capabilities, including Monte-Carlo models for transport of biological particles by acoustics, electrokinetics, electroosmosis, electrochemistry, and pH; (2) established quantitative assays for sample concentrations of *Escherichia coli* bacteria, *Saccharomyces cerevisiae* yeast, murine herpesvirus, adenovirus, and MS2, transfusion-transmitted, and Epstein-Barr viruses; (3) began nasal swab collection and analyzed samples with our assays for transfusion-transmitted virus, Epstein-Barr virus, and adenovirus; (4) designed, fabricated, and tested numerous configurations of microfluidic devices; and (5) optimized the probe hybridization and ligation protocols for a molecular inversion probe that resulted in a reduction of hybridization-ligation reaction time from 4 hours to 90 minutes. This represents the first time the molecular inversion probe technology has been modified for use on a bead-based microarray.

Biophysical Characterization of Pathogen Invasion

Amy L. Hiddessen 06-ERD-013

Abstract

A fundamental understanding of pathogen infection and host response is needed to develop new treatments for infectious disease. To address this challenge, a comprehensive description of key processes is needed, including data on the spatiotemporal expression of molecular effectors and regulators of signaling cascades. Using a model pathogen system having well-characterized pathogen ligands and host receptors, we will, for the first time, quantitatively characterize the virulent ligand-host cell receptor interaction and subsequent signal transduction using novel atomic force microscopy (AFM) that is combined with advanced optical methods. We also will develop a new capability for pathogen and host-cell characterization.



Investigations of the complex phenomenon of pathogenesis will provide valuable knowledge about the molecular causes of infectious disease, as well as new insights into cell-regulatory machinery and signaling pathways in host cells that may be used for new drug development. Successful completion of this project will provide the first detailed, quantitative picture of the initial process of infection with high spatiotemporal resolution. Moreover, this research will develop a novel single-cell platform for studying signal transduction in cellular systems and presymptomatic responses in the host cell. The results and tools generated in this work will be published and used to develop new capability for a high-throughput, multiplexed pathogen-characterization system.

Mission Relevance

By combining synergistic experimental techniques to better understand pathogen–host interactions, this work will support Laboratory missions in biosecurity, homeland security, and bioscience to improve public health.

FY07 Accomplishments and Results

In FY07, we (1) completed operational testing and development of the combined AFM–confocal instrument, including critical alignment of AFM cantilever with confocal optics; (2) functionalized bead and tip probes for delivering ligands to cells via AFM; (3) optimized the fluorescent labeling of actin filaments in live epithelial cells; (4) developed methods and performed preliminary tests of AFM-based ligand delivery to live cells in a controlled environment; and (5) incorporated optics into our instrument in preparation for visualizing and manipulating actin structures inside host cells. In addition, we recruited two new postdoctoral researchers to the Laboratory to work on the project.

Proposed Work for FY08

In FY08, we will (1) quantify ligand densities on beads coated with varying amounts of ligand; (2) deliver controlled ligand concentrations to fluorescently labeled live cells by attaching the ligand-coated beads to AFM tips; (3) determine rates of bead uptake and the threshold ligand concentration for induced internalization of the bead and associated cell signaling processes—for example, actin polymerization; (4) obtain time-lapse data of cell response and bead uptake as a function of ligand concentration using light and fluorescence confocal microscopy; (5) characterize the data using image acquisition and analysis software; and (6) begin to optimize parameters for laser disruption of actin filaments in cells to examine cytoskeletal involvement during uptake of ligand-coated beads.

Thermal-Fluidic System for Manipulating Biomolecules and Viruses

Kevin D. Ness 06-ERD-040

Abstract

We propose to develop a reconfigurable fluidic system that will simultaneously separate, concentrate, and purify biomolecules and viruses. Many pathogen detection, mitigation,

and protection applications require manipulation of biomolecules or viruses to accurately quantify the presence of a particular substance or synthesize and investigate the function of a molecule. We will achieve this manipulation in a single system by utilizing the novel microfluidic technology of temperature gradient focusing (TGF), an equilibrium gradient version of capillary electrophoresis that allows for the stationary fractionation and greater than 10,000 increase in concentration of target analytes based on bulk electrophoretic mobility.

Starting with the manually controlled proof-of-principle TGF system, we will develop a more robust system to concentrate and separate different virus strains as a final preparation for detection, and will demonstrate the capability to dramatically enhance the efficiency of processes for in vitro transcription/translation protein expression through protein purification and separation. Expected results include a demonstrated improvement of the TGF separation resolution using different capture chip designs for fluorescent dyes, e-tags, and proteins. Successful demonstration of these capabilities will also establish the system's utility for other biological applications.

Mission Relevance

The project will contribute to the Laboratory's national security and homeland security missions in biodefense by demonstrating the ability to preconcentrate viruses for fast identification and detection, and enhancing the production speed and efficiency of synthetic reagents for detection of threat toxins, bacteria, and viruses. Both applications support efforts that enhance the rapid detection of engineered or naturally emerging viruses, bacteria, or other toxins.

FY07 Accomplishments and Results

In FY07, we (1) demonstrated the successful capture and greater than 2,000-fold concentration of a small fluorescent dye in an automated sequential injection analysis system; (2) improved overall instrument performance by improving the stability of the applied electric field, linearity of the temperature field, increased magnitude of temperature gradient, and stability of flow rate; (3) used direct numerical modeling and multiphysic simulations to generate thermal gradients of 50°C/mm in a high-throughput microfluidic structure; (4) developed a Brownian dynamics simulation to solve for important analyte concentration profiles; and (5) extracted specific metrics such as peak height, peak width, throughput, and limit of detection to determine the efficiency and resolving power of the TGF approach.

Proposed Work for FY08

In FY08, we intend to move from fluorescent analytes to biological samples of viruses and proteins. We will (1) build a fluorescently labeled sample library of viruses and proteins to characterize system performance with biological samples, (2) organize the library into biologically relevant groupings, (3) determine how biologically relevant groupings of our sample library relate to electrophoretic mobility, and (4) demonstrate the ability to separate three spiked viral samples in a "simplified" background (and repeat with proteins) and purify a spiked virus from a "dirty" background of multiple contaminants (and repeat with proteins). We also intend to move on to experimentation with more relevant and complex sample matrixes such as nasopharyngeal and in vitro translated protein productions.

Viral Identification and Characterization

Christopher G. Bailey 06-ERD-064

Abstract

Humans are exposed daily to millions of viruses that are poorly understood. Identifying viruses is challenging, because many are impossible to culture and can evolve rapidly. This problem is compounded by our near total lack of knowledge about the number and types of nonvirulent viruses normally found in our environment—overcoming this barrier would enable huge leaps in our understanding of emerging diseases. We intend to use an approach similar to that of the Human Genome Project coordinated by the DOE and National Institutes of Health to create a translational measurement capability that will allow rapid, high-throughput viral screening. Instead of trying to selectively isolate a virus from a potentially complex media, we propose to analyze every virus in a representative sample.

If successful, this project will produce technology for rapidly obtaining a complete viral genomic profile from almost any type of sample (e.g., urine, blood, and saliva). Like the Human Genome Project, this capability will have an almost inestimable impact on the nation's science. By tackling the huge technological hurdles associated with this challenge and beginning viral prospecting to better understand respiratory infections, we will create a vital new competency for the nation. This work will lead to a small set of high-profile journal articles focusing initially on human respiratory diseases.

Mission Relevance

As the recent SARS viral respiratory illness outbreak demonstrates, the identification of a previously unknown or emerging viral pathogen can take months after a pandemic begins, despite Herculean efforts using existing technology. This work supports LLNL's national and homeland security missions by helping fill a critical gap in our nation's defense by creating the ability to rapidly and dynamically respond to such new biological threats, whether naturally arising or deliberately engineered.

FY07 Accomplishments and Results

Our work for FY07 focused on (1) bioinformatic design of multiplexed assays, which produced improved primer development for selectivity, minimization of multiplexing, complexity of banding structures, and compatibility of multiplexed reactions and specificity of short sequences; (2) experimental assay development, which optimized the conditions for multiplexed polymerase chain reaction (PCR) assays in extremely small volumes; and (3) microfluidic platform development that focused on the ability to carefully manipulate the introduction of samples, mix the sample stream with reagents, rapidly form droplets, conduct thermal cycling, detect PCR products, and sort droplets containing PCR products.

Publications

Beer, R., et al., 2007. "On-chip, real time, single-copy polymerase chain reaction in picoliter droplets." *Anal. Chem.* **79**(22), 8471. UCRL-JRNL-230233.

Serrated Light Illumination for Deflection-Encoded Recording (SLIDER)

John E. Heebne 07-ERD-017

Abstract

Ultrafast, high-fidelity, single-shot recording diagnostics are critical for high-energy-density (HED) physics. We propose a novel technique—serrated light illumination for deflection-encoded recording (SLIDER)—for high-dynamic-range recording in a temporal regime (1–100 ps) for which a strong technology base does not exist. Our technique avoids tradeoffs between dynamic range and resolution that would limit electron beam deflectors by implementing a photon beam deflection scheme in a waveguide. The deflection is achieved through the activation of an array of transient prisms. By illuminating a serrated mask with a pump beam, a prism array pattern is imprinted through nonlinear optical means onto the layer guiding the signal. Time-of-flight propagation through the prisms ensures a linear encoding of time to angle. A camera records the swept signal with high dynamic range.

If successful, we will demonstrate the world's first an all-optical streak camera capable of less than 10-ps temporal resolution with a dynamic range in excess of 8 bits and eventually scalable to higher fidelity. We place particular emphasis on the measurement of fusion burn histories and the characterization of ultrashort, petawatt radiographic capability pulses with heretofore unrealized dynamic range. This work leverages the Laboratory's long history of innovation in single-transient measurement technologies.

Mission Relevance

Single-shot measurement on a fast scale is crucial to experiments that support stockpile stewardship and HED physics. Fundamental questions in weapons design and HED physics have remained outstanding for decades because the technology required to record such data does not exist. The SLIDER technique, when coupled with technologies for imprinting x-ray signatures onto optical carriers, would help uncover these answers.

FY07 Accomplishments and Results

In FY07, we fabricated SLIDER deflectors in a gallium–arsenic planar-slab waveguide and performed lithographic patterning and deposition of a gold serrated mask. A deflector was mounted in a coupling stage incorporated within a newly constructed, ultrafast test bed. To validate the SLIDER concept, the prism-activating pump pulse and a test pulse pattern were derived from an amplified, mode-locked titanium:sapphire laser and optical parametric amplifier. Beam deflections were readily observed with magnitudes consistent with theoretical predictions. Overall, the proof-of-concept demonstration was very successful, displaying a

temporal resolution of less than 10 ps across a record length of 100 ps with a dynamic range in excess of 300.

Proposed Work for FY08

In FY08, we will conduct a full experimental exploration of the merits and drawbacks of the SLIDER concept. Close coupling of a detailed model with results from the first year will guide the refinement of device composition and geometry, mitigate technical issues, and solidify performance predictions. In addition, we will study the feasibility of replacing streak camera diagnostics on fusion-class lasers with an instrument consisting of SLIDER mated with an x-ray and optical encoder.

Ultraviolet-Visible Resonance Raman Studies of High Explosives, Impurities, and Degradation Products for Enhanced Standoff Detection

Jerry C. Carter 07-ERD-041

Abstract

Improvised explosive devices (IEDs) used by terrorists have proven a difficult weapon to counter. Our objective is to understand the fundamental resonance Raman (RR) spectroscopic properties of high-explosive (HE) materials with the ultimate goal of forming the science basis for a new IED standoff-detection technology. Our specific aims include characterizing and understanding the RR spectral signatures of HE materials, impurities, and degradation products and determining the optimal RR conditions, sensitivity, enhancement factors, and best target analytes for IED detection. We also will demonstrate the standoff RR measurement of HE materials at distances of tens of meters and with improved detection limits compared to normal Raman spectroscopy.

We expect to (1) determine optimal conditions at different wavelengths for the RR detection of HE materials; (2) determine RR sensitivity, selectivity, and response time for HE materials; (3) evaluate signatures from laser-induced sample degradation; (4) understand timescales and pathways of sample degradation for select HE materials; (5) understand background interference issues; and (6) demonstrate standoff RR HE identification at tens of meters.

Mission Relevance

This project supports LLNL's national security mission by developing the basis for a new standoff HE detection technology—an important capability needed to fill gaps in our ability to detect and mitigate the effects of IEDs and other terrorist-related threats. Furthermore, this technology also has potential applications in standoff aging and degradation studies of nuclear and conventional weapons. This work will create external strategic partnerships and help recruit new talent to LLNL.

FY07 Accomplishments and Results

In FY07, we experimentally quantified the RR enhancement for TNT using 244-nm excitation; preliminary results indicate an enhancement by several orders of magnitude. To achieve this, we first had to develop an in-house benchtop ultraviolet (UV) micro-Raman system, devise an internal standard measurement method to quantify UV RR, and measure the normal Raman spectra of HE materials. We also studied the RR enhancement effect as a function of laser power to study sample degradation and conducted 532-nm excitation studies of TNT both in terms of potential RR enhancement and sample degradation.

Proposed Work for FY08

For FY08, we will focus on (1) determining the RR signal enhancements relative to normal Raman on an expanded list of relevant HE materials; (2) measure, evaluate, and compare the RR spectral signatures of HE materials at different excitation wavelengths and determine optimal conditions for RR signal enhancement; (3) complete laser-induced degradation studies of TNT; (4) prepare for (and, if time allows, begin) the RR measurements of select HE materials at a standoff distance of tens of meters; and (5) modify the UV micro-Raman system to permit control of the raster scanning of samples.

Salicylic Acid Derivatives: A New Class of Scintillators for High-Energy Neutron Detection

Natalia P. Zaitseva 07-ERD-045

Abstract

The preferred method for detecting high-energy neutrons in the presence of strong gamma-radiation background is pulse-shape discrimination (PSD) using organic scintillators. However, single-crystal stilbene, the most effective known PSD material, is not only toxic but also has a very limited availability because of crystal growth difficulties. We are developing new organic materials for the detection of high-energy neutrons. Based on the studies of the PSD mechanism, we will identify nontoxic organic crystals with improved performance as alternatives to stilbene. Low-cost solution growth techniques will be used to survey the properties of many organic compounds. The candidates most suited to high-energy neutron detection will be selected for further development and growth of large crystals.

Our main deliverable will be identification and development of new efficient, readily available, low-cost scintillator materials that offer the sensitive neutron detection and effective gamma discrimination qualities needed for monitoring fissile materials. Systematic studies of many single crystals grown by solution techniques will produce scientific results important for developing a deeper understanding of the physics of scintillation processes, especially the composition, physical state (liquid or solid), crystallographic structure, and quality of materials. We also will apply our results to the development of technologies for production-scale growth of the crystals. Studies of growth mechanisms and optical characterization will generate publications and intellectual property.

Mission Relevance

By developing new compact, sensitive, low-cost, efficient scintillator materials for the detection and monitoring of fissile materials, this project supports LLNL's national and homeland security missions.

FY07 Accomplishments and Results

In FY07, we (1) grew submillimeter samples of 25 new compounds; (2) assessed the optical properties of the crystals to determine their ultraviolet- and radio-frequency-induced luminescence efficiency and emission decay time; (3) selected and grew the most efficient candidates as 3- to 5-mm single crystals, demonstrating that the scintillation efficiency of some salicylates (e.g., crystalline 2-aminopyridinium salicylate) is close to that of stilbene; (4) installed an experimental facility for PSD tests; (5) developed software for data analysis; and (6) started routine measurements of PSD with compounds (salicylic acid and 1,4-diphenyl-1,3-butadiene) never tested previously for neutron detection, showing that PSD in solution-grown crystals can be as good as in commercially produced stilbene single crystals.

Proposed Work for FY08

The main activity in FY08 will be growth of crystals of new materials based on their scintillation efficiency. Specifically, we will (1) grow crystals with improved optical quality at the larger sizes (1 to 3 cm) required for accurate PSD characterization; (2) use laser- and neutron-excitation sources to study the effects of different factors—such as crystal size and quality, surface conditions, and presence of oxygen—on the intensity of prompt and delayed components of fluorescence in organic materials; and (3) study the efficiency of PSD in relation to molecular and crystal structure, with a final goal of understanding PSD mechanisms to identify the best materials suitable for high-energy neutron detection.

Publications

Zaitseva, N., et al., 2007. "Germanium nanocrystals synthesized in high-boiling-point organic solvents." *Chem. Mater.* **19**, 5174. UCRL-JRNL-226844.

Ultrafast Laser Synthesis of Nanopore Arrays in Silicon for Voltage-Controlled Biomolecule Separation and Detection

Joseph W. Tringe 07-FS-001

Abstract

We propose to demonstrate the feasibility of creating large arrays of 1- to 100-nm-diameter tapered nanopores by using a novel process for direct patterning in silicon. These arrays will enable a new class of electronically controlled filters for biomolecule separation and synthesis, and for prescreening such molecules in advanced sensors. Uniform pore arrays

are not achievable over this diameter range with any other technique, and the shape of the pores will facilitate high flow rates, reducing the potential for fouling. Our new approach relies on interferometric pulsed-laser exposure, a technique that leverages LLNL's extensive capabilities with powerful, ultrafast lasers.

As a preconcentration and rapid-characterization component, the nanopore filter technology we develop can dramatically increase the throughput and selectivity of chemical and biological sensors with homeland security applications. In addition, once we demonstrate the capability of fabricating uniform, approximately 1-nm pores over a 1-mm² surface area, this filter structure will be immediately useful in LLNL's efforts to create a new high-throughput protein-synthesis capability. Additional applications for our laser-based fabrication method are in microelectronics, where a compelling need exists for advanced silicon nanofabrication tools, and in nanoporous thrusters capable of advanced station-keeping and mobility in small satellites.

Mission Relevance

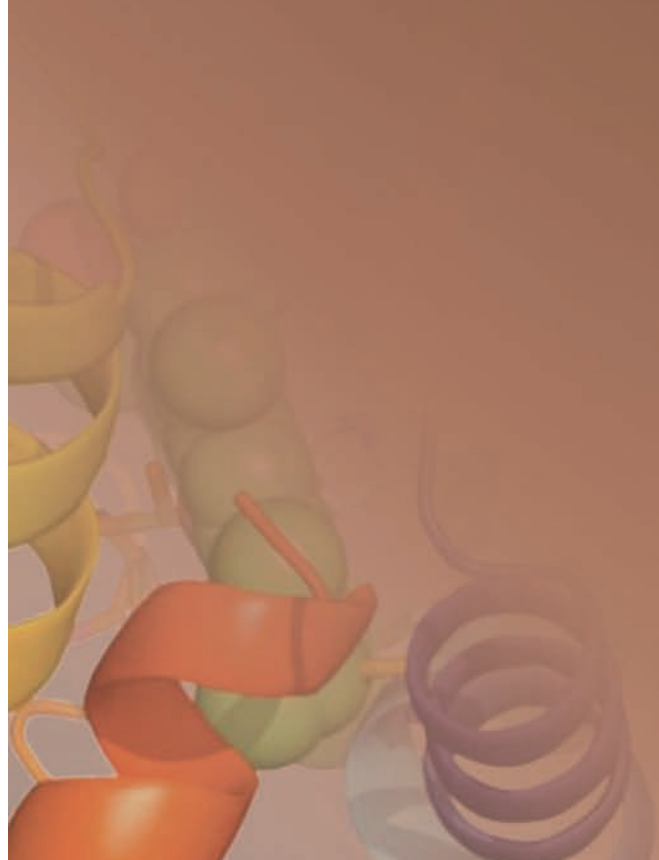
The project supports LLNL's national security mission areas of nonproliferation and homeland security, because the proposed filter would have direct application in chemical and biological sensor platforms. This work also supports the Laboratory's mission in bioscience to improve human health by creating a capability for high-throughput protein characterization and synthesis.

FY07 Accomplishments and Results

In FY07, we successfully created periodic microstructures in silicon by ablating with interferometrically defined patterns of femtosecond-pulsed laser light. We observed a critical energy for ablation and amorphization, and correlated pulse energy with the resultant microstructure. Achieving laser-beam spatial uniformity sufficient for creating a large array of uniform nanometer-scale structures was challenging. However, complementary experiments using photoresist exposed by interferometrically defined continuous-wave laser light demonstrated outstanding potential for this approach in creating effective pore arrays for biomolecule separation.

Laboratory Directed Research and Development

Biological Sciences





Multiprobe Investigation of the Proteomic Structure of Pathogens

Alexander J. Malkin 04-ERD-002

Abstract

We will investigate the proteomic structure, architecture, and function of bacterial spores through a combination of high-resolution, in vitro atomic-force microscopy (AFM) and AFM-based immunolabeling with threat-specific antibodies. This experimental approach will allow us to visualize, localize, and determine both surface and internal protein components of bacterial spores; model their complex architecture; and establish the relationship between pathogen structure and function. A more comprehensive understanding of the architecture and properties of bacterial spores would contribute significantly to a general understanding of their life cycle and may lead to advances in diagnostic and immunological aspects of *Bacillus anthracis* (anthrax) biodefense.

This work will provide a foundation for more efficacious diagnostic, chemotherapeutic, and immunological countermeasures for emerging diseases and for biodefense by elucidating the structurally related properties and function of bacterial spores and by modeling their architecture. It also will reveal the architecture, structure, and molecular-scale mechanisms of the therapeutic germination process of spores of the engineered bacterium *Clostridium novyi-NT* and will add new dimensions and insights to a pioneering bacteriolytic therapy for cancer treatment.

Mission Relevance

By developing AFM for evaluation of pathogenic morphological signatures and structural attributes, this project supports the Laboratory's national 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 modeling pathogen architectures for use in structural biology, in support of the Laboratory's mission in biosciences to improve human health.

FY07 Accomplishments and Results

In FY07, we (1) completed proteomic mapping of the spore coat and exosporium of several bacterial species, (2) demonstrated that bclA glycoprotein is the dominant epitope on the surface of *Bacillus anthracis* spores, (3) characterized the architecture of *B. anthracis* spores formulated under different processing conditions, and (4) completed transmission electron microscopy and AFM analyses of the structure of dormant and germinating *C. novyi-NT* spores and demonstrated the first case of nonmineral crystal growth patterns for a biological organism. The scientific basis and methodologies developed in this project will enable comprehensive studies of pathogen life cycles and provide a new capability for the development of attribution and forensic systems.

Publications

Malkin, A. J., et al., 2007. *Characterization and detection of biological weapons with atomic force microscopy*. Chemical and Biological Countermeasure Technologies for Homeland Security, Boulder, CO, Oct. 10–13, 2006. UCRL-PROC-224796.

Malkin, A. J., et al., in press. “Virus architecture probed by atomic force microscopy.” *Structure-Based Study of Viral Replication*. R .H. Cheng and T. Miyamura (Eds.), World Scientific Publishing Co., Hackensack, NJ. UCRL-BOOK-219826.

Plomp, M., et al., 2007. *In vitro high-resolution architecture and structural dynamics of single pathogens*. 2007 Materials Research Society Fall Mtg., Boston, MA, Nov. 26–30, 2007. UCRL-ABS-231840.

Plomp, M., et al., 2007. “In vitro high-resolution structural dynamics of single germinating bacterial spores.” *Proc. Natl. Acad. Sci. Unit. States Am.* **104**, 9644. UCRL-JRNL-226759.

Plomp, M., et al., 2007. “Spore coat architecture of *clostridium novyi*-NT spores.” *J. Bacteriol.* **189**, 6457. UCRL-JRNL-230805.

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

Lisa J. Stubbs 04-ERD-084

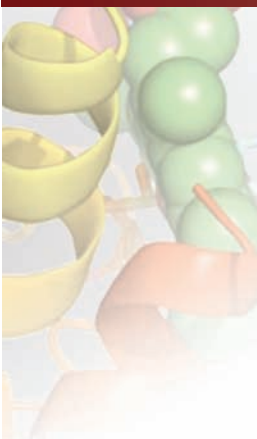
Abstract

This research seeks to establish new strategies and technologies for characterizing transcription factor (TF) proteins and the regulatory pathways in which they participate in living cells. Understanding TF proteins 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 a subclass of human TF proteins, we will develop a pipeline for characterizing them and their 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.

The project will develop novel methods for identifying TF protein binding sites and target genes in living cells. In addition, we will recruit and train several postdoctoral scientists with state-of-the-art expertise to add depth and breadth to genetics, comparative genomics, biochemical technology, and bioinformatics. We will also 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. By building expertise in regulatory biology, biochemistry, and network modeling, this project supports Laboratory



missions in biodefense, environmental management, and bioscience to improve human health.

FY07 Accomplishments and Results

In FY07, we met all major milestones for this project. Specifically, we (1) completed analysis of 20 TF proteins, (2) confirmed identified TF protein targets and pathways, (3) explored human and primate TF protein variation to identify genetic factors leading to individual and species-specific differences in development and disease susceptibility, and (4) completed development of a novel technology for TF protein tagging. The National Institutes of Health has validated our initial research with support for follow-on studies.

Publications

Hamilton, A. T., et al., 2007. "Evolutionary expansion and divergence in a family of primate-specific zinc finger genes." *Genome Res.* **16**, 584. UCRL-JRNL-215844.

Huntley, S., et al., 2007. "A comprehensive catalog of human KRAB-associated zinc finger genes: insights into the evolutionary history of a large family of transcriptional repressors." *Genome Res.* **16**, 699. UCRL-JRNL-216004.

Nowick, K., et al., 2007. *KRAB-Krüppel zinc finger transcription factors—a family of transcription factors with unusual high expression differences between human and chimpanzee brains*. The Biology of Genomes, Cold Spring Harbor, NY, May 8–12, 2007. UCRL-POST-230749.

Woo, Y., et al., 2007. *FRET monitored site-specific labeling of protein with small molecules in live cells via protein trans-splicing: method and application*. Symp. Protein Society, Boston, MA, July 21–25, 2007. UCRL-POST-232856.

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

Kristen Kulp 04-ERD-104

Abstract

Previous studies of microbial response to environmental stress have shown 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 to facilitate modeling of metabolite fluxes. TOF-SIMS is a mass spectrometry technique used to characterize chemical composition and can produce chemical maps of the distribution of small molecules with the spatial resolution needed to interrogate single cells. Our goal is to enhance bioanalytical 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 develop an unexplored area of imaging mass spectrometry: 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 accessible for 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 analysis of bacterial metabolites in single cells. This work 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 also is relevant to DOE's Genomics:GTL program by developing the capability to measure metabolites in a single bacterial cell, and complements efforts to study spectral signature definition and intracellular compound localization in mammalian cells.

FY07 Accomplishments and Results

In FY07, we established (1) our ability to detect small spectral differences in complex matrices, (2) the effectiveness of 2-dihydroxybenzoic acid as a matrix for enhancing the TOF-SIMS detectable mass range, (3) an effective cell preparation method for mammalian cells that will facilitate investigations of bacterial infections, and (4) the usefulness of six multivariate statistical and chemometric methods for analyzing increasingly complex biological data sets. During the course of this project we have increased instrument sensitivity by installing and applying a gold primary ion beam, developed effective sample preparation methods for both bacterial and mammalian cells, and established a hierarchy for applying multivariate statistical techniques to our spectral data sets.

Publications

Berman, E. S. F., in press. "Chemometric and statistical analyses of TOF-SIMS spectra of increasingly complex biological samples." *Anal. Chem.* UCRL-JRNL-235865.

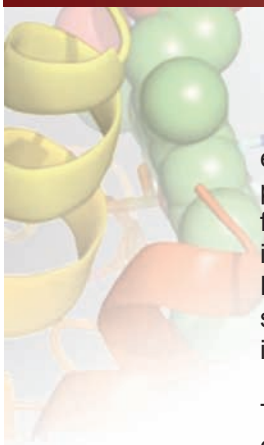
Berman, E. S. F., in press. "Preparation of single cells for imaging/profiling mass spectrometry." *J. Am. Soc. Mass. Spectrom.* UCRL-JRNL-235866.

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

Nan Liu 05-ERD-008

Abstract

The health effects of previously unidentified or unrecognized environmental contaminants are of prominent concern to 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 because of wastewater treatment facility and septic system discharges. The number and structural diversity of EDCs make it impractical to develop detection methods on a chemical-by-chemical basis. This project uses 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 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.

FY07 Accomplishments and Results

In FY07, we (1) finished analysis of FY06 microarray data for three remaining prototypical EDCs and identified unique sets of differentially expressed genes, (2) repeated Fourier-transform IR data from EDC-exposed cells and expanded work to characterize dose effects of EDC-induced changes in spectral regions of interest, (3) analyzed gene expression response of human cells to EDC mixtures using microarrays, (4) compared gene expression profiles from EDC mixtures to those developed from individual compounds to determine if a characteristic EDC gene expression response persists or differs from a mixtures exposure, and (5) tested the ability of gene microarrays to detect the presence of environmental EDCs.

Publications

Hall, L., et al., in press. "Estrogen-like effects of thyroid hormones on human breast cancer cell proliferation." *J. Steroid Biochem. Mol. Biol.* UCRL-JRNL-218037.

Innovative Copolymer Complex to Inhibit the Transport of Biological Aerosols

Paula W. Krauter 05-ERD-027

Abstract

We are developing a series of copolymer solutions for the purpose of immobilizing aerosolized biohazardous particles. The inhibition of secondary aerosolization and migration of biothreat particles has important implications for public health protection and contamination cleanup. We are evaluating film-forming copolymers with multiple functional groups capable of attracting and binding particles. These solutions are evaluated for their adhesion to biothreat agents in a series of wind-tunnel experiments in the laboratory and at a military base using highly refined aerosolized *Bacillus atrophaeus* spores. Using a 3,540-L aerosol chamber, we are assessing spore migration inhibition and the merits of adding decontamination agents to the formulation.

Expected results include (1) a copolymer-based solution formulated to be anionic to provide the 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 resuspension of contaminant particles through application of the copolymer. A successful completion of this project will also contribute to our basic understanding of particle shear stress and particle–copolymer adhesion strength under a variety of flow conditions.

Mission Relevance

This project addresses the Laboratory's national security mission to counter the proliferation and use of weapons of mass destruction by developing a new technology that inhibits effective dispersal and migration of biological weapon agents. The particle-suppression material developed in this project will increase the safety of emergency response personnel by abating breathing zone concentration, limit the potential for spore resuspension and migration, and aid in the restoration of contaminated areas.

FY07 Accomplishments and Results

An amphoteric acrylic copolymer solution was tested using a dry powdered biowarfare surrogate in an antistatic aerosol chamber designed and built by LLNL and U.S. Army scientists. The dissemination efficiency for a single release was determined to be 10 to 16%, and reaerosolization transport efficiency, without inhibitor application, ranged from 0.4 to 0.5%. However, the copolymer spray reduced reaerosolization transport efficiency to 0.03 to 0.0002%. Resuspension factors show that the copolymer solution inhibited spore resuspension by two orders of magnitude. Given the worst-case test environment (antistatic chamber and highly refined spores), this copolymer spray technique should further inhibit the resuspension of spores in a less-challenging environment.



Publications

Krauter, P. W., 2007. "Localizing and controlling BTA transport with polymer sprays." *Proc. 2007 Environmental Protection Agency Decontamination Workshop*. UCRL-PRES-231759.

Krauter, P. W., 2007. *Spore contamination: what concentration deposits, what resuspends, and can we inhibit its transport?* 2006 Workshop on Decontamination, Cleanup, and Associated Issues, Washington, DC, Apr. 26–28, 2006. UCRL-PRES-221011.

Krauter, P. W., 2007. "The problem with decontamination agent application: bioparticulate target—hit or miss?" *Proc. 2006 Denver Decon. Science and Technology Conf.* UCRL-POST-228248.

Krauter, P. W., and A. H. Biermann, 2007. "Reaerosolization of fluidized spores in HVAC systems." *J. Appl. Env. Microbiol.* **7**, 2165. UCRL-JRNL-224804.

Krauter, P. W., L. D. Larsen, and A. H. Biermann, 2007. *Novel approach towards elimination of inhalation hazards associated with reaerosolization of biothreat particles*. Am. Soc. Microbiology, Biodefense, and Emerging Diseases Res. Mtg., Baltimore, MD, Feb. 24–27, 2008. UCRL-POST-228248.

Krauter, P. W., 2007. "Inhibiting the transport of hazardous spores using polymer-based solutions." *J. Occup. Environ. Hyg.* **4**, 821. UCRL-JRNL-224651.

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 known proteins, yet many of these "hypothetical proteins" are important in nature. We propose to establish functions for a significant fraction of hypothetical proteins from an uncultivated microbial community by analyzing isolated proteins and the sequences that encode them. We will determine the distribution of proteins in a model community by fractionation and mass spectrometry analysis, identify source organisms and genome context, and measure protein abundance. To characterize unknown proteins, we will analyze protein sequences to predict functions, isolate proteins and multiprotein complexes, and test predicted functions using biochemical assays.

This research will advance understanding of how microbes act in their natural environment, particularly the molecular mechanisms of iron oxidation, biofilm formation, and evolution of adaptive mechanisms such as acid tolerance. We will develop analytical methods, find novel genes, and determine the function of previously unknown proteins, all of which will serve to accelerate genomics and environmental research and enhance the scientific basis for LLNL's focus on biosecurity. Important insights into protein function will result from the development of computational modeling tools. Furthermore, this project will establish unique expertise in the emerging area of proteogenomic analysis, and continue to build collaborations with researchers at the University of California at Berkeley and Oak Ridge National Laboratory.

Mission Relevance

This project will support LLNL's environmental management mission by developing new approaches to understanding natural microbial systems, particularly those that influence metal-contaminated environments. Work also supports the energy security mission because these microbial systems exacerbate environmental problems caused by mining principle energy sources such as coal and uranium. Research in this area is synergistic with DOE's Genomics:GTL program.

FY07 Accomplishments and Results

In FY07 we (1) extended our analyses to six distinctly different communities in an iron mine ecosystem and compared the proteins identified—results from this were reported in *Nature*, (2) expanded our repertoire of novel proteins from about 600 to over 2,000 and increased the number of protein structure predictions to over 350 using new high-throughput computational methods, (3) produced and developed several immunoreagents for detecting specific membrane proteins, (4) isolated genomic DNA from biofilm samples for clone library expression analyses and isolated DNA from several hot springs microbial communities for sequencing at the Joint Genome Institute, (5) isolated and biochemically characterized several new cytochrome proteins, and (6) hired two postdoctoral associates.

Proposed Work for FY08

For FY08, we will (1) finish our analyses of multiple protein targets and identify protein interactions within multiprotein complexes—this will require new chromatographic and mass spectrometry techniques, as well as biochemical assays of predicted functions and interactions; (2) move computational analyses to an array of high-performance processors for increased speed and accuracy of predictions; (3) develop tools to examine structural models for confidence, compare protein sequences from several acidophilic and neutrophilic organisms for specific structural differences, and analyze genomes for regulatory mechanisms that are relevant to novel proteins; and (4) submit three manuscripts for publication detailing our research results.

Publications

Banfield, J. F., et al., 2007. "Proteogenomic approaches for the molecular characterization of natural microbial communities." *OMICS: J. Integr. Biol.* **9**, 301. UCRL-JRNL-218040.

Beernink, P. T., et al., 2007. "Specificity of protein interactions mediated by BRCT domains." *J. Biol. Chem.* **280**, 30206. UCRL-JRNL-218035.

Lo, I., et al., 2007. "Strain-resolved community proteomics reveals that recombination shapes acidophilic bacterial genomes." *Nature* **446**, 537. UCRL-JRNL-225470.

Ram, R. J., et al., 2007. "Community proteomics of a natural microbial biofilm." *Science* **308**, 1915. 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 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. Our modeling tools to decompose the genome composition include non-negative matrix factorization, linear and logit models, and class-association rule mining, validated against published data.

The patterns we discover in gene composition across the spectrum of bacterial genomes will increase understanding of which genes, 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 cellular processes associated with its behavior, 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

By creating a predictive capability for genotype-to-phenotype mapping, this project is relevant to the goals of the DOE's Genomics:GTL program to gain predictive mastery of the microbial world, and it supports LLNL's missions in homeland security, environmental assessment and management, and biosciences to improve human health.

FY07 Accomplishments and Results

In FY07 we (1) compiled a phenotype database containing over 15,000 observations across 559 microbial organisms, (2) developed a novel phenotype prediction method, (3) analyzed six gene sets for phenotypes of interest, (4) adapted predictions to metagenomic data, and (5) investigated genome decomposition by class association rule data mining (and published a paper on the results), including developing a novel method to extrapolate phenotypes from evolutionary distance. Rather than regulatory analysis, we emphasized phenotype and metabolic analysis. Overall, this project achieved exciting results on microbial phenotypes. Follow-up work would include further application of our database and prediction tools, including possible integration in the Joint Genome Institute's Integrated Microbial Genomes database.

Publications

Light, S., T. Altman, and P. D'Haeseleer, 2007. *Assessment of phenotypic similarities between prokaryotes*. 3rd Intl. Conf. Metagenomics, La Jolla, CA, July 11–13, 2007. UCRL-POST-232548.

Tamura, M., and P. D'Haeseleer, 2007. *Comparative analysis of genome composition with respect to genotype-to-phenotype mapping by class association rule mining*. The 2007 Postdoctoral Program Symp., Livermore, CA, Sept. 6, 2007. UCRL-ABS-233733.

Tamura, M., and P. D'Haeseleer, 2007. *Comparative genome content analysis with respect to basic microbial phenotypes by class association rule mining*. 8th Intl. Conf. Systems Biology, Long Beach, CA, Oct. 1–6, 2007. UCRL-ABS-234209.

Developing and Integrating Novel Technologies for the Production and Characterization of Membrane Proteins

Paul D. Hoeprich 06-SI-003

Abstract

Membrane-associated proteins, although essential for mediating cellular processes, have received limited study because they are not amenable to conventional techniques used for isolating and characterizing soluble proteins. This project will develop robust, state-of-the-art methods to produce and characterize membrane proteins. Specifically, we will (1) produce membrane proteins by cell-free methods, (2) capture these proteins in nano-sized (10- to 20-nm) membrane mimetic structures or nanolipoprotein particles (NLPs), (3) optimize NLP production by creating semi-synthetic lipoprotein molecules, and (4) characterize and demonstrate unambiguous formation of membrane protein NLP constructs. Membrane proteins include G-protein-coupled receptors—for example, muscarinic receptors and hemagglutinins.

This project will result in generally applicable methods for the production, isolation, characterization, and functional reconstitution of membrane-bound proteins and protein complexes in NLPs. Success will significantly benefit life sciences in general and advance biological science at LLNL, contributing towards a better understanding of cell membrane-associated proteins. Following proof-of-principle studies, we will initially produce and capture membrane proteins relevant to biodefense. Subsequent work will focus on proteins that play an important role in DOE's strategies for environmental management. The capabilities we develop for membrane proteins will be adaptable to biodefense, human health research, bioenergy research, and environmental biology.

Mission Relevance

This work supports LLNL missions in biodefense, energy security via hydrogen generation, and environmental management through uranium bioremediation. It also will support Laboratory efforts in fundamental life sciences and biotechnology to improve human health by providing an understanding of membrane biochemistry, which is essential to the understanding of complex living systems.



FY07 Accomplishments and Results

In FY07, we (1) performed biochemical characterization of membrane proteins and NLP complexes; (2) scaled up scaffold protein production; (3) developed the use of lipophorins and related proteins as scaffold proteins; (4) achieved functionally active NLP production; (5) developed multiple methods for incorporating membrane proteins, such as rhodopsin, into NLPs; (6) developed proprietary cell-free synthesis of membrane protein expression with simultaneous NLP formation; and (7) filed two provisional patent applications. Overall, these activities are leading to state-of-the-art methods to produce and characterize membrane proteins.

Proposed Work for FY08

In FY08, we will (1) expand our efforts in cell-free protein synthesis for preparation of NLP–target protein constructs; (2) expand membrane proteins of interest to include *Yersinia pestis* membrane proteins, G-protein-coupled receptors, West Nile virus envelop proteins, toll-like receptors and, possibly, some plant cellulases; (3) perform specific fluorescent tagging of reactants to understand assembly; (4) study semi-synthetic apolipoproteins and their impact on NLP formation; and (5) develop an approach to NLP-based therapeutic and vaccine countermeasures.

Publications

Chromy, B. A., et al., in press. “Different apolipoproteins impact nanolipoprotein particle formation.” *J. Amer. Chem. Soc.* UCRL-JRNL-232256.

Development of Single-Cell Raman Spectroscopy for Cancer Screening and Therapy Monitoring

James W. Chan 06-ERD-051

Abstract

Current methods for determining cancer prognosis and monitoring therapy response are nonspecific and invasive, making the development of better methods for early detection of cancer and its recurrence vital for improved survival rates. Our objective is to develop new techniques for early cancer detection and therapy monitoring using single-cell Raman spectroscopy. We plan to identify new Raman markers associated with cancer cells, drug-resistant cancer cells, and cancer cell response to treatment conditions (e.g., chemotherapy and radiation) that can be used to improve detection and therapy response. To understand the biology of these markers, we will acquire data on different leukemia cell types and cellular processes, apply algorithms for analysis, and correlate results to clinical parameters.

We expect to (1) demonstrate the ability of laser-tweezers Raman spectroscopy to identify single cancer cells based on spectral markers, (2) identify spectral markers associated with dynamic cell response to chemotherapy and radiation treatment, (3) determine this technique’s accuracy and sensitivity, (4) achieve a fundamental understanding of the

biological relevance of these spectral markers, and (5) develop an optical-trapping nonlinear coherent anti-Stokes Raman spectroscopy method that should significantly speed up single-cell analysis. The cancer signatures and their dynamic changes to stimuli, as well as the data algorithms and Raman technology developed, will lead to a novel clinical tool that will replace or complement existing technology to improve early detection and diagnosis.

Mission Relevance

This project supports the Laboratory's mission in bioscience to improve human health. By developing multiplexed assays and Raman-based optical techniques for cancer diagnostic applications, this project will enable more cost-effective, highly specific, early disease detection technology and biomedical instrumentation.

FY07 Accomplishments and Results

In FY07, we (1) expanded our analysis to a larger number of normal and cancer cells and showed high reproducibility and detection sensitivity; (2) performed chemometric analysis to identify different cancer subtypes; (3) showed high discrimination of normal and cancer monocytes; (4) demonstrated that Raman spectroscopy can monitor a dynamic cell process (i.e., protein expression); (5) developed microfluidic channels and arrays for rapid, multimodal analysis of cells; (6) validated our data by comparing the results to clinical parameters (flow cytometry); and (7) applied Raman spectroscopy on activated/apoptotic cells to determine the biology of the markers and their effect on cancer detection.

Proposed Work for FY08

In FY08, we propose to focus on therapy monitoring. We will (1) use Raman spectroscopy to monitor cell response to chemical and radiation treatment and to identify subsets of cancer cells responsible for cancer growth, (2) expose cells to different levels of radiation and cancer drugs (doxorubicin and taxol) and monitor their response in real time, (3) sample cells from patients at different points during treatment and correlate results to biological data using the multiwell arrays developed in FY07, (4) characterize the spectra of drug-resistant cells sorted according to surface markers and drug-efflux phenotype prior to Raman analysis, and (5) monitor the cells' efflux dynamics by using drug Raman markers to measure concentrations.

Publications

Chan, J. W., et al., in press. "Laser tweezers Raman spectroscopy of single lymphocytes reveals distinct differences in cells after chemical fixation." *Anal. Chem.* UCRL-JRNL-234574.

Chan, J. W., et al., 2007. "Monitoring dynamic protein expression in single living *E. coli* bacterial cells by laser tweezers Raman spectroscopy." *Cytometry Part A* **71A**, 468. UCRL-JRNL-227444.

Chan, J. W., et al., in press. "Non-destructive identification of individual leukemia cells by optical trapping Raman spectroscopy." *Canc. Res.* UCRL-JRNL-229152.



***Francisella Tularensis*: Understanding the Host–Pathogen Interaction**

Amy Rasley 06-ERD-057

Abstract

The highly infectious nature of *Francisella tularensis*—the agent that causes tularemia in humans—highlights a need for continued research efforts to understand the interactions of this organism with host immune defenses. Very little is known about how this pathogen defeats host immune responses to cause disease, and no licensed vaccine is currently available. This project will study the host–pathogen interaction with respect to pathogenesis and environmental persistence. Our goals are to uncover global host-response patterns that may be used for early detection of exposure to this potential biowarfare pathogen and to understand how *F. tularensis* evades innate immune defenses to cause disease and how it persists in nature.

If successful, this project will identify the genes and gene pathways that are important during *F. tularensis* infection and define the role that host pattern-recognition receptors play in detecting *F. tularensis* and initiating innate immune responses. The proposed research could ultimately lead to the identification of target genes that could be used to develop efficacious therapies to combat tularemia and may help us identify potential early-warning markers for biowarfare agent exposure. In addition, we hope to identify factors involved in environmental persistence by studying the interaction between *F. tularensis* and environmental amoeba—findings that may improve detection efforts and aid in characterization of environmental samples.

Mission Relevance

Developing the ability to effectively detect and treat exposure to the highly infectious, potential biowarfare agent *F. tularensis* first requires gaining an understanding of how this pathogen interacts with host cells to cause disease. Moreover, by defining this previously unknown pathway of pathogen infection and linking it to the human genomic sequence, we will be advancing the detection of emerging and engineered threats. We will also understand how this bacterial persists in nature. Consequently, this research directly supports LLNL's national security and homeland security missions in the area of biodefense.

FY07 Accomplishments and Results

In FY07, we (1) expanded our study to include analyses of *F. tularensis*–amoeba interactions, demonstrating that pathogenic strains of *F. tularensis* induce rapid amoeba encystment mediated by a secreted factor; (2) finalized a list of host genes differentially expressed in mouse macrophages during *F. tularensis* infection and compared this list with preliminary gene expression data from human macrophages; (3) characterized the ability of different strains of *F. tularensis* to survive and replicate within phagocytic cells (human and mouse); (4) isolated RNA from bacteria grown in phagocytes to identify genes involved in intracellular survival; and (5) analyzed toll-like receptor protein expression by fluorescent-activated cell-sorting analyses.

Proposed Work for FY08

In FY08, we will (1) finalize microarray data from bacteria grown inside host phagocytic cells (mouse and human macrophages and amoeba)—data that may shed light on the dramatic differences observed between strains in their ability to survive intracellularly and may indicate why the live vaccine strain is attenuated and unable to cause human disease; (2) use microscopy to determine how this pathogen traffics through phagocytic cells; (3) develop a mechanistic understanding of the role of toll-like receptors during *F. tularensis* infection; and (4) identify the soluble factors secreted by pathogenic *F. tularensis* strains that cause rapid amoeba encystment to shed light on factors involved in environmental persistence.

Characterization and Quantification of Dynamic Robustness in Biological Systems

Eivind Almaas 06-ERD-061

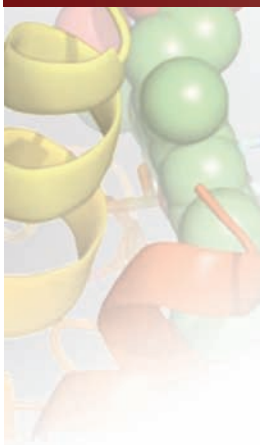
Abstract

Organisms—even single-celled ones—are extremely complex dynamic systems that must maintain functional stability despite being constantly impacted by destabilizing forces. Recent discoveries show that a major destabilizing force lies in the stochastic nature of the cellular machinery itself. The aim of this proposal is to (1) develop analytical and computational methods to identify elements in whole-cell network architectures that increase the robustness of that cellular machinery, (2) analyze weaknesses and failure modes of these robustness-conferring elements, (3) develop new constraint-based approaches to studying cellular networks, and (4) establish and test new metabolic network models that incorporate regulatory programs for *Yersinia pestis*, the organism that causes bubonic plague.

By coupling experimental information on whole-cell organization with analytical and computational predictions and models, we expect to develop a comprehensive understanding of the levels of environmental and inherent internal variation that microbes can tolerate. We hope to infer general principles of how individual microorganisms and communities of microorganisms manage both internal and external stochastic variations to avoid the multitude of potential failure modes. We aim to understand and predict the mechanisms that determine observed failure rates and gain a similar understanding of the dominant failure modalities. For *Y. pestis*, we also expect to identify new, network-based antimicrobial approaches.

Mission Relevance

By identifying possible genetic targets for antibiotics to use against *Y. pestis*, this project supports Lawrence Livermore's national and homeland security missions in the area of biodefense as well as environmental stewardship. More broadly, this project supports LLNL missions in breakthrough science by developing new expertise for the study of stochastic noise propagation in biological networks and the robustness of cellular organisms.



FY07 Accomplishments and Results

In FY07, we (1) developed our first version of a metabolic model for *Y. pestis*, with 1,099 metabolites and 1,267 reactions; (2) developed a stochastic model for a small gene-regulatory circuit with two interlinked positive-feedback loops and analyzed noise-processing properties; (3) completed computational predictions for epistasis in *Escherichia coli* and yeast and predicted numerous lethal gene knockouts, many of which were confirmed through single-gene inhibition experiments conducted in collaboration with Harvard Medical School; (4) published several journal articles and a book chapter; (5) gave a number of invited and contributed talks; and (6) hired two postdoctoral researchers.

Proposed Work for FY08

In FY08, we will (1) complete stochastic analysis of multiple small gene-regulatory circuits; (2) complete development and testing of a new constraint-based approach (dynamic flux balance) to analyze noise propagation in whole-cell networks; (3) include virulence production into the *Y. pestis* metabolic model, as well as at least one additional regulatory module; (4) validate our model against experimental results with gene expression arrays; (5) complete models for the three *Y. pestis* variants and compare their metabolic performance; and (6) analyze the robustness of the *Y. pestis* metabolic model.

Publications

Almaas, E., 2007. "Biological impacts and context of network biology." *J. Exp. Biol.* **210**, 1548. UCRL-JRN-227407.

Almaas, E., 2007. "Optimal flux patterns in cellular metabolic networks." *Chaos* **17**, 026107. UCRL-JRN-227543.

Almaas, E., A. Holder, and K. Livingstone, in press. "An introduction to systems biology for mathematical programmers." *Optimization in Medicine and Biology*. Taylor & Francis, New York, NY. UCRL-BOOK-227138.

Deri, J., and E. Almaas, 2007. *Dynamic modules in complex biological networks*. UCRL-POST-233427.

Ghim, C.-M., and E. Almaas, 2007. *Combinatorial principles of feedback control in biochemical networks*. 11th Ann. Intl. Conf. Research in Computational Biology, Oakland, CA, Apr. 21–25, 2007. UCRL-ABS-227858.

Ghim, C.-M., and E. Almaas, 2007. *Quantitative and functional characterization of genetic regulatory circuits in vivo*. UCRL-ABS-233717.

Ghim, C.-M., and E. Almaas, 2007. *Structural and functional characterization of combined regulatory network motifs*. 8th Intl. Conf. Systems Biology, Long Beach, CA, Oct. 1–6, 2007. UCRL-ABS-234207.

Jin, R., S. McCallen, and E. Almaas, in press. "Trend motif: a graph mining approach for analysis of dynamic complex networks." *Proc. 7th IEEE Intl. Conf. Data Mining*. UCRL-PROC-231357.

Navid, A., and E. Almaas, 2007. *Analysis of systems properties of the Yersinia pestis biovars using a genome-scale constraint based model*. 11th Ann. Intl. Conf. Research in Computational Biology, Oakland, CA, Apr. 21–25, 2007. UCRL-ABS-227857.

Navid, A., and E. Almaas, 2007. *Analysis of the metabolic properties of Yersinia pseudotuberculosis and Yersinia pestis biovars using genome-scale constraint based models*. UCRL-ABS-233747.

Navid, A., and E. Almaas, 2007. *System-level analysis of metabolism in Yersinia pseudotuberculosis and the biovars of Yersinia pestis using a genome-scale mathematical model*. 8th Intl. Conf. Systems Biology, Long Beach, CA, Oct. 1–6, 2007. UCRL-PROC-234174.

Redox Proteins in Environmentally Relevant Bacteria

Harry R. Beller 06-ERD-063

Abstract

This project adopts a systems biology approach combining genomics, transcriptomics, and proteomics to explore novel, metal-oxidizing reactions that can mediate the behavior of uranium in the subsurface. Our focus is on redox-active proteins that can abstract electrons from minerals such as uraninite. This is particularly pertinent to a remediation process of great interest to DOE: the in situ, reductive immobilization of metals, including radionuclides. This project will utilize collaborations with the DOE Joint Genome Institute and leverage LLNL's existing capabilities in genomics, microbiology, protein biochemistry, and geochemistry in a significantly new research direction for bioscience.

Expected results include (1) new knowledge about redox-active, outer membrane proteins in the bacterium *Thiobacillus denitrificans*, which carries out the anaerobic, nitrate-dependent oxidation of metals; (2) a complementation system in *T. denitrificans* that can be used to confirm the results of targeted gene mutation (knockout) studies; and (3) a better understanding of the genetic and biochemical basis of anaerobic metal oxidation. These results will be documented through publications in high-visibility journals.

Mission Relevance

This project furthers LLNL's mission in environmental management, particularly the enduring national need for remediation of uranium contamination. The subject of this project—microbial processes active in the near-surface environment—is also relevant to an important national security issue, namely radionuclide contamination of groundwater and the safe disposition of contaminated waste.

FY07 Accomplishments and Results

Our FY07 accomplishments include (1) fractionation of the outer membrane (OM) proteins of *T. denitrificans* by sucrose density gradient ultracentrifugation for different conditions of



induction (including iron carbonate oxidation and thiosulfate oxidation), (2) identification of the heme-containing proteins in the OM fraction by heme visualization after separation by gel electrophoresis, (3) de novo protein sequencing of the relevant OM proteins, (4) generation of knockout mutants based on the protein work and assay of these mutants for U(IV) oxidation, (5) completion of a genetic complementation system for *T. denitrificans*, and (6) publication of our genetic system for *T. denitrificans* in a high-profile journal.

Publications

Letain, T. E., et al., 2007. "Development of a genetic system for the chemolithoautotrophic bacterium *Thiobacillus denitrificans*." *Appl. Environ. Microbiol.* **73**, 3265. UCRL-JRNL-226908.

Developing a New Accelerator Mass Spectrometry Assay for Quantitation of Platinum DNA Adducts for Response to Platinum-Based Chemotherapy

Paul Henderson 06-LW-023

Abstract

Platinum-based drugs are the most successful class of compounds for the treatment of cancer. These drugs kill cancer cells through toxic DNA damage. However, many patients are unresponsive to treatment or acquire drug resistance. We will address this problem using carbon-14-labeled carboplatin and carbon-14-labeled oxaliplatin, both platinum-based anticancer drugs, and accelerator mass spectrometry (AMS), the most sensitive method for studying long-lived isotopes. We will measure platinum–DNA adducts in cultured cancer cells and mice exposed to the compounds. This study aims to develop an assay for determining which patients will benefit from carboplatin treatment and which will be resistant. This project is being conducted in collaboration with researchers from the University of California Davis Cancer Center.

This project will develop robust assays for quantitation studies on platinum–DNA adduct formation and repair using AMS detection of radiolabeled tracers for ultimate use in human studies. The unique analytical method developed is expected to provide the scientific proof-of-principle framework for the application of AMS to drug metabolism and personalized medicine for platinum-based drugs. We expect AMS data from cells and mice dosed with carboplatin to clearly differentiate resistant from sensitive tumors. Such differences may include rates of accumulation in cells and DNA of the radiolabeled drug and different rates of DNA repair. Applications of the resulting methodology to chemotherapy in humans will be proposed if we are successful.

Mission Relevance

The ability to track pharmacokinetics at exceedingly low isotopic doses will directly contribute to LLNL's mission in biosciences to improve human health and has potential applications in biothreat detection for LLNL's missions in national security and homeland security.

FY07 Accomplishments and Results

We exposed several human cancer cells lines to carbon-14-labeled platinum drugs. The cells and DNA became radiolabeled, and we observed the drug bounding to the DNA in two phases: monoadducts attached to a single strand of DNA, after which diadducts formed from crosslinking of the drug between two sites in the DNA. The sensitive cells always accumulated more radioactivity than the resistant cells, indicating a strategy for predicting which tumors will respond to therapy in humans. These achievements resulted in two peer-reviewed papers and a U.S. patent application. The next step would be a human clinical study with AMS.

Publications

Hah, S. S., and R. A. Sumbad, 2007. *Biomedical applications of accelerator mass spectrometry: kinetics of oxaliplatin–DNA binding in genomic DNA and human cancer cells*. American Association for Cancer Research Ann. Mtg., Los Angeles, CA, Apr. 14–18, 2007. UCRL-ABS-226362.

Hah, S. S., et al., 2007. *Characterization of oxaliplatin–DNA adduct formation in naked DNA and cancer cells using accelerator mass spectrometry*. UC Davis Cancer Center 13th Ann. Symp., Davis, CA, Sept. 24, 2007. UCRL-POST-229855.

Hah, S. S., et al., 2007. *Quantitation of oxaliplatin–DNA binding in genomic DNA and human cancer cells using accelerator mass spectrometry*. 2007 American Association of Pharmaceutical Sciences Natl. Biotechnology Conf., San Diego, CA, June 24–27, 2007. UCRL-ABS-228094.

Hah, S. S., et al., in press. “Characterization of oxaliplatin–DNA adduct formation in DNA and differentiation of cancer cell drug sensitivity at microdose concentrations.” *Chem. Res. Toxicol.* UCRL-JRNL-229554.

Analysis of the Mucin Membrane Protein by Cryo-Electron Microscopy and Computational Image Processing

Joseph B. Pesavento 06-LW-064

Abstract

We propose to express, purify, image, and structurally analyze the membrane-associated portions of a mucin protein (MUC1) using a combination of cryo-electron microscopy (cryo-EM) and computational image processing. MUC1 is linked to the survival of aberrant cell types with respect to invasion and metastasis in cancer. Our results will provide researchers with a better understanding of both the normal and abnormal functions of mucin, which should lead to improvements in diagnostics and therapeutics for cancer treatment. Imaging will be conducted using existing cryo-EM facilities at the University of California at Davis and a new instrument at Livermore, and processing will occur on computers at Livermore.



We expect our results will be twofold. First, we will deliver a new method for producing membrane proteins under conditions that facilitate their incorporation into lipid bilayers and imaging by cryo-EM, followed by three-dimensional reconstruction and visualization. A paper describing these results—specifically the cell-localization behavior of the protein and cell-surface characteristics—will be submitted for publication to a peer-reviewed journal. Second, our work will result in a parallelized code and image-processing workflow for large image sets of cryo-EM, protein, and macromolecular complex data processing and structural reconstructions. This project will help determine the biochemical and structural basis for mucin's cancer-causing properties.

Mission Relevance

Our project contributes to the Laboratory's national and homeland security and biosciences missions. This technology can be used for research on microbial pathogens such as *Yersinia pestis* (plague) and *Bacillus anthracis* (anthrax), as well as viruses such as coronavirus (SARS), vaccinia, and hemorrhagic fever.

FY07 Accomplishments and Results

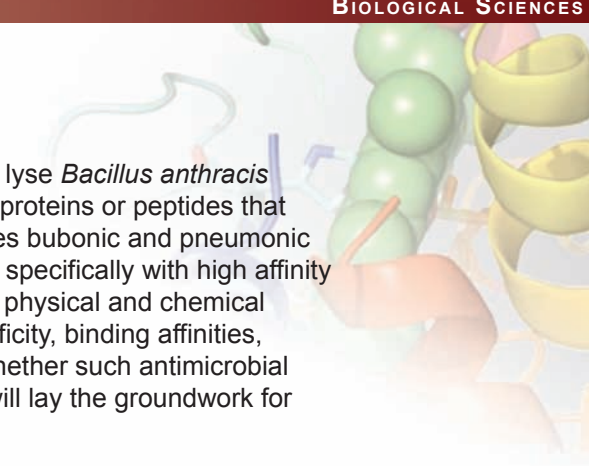
In FY07, we (1) cloned mucin purified from tissue-culture cells into an expression vector, (2) successfully tested our in vitro translation/cryo-EM chamber using mucin and bacteriorhodopsin and submitted the technology as a record of invention, (3) compiled cryo-EM image-processing algorithms and tested them on Livermore supercomputers to conduct image processing of large data sets, (4) prepared manuscripts detailing the speed gains made using the supercomputers, and (5) generated a successful reconstruction of the *Hepatitis E* virus G4 data from University of California at Davis—this reconstruction was paired with a homology model generated at Livermore and the results prepared for publication.

Development of Novel Antimicrobial Proteins and Peptides Based on Bacteriophage Endolysins

Paul J. Jackson 07-ERD-025

Abstract

Antibiotics, the primary tool for fighting bacterial diseases, are limited by microbial resistance and secondary health effects. New antimicrobial agents are needed. We will investigate endolysins, bacteriophage-produced proteins that burst bacterial cells, as antimicrobial and detection agents that target specific microbes. We will identify different phage endolysin genes and use these genes and segments thereof for in vitro production of different endolysins and endolysin fragments. We will measure the binding and lytic (bacterial cell-bursting) function of the resulting proteins and peptides to determine specificity and impact on bacterial targets. Based on these initial studies, we will refine our fragment design to optimize bacterial binding and lytic traits of small endolysin protein fragments.



We plan to produce antimicrobial proteins or peptides that rapidly lyse *Bacillus anthracis* and related pathogenic bacilli. We will also produce antimicrobial proteins or peptides that can rapidly lyse *Yersinia* species, including *Y. pestis*, which causes bubonic and pneumonic plague. We also intend to produce one or more proteins that bind specifically with high affinity to each of these threat agents. The knowledge we gain about the physical and chemical properties of these proteins and peptides, their lytic activity, specificity, binding affinities, and stability under different conditions will provide insights into whether such antimicrobial proteins can be used to control other pathogenic microbes, and will lay the groundwork for producing large quantities of these agents.

Mission Relevance

This project will develop a novel approach to the in vitro production of a new class of antimicrobial and microbe-detection compounds in support of LLNL's national and homeland security missions.

FY07 Accomplishments and Results

For FY07, we (1) characterized genes encoding two endolysins and expressed these proteins in vitro; (2) designed, constructed, and expressed genes encoding the N- and C-terminal portions of these proteins and tested the lytic and binding properties of the full-sized and truncated proteins; (3) sequenced two additional phage genomes to identify additional endolysin genes; (4) identified several additional endolysin gene sequences from published sequences of phage that infect *B. anthracis* and *Y. pestis* and from prophages (inserted phage genomes) in these bacterial genomes and began the process to amplify and express these genes; and (5) developed a more rapid, quantitative assay to measure the lytic activity of endolysins.

Proposed Work for FY08

In FY08, we will (1) continue to characterize the properties of the first two endolysins, including their N- and C-terminal regions, to understand their efficacy, target range, and physical and enzymatic characteristics; (2) characterize additional phage endolysin genes and endolysins; (3) identify additional endolysin genes by sequencing additional phage genomes; (4) optimize the endolysin assay and study endolysin binding to target bacteria in the absence of lysis; (5) continue computer alignment studies to identify the specific endolysin protein sequences responsible for lytic activity and target recognition, and use this information to design and produce even more-active, specific protein fragments; and (6) test these proteins to determine their impact on the two pathogenic bacteria.



Development of Novel Transgenic Technologies to Study Genome Regulation and Architecture

Gabriela G. Loots 07-ERD-046

Abstract

We propose to develop novel transgenic technologies to identify and characterize the biological function of noncoding sequences in genomes. We intend to test some of the known transposable elements including Tol2 and piggyBac for the potential to enhance transgenesis in the frog *Xenopus tropicalis*. The advantage of developing transposon-mediated transgenesis in frogs is that foreign DNA is delivered to a fertilized embryo via microinjection, a procedure that is quick and easy because the fertilized egg is large and tolerant to manipulations. Our approach will blend state-of-the-art computing capabilities with cutting-edge technologies in genomic research.

We intend to develop a new set of experimental technologies that will allow us to identify the function of putative regulatory elements *in vivo*, using the frog as an efficient experimental system. We will establish novel transgenic technologies that will bypass most of the problems associated with current methodologies, and develop a universal approach to study genome regulation and architecture on a global scale. Specifically, we will create methods to deliver large fragments of DNA into the frog genome in a site-directed manner. This will permit us to associate regulatory element structure with function, and significantly advance the understanding of regulatory mechanisms in complex genomes.

Mission Relevance

This project supports LLNL's mission in biotechnology by developing new tools for basic biological research with application to prevention of disease from both natural and bioterrorist threats. This work also supports efforts in Laboratory outreach and recruitment through its strong potential to attract top-notch university collaborators, students, and postdoctoral fellows to the Laboratory.

FY07 Accomplishments and Results

In FY07, we (1) tested Tol2 and determined that it is suitable for frog transgenesis—we have obtained promising results, but need to further optimize the procedure to enhance its potential for high-throughput transgenic technology development; (2) developed a method for predicting tissue-specific enhancers, which was featured on the cover of the February 2007 issue of *Genome Research*; (3) used this method to predict 30 tissue-specific enhancers (liver and skeletal elements) for testing and characterization in transgenic animals; and (4) generated loxP constructs for identification of unidirectional gene delivery.

Proposed Work for FY08

In FY08, we will (1) develop an *in vivo* transgenic method that utilizes a universal reporter based on the transposable element system (Tol2 or piggyBac) and that works best in frogs,

(2) examine position effects and germline transmission, (3) determine the loxP site pairs that promote *in vivo* integration in frogs, (4) use homologous recombination to engineer a bacterial artificial chromosome by inserting green fluorescent protein as a reporter and transposable element, and (5) test this method in transgenic frogs to determine if position effects are eliminated and different alleles of the same bacterial artificial chromosome can be analyzed for impact on green fluorescent protein expression.

Publications

Leupin, O., et al., 2007. "Control of the SOST bone enhancer by PTH via MEF2 transcription factors." *J. Bone Miner. Res.* **22**(12), 1957. UCRL-JRNL-233272.

Pennacchio, L. A., et al., 2007. "Predicting tissue-specific enhancers in the human genome." *Genome Res.* **17**, 201. UCRL-JRNL-216861.

Microarrays + NanoSIMS: Linking Microbial Identity and Function

Jennifer Pett-Ridge 07-ERD-053

Abstract

This project will develop a next-generation microarray for the rapid genetic identification of biothreat agents, then combine this technology with the nanoscale high-resolution secondary ion mass spectrometer (NanoSIMS) to address issues in energy generation, human disease, and bioremediation. We will achieve these goals by leveraging LLNL's unique combination of microarray expertise and mass spectrometry capability. We will use the high resolution and sensitivity of NanoSIMS to detect isotopic enrichment in ribosomal RNA (rRNA) hybridized in a microarray. This novel technique will allow links between microbial identity and function in multiple applications and will substantially improve our ability to predict environmental microbial activity and engineer microbes to produce biofuels or degrade contaminants.

Against a background of nonenriched genes, we will identify isotopically enriched rRNA in bacteria extracted from cellulose-degrading biofilms, human tissues, and contaminated soils and labeled with carbon-13 (^{13}C). Comparing these data to the standard microarray fluorescence analysis, we will identify key microbial phylogenies and their functional roles. This knowledge could enable bioengineering of organisms that efficiently degrade pollutants or produce biofuels (hydrogen and methane). It also may be used to define pathways of infection by human pathogens during disease progression, because understanding specific requirements of pathogenic species may enable development of medical treatments. This technique will enable a long-sought means to link structure and function in unculturable microbes.

Mission Relevance

This project supports LLNL's national security mission by delivering an improved array methodology for biosecurity research; supports the energy security mission by furthering the



development of biofuels, fuel cell bioreactors, and secure energy sources; and supports the environmental management mission by furthering the remediation of contaminated sites and enabling modeling of microbial roles in carbon sequestration and global climate change.

FY07 Accomplishments and Results

In FY07 we (1) manufactured slides from several polyolefinic copolymers, (2) used chemical vapor deposition to coat them with tin-doped indium oxide (ITO) and showed that these slides ionize well and are conductive in the NanoSIMS device, (3) functionalized the ITO surfaces with alkyl phosphonates, (4) successfully synthesized a fiducial-only array with an alkyl-phosphonate-functionalized surface on ITO glass, and (5) began proof-of-concept tests of the technique combining NanoSIMS with stable isotope probing (NanoSIP) by generating ^{13}C bacteria, extracting their ^{13}C -rRNA, designing appropriate oligonucleotide probes, and hybridizing the rRNA onto our new arrays. Our initial experiments were conducted with human oral biofilm and soil bacteria.

Proposed Work for FY08

In FY08, we will finalize our new array and then test the NanoSIP technique in two real-world applications. First, we will use rRNA extracts from the DOE Hanford site, where ^{13}C -lactate has been added to stimulate chromium reduction. Extracts will be hybridized to ITO-alkyl phosphonate arrays printed with a probe set for all community members, which were previously identified. We will then test the capability of NanoSIMS to resolve ^{12}C -rRNA from ^{13}C -rRNA probe spots, allowing us to identify which organisms co-metabolized ^{13}C -lactate. Second, we will label human oral TM7 bacteria (associated with periodontal disease) with nitrogen-15-labeled amino acids and ^{13}C -oligosaccharides and use the NanoSIP approach to investigate their physiology.

Publications

Pett-Ridge, J., et al., 2007. *Microarrays + nanoSIMS: linking microbial identity and function*. Gordon Research Conf. Applied and Environmental Microbiology, South Hadley, MA, July 15–20, 2007. UCRL- POST-228479.

Identification of Pathways Critical to Quorum Sensing and Virulence Induction

Ted Ognibene 07-ERI-001

Abstract

Therapeutic compounds that interfere with quorum sensing in pathogens should either increase the window of opportunity for more traditional antibiotic intervention or attenuate the pathogen such that the host immune system can clear the infection. We propose to utilize carbon-14 labeling combined with accelerator mass spectrometry (AMS) detection to

quantitate the autoinducer molecules—acylhomoserine lactones (AHLs)—derived from the in vivo carbon-14 labeling of S-adenosylmethionine in *Yersinia pestis* and *Vibrio harveyi* (both disease-causing bacteria) to identify pathways critical to quorum sensing and to ascertain the role that quorum sensing may play in virulence induction. This will establish a platform in which the efficacy of novel therapeutics can be assessed.

We will demonstrate the effectiveness of using isotope labeling and quantitation by AMS to study biochemical pathways. We will provide a platform in which quorum-sensing molecules can be quantitated in bacteria. We will use AMS to identify pathways in *Y. pestis* that are critical to quorum sensing. The success of this project will lay the groundwork for AMS-based metabolite analysis of pathogens and also have possible applications to biodefense.

Mission Relevance

This project supports the Laboratory's national and homeland security missions by creating a capability to quantitate autoinducers in potential biowarfare agents. Autoinducer quantitation will help identify the pathways critical to quorum sensing and pathogen viability and could assist in identification of novel therapeutic targets for proteins associated with those pathways, as well as the assessment of therapeutic efficacy.

FY07 Accomplishments and Results

In FY07, we (1) developed and refined our analytical procedures for the separation and identification of AHLs through the use of commercially available standards; (2) separated a set of ten known AHLs using liquid chromatography and detected them by Fourier-transform ion cyclotron resonance mass spectrometry (FTICR); (3) identified each AHL by FTICR accurate-mass analysis of both the protonated ion and the sodium and potassium adducts; (4) established the conditions necessary for ionization and gas-phase transfer at the electrospray interface, as well as the ionization characteristics of molecules in electrospray ionization; and (5) cultured *Yersinia pestis* cells under different growth conditions and identified AHLs using our newly developed methods.

Proposed Work for FY08

In FY08, we will (1) continue culturing cells in defined media in the presence of carbon-14-labeled and unlabeled methionine; (2) quantitate AHLs in extracts from the carbon-14-labeled methionine, while the unlabeled cultures will be used for AMS identification of compounds separated by high-performance liquid chromatography; and (3) harvest *Y. pestis* cells at specific culture densities under both vector-like and host-like conditions. These results will demonstrate our platform's ability to identify and quantitate quorum-sensing molecules in bacteria and lay the groundwork for future experiments.



Quantification of Radiation-Induced Protein Expression

Matthew A. Coleman 07-LW-043

Abstract

This project will provide a new research tool, based on accelerator mass spectrometry (AMS), for the quantitative analysis of protein expression in cells induced by ionizing radiation (IR). Living systems have evolved mechanisms to detect and repair damage. The biological response to low-dose IR is difficult to quantify. Many IR-induced genes have been identified, but messenger RNA (mRNA) expression correlates poorly with protein levels. Response to radiation is therefore better quantified by protein expression, rather than the direct damage to genomic DNA or the mRNA. The high sensitivity of AMS combined with the specificity of antibodies against specific putative IR-induced proteins will enable quantitation of protein expression after radiation exposure at low levels (i.e., attamole).

First, we expect to demonstrate the quantitative ability of the affinity capture of proteins to selectively isolate expressed proteins that are metabolically isotope-labeled and quantified by AMS. We will subsequently quantify protein expression response to ionizing radiation using human-derived cell lines, pulse labeling with carbon-14 amino acids, affinity capture of targeted proteins, and AMS as functions of radiation dose and time. Quantified protein expression will be compared with mRNA expression to validate protein biomarkers of radiation. These approaches will be crucial for expanding the sensitivity of measuring proteins within a single cell.

Mission Relevance

By providing a significant new research tool for quantitative protein expression that bridges the gap between large proteomic data sets and the interactions of DNA-damage proteins, this project contributes to the development of better tools and capabilities for the Laboratory's homeland security and national security missions. The sensitivity of this AMS tool will also be applicable to low-dose radiation research and general investigations of proteins and their interactions.

FY07 Accomplishments and Results

In FY07, we (1) demonstrated a method for protein arraying using a Mylar substrate attached to glass surface that is compatible with AMS and that allows samples to be easily manipulated; (2) demonstrated the ability to quantify the expression of the transcription factor p53 in both TK6 and NH32 cells, verifying in measurements a dose response for p53 at low and high doses in the TK6 cell line that contains the functional p53; (3) used isotopically labeled proteins to demonstrate our ability to label tyrosines for specific quantification by AMS; and (4) compared the quantitative recovery and reproducibility of proteins captured by antibodies immobilized on low-density arrays using both the enzyme-linked immunosorbent assay and AMS.

Proposed Work for FY08

In FY08, we plan to (1) quantify protein expression response to ionizing radiation using the human-derived cell lines grown to confluence (i.e., cell cycle arrested) over time for specific targeted proteins using the optimized methods developed in FY07; (2) use AMS to quantify sample responses as functions of radiation dose and time; and (3) compare these data with mRNA expression to validate protein biomarkers of ionizing radiation, which will then be compared to existing knowledge of cancer-related pathways. We will also prepare results for publication in a high-profile, peer-reviewed journal.

Stem Cell Fate Decisions

Amy L. Hiddessen 07-LW-098

Abstract

We seek to understand the biological functions of stem cells to harness their power for tissue regeneration. Currently, internal and external factors that regulate stem cell fate are poorly understood. We aim to determine the roles and significance of immobilized factors like extracellular matrix and signaling peptides, synergistic and opposing soluble factors and signals, and cell–cell communication, in stem cell fate decisions. Cell arrays will be developed to capture a single cell or clusters of cells onto substrate-bound signals and proteins. Commitment to proliferation and/or differentiation will be assessed using time-lapse microscopy and fluorescent cell surface and intracellular markers during exposure to immobilized and soluble signals.

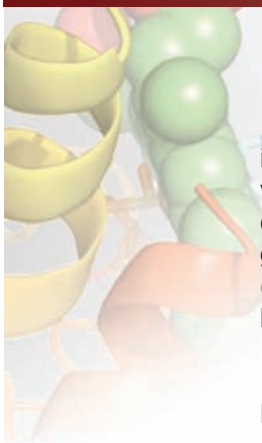
We expect to provide a greater understanding of how the cell microenvironment controls fate and signaling, information that can shed light on disease states and the therapeutic potential of stem cells in regenerative medicine. Single-cell data from our research will contribute to development of computational models for predicting cell behavior. Developmental and cancer biology will benefit from insights into the function of stem cells, and we expect our research to be aligned with National Institutes of Health and National Aeronautics and Space Administration interests, as well as with California's three-billion-dollar stem cell initiative, among others. In addition, this effort will help advance a larger stem cell research effort at LLNL, attracting top scientists in the field.

Mission Relevance

This research will support DOE and LLNL missions in biosciences to improve public health by advancing technologies for investigating cell signaling and response as well as medical treatments based on tissue regeneration. This work will apply and build upon technologies and techniques previously developed at LLNL, thus leveraging prior efforts and investments.

FY07 Accomplishments and Results

In FY07, we (1) adapted and developed chemical patterning approaches for arraying a single cell to clusters of model epithelial cells, (2) established functionalization protocols to



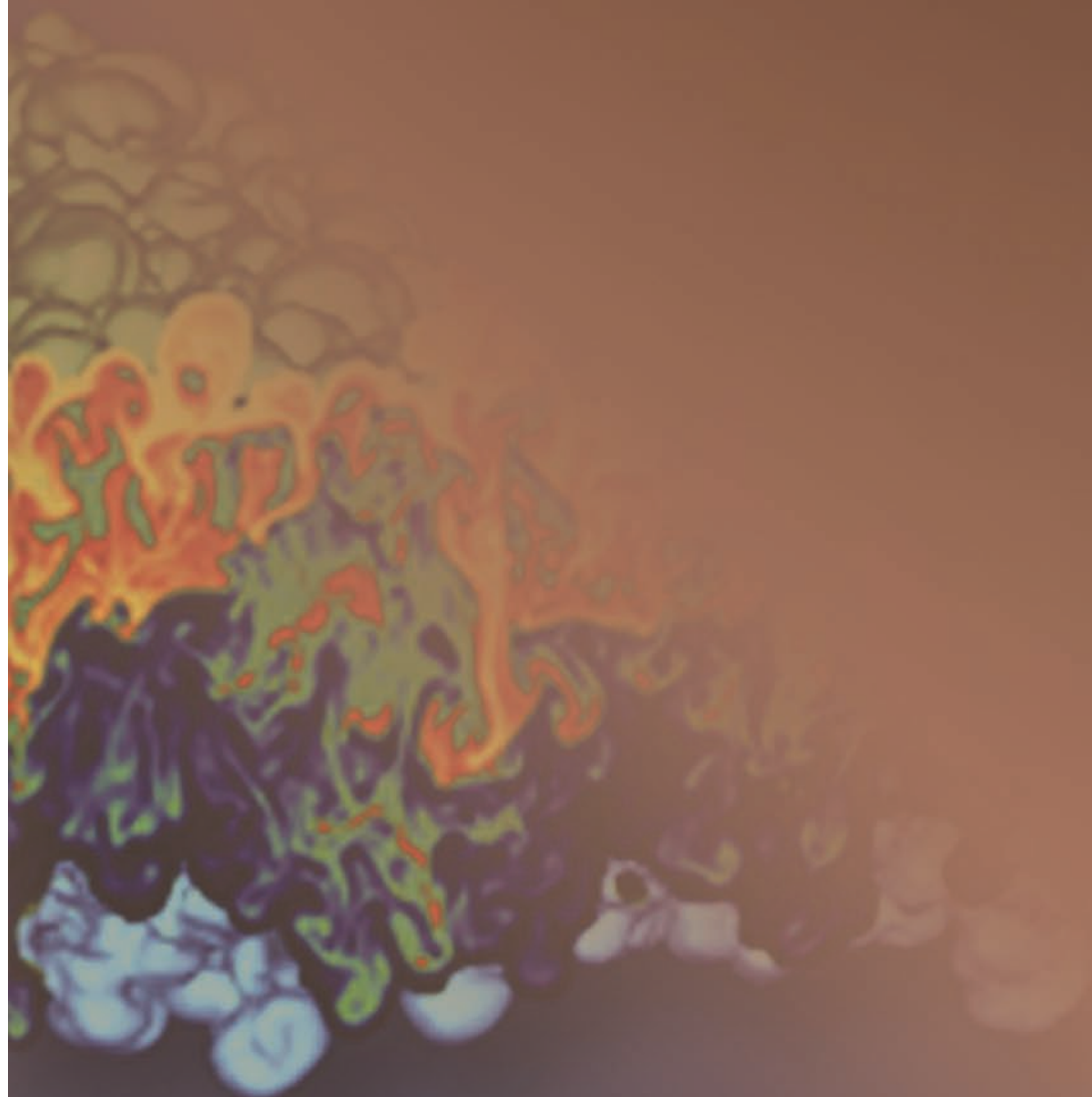
immobilize peptides on the array, (3) optimized and developed methods for extending cell viability on the array, (4) assessed baseline cell behavior as function of generic patterning chemistry (no signal peptides) and physical pattern parameters, (5) initiated development of a green fluorescent protein reporter needed to visualize and quantify signaling in live cells, and (6) characterized phenotypes of cells induced to differentiate by interactions with substrates having different extracellular matrix peptides.

Proposed Work for FY08

For FY08, we will (1) continue development of off-array cell assays for measuring fate-related signaling pathways; (2) assess, using assays developed in FY07, baseline behavior and state of cells on signal peptide-presenting arrays; and (3) begin measuring responses for epithelial stem cells to signal peptides using assays developed off-array.

Laboratory Directed Research and Development

Chemistry



New Fragment Separation Technology for Superheavy Element Research

Dawn A. Shaughnessy 04-ERD-085

Abstract

This project will investigate the topography of the western edge of the Island of Stability, with the intent of answering the important question of whether or not the next closed spherical proton shell is located at 114 protons. We will accomplish this via two paths. First, we will fabricate a thick plutonium ceramic target that will be bombarded with calcium-48 ions to produce long-lived element 114 isotopes that will be separated in the Mass Analyzer for Super Heavy Atoms (MASHA) mass separator in Dubna, Russia, to identify the atomic mass of the element 114 isotopes produced. Second, we will irradiate californium-249, curium-245 and -248, and americium-243 targets with the intent of producing isotopes of elements 118, 116, and 115, which will further clarify the western edge of the Island of Stability.

If these experiments prove to be successful, we will discover yet another new element (element 118), 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. We expect that the results of these experiments will extend and further improve the theoretical models used to calculate the decay properties and nuclear shapes of the heaviest elements.

Mission Relevance

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

FY07 Accomplishments and Results

The development of MASHA in Dubna was delayed so we focused on using physics and chemistry to study the Island of Stability. A new isotope of element 113 was discovered, further defining the western edge of the Island of Stability. Samples from chemical separations of element 105 performed in FY05 were extensively analyzed, and the results strengthen our claim that fission events observed in these samples were from the element 105 decay daughter of element 115. We participated in experiments that evaluated gas-phase chemistry of elements 112 and 114. The results from this project include discovery of a new chemical element (118) and several new isotopes, as well as elucidating the chemistry of single atoms of element 105, which showed ekatantalum behavior.

Publications

Oganessian, Y. T., et al., 2007. "Synthesis of the isotope $^{282}113$ in the Np-237+Ca-48 fusion reaction." *Phys. Rev. C*. **76**, 011601. UCRL-JRNL-230346.

Oganessian, Y. T., et al., 2007. "Synthesis of the isotopes of elements 118 and 116 in the Cf-249 and Cm-245+Ca-48 fusion reactions." *Phys. Rev. C*. **74**, 044602. UCRL-JRNL-220958.

Shaughnessy, D. A., 2007. *Future automation challenge—chemistry of element 114*. 233rd American Chemical Society National Mtg., Chicago, IL, Mar. 25–29, 2007. UCRL-ABS-229088.

Stoyer, M. A., 2007. *An examination of the experimental evidence for spherical proton shell closure in the heaviest elements*. 3rd Intl. Conf. Chemistry and Physics of the Transactinide Elements, Davos, Switzerland, Sept. 23–28, 2007. UCRL-PRES-234778.

Stoyer, M. A., 2007. *Superheavy elements and radioactive ion beams*. 234th American Chemical Society National Mtg., Boston, MA, Aug. 19-23, 2007. UCRL-ABS-229643.

Stoyer, N. J., et al., 2007. "Chemical identification of a long-lived isotope of dubnium, a descendant of element 115." *Nucl. Phys.* **787**, 388C. UCRL-PROC-224777.

Wilk, P. A., et al., 2007. "Separation of group five elements by reversed-phase chromatography." *J. Radioanal. Nucl. Chem.* **275**, 651657. UCRL-JRNL-228727.

Heterogeneous Processes at the Intersection of Chemistry and Biology

I-Feng W. Kuo 05-ERD-021

Abstract

Phenomena in heterogeneous environments are important processes found 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 non-bulk environments and to incorporating reactivity at no additional cost.

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

FY07 Accomplishments and Results

For FY07, we completed our large-scale quantum mechanical/molecular mechanical calculations of ODC-ase with its substrate OMP (orotidine 5'-monophosphate) and mapped out the full free-energy surface for common reaction mechanisms. The main results were published in a peer-reviewed journal, as was a paper on the validation of semi-empirical methods implemented in the code CP2. In addition, we made progress towards understanding atmospheric oxidation on the surface of sea salt aerosols using ab initio potential.

Publications

D'Auria, R., I. F. W. Kuo, and D. J. Tobias, in press. "An ab initio molecular dynamics study of the solvated OHCl-complex: implications for the atmospheric oxidation of (Cl⁻)_{aq} to (C₂)_{gas}." *J. Phys. Chem.* UCRL-JRNL-233308.

McGrath, M. J., et al., 2007. "Spatial correlation of dipole fluctuations in liquid water." *Mol. Phys.* **105**, 1411. UCRL-JRNL-226550.

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 have been designed by an adversary to be either incapacitating or lethal chemical and biological weapon (CBW) agents. From this information, detection and countermeasure strategies can be designed. We have sought to define, develop, and implement a comprehensive scientific approach coupling cutting-edge computational chemistry and new synthetic methods to enhance our understanding of the threats posed by the potential development and use of novel CBW agents. The efforts outlined here have led to high-quality results intended to cue U.S. defensive efforts by providing guidance in creating new detection and countermeasures programs.

Mission Relevance

The work described in this proposal 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, this work directly supports the LLNL mission of enhancing national

security and facilitating efforts to halt and reverse the proliferation of weapons of mass destruction.

FY07 Accomplishments and Results

During FY07, we continued exploration of synthesis and computational modeling as methods for characterizing the physiological capabilities and properties of candidate compounds that could be designed by an adversary as either incapacitating or lethal CBW agents. Over its lifetime, this project successfully demonstrated the usefulness of a combined computational-experimental program in the areas of chemical synthesis and biological interactions. This work has generated a great deal of interest in the defense and intelligence communities, including interest in follow-on work in both the computational and experimental aspects.

Discovering the Folding Rules that Proteins Obey

Olga 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. We will develop a robust microfluidic mixing device, conduct long-time simulations on supercomputers, and perform measurements on the systems that exhibit a fast folding and hydrophobic collapse. Our results will be used to answer the following questions: Can kinetic traps and intermediate states in the protein folding process 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 projects at LLNL. This research will establish and strengthen collaborations between LLNL scientists and leading researchers in academia and industry. Because of the fundamental nature of this project, it should result in publications in high-impact, peer-reviewed journals.

Mission Relevance

By preparing 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 also will support the Laboratory's mission in bioscience to improve human health.

FY07 Accomplishments and Results

In FY07, we (1) further shortened the mixing time in the microfluidic volume to 1 μ s; (2) fully characterized the mixers, fabricated them in fused silica and silicon, and made them available for experiments; (3) developed a capability for conducting mixing experiments using synchrotron radiation circular dichroism; (4) fabricated mixers compatible with direct ultraviolet excitation, observing the ultraviolet fluorescence spectrum from naturally occurring tryptophans in three well-studied proteins—cytochrome c, apomyoglobin, and lysozyme—as a function of time; (5) performed simulations with experimentally relevant starting conformations; and (6) focused simulations on a rapidly folding variant of the DNA binding domain from the lambda repressor that can also be investigated experimentally.

Proposed Work for FY08

In FY08, we will (1) perform computer simulations to accurately reproduce kinetic information on the folding of small proteins from experimentally relevant starting conformations, (2) address proteins of a size and folding rate that can also be addressed experimentally and focus on a fast-folding variant of the DNA binding domain from lambda repressor fragment 6-85, and (3) perform measurements with submicrosecond time resolution on lambda repressor 6-85 using both fluorescence resonance energy transfer and ultraviolet fluorescence spectroscopies, then compare experimental data with simulation results.

Publications

Hoffmann, A., et al., 2007. "Mapping protein collapse with single molecule fluorescence and kinetic synchrotron radiation circular dichroism spectroscopy." *Proc. Natl. Acad. Sci. Unit. States Am.* **104**, 105. UCRL-JRNL-225980.

Lapidus, L. J., et al., 2007. "Protein hydrophobic collapse and early folding steps observed in a microfluidic mixer." *Biophys. J.* **93**, 218. UCRL-JRNL-217804.

Yao, S., and O. Bakajin, 2007. "Improvements in mixing time and mixing uniformity in devices designed for studies of protein folding kinetics." *Anal. Chem.* **79**, 5753. UCRL-JRNL-229290.

Conversion of Plutonium and Enriched Uranium

Thomas W. Trelenberg 06-ERD-012

Abstract

This project seeks to determine the mechanisms governing hydride and oxide conversions of uranium and plutonium. The unpredictable nature of these conversion processes raises long-term storage and retrieval concerns for these materials and highlights a lack of understanding of how impurities serve to catalyze the reactions and their impact on archived scientific samples. Surface-science photoemission experiments will be used to characterize the reaction rates (with and without catalysts) and mechanisms that convert plutonium and uranium into their respective hydrides and oxides. Computer simulations using relativistic

quantum models will guide the experimental program. In turn, data collected for this project will help verify the modeling results.

The integration of our experimental and theoretical findings will lead to a predictive capability for these reactions, and begin the process of defining the mechanisms governing these conversions, providing insight into their proper application to the storage and retrieval of plutonium and uranium.

Mission Relevance

Actinide compounds such as hydrides or oxides are of interest for the storage, recovery, and retrieval of nuclear materials in support of the Laboratory's national security missions in stockpile stewardship and nonproliferation.

FY07 Accomplishments and Results

In FY07, we (1) updated calculations to include impurity atoms by examining the effect of a tungsten impurity on both uranium and plutonium, (2) used data from our archived test-sample studies and fit-to-model parameters from our calculations to predict optimal temperature for hydride recovery operations, (3) constructed an apparatus for hydriding studies on uranium samples to duplicate the plutonium experiments, (4) constructed and began using an independent system to investigate how to eliminate contaminants present in the reclaiming process (without actinides present), and (5) began building a glove box and sample-transport system for the surface science equipment.

Proposed Work for FY08

In FY08, we will (1) complete calculations including the catalysts identified in FY07 sample experiments using our modified computer code; (2) continue modifications to the test sample instruments and begin advanced experiments with catalysts; (3) conduct work with the uranium sample apparatus, paralleling work done with plutonium; (4) continue to evaluate the role of contaminants in the retrieval process; and (5) begin experimental work with plutonium and uranium with the surface science instruments.

Publications

Balasubramanian, K., et al., 2007. "Atomistic level relativistic quantum modeling of the plutonium hydrogen reaction." *J. Alloy Comp.* **444–445**, 447. UCRL-JRNL-222424.

Long-Time-Scale Shock Dynamics of Reactive Materials

Nir Goldman 06-ERD-037

Abstract

We are studying the long-time-scale effects of shocks on highly reactive materials, using a novel shock dynamics technique, the multiscale shock method (MSSM), implemented in an ab initio molecular dynamics code. Until recently, existing techniques for quantum simulations of shocked materials have been prohibitively expensive. The combination of MSSM with an ab initio molecular dynamics code will elucidate answers to key questions regarding materials decomposition. Our simulations of shock-compressed water show that the ionic conductivity near the Neptune isentrope is because of an ensemble of short-lived ionic species. We are currently starting simulations of shocked astrochemical mixtures of prebiotic molecules that are found in comets and other celestial bodies.

Our studies represent the first use of a state-of-the-art quantum mechanical simulations code to study shocked, highly reactive materials with an atomic number greater than one. Our simulations allow us to determine the exact kinetic and thermodynamic nature of chemical reactivity at high temperature and pressure, such as the dynamic ionization of water within large planets. We continue to gain important insight into how simple molecular systems are influenced by rapid compression and how this pertains to geochemical and planetary processes. We have already published in a peer-reviewed journal and have several more publications in preparation.

Mission Relevance

Knowledge of highly reactive materials decomposition under conditions of high temperature and pressure is essential for a complete understanding of liquids in the interiors of Earth and large planets, and is central to the Laboratory's stockpile stewardship mission. The quantum simulations algorithms and codes developed in this project will be applicable to several national security missions. In addition, this high-profile scientific research supports LLNL's mission in fundamental science and technology and will attract new talent to the Laboratory.

FY07 Accomplishments and Results

In FY07 we (1) completed our simulations of the shock Hugoniot of water—results that provide a simple chemical picture of the large ionic conductivities within Neptune and Uranus; (2) computed x-ray scattering intensities for water at these extreme conditions, which allows for direct comparison to experiment; and (3) developed a metadynamics simulation capability for use in shock ensembles, which will allow us to calculate the melting lines for ice VII and superionic water. These results are vital for equation-of-state models for planetary science and energetic materials. We have now established our ability to study any number of shocked materials.

Proposed Work for FY08

In FY08 we will conduct MSSM—metadynamics simulations of shocked interstellar ices, initially focusing on fundamental chemistry, such as the lifetimes and concentrations of peptide-bonded species formed under such shock conditions. We will then alter initial shock conditions and use metadynamics to investigate changes in the kinetics and free-energy surface for peptide-bond formation. We will also investigate compositional effects, such as varying the ice composition and placing the interstellar ice in reducing and oxidizing conditions.

Publications

Goldman, N., and L. E. Fried, 2007. “X-ray scattering intensities of water at extreme pressure and temperature.” *J. Chem. Phys.* **126**. UCRL-JRNL-227048.

Goldman, N., et al. , 2007. *Ab initio molecular dynamics simulations of shock water*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kona, HI, June 24–29, 2007. UCRL-ABS-225649.

Goldman, N., et al. , 2007. *Ab initio molecular dynamics simulations of shock water*. 223rd Am. Chem. Soc. Natl. Mtg., Chicago, IL, Mar. 25–29, 2007. UCRL-ABS-225649.

Goldman, N., et al., in press. “Ab initio molecular dynamics simulations of water under static and shock compressed conditions.” *Proc. 15th APS Topical Conf. Shock Compression of Condensed Matter*. UCRL-PROC-233148.

Goldman, N., et al., 2007. *Transforming graphite to diamond: an ab initio molecular dynamics study of graphite under shock compression*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kona, HI, June 24–29, 2007. UCRL-ABS-228457.

Biologically Driven Fabrication of Complex Nanostructures with Nanoscale Chemical Templates

Sung-Wook Chung 06-LW-051

Abstract

The ability to deposit biomolecules such as RNA onto specific sites or into ordered arrays would facilitate determination of protein structure, a major challenge in proteomics. Perhaps most intriguing is the idea of combining biomolecules with inorganic nanostructures to form hierarchical multicomponent structures. This project proposes to use nanotemplated assembly of biomolecules to fabricate biological–inorganic nanostructures, and understand the physical forces driving nanostructure formation. Scanned-probe nanolithography will be used to create patterned structures as templates for organizing biomolecules. Initial experiments will pattern RNA on metallic surfaces. Then, we will study the RNA-mediated formation of metallic nanoparticles on these patterned surfaces using various types of microscopy.

Multicomponent biological–inorganic hybrid nanostructures are expected to result from this project, along with a better understanding of the physical forces at the bioinorganic interface that influence nanostructure formation. Leveraging the unique materials-synthesis properties of biomolecules such as these RNA catalysts with controlled two-dimensional (2D) and 3D organization of biomolecules on pre-patterned nanoscale templates is expected to lead to the formation of entirely new inorganic nanostructures of unique size, shape, and morphology. These unique characteristics are likely to result in novel properties that conventional, solution-based synthetic approaches cannot produce.

Mission Relevance

As a means for fabricating arrays of multicomponent hybrid nanomaterials, the biotemplated synthesis technology developed in this project has potential applications in fabricating novel thin films and materials for stockpile stewardship experiments on future fusion-class lasers. The combination of small size, high density, and biological integration could also enable high-sensitivity, high-selectivity pathogen detection for counterproliferation and counterterrorism missions. Finally, this technology has the potential for use in repairing defects or broken metallic electrodes in the circuits of custom electronic devices used in military and space applications.

FY07 Accomplishments and Results

In FY07 we (1) developed a synthetic strategy for preparation and selection of RNA with a variety of bioconjugable linker chemistries; (2) demonstrated the ability to fabricate nanotemplate patterns for RNA with scanned-probe nanolithography, a linker library, and surface chemistry; (3) grew palladium nanostructures on an RNA nanotemplate patterned with scanned-probe nanolithography; (4) demonstrated the ability to investigate time-dependent kinetics of palladium nanostructure formation in solution using ex situ transmission electron microscopy; (5) finished investigating the growth process of palladium structures on RNA templates using in situ atomic force microscopy; and (6) measured the interaction between RNA and palladium formed on RNA templates using chemical force microscopy.

Publications

Chung, S.-W., et al., 2007. *Growth of metal nanostructures on templates of RNA-aptamer catalysts formed by scanned probe nanolithography*. Materials Research Society Spring Mtg., San Francisco, CA, Apr. 9–13, 2007. UCRL-PRES-229795.

Chung, S.-W., et al., 2007. *Growth of nanostructures on templates of RNA-aptamer catalysts formed by scanned probe nanolithography*. American Chemical Society Spring Mtg., Chicago, IL, Mar. 25–29, 2007. UCRL-ABS-233530.

Novel Approach to Investigate the Mechanism of *Yersinia Pestis* Pathogenicity in Real Time and at the Single-Cell Level

Julio A. Camarero 07-ERD-050

Abstract

The main objective of this proposal is to study the pathogenicity of *Yersinia pestis*—the agent that causes plague—in real time and at the single-cell level. A key to our approach is development of new molecular tools based on protein splicing, which will allow reconstitution and site-specific labeling of cytotoxic proteins inside the host cell with total temporal and spatial control. This approach also will be used for the simultaneous, multicolor site-specific labeling of *Y. pestis* cytotoxic effectors and their target proteins in vivo. This will allow, for the first time, study of multiple interactions in vivo between the *Y. pestis* cytotoxic proteins and their host target proteins to better understand the virulence mechanisms of *Yersinia* during infection of its natural hosts.

This truly multidisciplinary project will allow the development of a new set of biomolecular tools for study of proteins and their interactions in vivo with minimal perturbation to the protein function, as well as determine primary factors of total temporal and spatial control. This set of tools will be used for the first time to elucidate the action mechanism of YopM, a key effector protein for *Yersinia* pathogenicity, within the host cell. We will address key questions regarding the pathogenicity of YopM. Specifically, is the tetrameric structure of YopM required for nuclear delivery and biological activity? What is the temporal mechanism for RSK1 and PRK2 *Yersinia*-outer-protein-mediated activation? What other proteins interact with YopM in the cytoplasm?

Mission Relevance

Successful completion of this project will provide a completely new way to study pathogenicity in living cells with temporal and spatial control. It will enable real-time, single-molecule level studies on host–pathogen interaction biology. Moreover, the project is extremely well aligned with the Laboratory’s missions in homeland security and in biosciences to improve human health and with the National Institute of Allergy and Infectious Diseases strategic plan for biodefense research.

FY07 Accomplishments and Results

In FY07, we (1) replicated and expressed a gyrase intein YopM (*Y. pestis* strain CO92) fusion protein, which we then began evaluating in vitro for biological activity (tetramerization ability) using a combination of gel permeation chromatography and fluorescence; (2) synthesized DnaE and DnaB C-inteins by solid-phase peptide synthesis; (3) replicated several model proteins (enhanced green fluorescent and maltose binding proteins) to the DnaE and DnaB N-inteins and began testing the proteins for orthogonal labeling in vitro; and (4) tested the translocation potential of DnaE and DnaB C-inteins in human HeLa and U2OS cells.

Proposed Work for FY08

In FY08 we expect to (1) express and activate YopM in vitro and in vivo using photomodulated protein trans-splicing, (2) perform in vivo studies using first-model human cells and later-macrophage cell lines, and (3) clone RSK1 and PRK2 enzymes fused to the DnaE and DnaB C-intein polypeptides to demonstrate orthogonal labeling with fluorescent dyes—this will be accomplished first in vitro and later in vivo using the same human cells as before.

Publications

Camarero, J. A., et al., 2007. "Biosynthesis of a fully functional cyclotide inside living bacterial cells." *ChemBioChem* **8**, 1363. UCRL-JRNL-230072.

Camarero, J. A., 2007. "Protein microarrays—without a trace." *BIOforum Eur.* **5**, 30. UCRL-JRNL-230183.

Camarero, J. A., in press. "Recent developments in the site-specific immobilization of proteins onto solid supports." *Biopolymers*. UCRL-JRNL-228513.

Kimura, R. H., E. R. Steenblock, and J. A. Camarero, 2007. "Development of a cell-based fluorescence resonance energy transfer reporter for *Bacillus anthracis* lethal factor protease." *Anal. Biochem.* **369**, 60. UCRL-JRNL-230670.

Woo, Y. H., A. R. Mitchell, and J. A. Camarero, 2007. "The use of aryl hydrazide linkers for the solid phase synthesis of chemically modified peptides." *Int. J. Pept. Res. Therapeut.* **13**, 181. UCRL-PRES-226746.

Woo, Y. H., et al., 2007. *FRET monitored site-specific labeling of protein with small molecules in live cells via protein trans-splicing: method and application*. 21st Ann. Symp. Protein Society, Proteins: From Birth to Death, Boston, MA, July 21–25, 2007. UCRL-POST-232856.

The Structure and Transport of Water and Hydrated Ions within Hydrophobic, Nanoscale Channels

Jason K. Holt 07-LW-056

Abstract

Although studies have recently emerged on fluid and ion confinement in nanoscale materials, there is no consensus regarding the length scale below which nanoscale effects become important, nor is the interplay between ordering and transport clear. Because of their size and hydrophobic nature, carbon nanotubes (CNTs) are nanoscale materials that can serve as analogues for biological channels that regulate water and ion conduction in cells. We plan to study the structure and transport of water and hydrated ions in CNTs using nuclear magnetic resonance (NMR), x-ray absorption spectroscopy (XAS), and molecular simulations. Our goal is to define length scales and probe the consequences for molecular transport. Answering

these and other questions are of fundamental scientific interest and also will impact the future generation of filters and other devices that operate on this scale.

We intend to probe the dynamics of water in the unique CNT environment with the use of NMR and determine how ion structure is altered by their interaction with CNTs. The interest in these phenomena stems from our recent observations of enhanced flow in CNTs. We will determine if the enhanced flow can be attributed to molecular ordering within the channel. This research will enhance our understanding of transport in CNT systems and also will aid in development of high-throughput membranes for applications such as desalination.

Mission Relevance

This project brings cutting-edge science to the Laboratory at the intersection of chemistry, materials science, and biology. The project also will foster collaboration with the Stanford Synchrotron Radiation Laboratory, another DOE lab. The research will generate publications in journals such as *Science*, increasing Laboratory visibility. Finally, the technologies developed will enable new research opportunities of value to biosecurity in support of the Laboratory's homeland security mission, and water purification in support of environmental management and remediation.

FY07 Accomplishments and Results

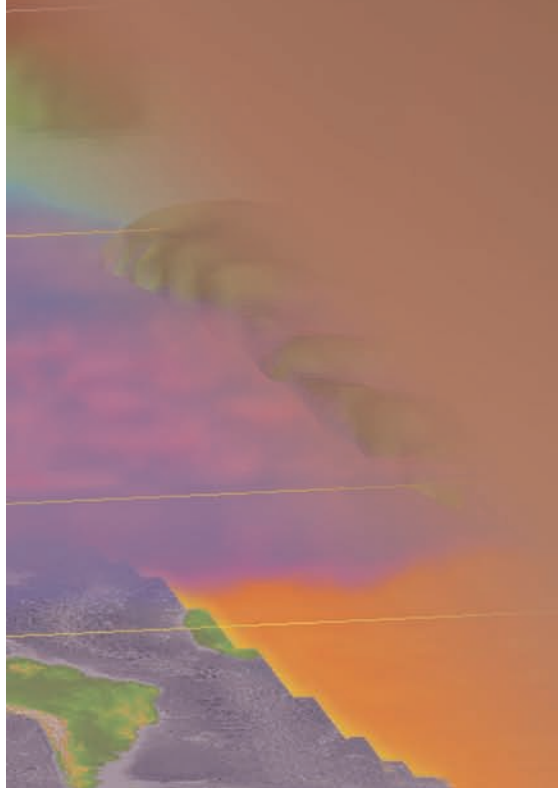
In FY07, we (1) identified a confined water signature in CNTs down to 1 nm in diameter through water-adsorption NMR measurements; (2) carried out XAS measurements on rubidium bromide-treated CNTs and modeled the resulting data, showing a unique structure not previously reported in the literature; (3) began studies to determine growth conditions needed for CNT growth in the 1-nm-to-subnanometer range; and (4) began molecular simulations of bromide solutions in CNTs to compare with experimental XAS measurements.

Proposed Work for FY08

In FY08, we propose to (1) carry out variable temperature proton/deuterium NMR measurements of water in CNTs from 1 to 15 nm in diameter, and probe the dynamics of the different water environments in the CNTs; (2) perform pulsed-field gradient NMR measurements to determine water self-diffusivity in CNTs, which should be a strongly size-dependent parameter; (3) continue simulations of ion structure in CNTs and calculate NMR and XAS spectra, which will aid in interpretation of experimental results; (4) continue XAS measurements on CNTs treated with other ions to understand how they are structured at the CNT interface; and (5) publish the results of the NMR, XAS, and simulation studies.

Laboratory Directed Research and Development

Earth and Space Sciences



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, and are an unsolved, multibillion-dollar problem in remediating groundwater contamination. We will investigate a process for chemically shrinking and cracking the sediments, and then flushing out the trapped contaminants using benign chemical cracking agents and co-solvents such as ethanol. We will perform laboratory experiments on synthetic clays to determine constitutive properties, crack propagation velocities, crack spacings and patterns, and methods for cost-effectively delivering and recovering the injected chemicals. Using these results, we will develop predictive models and computer codes for designing field remediation.

This project will develop models to reliably predict the performance of this novel chemical clay-cracking process and define its design limits. We will build on results of a previous project that demonstrated 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. The ultimate result will be development of a technology for remediating contaminants entrapped in fine-grained sediments. This is a common multibillion-dollar problem for which currently there is no other technically feasible, cost-effective solution.

Mission Relevance

This research directly supports the Laboratory's environmental management mission by developing an effective method of remediating contaminated fine-grained sediments. It addresses a serious threat to national water resources and furthers the remediation of previously intractable contaminant sources at several DOE sites. Enhanced understanding of chemically induced cracking enables reliable design of waste-containment facilities, prediction of the performance of clay seals in nuclear waste repositories, and evaluation of the integrity of cap-rock seals in geologic sequestration of carbon dioxide. Most importantly, this research breaks new ground in modeling and measuring generic coupled transport and chemo-mechanical processes leading to material failure by shrinkage-induced fracturing.

FY07 Accomplishments and Results

In FY07, we (1) continued crack network experiments, chemically inducing crack networks in clay in a large 50-cm-diameter flow cell; (2) photographed crack spacings and widths and verified that they agreed with predictions based on material properties; (3) developed approximate discrete-crack and dual-continuum models to design and analyze ethanol-water flow, clay-cracking, and contaminant transport in a full remedial-process experiment performed in a second large flow cell in which colored dyes were emplaced in six clay layers to mimic entrapped contaminants and enable the penetration depths of cracks to be estimated from dye release; and (4) devised Raman, absorbance, and fluorescence methods to measure ethanol and dye concentrations. During the course of this project, we

measured constitutive properties and crack velocities, spacings, and widths and developed a full cracking, flow, and transport experiment. We also developed models and approximate computer codes for experiment design and analysis, and verified the concept and its implementation. In the future, we hope to demonstrate this technology in the field.

Developing a Reactive Chemistry Capability for the Operational Model of the National Atmospheric Release Advisory Center

Philip J. Cameron-Smith 05-ERD-050

Abstract

Atmospheric chemistry can significantly alter the impact of many chemical releases, such as chlorine and nerve agents. To address the current imbalance between the chemistry and dispersion capabilities of LODI, the operational response model used by Lawrence Livermore's National Atmospheric Release Advisory Center (NARAC), we are adding a full reactive chemistry and aerosol capability to account for an arbitrary network of chemical reactions and evaporation and condensation of aerosols. Improvements will include the ability to read time- and space-dependent ambient concentrations of relevant species from IMPACT, the Laboratory's global atmospheric chemistry and aerosol model.

The addition of a full reactive chemistry capability to LODI, as well as interfacing with the IMPACT global atmospheric chemistry code, will create a unique, emergency-response capability that will greatly enhance our ability to respond to terrorist attacks and industrial accidents that involve reactive chemistry, including many chemical agents and toxic industrial chemicals. This work also will facilitate development of detection and monitoring plans for factories and laboratories, including clandestine facilities. The resulting model also will have a dual use in local and regional air-quality studies.

Mission Relevance

By improving emergency response capabilities, 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. Chemical fate also has an important role in understanding proliferation signatures as well as pre- and post-strike consequence evaluation of military targets.

FY07 Accomplishments and Results

In FY07, we implemented in LODI (1) our semi-Lagrangian advection scheme, (2) our aerosol dynamics capability, (3) a GEAR chemistry solver, and (4) our chemical mechanisms for chlorine, oleum, ozone, and nerve agents. As an incidental benefit, we improved the multiprocessor scaling and memory usage of LODI to expedite the simulations for emergency response. We provided the NARAC development team with extensive training on the principles and capabilities we developed, and helped them to implement our changes into the main LODI code base. Overall, this project has given NARAC a general capability for scenarios requiring reactive chemistry, aerosol production and evolution, and chemical heat liberation for an arbitrary number of plumes and the background atmosphere.

The Physics of Recombining Plasmas in Celestial Sources

Gregory V. Brown 06-ERD-010

Abstract

Radiative recombination and charge-exchange x-ray production by recombination of highly charged ions with free or bound electrons plays an important role in essentially all laboratory plasmas, and is believed to be the dominant x-ray line formation process in many celestial sources. Interpretation of the x-ray spectral signatures from charge exchange in complex sources, however, has been challenging because little targeted laboratory data are available. By taking advantage of instrumentation unique to LLNL, including the super electron beam ion trap, gas- and laser- injection systems, and a high-resolution microcalorimeter array from the National Aeronautics and Space Administration Goddard Space Flight Center, we will determine the x-ray spectral signature of charge-exchange recombination under controlled laboratory conditions.

We will provide the first-ever, high-resolution x-ray spectra produced by charge-exchange recombination between neutral atoms and bare and hydrogenic ions. In addition, we are implementing the new Photon Clean Method for analyzing spectra, which uses a novel Monte Carlo approach to spectral fitting that raises the probability of discovering weak x-ray production mechanisms that may contribute line emission to a spectrum. Our results will be used to benchmark theoretical models and largely improve the ability to accurately diagnose highly complex laboratory and celestial plasmas, such as those from tokamaks, the aurora of Jupiter, supernova remnants, cometary atmospheres, and the Earth's magnetosheath.

Mission Relevance

Because the thermonuclear phenomena found in stars are the same as those that occur in nuclear explosions and in fusion reactors, this project will benefit numerical simulation and contribute to fundamental experimental research that supports the Laboratory's national and energy security missions. In addition, it extends the Laboratory's core competency in atomic physics and will attract talented scientists to the Laboratory.

FY07 Accomplishments and Results

In FY07 we have continued our measurements of high-resolution x-ray spectra produced by charge exchange between bare and hydrogenic iron and argon target ions interacting with neutral molecular hydrogen, helium, and nitrogen projectiles. Our measurements show a distinct shift in the principal quantum number of maximum capture between molecular hydrogen and helium donors. The shift is a result of the fact that the value of the maximum quantum number decreases as the donor's ionization potential increases. This trend is predicted by theory—however, theory predicts significantly lower quantum numbers. We also measured charge-exchange spectra as a function of collision energy, and investigated the applicability of the Photon Clean Method to problems facing the Department of Homeland Security, namely the detection of man-made radioisotopes.

Proposed Work for FY08

During FY08, we will measure the charge-exchange spectrum from bare and hydrogenic ions reacting with atomic hydrogen. These results will provide high-visibility benchmarks for atomic theory and determine if the discrepancy between experiment and theory is a result of the fact that earlier measurements involved complex gases instead of atomic hydrogen as donors. Our measurements will include ions relevant to the interpretation of the x-ray spectra from a plethora of celestial sources, notably the Earth's magnetosheath and the atmosphere of Jupiter, both recently measured by the Suzaku x-ray observatory, and also the Bright Eastern Knot of the Puppis A supernova remnant to be measured by the Massachusetts Institute of Technology high-resolution spectrometer on the Micro-X suborbital rocket experiment.

Publications

Beck, B., et al., 2007. "Energy splitting of the ground-state doublet in the nucleus ^{229}Th ." *Phys. Rev. Lett.* **98**, 142501. UCRL-JRNL-223439-DRAFT.

Bhardwaj, A., et al., 2007. "X-rays from solar system bodies." *Planet. Space Sci.* **55**, 1135. UCRL-JRNL-218823.

Otranto, S., et al., 2007. "Cometary x-rays: line emission cross sections for multiply-charged solar wind ion charge exchange." *J. Phys. B Atom. Mol. Phys.* **40**, 1755. UCRL-JRNL-227014.

Otranto, S., et al., 2007. "X-ray emission cross sections following charge exchange by multiply-charged ions of astrophysical interest." *Phys. Rev. A* **73**, 022723. UCRL-JRNL-215642.

Urban Atmospheric Turbulence: Improved Turbulence Closure Models through Observations and Simulations

Julie K. Lundquist 06-ERD-026

Abstract

We propose to test and integrate advanced turbulence models and numerical methods for urban simulations into a numerical model that is widely used for weather prediction and transport as well as dispersion prediction. This project is motivated by the inadequacy of current turbulence models (compared to observations) and the demand for complex time- and space-varying boundary conditions for urban-scale models. Improved models are essential for national security purposes in the case of accidental or intentional urban atmospheric release of hazardous materials. Project success will be determined by comparing simulations to a refined field dataset from the Joint Urban 2003 field study.

New, more accurate turbulence models for urban areas developed in this project will provide improved plume predictions for national security applications. These improved numerical methods will allow the seamless integration of urban-scale simulations with appropriate coarser-scale simulations to provide the best inputs to urban scale models. This work will

ensure that new models for emergency response, sensor siting, and forensic studies in urban areas are of the greatest fidelity possible for protection of urban populations. The project also provides improved access to renewable energy—our improved simulation capability will allow more accurate forecasts of available wind resources in complex terrain and will, therefore, promote more efficient siting of turbines.

Mission Relevance

Our project supports LLNL's national and homeland security missions by improving capabilities to provide emergency response for atmospheric releases of hazardous materials. Our project also supports LLNL's mission to provide the nation with abundant, reliable energy as well as a clean environment by providing reliable and accurate predictions of wind-energy resources.

FY07 Accomplishments and Results

In FY07, we (1) implemented a suite of improved turbulence models for the Weather Research and Forecasting (WRF) code, validated the models with canonical case studies, and prepared to execute simulations on 100-m scales that will document improved predictions of winds and turbulence fields; (2) developed a new approach to implementing the immersed boundary method (IMB) for urban and complex geometries and began rigorously testing and documenting the approach before moving on to urban-scale simulations; (3) contacted members of the WRF community to include our improved version of the code in the official distribution; and (4) documented our analysis of turbulence kinetic-energy budgets of the Joint Urban 2003 experiment for publication in a peer-reviewed journal.

Proposed Work for FY08

In FY08, we will (1) complete implementation of the IMB, (2) carry out extended nested simulations with the dynamic reconstruction model (DRM) to determine appropriate scale transitions between DRM and traditional turbulence parameterizations, (3) conduct extended simulations in regions other than the Joint Urban 2003 domain, (4) submit DRM turbulence parameterization to the atmospheric science community via a new WRF distribution, (5) simulate flow and dispersion using DRM turbulence closure models and IMB at building scales (tens of meters) for canonical cases, and (6) publish a paper in a peer-reviewed journal quantifying the improvement in dispersion, wind field, and turbulence calculations afforded by the DRM and IMB in nested and building-scale simulations.

Publications

Lundquist, J. K., and J. D. Mirocha, in press. "Interaction of nocturnal low-level jets with urban geometries as seen in Joint Urban 2003 data." *J. Appl. Meteorol. Climatol.* UCRL-JRNL-224264.

Lundquist, J. K., and S. T. Chan, 2007. "Consequences of urban stability conditions for computational fluid dynamics simulations of urban dispersion." *J. Appl. Meteorol. Climatol.* **46**, 1080. UCRL-JRNL-217489.

Lundquist, J. K., et al., 2007. *An improved WRF for urban-scale and complex-terrain applications*. American Meteorological Society 7th Symp. Urban Environment, San Diego, CA, Sept. 10–13, 2007. UCRL-CONF-234277.

Lundquist, K. A., et al., 2007. *Improving atmospheric dispersion simulations with an immersed boundary method to represent urban terrain*. American Meteorological Society 7th Symp. Urban Environment, San Diego, CA, Sept. 10–13, 2007. UCRL-CONF-234276.

Mirocha, J. D., et al., 2007. *Improved subfilter turbulence modeling for large eddy simulation using WRF*. American Meteorological Society 7th Symp. Urban Environment, San Diego, CA, Sept. 10–13, 2007. UCRL-CONF-234278.

Atmospheric Carbon Dioxide-14 Constraints on and Modeling of Net Carbon Fluxes

Thomas P. Guilderson 06-ERD-031

Abstract

Carbon dioxide (CO₂) is the most important anthropogenic greenhouse gas influencing global climate. Sources and sinks impart their signature on the distribution, concentration, and isotopic composition of CO₂, while spatial and temporal variability provide information on net surface fluxes. Observations of carbon and oxygen isotope tracers and their rate of change in the atmosphere can be used to constrain global and regional contributions of different carbon sources and sinks, because each bears a different isotope or elemental ratio signature. We propose to measure ¹⁴CO₂ samples from a suite of clean-air locations and use these results in inversion estimates to constrain unidirectional carbon fluxes.

This project is expected to produce the best possible estimates of surface carbon sources and sinks on a global scale using atmospheric observations of CO₂ partial pressure, carbon-13, carbon-14, and oxygen/nitrogen ratios; atmospheric transport based on the range of Atmospheric Tracer Transport Model Intercomparison (TransCom) results; and the best possible representation of isotopic composition of exchangeable carbon pools and associated isotopic fractionation factors.

Mission Relevance

By investigating size and variability of the dynamic reservoirs and fluxes of carbon within the Earth system, this research supports Laboratory missions in energy security. The synergy between field data and computational models offered by this project will contribute to DOE's missions in energy security and environmental management (specifically, carbon management).

FY07 Accomplishments and Results

We (1) participated in the TransCom project to validate CO₂ transport in our global model (IMPACT), which also raised our visibility in the community; (2) used IMPACT to generate

global basis functions for carbon isotopes carbon-12, carbon-13, and carbon-14 from 22 regions and the stratosphere, which we used to test the retrieval of global fossil fuel emissions from synthetic data for the seven sites where we are measuring carbon-14; (3) implemented an algorithm for optimal measurement network design; (4) analyzed Scripps Institution of Oceanography (SIO) clean-air samples and samples collected under the Mid-Continent Intensive Experiment; (5) continued to collect SIO samples; and (6) presented our results at conferences of the National Oceanic and Atmospheric Administration and North American Carbon Program.

Proposed Work for FY08

In FY08, we will (1) measure carbon-14 in SIO pole-to-pole clean-air samples, in samples collected at the Oklahoma site of the Atmospheric Radiation Measurement Program and in stratospheric samples; (2) continue our modeling effort, leveraging collaborations with Purdue and the University of Colorado (which are responsible for the TransCom project) and focusing on estimates of an optimal flask sample network that incorporates carbon-13, carbon-14, and CO₂ partial pressure on sensitivity to an independent fossil-fuel emission estimate and on terrestrial carbon fluxes; and (3) verify the stratospheric input function through comparisons with stratospheric and upper troposphere ¹⁴CO₂.

Publications

Graven, H. D., et al., 2007. "Identifying CO₂ sources with vertical profiles of delta ¹⁴CO₂ above Colorado." *Eos Trans. AGU* **87**, B41G-08. UCRL-JRNL-225392.

Graven, H. D., T. P. Guilderson, and R. F. Keeling, 2007. *Delta ¹⁴CO₂ from the Scripps CO₂ program*. 14th Intl. CO₂ Experts Mtg., Helsinki, Finland, Sept. 10–13, 2007. UCRL-CONF-234905.

Graven, H. D., T. P. Guilderson, and R. F. Keeling, 2007. "Methods for high precision ¹⁴C AMS measurement of atmospheric CO₂ at LLNL." *Radiocarbon* **49**(2), 349. UCRL-JRNL-225389.

Guilderson, T. P., H. D. Graven, and R. F. Keeling, 2007. *¹⁴CO₂ measurements in clean air samples: a record of the carbon cycle and anthropogenic influences*. NOAA Carbon Cycle Mtg., Silver Springs, MD, Sept. 10–11, 2007. UCRL-CONF-234691.

Regional Climate

David C. Bader 06-ERD-066

Abstract

Understanding regional climate depends on gaining knowledge about climate variability at similar scales. The proposed research will contribute to this understanding by developing an integrated, multimodel capability for regional climate change impact and adaptation studies and by using the model to examine climate change impacts on regions as small as watersheds and cities. The emphasis of this project is to develop and establish within Lawrence Livermore a scientifically strong and peer-accepted regional climate modeling

capability through studies focusing on the effects of climate change on California. Our first focus area is the effect of climate change on water resources and our second is the relationship between climate change and air quality.

This project will use observations of atmospheric variables to examine the ability of climate models to simulate this variability. We will leverage LLNL's climate-modeling expertise to further analysis of California climate change. The modeling capability developed in this project will enable the Laboratory to design and execute simulations of important climate change impacts such as the frequency and intensity of flooding in regions as small as individual cities or watersheds. In addition, our efforts complement global modeling used for forming national and international energy, air quality, and water management policies.

Mission Relevance

In support of LLNL's missions in energy and water security and environmental management, we will develop an integrated, multimodel capability for regional climate change analysis and for impact and adaptation studies. This suite of simulation tools will provide a new capability for managing regional water resources in California that can be applied to other regions in the U.S.

FY07 Accomplishments and Results

In FY07, we compared the effects of resolution on climate simulation of surface temperature and precipitation using results from three models. The first was a moderate-resolution (~150-km horizontal grid spacing) global coupled atmosphere–ocean model. The second was a global atmospheric model at double resolution forced by the ocean temperatures from the coupled run. The third was a regional model with 12-km grid spacing forced at the boundaries by the global coupled model fields. We also replaced the Coupled Ocean–Atmosphere Mesoscale Prediction System (COAMPS) regional model with the community Weather Research and Forecasting (WRF) modeling system. Results show that precipitation, especially low-probability high-rainfall events, are particularly sensitive to model resolutions. Several short simulations of one-month duration were executed to select the best cloud microphysics parameterization option in the WRF model to correctly simulate wintertime precipitation that dominates California climate. We found that WRF is superior to COAMPS in its simulation of California precipitation, producing more realistic patterns of weather phenomena caused by physical geography and total precipitation amounts.

Proposed Work for FY08

In FY08, we will focus on two tasks. The first and primary effort will be to compare the results from the 40-year simulation of WRF with observations of precipitation in California with an emphasis on extreme precipitation months (probability less than 5%). Further, we will collaborate with researchers at Scripps Institution of Oceanography to compare and contrast their techniques to “downscale” global climate model runs statistically with our dynamically downscaled results. Once these analyses are complete and we are satisfied with the results, we expect to extend the length of the simulation to 500 years, to examine a more complete climatology of extreme rainfall events that lead to flooding in California.

Development of Integrated Microanalysis of Nanomaterials

John P. Bradley 06-ERI-001

Abstract

In recent years significant advances have been made in detection and imaging capabilities using electron microscopy and ion microprobe techniques. We propose to exploit LLNL's significant investments in state-of-the-art resources to elevate analytical interrogation of both natural and man-made nanomaterials to new levels. Samples of interest include comet grains recently returned to earth by the Stardust mission, as well as surrogate nuclear and bioforensics-related materials. Initial developments have focused on Stardust nanomaterials captured at hypervelocity speeds with low-density silica aerogels. The research is synergistic with multiple mission-relevant analytical needs.

Stardust samples are the first solid-matter materials to be returned to Earth since the Apollo missions of the 1970s. Determining the relationship between the star samples and other classes of meteoritic materials—such as meteorites, micrometeorites, and interplanetary dust particles from comets—is one of the key science goals of Stardust's capture of a comet sample from comet Wild 2, a known Kuiper Belt body. We expect to be able to draw a definitive conclusion about this relationship. Our studies indicate the Wild 2 sample resembles meteorites from the asteroid belt and does not resemble cometary dust particles collected in the stratosphere.

Mission Relevance

The proposed work will develop and investigate new analytical techniques for nanoscale materials characterization that are directly applicable to the Laboratory's missions in stockpile stewardship and homeland security. The project also supports the Laboratory's mission in breakthroughs in fundamental science and applied technology, specifically in the area of astrophysics and space science, which includes the exploration of Kuiper Belt objects such as Comet Wild 2.

FY07 Accomplishments and Results

We have (1) completed our comparison of Wild 2 dust with other classes of meteoritic materials, (2) prepared surrogate nuclear forensics materials using small-particle mounting techniques developed for Stardust and analyzed them for trace elements using synchrotron x-ray fluorescence at the Stanford Linear Accelerator Center, (3) developed a procedure to stabilize transmission electron microscopy specimens so they can be cycled repeatedly between scanning and transmission electron microscopy and nanometer-scale secondary-ion mass spectroscopy instruments, and (4) completed installation of the 300-kV Titan super-scanning transmission electron microscope at LLNL.

Proposed Work for FY08

In FY08, we aim to (1) further characterize the mineralogical, chemical, and isotopic properties of Wild 2 dust; (2) assess the plethora of new avenues of planetary science

investigations opened up by the Wild 2 samples; (3) continue to develop and refine manipulations and analysis procedures for nanogram-mass samples; and (4) begin to use the newly commissioned, super-scanning transmission electron microscope for other mission-relevant applications.

Publications

Bringa, E. M., et al., 2007. "Energetic processing of interstellar silicate grains by cosmic rays." *Astrophys. J.* **662**, 372. UCRL-JRNL-229546.

Ishii, H. A., et al., in press. "Refining the quantitative elemental composition of Comet Wild 2 dust in aerogel." *Meteorit. Planet. Sci.* UCRL-ABS-227184.

Kearsley, A. T., et al., 2007. "Analytical scanning and transmission electron microscopy of laboratory impacts on Stardust aluminum foils: interpreting impact crater morphology and the composition of impact residues." *Meteorit. Planet. Sci.* **42**, 191. UCRL-JRNL-225844.

The Chemistry of Core Formation

Frederick J. Ryerson 06-ERI-002

Abstract

Core formation is the major chemical differentiation event for terrestrial planets. Generation of the Earth's magnetic field is related to core formation, and is important in establishing planetary habitability. The depth and temperature of core segregation controls its chemistry, and high-pressure experiments can be used to simulate this process. We are developing methods that link the diamond anvil cell with analytical systems such as a secondary-ion mass spectrometer, a focused ion-beam system, and a transmission electron microscope to determine the partitioning of elements such as sulfur, silicon, oxygen, carbon, hydrogen, vanadium, tungsten, molybdenum, ruthenium, and lead at extreme conditions relevant to the Earth's lower mantle.

Results of this research will aid in explaining the low density of the Earth's core relative to pure iron and the overabundance of siderophile elements (i.e., chemical elements that partition strongly into a metal-rich phase) in the silicate mantle. This explanation will constrain the potential range of core-forming processes for Earth, which ultimately constrains generation of the Earth's magnetic field. Constraining composition of the core may provide clues to the absence of magnetic fields on other terrestrial planets and the influence of magnetic fields on planetary habitability.

Mission Relevance

The extreme conditions of temperature and pressure present in the Earth's core overlap those found in the stars and in exploding nuclear weapons. The microanalytical methods, materials characterization techniques, microanalysis, and high-pressure experimentation that will be developed in this project to understand the Earth's core are applicable to LLNL's

stockpile stewardship mission and to forensics efforts for the nonproliferation mission. This project also contributes to the Laboratory's mission in basic science.

FY07 Accomplishments and Results

In FY07, we extended our previous low-pressure experiments to higher pressures (up to 25 GPa) to better constrain the effects of pressure on core formation. We also investigated the self-oxidation of silicate perovskite to pressures up to 130 GPa. Transmission electron microscope analysis of these samples indicates the presence of iron metal produced by a reaction in which divalent iron disproportionates to produce trivalent iron and iron metal. Dissolution of the ferric perovskite component in a terrestrial magma ocean provides a mechanism for progressive oxidation of the earth's mantle. Self-oxidation had been previously observed in aluminum-bearing perovskite at 25 GPa, but not for aluminum-free samples at core-mantle boundary pressures.

Proposed Work for FY08

In FY08 we will (1) extend our understanding of the pressure effect on siderophile element partitioning using both the multi-anvil press and the laser-heated diamond anvil cell; (2) combine these results with our temperature-dependence data to model iron segregation in a magma ocean; (3) develop analytical methods to determine the concentrations of light elements (oxygen, carbon, silicon, and sulfur) in previously synthesized materials with metal-silicate pairs; and (4) equilibrate perovskite/ferropericase and metal in the laser-heated diamond anvil cell to determine light-element concentrations imposed by solid-phase assemblages.

Publications

Auzende, A.-L., et al., in press. "Element partitioning between magnesium silicate perovskite and ferropericase: new insights into bulk lower-mantle geochemistry." *Earth Planet. Sci. Lett.* UCRL-JRNL-232988.

Badro, J., et al., in press. "Chemical imaging with nanoSIMS: a window into deep-earth geochemistry." *Earth Planet. Sci. Lett.* UCRL-JRNL-228823.

Feineman, M. D., et al., in press. "Mineral-fluid partitioning of trace elements in phlogopite and clinohumite at subduction zone pressures and temperatures: implications for fluid transport in the mantle wedge." *Chem. Geol.* UCRL-JRNL-227472.

Feineman, M. D., et al., 2007. "Zoisite-aqueous fluid trace element partitioning with implications for subduction zone fluid composition." *Chem. Geol.* **239**, 250. UCRL-JRNL-227468.

Putirka, K. D., et al., 2007. "Ambient and excess mantle temperatures, olivine thermometry, and active vs. passive upwelling." *Chem. Geol.* **241**, 177. UCRL-JRNL-217967.

Evidence for Stratospheric Downwelling Associated with High-Elevation Topography

Robert C. Finkel 06-ERI-005

Abstract

The continued presence of elevated chlorine-36 in Sierra Nevada streams is commonly interpreted as residual nuclear fallout, but this prolonged storage contradicts accepted hydrologic models, which indicate much less short-term groundwater storage. Here we test the hypothesis that the chlorine-36 source is stratospheric downwelling during high-intensity storms by measuring beryllium-7 and -10, sodium-22, and chlorine-36 in precipitation, lake, and soil samples. These nuclides are produced in abundance in the stratosphere and, except for chlorine-36, did not occur as nuclear fallout. This project will either substantially change hydrologic models or reveal an unrecognized pathway for stratosphere-troposphere exchange. Either result will have substantial scientific impact.

The test of the hypothesis is straightforward. If stratospheric downwelling occurs to the extent indicated by observed Sierran chlorine-36 levels, it should be detectable by elevated levels of beryllium-7 and -10, sodium-22, and chlorine-36 in storm precipitation. Samples will be collected from an established array of sampling locations. If elevated levels of these nuclides are not found, it would cast severe doubt on the hypothesis. In this case, Sierran hydrologic models will have to take into account high levels of long-term groundwater storage. If elevated levels of these nuclides are found in Sierran precipitation, it could only be from stratospheric input, both because nuclear fallout is no longer occurring and because beryllium-7 and -10 and sodium-22 are not produced by atmospheric nuclear tests.

Mission Relevance

This project supports the Laboratory's national security mission because of its relevance to atmospheric dispersal monitoring for nuclear and other releases. The work also supports the environmental-management mission areas of atmospheric and climate modeling and atmospheric protection from ozone-depleting and greenhouse gases. An ability to characterize certain types of stratosphere-troposphere exchange pathways using relatively inexpensive ground level measurements, as opposed to airborne tropopause measurements, would enhance LLNL's capabilities for testing regional atmospheric and climate models.

FY07 Accomplishments and Results

In FY07, we (1) continued installation of a precipitation- and soil-sampling network in collaboration with scientists at the U.S. Geological Survey, the National Park Service, and the University of California at Merced; (2) collected and began analyzing a suite of late-year soils for the stratospheric nuclides beryllium-7 and -10, and collected and analyzed snow samples; (3) constructed and deployed 15 unattended rain samplers and analyzed the initial samples; and (4) collected stream and lake samples throughout the year to monitor the relationship between precipitation input and elevated chlorine-36 levels.

Proposed Work for FY08

During FY08, we will (1) continue to monitor the network of precipitation samplers installed in FY07 in collaboration with colleagues from University of California at Merced and the U.S. Geological Survey; (2) continue to deploy precipitation samplers; (3) conduct spring and autumn monitoring of high-elevation lakes and streams; (4) determine chlorine-36 and beryllium-10 levels in rainwater, snow, lake, and stream samples; (5) measure sodium-22 and beryllium-7 in selected rainwater, snow, lake, and stream samples by gamma counting; (6) quantify chlorine-36 and beryllium-10 in soil samples, if this is shown to be appropriate; (7) begin compiling results and drawing conclusions about the validity of the downwelling hypothesis; and (8) present an update of results at the Yosemite National Park Hydroclimate Conference.

Broad-Area Search for Proliferant Infrastructure

David B. Harris 07-ERD-011

Abstract

This project will determine the feasibility of using long-term observations of ambient noise to detect, locate, and characterize industrial infrastructure. We will use very long-term (1- to 100-day) estimates of coherent noise structures across networks of sensors to detect and locate infrastructure and vehicular and rail traffic that radiate continuous noise-like signals. The ultimate application of this technology is to detect and locate proliferant infrastructure and traffic supporting possible sites of weapons of mass destruction (WMD) development.

We propose to develop (1) methods to detect and enumerate narrowband sources, (2) robust methods to locate narrowband sources, (3) methods to extract source time histories for characterization, and (4) calibration methods for matched field processing to track traffic. We expect to demonstrate the ability to detect and locate the larger components of infrastructure that operate continuously over long intervals and to provide a calibrated capability to track vehicle movement with sensor networks. Examination of long-term ambient noise covariance over sensor networks may select for such sources.

Mission Relevance

This project supports LLNL's nonproliferation mission by providing techniques to implement an indirect strategy of broad-area search for WMD development facilities through detection and location of supporting infrastructure (e.g., power generation). While not likely directly to identify WMD production processes, the techniques could assist in the development of target lists for more specific, shorter-range process-identification methods. These techniques also could help to characterize whether a specific process, once identified, is in operation at a given time and provide some information on power level from relatively far-field observations (typically tens of kilometers).

FY07 Accomplishments and Results

We successfully located and identified the source of an example persistent spectral signature in the San Francisco Bay Area using seismic network data. We tracked the 3.75-Hz spectral line, persistent for over 18 months, back to its source at the Harvey O. Banks Pumping Plant in Tracy. We successfully characterized the duty cycle (on–off operation) of the source and consulted with the plant operator to narrow it to a specific pump unit at the facility. In addition, we laid the groundwork for FY08 research on detecting, locating, and characterizing industrial sources in an urban setting and a remote location using a 12-element seismic array at Site 300 in the Tracy hills as well as two remote-area arrays. We also upgraded one remote-area array and collected critical ground truth information.

Proposed Work for FY08

In FY08, we will develop and test array beam-forming and matched-field processing methods to extend the ranges of detecting and characterizing industrial infrastructure. Our characterization methods will extract time histories for weak sources to infer periods of operation and power levels. We will also investigate deployment of a third array at our remote test location at a greater distance than the existing arrays to test the feasibility of beam forming and characterization from a greater standoff distance. We will complete work on methods to locate industrial sources using the arrays deployed at the remote site. Time permitting, we will also demonstrate a sophisticated seismic technique for vehicle tracking using the same array data.

Dense Gas Transport in Complex Environments

Branko Kosovic 07-ERD-020

Abstract

The potential for intentional or accidental atmospheric releases of hazardous, dense gas is increasing with the global risk of terrorism, renewed interest in liquefied natural gas (LNG) as fuel, and carbon dioxide sequestration. There currently exists no general way to accurately predict how dense gases will behave in an urban environment because models do not explicitly resolve the effects of buildings and gas density, which can produce misleading predictions of plume magnitude, location, or even direction. We propose to build an urban dispersion-modeling capability that will include both the appropriate dense gas physics and effects of buildings and complex terrain on atmospheric transport and dispersion, which we will validate using field data.

Dense gases suppress local atmospheric turbulence, whereas buildings and other obstacles generate turbulence. Furthermore, heat transfer, affected by phase change, may amplify or reduce turbulence. A new, validated denser-than-air simulation capability will establish a new capability for simulation of fate and transport of dense gas in urban and complex terrain, which will enable a more effective response to hazardous releases involving dense gases. There is considerable interest in a dense gas model for hazard assessment, planning, and emergency response. Potential applications include assessing risks associated with the transportation of toxic chemicals, LNG storage, and carbon dioxide sequestration.

Mission Relevance

This project supports the Laboratory's missions in homeland security, environmental management, and energy security, with respect to response to intentional or accidental releases of hazardous chemicals. This capability will enable a more effective response to both terrorist attacks and industrial accidents. For energy, there are specific applications to carbon dioxide sequestration, LNG spills and leaks, terminal safety, and transportation.

FY07 Accomplishments and Results

In FY07, we (1) added isothermal and nonisothermal density effects to the flow and transport equations in the urban dispersion model using a generalized anelastic formulation of the Navier–Stokes equations to permit large density variations in space and time, (2) evaluated potential dense gas scenarios to identify source requirements for our capability, (3) tested models for an instantaneous vapor release (such as a catastrophic tank breach) and a constant flux (such as an evaporating pool), and (4) began validating our model with data from the Burro-series LNG-spill field experiments.

Proposed Work for FY08

In FY08, we will develop physics-based models to better resolve fast-transient effects from building-induced turbulence and gravity-induced dense gas flows. Specifically, we will (1) refine turbulence modeling with large-eddy simulations including state-of-the-science, sub-grid-scale parameterization; (2) refine our phase-change model; (3) continue code validation with field data; and (4) continue assessing source-term needs.

Publications

Kosovic, B., et al., 2007. *Building-resolving simulations of dense gas dispersion*. 7th Symp. Urban Environment, San Diego, CA, Sept. 9–13, 2007. UCRL-PRES-234505.

Cosmochemical Forensics

Ian D. Hutcheon 07-ERI-005

Abstract

This project uses nuclear forensics and investigates correlated isotopic anomalies associated with short-lived radionuclides (SLNs) and with oxygen-16 in calcium–aluminum-rich inclusions (CAIs) to carry out four cutting-edge scientific tasks: (1) refine the initial abundance of aluminum-26 in the solar system, (2) determine whether aluminum-26 and oxygen-16 originated from a single supernova source, (3) use correlations between beryllium-10 and chlorine-36 to evaluate the extent of spallation reactions in the nascent solar nebula, and (4) compare the signatures of SLNs and oxygen-16 in cometary materials containing normal

CAIs. The results will constrain the time scales of formation during the first 3 million years of solar system history and elucidate astrophysical and cosmochemical models of the evolution of Sun-like stars.

We will (1) define the initial abundances of three SLNs—aluminum-26, chlorine-36, and manganese-53; (2) determine whether SLNs are correlated with oxygen-16; (3) constrain the relative contributions of stellar and local sources to the SLN inventory; (4) elucidate the evolution of nebular oxygen isotope reservoirs; and (5) determine how refractory inclusions in comet Wild 2 samples are related to CAIs in meteorites. This project addresses cutting-edge issues in cosmochemistry and contributes to a better understanding of the earliest history of the solar system and the sequence of events extending from condensation in a hot gaseous nebula to the formation of terrestrial planets. The project also serves as a mechanism to enhance and refine a suite of microanalytical techniques needed for national security applications.

Mission Relevance

The project develops and enhances advanced microanalytical capabilities in support of Lawrence Livermore's national security mission and enhances its mission in basic science. Nonproliferation and homeland security specifically benefit most from advancing forensic capabilities under this project. Furthermore, the techniques developed here for high-accuracy and high-sensitivity analyses on a nanometer scale have broad applicability throughout the laboratory.

FY07 Accomplishments and Results

We determined the first oxygen and nitrogen isotope data for Wild 2's "Inti," an oxygen-16-rich cometary CAI similar to meteoritic CAIs. Oxygen-16-rich compositions occur in rare stratospheric interplanetary dust particles, but the relationship between these particles and comets has never been thoroughly investigated. The isotopic compositions of carbon, nitrogen, and oxygen demonstrated that Wild 2 is an unequilibrated aggregate of material from disparate sources, suggesting contributions from several isotopically fractionated minor components that indicate chemical reactions at very low temperatures, such as those characteristic of molecular clouds or Edgeworth–Kuiper Belt environments. These data were inconsistent with the origin of Jupiter-family comets by accretion of cold materials at the edge of the solar nebula.

Proposed Work for FY08

We will explore the paradigm shift created by recent results from the Stardust mission. Specifically, we will characterize carbon, nitrogen, oxygen, and magnesium isotope abundances in Wild 2 samples and correlate these abundances with transmission and scanning electron microscopy and mineralogical studies to determine whether comets are composed of unprocessed debris left over from the earliest epoch of solar system history or contain predominantly recycled material, similar to asteroids. We will also determine the abundances of the short-lived radionuclides chlorine-36, aluminum-26, and manganese-53 in primitive CAIs. These abundances are key to evaluating the hypothesis that chlorine-36 was

produced by energetic particle irradiation within the solar nebula, whereas aluminum-26 was produced by nucleosynthesis in asymptotic giant branch stars or supernovae.

Publications

McKeegan, K. D., et al., 2007. "Isotopic compositions of cometary matter returned by Stardust." *Science* **314**, 1724. UCRL-JRNL-225917.

Simon, S. B., et al., 2007. "Formation of spinel-, hibonite-rich inclusions found in CM2 carbonaceous chondrites." *Am. Mineral.* **91**, 1675. UCRL-JRNL-216859.

Yin, Q. Z., et al., 2007. "Toward consistent chronology in the early solar system: high-resolution ^{53}Mn – ^{53}Cr chronometry for chondrules." *Astrophys. J.* **662**, L43. UCRL-JRNL-230698.

Laboratory Directed Research and Development

Energy Supply and Use





Separation of Carbon Dioxide from Flue Gas Using Ion Pumping

Roger D. Aines 06-ERD-014

Abstract

Cost-effective separation of carbon dioxide from combustion sources is the main limitation to lowering carbon emissions generated during energy production. In this project, we propose to separate carbon dioxide from flue gas by ionic pumping of carbonate ions dissolved in water. The ion pump dramatically increases dissolved carbonate ion in solution and hence the overlying vapor pressure of carbon dioxide gas, allowing its removal as a pure gas. This novel approach to increasing the concentration of extracted gas permits new approaches to treating flue gas, because the slightly basic water used as the extraction medium is impervious to trace acid gases that destroy existing solvents used in conventional methods, and no pre-separation is necessary. We will demonstrate the chemistry of the method through modeling and laboratory experiments.

We anticipate that our method will compete favorably with current chemical stripping systems used for carbon dioxide separation at power plants, which incur a 35% energy penalty. Thus we expect to offer a dramatically improved solution for removing carbon from hydrocarbon combustion. Our method can be demonstrated on small sources, which will enable us to conduct the demonstrations required to build confidence in the method. If successful, we will be in a position to advance a follow-on proposal for a demonstration at the 10-MW scale.

Mission Relevance

This project supports energy security and environmental missions by enhancing currently proposed carbon management options such as technology development for fuel efficiency via fuel cells, hydrogen fuel, and other methods; fossil-fuel recovery; and carbon dioxide sequestration.

FY07 Accomplishments and Results

In FY07 we achieved two major milestones. We addressed the significant research question of the process working at flue-gas concentrations by designing and executing a successful demonstration of the technology. We also addressed another significant hurdle for large-scale implementation by establishing that the process works with all three water desalinization methods commonly in use today—that is, reverse-osmosis, electrodialysis, and thermal distillation systems. These two accomplishments are major steps toward validating the economic utility of our approach.

Proposed Work for FY08

In FY08 we propose to focus on the application of our ion pump as a carbon dioxide concentrating mechanism. We will test gas and fluid flow in multiple-plate ion pumps and determine if plate materials can be optimized for the carbon separation process. A major goal is to determine the parameters necessary for a pilot-scale test of the technology. We will also address a particularly interesting possibility by investigating if it would be feasible to produce drinking water during the separation process, thereby increasing its economic viability.

Fossil-Fuel Emission-Verification Capability

John P. Knezovich 07-ERD-064

Abstract

The capability to quantify and verify carbon emissions to ensure adherence to emission limits is vital to California as well as national and global environmental management and energy security efforts. Expertise in carbon isotope analysis, atmospheric modeling, and computational physics make Lawrence Livermore uniquely qualified to supply independent verification of carbon dioxide emissions from fossil-fuels. Accurate assessment of fossil-fuel carbon emissions require highly precise measurements of car emissions and a sampling program with geographic and temporal resolution adequate to determine regional emission profiles. This project will provide the basis for a measurement program that will be intimately coupled with advanced atmospheric transport and inversion models to provide a transparent and independent capability for verification of fossil-fuel carbon dioxide emissions in California.

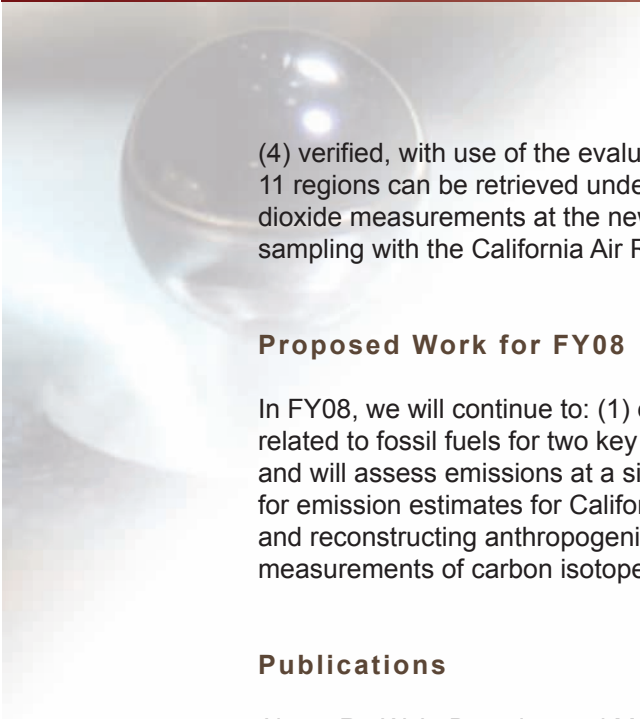
This project will establish the fossil-fuel emission (carbon-14 dioxide) variability for a metropolitan region in California and create a simulation-based framework for locating and tracking fossil-fuel emissions for California. Completing these two tasks will provide the basis for a fossil-fuel emission verification program for California and establish LLNL as a key resource for implementation of national carbon emission-verification programs.

Mission Relevance

This project supports LLNL's missions in environmental management and energy security by promoting advancements in carbon and climate research. In addition, DOE's Office of Biological and Environmental Research has a vested interest in understanding carbon-cycle dynamics. This project is also closely aligned with LLNL efforts to establish initiatives relevant to needs within California.

FY07 Accomplishments and Results

Initiated in midyear FY07, our effort was directed toward creating a retrieval scheme to calculate the degrees of freedom provided by existing data. Specifically, we (1) adapted the stochastic Bayesian event reconstruction algorithm to applications involving multiple distributed sources; (2) successfully carried out an intercomparison of the algorithms using synthetic data for 11 California air basins; (3) assessed global sources of carbon dioxide and stratospheric carbon-14 using global and regional air-quality forecasting models;



(4) verified, with use of the evaluated algorithms, that fossil-fuel emissions from the 11 regions can be retrieved under an idealized scenario; and (5) initiated continuous carbon dioxide measurements at the new Sacramento sampling tower, and coordinated Los Angeles sampling with the California Air Resources Board staff.

Proposed Work for FY08

In FY08, we will continue to: (1) establish the variability for carbon-14 dioxide emission related to fossil fuels for two key regions in California (i.e., Sacramento and San Diego) and will assess emissions at a site in Los Angeles; (2) create a prototype retrieval scheme for emission estimates for California; and (3) demonstrate two methodologies for retrieving and reconstructing anthropogenic sources of carbon dioxide in California based on in situ measurements of carbon isotopes.

Publications

Aines, R., W. L. Bourcier, and M. R. Johnson, 2006. "Separation of carbon dioxide from flue gas using ion pumping." *Proc. Eighth Intl. Conf. Greenhouse Gas Control Technologies*. UCRL-CONF-220812.

Laboratory Directed Research and Development

Engineering and Manufacturing Processes



Study of Transport Behavior and Conversion Efficiency in Pillar Structured Neutron Detectors

Rebecca J. Nikolic 06-ERD-067

Abstract

An efficient radiation detection device that can be easily fielded is vital to national security efforts. In this project, we will demonstrate technology that could lead to a device with over 70% thermal neutron detection efficiency. By applying microtechnology methods to neutron detection, we expect to make revolutionary improvements in device efficiency and field usability. We will take advantage of recent advancements in material science, charged carrier transport, and neutron-to-alpha conversion dynamics to fabricate semiconductor pillars in a three-dimensional matrix in which the neutron-to-alpha conversion material has adequate density to capture the full neutron flux.

With this project, we intend to develop and demonstrate a proof-of-principle device, and we will devise a roadmap for scaling the device to optimal efficiency. This is significant because current technology suffers from poor efficiency and adaptability to field use, high voltage, sensitivity to microphonics, a large device footprint, and high pressure, resulting in significant complications in air transport and deployments. The advances we propose in microfabrication and nanofabrication methods are applicable to many other fields, including biochemical detection, communications, and computations.

Mission Relevance

Our project supports the Laboratory's national security mission by advancing technology for detection of special nuclear materials and radiological dispersal devices. If our demonstration device meets the requirements for high efficiency and demonstrates suitability for field use, it would pave the way to manufacturing field-ready devices in partnership with an industrial collaborator.

FY07 Accomplishments and Results

In FY07, we (1) achieved a boron-10 fill factor of greater than 95% for pillar heights of 12 μm ; (2) completed a fully functional alpha conversion efficiency setup to characterize the boron-10 films; (3) developed devices for high-energy alpha testing; (4) further developed a pillar detector model by determining the energy loss within the semiconductor pillar (in FY06 we had used the Monte Carlo MCNP code to simulate a nearby neutron point-source interaction with the structure, after which the range of the charged particles within the boron-10 was calculated using the code IRMA); and (5) presented conference papers to the American Chemical and Materials Research Societies.

Proposed Work for FY08

Our primary milestone for FY08 will be characterization and modeling of a 20- μm pillar detector. We will develop (1) methods to etch high-aspect-ratio features with 2- μm feature

size and deposit boron materials in these features with a high fill factor; (2) neutron-to-alpha conversion efficiency in high-resistivity, chemical-vapor-deposited boron films; (3) electron and hole transport in vertical high-aspect-ratio features; and (4) a model of the pillar detector that includes both nuclear physics and semiconductor device physics.

Publications

Deo, N., et al., 2007. *Chemical vapor deposition of boron*. Materials Research Society Spring Mtg., San Francisco, CA, Apr. 9–13, 2007. UCRL-PRES-229731.

Deo, N., et al., 2007. *Growth of boron films by LPCVD*. 41st American Chemical Society Midwest Regional Mtg., Quincy, IL, Oct. 25–27, 2006. UCRL-PRES-225482.

Standing-Wave Probes for Micrometer-Scale Metrology

Richard M. Seugling 07-ERD-042

Abstract

The objective of this project is to develop a low-force, high-aspect-ratio contact probe for the nondestructive characterization of components and assemblies developed for fusion-class laser targets. The key concept for the standing-wave probe is correlation between the dynamic response of an oscillating, slender, cantilever rod and its interaction with a material surface. The goal of this project is to provide information about surface topography and localized material properties, specifically in low-density foams. A principal component is a detailed analysis of the fundamental contributions to the uncertainty of the measurement process, including the surface science of probe–sample interaction, dynamic response of the probe, and limitations of scale.

The expected results of this project include (1) a functional probe system capable of determining surface location, material properties, and possible surface modification; (2) a fundamental understanding of the probe–surface interactions and the resulting uncertainty related to the measurement based on analytical models and experimental results; (3) an assessment of fundamental limitations of this technology as scale decreases; and (4) an evaluation of expanding probe sensitivity to multiple dimensions. It is the goal of this work to develop a fundamental understanding of how surface–probe interactions affect the dynamic response of the probe system and how these variations in dynamic response can be attributed to parameters such as surface location, hardness, and elastic modulus.

Mission Relevance

Two specific areas of relevance for this work are stockpile science and technology and high-energy-density science and technology as they relate to characterization of high-power laser target parts and assemblies. Immediate applications include dimensional characterization of target geometry with micrometer features, with future applications in dimensional as well as material characterization of low-density materials such as aerogels and metallic foams.

FY07 Accomplishments and Results

Our accomplishments for FY07 include (1) implementing a probe system and test platform including upgraded electronics and mechanics for improved sensitivity and ease of use; (2) performing sensitivity analysis of the new probe on characterized samples; (3) developing a model of the probe dynamics including nonlinear contact conditions and theoretical approximations of near-surface (van der Waals and electrostatic) and contact forces (impact, adhesion, and meniscus); (4) quantification of surface forces measured using a modified atomic-force microscope tip on multiple substrates; and (5) establishing collaborations with the University of North Carolina at Charlotte and a private-industry partner.

Proposed Work for FY08

In FY08, we will focus on leveraging our understanding of probe response to reduce the scale of the probe, investigate combinations of drive methods to create multidirectional sensitivity, and evaluate the ability to determine material properties and make any necessary surface modifications. Our modeling effort will transition to considering the “inverse problem.” That is, we will strive to infer material surface properties based on measurement data. Because properties such as hardness and elastic constants are natural extensions of nano-indentation work, we will also consider aerogel properties (fracture toughness) and adhesion properties (using various tip materials) and potentially search for bond failures in foil targets.

Ultrahigh-Velocity Railgun

Jerome M. Solberg 07-ERD-055

Abstract

Previous efforts to develop ultrahigh-velocity railguns for shock physics experiments were limited by the lack of appropriate simulation tools for the three-dimensional nature of plasma in the armature. However, the availability of a new, three-dimensional magnetohydrodynamic code, along with railgun components available at LLNL, allows us to gain a detailed understanding of armature plasma behavior. We propose a combination of simulations and experiments aimed at creating a class of ultrahigh-velocity railguns utilizing a hybrid armature of solid and plasma brushes. Such a railgun would provide a means for equation-of-state (EOS) research at pressures unattainable by gas guns.

If successful, this project will provide an experimentally and computationally supported understanding of ultrahigh-velocity railguns. Such an understanding may lead to designs which would be useful for EOS research at pressures unattainable by gas guns. In addition, the knowledge of the underlying physics is applicable to other railgun applications important to the DOE, Department of Defense, and National Aeronautics and Space Administration, such as fusion-reactor pellet refueling, kinetic-energy weapons, asteroid impact testing, and space launch.

Mission Relevance

This project supports future applications in support of LLNL's stockpile stewardship mission by designing a device capable of reaching, in a controlled fashion, physical states not easily attainable by any other technology. In addition, the simulation and experimental capabilities developed in this research will be transferable to a number of other national security efforts involving explosive pulsed power and high-energy physics, such as magnetic flux compression.

FY07 Accomplishments and Results

In FY07, we (1) completed the primary mechanical, electrical, and diagnostic design of the fixed-armature experiment; (2) performed simple runs of the experimental design using the code ALE3D; (3) performed simulations of the plasma physics for the experiment with the code CALE, which assisted us in the design and suggested variations (e.g., an increase in the armature gap to ensure blackbody conditions and a longer armature to minimize plasma mass loss after the foil explosion) and provided a simulation baseline; (4) incorporated plasma conductivity combined radiation–conduction heat transfer for plasmas into ALE3D via EOS data library tables; and (5) began fixed-armature experiment data collection.

Proposed Work for FY08

In FY08, we will (1) continue simulations of and perform additional experiments on the fixed-armature device aimed at increasing code–experiment correlation; (2) develop simulations of the HELEOS EOS railgun experiments, which utilized a unique hybrid concept incorporating plasma brushes with a solid armature; (3) compare these simulation results with actual data recovered from archives; (4) utilize this capability to design plasma and hybrid-armature experiments for future test on a reconstituted railgun; and (5) use the fixed-armature experiment to design a diagnostic package sufficient to compare with the simulations, which will include high-resolution information about plasma position, pressure, and electrical and optical properties.

Laboratory Directed Research and Development

Materials Science and Technology





Target Fabrication Science and Technology: An Enabling Strategic Initiative

Peter A. Amendt 05-SI-005

Abstract

This project established a new, major capability for fabricating targets for fusion ignition with applications in homeland security and nanotechnology. To this end, we developed the science and technology for (1) nanocrystalline grains, (2) graded-dopant materials, (3) high-strength diamond shells, (4) nanoporous metals, (5) high-strength aerogels, (6) atomic-layer deposition on aerogels, (7) interference and advanced lithography, and (8) joining techniques for precision microassembly. We applied and integrated the various elements to demonstrate the capability to produce a prototype ignition double-shell target.

This project advanced nanoscience techniques for fabricating targets and other structures by developing (1) nanostructured alloys with extraordinary strength, ductility, 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 (3D) nanofabrication on large areas and complex surfaces with nanoscale-relief structures. Insight into all of these issues has led to a new class of materials with novel properties.

Mission Relevance

This work supports LLNL's missions in national and energy security by developing the target-fabrication technology for the increasingly complex targets needed in stockpile stewardship and fusion ignition.

FY07 Accomplishments and Results

In FY07 we demonstrated the capability to fabricate an ignition-ready, double-shell prototype target integrating the world-class material science advances achieved in the previous two years of this project. Specifically, we demonstrated (1) a nanocrystalline ignition-scale gold-copper shell, (2) native root-mean-square (RMS) surface smoothness of less than 1 μm on the outer surface of the bimetallic nanocrystalline inner shell, (3) equivalent inner-shell strength to contain 800 atm of hydrogen fuel at room temperature, (4) nanoporous metallic foams ($<100 \text{ mg}/\text{cm}^3$) with submicrometer pore size formed into a shell, (5) hemispherical diamond shells and greater than 50-nm RMS surface roughness, and (6) the viability of using glass mandrels as radiochemical tracers in an ignition double shell for mix studies.

Publications

Amendt, P., et al., 2007. "Assessing the prospects for achieving double-shell ignition on the National Ignition Facility using vacuum hohlraums." *Phys. Plasma* **14**, 056312. UCRL-JRNL-225805.

Biener, J., et al., 2007. "Ruthenium/aerogel nanocomposites via atomic layer deposition." *Nanotechnology* **18**, 055303. UCRL-JRNL-226605.

Biener, J., et al., 2007. "Size effects on the mechanical behavior of nanoporous Au." *Nano Lett.* **6**, 2379. UCRL-JRNL-221479.

Bono, M, et al., 2007. "Fabrication of double shell targets with a glass inner capsule supported by SiO₂ aerogel for shots on the Omega Laser in 2006." *Fusion Sci. Tech.* **51**, 611. UCRL-JRNL-225802.

Hayes, J. R., et al., 2007. "Monolithic nanoporous copper by dealloying Mn–Cu." *J. Mater. Res.* **21**, 2611. UCRL-JRNL-218835.

Hayes, J. R., et al., 2007. "Synthesis of bi-modal nanoporous Cu, CuO and Cu₂O monoliths with tailored porosity." *Nanotechnology* **18**, 275602. UCRL-JRNL-221538.

Hodge, A. M., et al., 2007. "Scaling equation for yield strength of nanoporous open-cell foams." *Acta Mater.* **55**, 1343. UCRL-JRNL-221840.

Jankowski, A. F., C. K. Saw, and J. P. Hayes, 2007. "The thermal stability of nanocrystalline Au–Cu alloys." *Thin Solid Films* **515**, 1152. UCRL-JRNL-219074.

Jurgens, B., et al., 2007. "New gold and silver-gold catalysts in the shape of sponges and sieves." *Gold Bull.* **40**, 142. UCRL-JRNL-229166.

Kucheyev, S. O., et al., 2007. "Structure of low-density nanoporous dielectrics revealed by low-vacuum electron microscopy and small-angle x-ray scattering." *Langmuir* **23**, 353. UCRL-JRNL-221923.

Nyce G. W., et al., 2007. "Synthesis and characterization of hierarchical porous gold materials." *Chem. Mater.* **19**, 344. UCRL-JRNL-225648.

Smith R. F., et al., 2007. "Graded-density reservoirs for accessing high stress low temperature material states." *Astrophys. Space Sci.* **307**, 269. UCRL-JRNL-220822.

Wang Y. M., A. F. Jankowski, and A. V. Hamza, 2007. "Strength and thermal stability of nanocrystalline gold alloys." *Scripta Mater.* **57**, 301. UCRL-JRNL-228995.

Zepeda-Ruiz, et al., 2007. "Mechanical response of freestanding Au nanopillars under compression." *Appl. Phys. Lett.* **91**, 101907. UCRL-JRNL-226968.

Zielasek, V., et al., 2007. "Gold catalysts: nanoporous gold foams." *Angew. Chem. Int. Ed.* **45**, 8241. UCRL-JRNL-222533.



The Structure and Properties of Nanoporous Materials

Anthony W. van Buuren 05-ERD-003

Abstract

Although much research has been devoted to the physical properties of porous structures, it is not clear whether the theoretical models developed to date can be extended to nanoscopic length scales. In this project, we are measuring how the structure of highly porous metal and metal oxide foams changes with temperature, pressure, and surface environment. How these porous materials react mechanically and structurally under such conditions will be examined using 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 anticipate several publications in peer-reviewed scientific journals.

Mission Relevance

The ability to characterize and model nanoporous materials developed in this project will further applications in stockpile stewardship and high-energy-density science, in support of the national security mission.

FY07 Accomplishments and Results

One of the main deliverables in FY07 was to quantify the pore structure of metal and metal oxide foams. We used SAXS to measure the correlation range (CR)—the distance between ligaments in an aerogel—as a function of sample preparation. In tantalum oxide aerogel, the inverse of the CR scaled linearly with density until 100 mg/c³; below this the density decreased with no corresponding change in CR. Measurement of scattering at a low scattering vector showed evidence of greater than 800-nm holes in some samples. For samples below 100 mg/c³ the CR remained constant, and the decreased density was attributable to the formation of holes. We also measured deformation of the pore structure of an aerogel immersed in liquid nitrogen. X-ray tomography was used to measure density fluctuation in the metal oxide foams at better than 1- μ m resolution. In summary, the small-angle scattering capability developed in this project has successfully led to capabilities that support of various mission-relevant applications at LLNL, such as measurement of the void structure in highly energetic materials, the study of pore structure and volume in beryllium, and the in situ SAXS study of carbon aerogels immersed in liquid hydrogen.

Publications

Barty, A., et al., in press. "Three-dimensional coherent x-ray diffraction imaging of a ceramic nanofoam: determination of structural deformation mechanisms." *Phys. Rev. Lett.* UCRL-JRNL-231416.

Characterization and Control of Laser-Induced Modifications in Potassium Dihydrogen Phosphate and Deuterated Potassium Dihydrogen Phosphate Crystals

Stavros G. Demos 05-ERD-016

Abstract

The objective of this research is to reveal fundamental mechanisms involved in the laser-induced defect reactions leading to bulk damage and laser conditioning in potassium dihydrogen phosphate (KDP) and deuterated KDP (DKDP) crystals. The interaction of laser light with defect structures in KDP and DKDP crystals can lead either to modification to a less-absorbing species or to initiation of damage. The precise conditions controlling these behaviors are not currently known. We propose to understand both the nature of the interactions of laser light with defect structures and how to control the results. KDP and DKDP are the only materials available for use in large-aperture lasers as frequency converters.

We intend to 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 to laser irradiation while the laser parameters for conditioning and damage testing are varied. This will lead to predictive models of the behavior of materials at operational conditions, methods to optimize performance, and possibly, the characterization of damage-initiating defects. 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 stockpile stewardship mission by providing novel diagnostic tools to quantify and predict damage characteristics of optical materials for large-aperture laser systems, and by offering basic knowledge to optimize conditioning protocols for frequency-doubling and frequency-tripling crystals. In the long term, this project will lead to novel technologies for materials characterization at future large-scale lasers.

FY07 Accomplishments and Results

In FY07, we completed our study of the dependence of damage performance on (1) materials pre-exposed to laser pulses over a wide range of parameters (wavelength, fluence, number of pulses, and time resolved pump-probes); (2) x-ray irradiation that leads to formation of known transient defects species; and (3) growth parameters to identify those that correlate to increased density of precursors. The results allowed us to reveal characteristic behavior of the damage precursors, which helped develop a detailed model of their electronic structure and the excitation pathways leading to conditioning and damage initiation. We used this information to determine a particular subset of defects that have many elements predicted to govern the interaction of laser pulses with damage precursors.

Publications

DeMange, P., et al., 2007. "Differentiation of defect populations responsible for bulk laser-induced damage in potassium dihydrogen phosphate crystals." *Opt. Eng.* **45**, 104205. UCRL-JRNL-217769.

DeMange, P., et al., 2007. "Laser-induced defect reactions governing damage performance in KDP and DKDP crystals." *Proc. SPIE* **6103**, 610305-1. UCRL-CONF-218739.

DeMange, P., et al., 2007. "Understanding and predicting the damage performance of $\text{KDxH}_2\text{-xPO}_4$ crystals under simultaneous exposure to 532- and 355-nm pulses." *Appl. Phys. Lett.* **89**, 181922. UCRL-JRNL-214882.

Negres, R. A., et al., 2007. "An expedited approach to evaluate the importance of different crystal growth parameters on laser damage performance in KDP and DKDP." *Proc. SPIE* **6403**, 64031S-1. UCRL-PROC-225780.

Negres, R. A., et al., 2007. "Evaluation of UV absorption coefficient in laser-modified fused silica." *Appl. Phys. Lett.* **90**, 061115. UCRL-JRNL-224110.

Negres, R. A., et al., 2007. "Expedited laser damage profiling of $\text{KDxH}_2\text{-xPO}_4$ with respect to crystal growth parameters." *Optic. Lett.* **31**, 3110. UCRL-JRNL-220921.

Negres, R. A., et al., 2007. "Thermal imaging investigation of modified fused silica at surface damage sites for understanding the underlying mechanisms of damage growth." *Proc. SPIE* **6403**, 640306-1. UCRL-PROC-225830.

Ceramic Laser Materials

Thomas F. Soules 05-ERD-037

Abstract

Our objective is to evaluate the utility of using transparent ceramics for optical components of high-power lasers, including gain media. Ceramics made by sintering powders at temperatures below their melting point can 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 manufacture, potential multifunctionality, higher and more uniform activator concentrations, and reduced brittleness. We plan to make small pieces of transparent ceramics using nanoparticle material and to evaluate larger transparent ceramic pieces obtained commercially in actual high-power laser configurations.

If successful, we will demonstrate that LLNL can fabricate transparent ceramic laser materials and that these materials are suitable for use in high-power lasers. We will further implement transparent ceramic slabs, where applicable, in LLNL high-powered lasers and design several laser architectures that are possible only with ceramic materials, including large-aperture slabs for a solid-state heat-capacity laser and monolithic transparent pieces with multiple functionalities. For example, they could serve as laser gain media while also acting as light guides for optical pumping, amplified spontaneous emission suppression, and heat

extraction. Finally, we expect to establish an industrial source of transparent ceramics for LLNL.

Mission Relevance

This project supports the national security mission by providing ceramic laser-gain slab technology for solid-state heat capacity lasers, which can provide the power (~30 kW) needed for defense applications such as destroying mortar shells, rockets, and land mines. Current single-crystal slabs are very difficult to grow, and larger slabs may not be possible, whereas the larger slab sizes enabled by ceramic slabs would increase available laser power.

FY07 Accomplishments and Results

In FY07 we (1) fabricated transparent pieces of a new material (strontium fluoride) of importance to storage lasers, (2) developed slip casting and extrusion techniques, (3) successfully fabricated neodymium:yttrium–aluminum–garnet (Nd:YAG) transparent samples using nano-sized powders subjected to flame spray pyrolysis, and (4) characterized the optical properties of the ceramic ytterbium–ytterbium sesquioxide, a potential long-lifetime laser material. In summary, this project developed robust synthesis methods for creating transparent ceramics and, in collaboration with an industrial partner, developed large-aperture Nd:YAG ceramics and a novel ceramic edge cladding that suppresses amplified spontaneous emission and enables edge pumping. Follow-on work would include the development of tailored ceramic laser parts.

Publications

Soules, T. F., 2007. *Transparent ceramics for laser amplifiers*. 31st Intl. Cocoa Beach Conf. and Exposition Advanced Ceramics and Composites, Daytona Beach, FL, Jan. 21–26, 2007. UCRL-ABS-342692.

Soules, T. F., 2006. *Ceramic Laser Materials*. OSA Frontiers in Optics, Rochester, NY, Oct. 8–12, 2006. UCRL-PRES-339369.

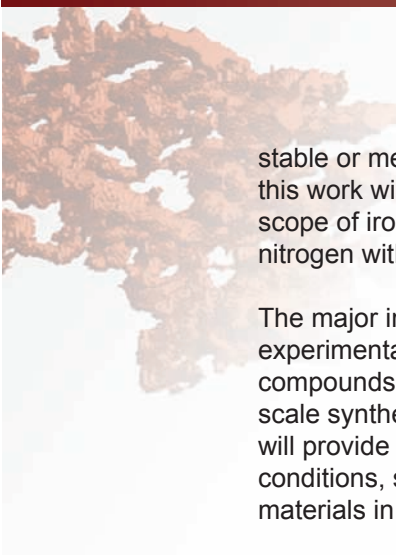
Soules, T. F., 2007. *Transparent Laser Ceramics at Lawrence Livermore*. 3rd Laser Ceramics Symp., Paris, France, Oct. 8–10, 2007. URCL-PROC-349510.

Determination of the High-Pressure Melting Curve of Iron

Jonathan C. Crowhurst 05-ERD-039

Abstract

The original aim of this project was to determine, with unprecedented precision, the melting curve of iron at geophysically relevant pressures. In the course of developing the technology and techniques required to obtain this information, we encountered and studied novel chemical reactions whose products—nitrides of the noble metals platinum and iridium—are



stable or metastable under ambient conditions. We believe the scientific impact of continuing this work will be greater than that of the original goals. Therefore, we propose to reduce the scope of iron melting and make our primary focus the chemistry under extreme conditions of nitrogen with other platinum-group metals.

The major impact of this work will be the elucidation of new noble metal chemistry. Our experimental results provide a rigorous test of first-principles theoretical approaches. Once compounds with useful properties have been shown to exist under ambient conditions, large-scale synthesis techniques (e.g., nitrogen ion irradiation) can be considered. This project will provide the Laboratory with wide-ranging achievements in chemistry under extreme conditions, such as an enhanced ability to perform Raman measurements on relevant materials in the Chapman–Jouguet state and to identify reaction products.

Mission Relevance

By offering a practical and accurate means for determining 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.

FY07 Accomplishments and Results

We observed iron melting at an approximate pressure of 25 GPa, using nuclear forward scattering as a diagnostic to determine the onset of melting. We also determined the structure of platinum nitride and synthesized the entirely novel compounds iridium and palladium nitride. We published two papers on the use of cubic boron nitride as an in situ high-pressure, high-temperature pressure sensor and two papers on novel nitride synthesis, including an article in *Science*. This project significantly advanced our basic understanding of these novel systems and directly impacted stockpile-relevant material.

Publications

Crowhurst, J. C., et al., in press. "Synthesis and characterization of nitrides of iridium and palladium." *J. Mater. Res.* UCRL-JRNL-229796.

Crowhurst, J. C., et al., 2007. "Synthesis and characterization of the nitrides of platinum and iridium." *Science* **311**, 1277. UCRL-JRNL-216422.

Goncharov, A. F., 2007. "Thermal equation of state of cubic boron nitride: implications for a high-temperature pressure scale." *Phys. Rev. B* **75**, 224114. UCRL-JRNL-225104.

Goncharov, A. F., et al., 2007. "Raman spectroscopy of cubic boron nitride under extreme conditions." *Phys. Rev. B* **72**, 100104. UCRL-JRNL-218266.

Mitigation of Damage Sites on Ultraviolet Optics

Vaughn G. Draggoo 05-ERD-066

Abstract

Our objective is to explore and further develop the concept that laser-induced damage on surfaces of an optic can be mitigated to extend its lifetime in high-fluence laser systems. We will explore mitigation processes utilizing laser, mechanical, and chemical techniques as well as characterize the size and shapes of the resulting divots in terms of their acceptability with respect to downstream beam intensification. Advanced materials characterization tools will be used to investigate physical properties of the mitigated regions. The proposed mitigation efforts will focus on fused silica and frequency-conversion potassium dihydrogen phosphate (KDP) crystals, and will establish a fundamental understanding of the physical mechanisms of mitigation processes to extend their applicability.

We will develop robust mitigation protocols for fused silica capable of treating damage sites varying in size from 0.02 to greater than 1 mm in diameter. Several options will be explored, including (1) varying the optical penetration depth by appropriate choice of laser wavelength, (2) controlling end-state material properties by controlling surface and bulk temperature, and (3) varying the mitigation laser-output properties (e.g., pulse width and intensity). For KDP crystals, we propose to examine methodologies based on micromachining and chemical etching. If successful, the application of mitigation techniques will significantly extend the useful lifetime of an optic in a high-fluence laser system, and drastically reduce the associated operational costs.

Mission Relevance

Development of mitigation techniques that arrest damage growth in optical components would significantly benefit efforts to field powerful, fusion-class laser systems by allowing these systems to operate efficiently, reliably, and cost effectively at or above their design specifications. Large, fusion-class laser systems are essential tools for studying weapons physics for stockpile stewardship and inertial confinement fusion, in support of LLNL's national and energy security missions.

FY07 Accomplishments and Results

In FY07, we (1) correlated light-propagation simulations with actual mitigation divot morphology to minimize downstream intensification, (2) evaluated mitigation processes combining galvo-steered and frequency-doubled carbon-dioxide lasers for silicon-dioxide optics, (3) evaluated the degree of damage mitigation achieved in KDP through micromachining and scanned-probe-based recrystallation techniques, and (4) employed advanced optical materials characterization techniques with high spatial and temporal resolution, such as confocal photoluminescence microscopy and microphotoelasticity, to identify conditions causing damage initiation and growth. Optical coherent tomography provided high-resolution, three-dimensional images of crack morphology healing during mitigation.

Publications

Guss, G. M., et al., 2007. *High-resolution 3-D imaging of surface damage sites in fused silica with optical coherence tomography*. Laser Induced Damage in Optical Materials, 39th Annual Boulder Damage Symp., Boulder, CO, Sept. 24–26, 2007. UCRL-PRES-234640.

Matthews, M. J., and M. D. Feit, 2007. *Effect of random clustering on surface damage density estimates*. Laser Induced Damage in Optical Materials, 39th Annual Boulder Damage Symp., Boulder, CO, Sept. 24–26, 2007. UCRL-PRES-234637.

Matthews, M. J., et al., 2007. *Downstream intensification effects associated with CO₂ laser mitigation of fused silica*. Laser Induced Damage in Optical Materials. 39th Annual Boulder Damage Symp., Boulder, CO, Sept. 24–26, 2007. UCRL-POST-234882.

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 develop a solid scientific understanding of the creation and characteristics of surface fractures formed during the grinding and polishing of brittle materials, specifically glass. We have experimentally characterized various surface cracks as a function of a number of typical finishing processes. In addition, the effect of key process parameters on the surface damage of fused silica has been measured to develop a global working model to predict surface damage. This project has greatly advanced the scientific knowledge of microscopic mechanical damage caused by grinding and polishing. This knowledge base has enabled the optimization of surface finishing processes.

The work has produced a body of materials research and enhanced manufacturing fabrication processes, which benefit efforts to field high-energy, high-power laser systems and allow them to operate efficiently, reliably, cost effectively, and at or above their design specifications. In addition, this effort has 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 Stockpile Stewardship Program's ability to understand weapons physics and materials under extreme conditions of temperature, pressure, and strain rate.

FY07 Accomplishments and Results

In FY07, we characterized and modeled the effects of rogue particles present during polishing, including their effect on subsurface damage and scratch characteristics. The results of our experiments were complimented by an offline experimental setup to make individual controlled scratches as a function of load, velocity, indenter shape, indenter stiffness, and lubrication. We also measured crack distributions and correlated measured etching behavior with that predicted by our etching model. Follow-on work would include further experiments and modeling to enhance the understanding of etching on subsurface damage.

Publications

Menapace, J., 2007. "Utilization of magnetorheological finishing as a diagnostic tool for investigating the three-dimensional structure of fractures in fused silica." *Proc. SPIE* **5991**(599102). UCRL-PROC-217132.

Miller, P., et al., 2007. "The distribution of subsurface damage in fused silica." *Proc SPIE* **5991**. UCRL-PROC-217443.

Suratwala, T., et al., 2007. "Subsurface mechanical damage distributions during grinding of fused silica." *J. Non-Crystalline Solid*. **352**, 5601. UCRL-JRNL-217445.

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 (up to 1-ns) laser parameters affect the conditioning and initiation of bulk damage sites in potassium dihydrogen phosphate (KDP) and deuterated KDP (DKDP) crystals and surface damage in fused silica (SiO_2). We will also develop a short-pulse laser with the desired operating parameters for devising optimum protocols for conditioning crystals and the pre-initiation of damage precursors in SiO_2 . Conditioning, pinpoint damage, pinpoint size, and pinpoint morphology will be measured. 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 the laser parameters responsible for the conditioning effect, we will use this information to develop offline, laser-based conditioning protocols for crystalline KDP and DKDP and fused silica. 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 in 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.

FY07 Accomplishments and Results

In FY07 we (1) determined the optimal conditioning parameters for second-harmonic-generative crystals, (2) studied the morphology of damage sites produced on SiO_2 with pulse durations between 0.3 and 10 ns, (3) studied the effect of pulse duration on growth, and (4) developed a model describing laser conditioning in KDP. This project developed a subnanosecond laser which we used to experimentally determine the best-available conditioning protocol for second- and third-harmonic generation crystals. We also developed a model describing laser conditioning, measured the effect of pulse duration on initiation and growth for SiO_2 optics, and developed a model describing how pulse duration affects the mechanics of damage site formation.

Publications

Adams, J. J., et al., 2007. *Comparison between S/1 and R/1 tests and damage density vs. fluence ($r(f)$) results for unconditioned and sub-nanosecond laser-conditioned KD_2PO_4 crystals*. 39th Ann. Symp. Optical Materials for High-Power Lasers, Boulder, CO, Sept. 24–26, 2007. UCRL-PRES-234881.

Norton, M. et al., 2007. *Growth of laser damage in fused silica: diameter to depth ratio*. 39th Ann. Symp. Optical Materials for High-Power Lasers, Boulder, CO, Sept. 24–26, 2007. UCRL-PRES-234639.

Molecular Transport in One-Dimensional Lipid Bilayers: A Biological “Smoke Alarm”

Aleksandr Noy 05-LW-040

Abstract

This research centers on one-dimensional (1D) bilayers—new biomimetic nanostructures that consist of a lipid bilayer wrapped around a single-carbon nanotube transistor. The lipid bilayer insulates the nanotube from the solution and serves as a matrix for membrane proteins. Use of biomimetic components in a nanoelectronic device allows direct conversion of ion transport events into electrical signals. We will synthesize 1D lipid bilayers and use these structures to study ion transport through a single protein pore.

This project will not only create a new class of functional biocompatible nanostructures and use them to perform fundamental studies of molecular transport in these systems, but

also will develop a prototype nanodevice for ion flux detection. In addition to addressing important scientific questions such as synthesis of 1D organic nanostructures and studies of mass transport in 1D environments, this research also establishes an important capability in developing nanowire-based biological sensors for homeland security. These studies have the potential to enable several important biophysical applications, such as creation of subcellular-size, ion-selective biocompatible electrodes or analyzing mass transport through individual protein pores.

Mission Relevance

This project is intended to develop a set of novel and important capabilities in biodetection and scientific capabilities in biophysical research: the synthesis and characterization of organic nanostructures. It supports the Laboratory's mission in homeland security and bioterrorism prevention, and biophysical aspects of this research are relevant to the DOE's Genomics:GTL program. In addition, the project contributes to LLNL's mission in breakthroughs in fundamental science.

FY07 Accomplishments and Results

In FY07, we (1) constructed nanodevices that incorporate 1D lipid bilayers, (2) showed that the devices retain single-wire transistor electrical properties, (3) performed electrochemical measurements that verified electrical shielding properties of the bilayer adsorbed on the silicon nanowire platform, (4) used the devices to measure ion current through a biological pore—this result marks the first demonstration of the operation of a 1D bilayer device, and (5) studied the curvature effect on mobility of the lipid bilayer molecules. For curvatures up to 50 nm, the bilayer mobility follows the free-space diffusion model, but at higher curvatures the mobility shows an unexpected deviation, indicating significant rearrangements.

Publications

Artyukhin, A. B., et al., 2007. "Controlled electrostatic gating of carbon nanotube FET devices." *Nano Lett.* **6**, 2080. UCRL-JRNL-222278.

Artyukhin, A. B., et al., in press. "Fabrication and characterization of suspended carbon nanotube devices in liquid." *Intl. J. Nanotech.* UCRL-JRNL-225782.

Artyukhin, A. B., et al., 2007. "Functional one-dimensional lipid bilayers on carbon nanotube templates." *J. Am. Chem. Soc.* **127**, 7538. UCRL-JRNL-205932.

Artyukhin, A. B., et al., 2007. "Layer-by-layer electrostatic self-assembly of polyelectrolyte nanoshells on individual carbon nanotube templates." *Langmuir* **20**, 1442. UCRL-JRNL-200548.

Huang, S.-C. J., et al., 2007. "Formation, stability, and mobility of one-dimensional lipid bilayers on polysilicon nanowires." *Nano Lett.* **7**(11), 3355. UCRL-JRNL-229513.

Huang, S.-C. J., et al., 2007. "Persistence length control of the polyelectrolyte layer-by-layer self-assembly on carbon nanotubes." *J. Am. Chem. Soc.* **127**, 14176. UCRL-JRNL-214130.



Transformational Materials Initiative

Randall L. Simpson 06-SI-005

Abstract

The goal of this project is to provide the underlying science and technology for converting the U.S. nuclear weapons complex to one that is smaller, safer, and more agile. We will create new materials, processes, and diagnostics to facilitate the nuclear weapons complex transformation by (1) reducing the cost and time required to produce and maintain the stockpile, (2) enhancing weapon safety, (3) ensuring future stockpile longevity, and (4) optimizing stockpile performance. This multidisciplinary team combines capabilities in materials synthesis, characterization, theory, and modeling to deliver cutting-edge advances in high explosives, multifunctional materials, metals, and sensing.

The project will create basic scientific and technical capabilities that, if successful, will help transform the nuclear weapons complex. Success will make it more efficient and effective by achieving fundamental scientific advances in synthesis chemistry, metallurgy, dynamic experiments, and molecular design.

Mission Relevance

This project will achieve enhanced reliability, improved safety, easier manufacturing, and reduced surveillance requirements for the nuclear weapons complex and help ensure the continued success of the Stockpile Stewardship Program.

FY07 Accomplishments and Results

For FY07, we (1) prepared a new explosive composite with improved quality; (2) completed new simulations of shocked high explosives, which revealed a strong interaction of the shock wave loading with the volume collapse from a phase change in bismuth; (3) manufactured test specimens of advanced metals by both proposed processing routes; and (4) developed three sensing technologies and conducted preliminary testing of a capability to monitor criteria of interest. Our high-explosive loading results showed novel and promising results for their ductility. To develop multifunctional materials, we synthesized a number of lead–polyhedral oligomeric silsesquioxane–polydimethylsiloxane copolymer composites and created polymer–catalyst blends that demonstrate both catalytic activity and reinforcement properties.

Proposed Work for FY08

We plan to (1) scale up the manufacture of our new high explosives to the kilogram level to test detonation and low-temperature mechanical properties, (2) extend our first-principles studies on the detonation of nitromethane to triamino-trinitro-benzene, (3) more fully characterize the chemical purity of recrystallized triamino-trinitro-benzene, (4) demonstrate the capability to discriminate multiple species in a complex mixture of gases in our prototype Raman and mid-infrared spectrometers, (5) demonstrate communication to a microelectromechanical

systems-based load sensor within realistic materials, (6) continue to refine our understanding of the effects of processing parameters on high-explosives loading response and continue to refine our simulations, and (7) synthesize a unified, catalytically active polymer nanocomposite.

Publications

Colvin, J., in press. "Modeling dynamic ductility: an equation of state for porous metals." *Proc. 15th APS Topical Conf. Shock Compression of Condensed Matter*. UCRL-CONF-233170.

Gee, R., et al., 2007. "Molecular dynamics investigation of adhesion between TATB surfaces and amorphous fluoropolymers." *Macromolecules* **40**, 3422. UCRL-JRNL-227533.

Giuliani, J., et al., in press. "Thermal degradation in endlinked, nonstoichiometric reacted PDMS networks by 1H multiple quantum NMR." *J. Phys. Chem.* UCRL-JRNL-231546.

Gjersing, E., et al., 2007. "Investigation of network heterogeneities in endlinked, nonstochiometric reacted PDMS networks by 1H multiple quantum NMR." *Macromolecules* **40**, 4962. UCRL-JRNL-229373.

Letant, S., et al., in press. "POSS-stabilized palladium nanoparticles as a hydrogenation catalyst." *Catalyst Comm.* UCRL-JRNL-227434.

Maiti, A., et al., 2007. "Hydrogen catalysis and scavenging action of Pd-POSS nanoparticles." *Chem. Phys. Lett.* **440**, 244. UCRL-JRNL-227837.

Martin, L. P., et al., in press. "Compaction behavior and mechanical properties of uniaxially pressed Bi-W composites." *Met. Transactions*. UCRL-JRNL-231733.

Martin, L. P., et al., 2007. "Compaction behavior of uniaxially cold-pressed Bi-Ta composites." *Scripta Mater.* **57**(3), 229. UCRL-JRNL-228578.

Orme, C., et al., 2007. "The use of scanning probe microscopy to investigate crystal-fluid interfaces." *Perspectives on Inorganic, Organic, and Biological Crystal Growth*. Springer, Secaucus, NJ. UCRL-BOOK-230189.

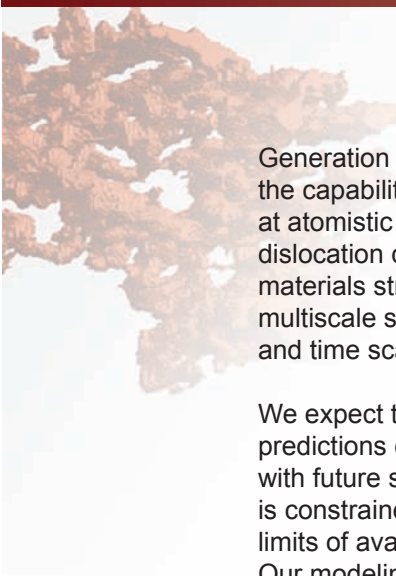
Reed, E., et al., in press. "A transient semi-metallic layer in detonating nitromethane." *Nat. Phys.* UCRL-JRNL-230354-DRAFT.

Critical Materials Issues for Generation IV Reactors

Magdalena A. Serrano De Caro 06-ERD-005

Abstract

In this project, we will develop predictive tools to calculate structural and mechanical properties of iron–chromium-based alloys that form the base matrix of advanced ferritic-martensitic steels that would be required for fuel cladding and structural components in



Generation IV reactors. Using the paradigm of Multiscale Materials Modeling, we will develop the capability to predict hardening, swelling, and embrittlement. We will link approaches at atomistic and mesoscale levels to create an integrated modeling platform that relates dislocation dynamics to polycrystal plasticity. This will be used to develop engineering-scale materials strength models for irradiated alloys. For validation purposes, our physically based multiscale study will be compared with the existing experimental data over a range of length and time scales.

We expect to provide a detailed physical understanding of iron–chromium alloy behavior and predictions of its mechanical properties critical for resolving technological issues associated with future sources of nuclear energy. Progress in developing advanced reactor concepts is constrained by the behavior of materials under irradiation. Our methodology expands the limits of available science on the thermodynamic and mechanic behavior of metallic alloys. Our modeling results contribute to an understanding of the performance of nuclear materials and the development of new high-temperature, radiation-resistant materials.

Mission Relevance

This project supports Laboratory and DOE missions in long-term energy security by developing scalable materials-science codes to predict nuclear materials properties and their performance under extreme conditions. It will assist efforts to develop future reactor concepts such as the Advanced Burner Reactor envisioned by the DOE's Global Nuclear Energy Partnership initiative. In addition, materials properties codes developed in this project can be used to predict radiation damage to materials important for LLNL's stockpile stewardship mission.

FY07 Accomplishments and Results

We (1) conducted molecular dynamics Monte Carlo studies of self-interstitial transport, vacancy, and point-defect clusters in concentrated iron–chromium alloys; (2) described defect microstructures evolution, local chemistry changes, grain boundary segregation, and alpha-prime precipitation; (3) used a recently developed parallel kinetic Monte Carlo code to demonstrate that grain boundary, dislocations, and free surfaces are not preferential sites for alpha-prime precipitation; (4) obtained dislocation mobility for iron–chromium; (5) developed a nodal dislocation dynamics model to simulate plastic processes in face-centered-cubic crystals that demonstrated our ability to tread dislocation interactions with complex defects; and (6) connected meso- and micro-scales in a self-consistent way to enable prediction of mechanical property degradation.

Proposed Work for FY08

We expect to (1) finish our short-range order and precipitation studies in iron-rich iron–chromium alloys and determine dislocation interaction with precipitates in iron–chromium alloys with a description to be used in dislocation dynamics codes, (2) determine the dislocation mobility in iron–chromium as a function of short-range order, (3) finish developing a continuum formulation based on Eshelby's theory for elastic inclusions, (4) obtain strength maps as a function of body-centered-cubic defect size and interaction orientation, and

(5) benchmark dislocation dynamics simulations against molecular dynamics simulations and experiments. If time permits, we will study the dislocation density dependence of plastic strain, feed a macromechanical model of flow stress to be used in finite-element method calculations of tensile tests, and compare these simulation results to those of experiments.

Publications

Caro, A., M. Caro, and E. M. Lopasso, 2007. "Implication of ab-initio energetics on the thermodynamics of Fe-Cr alloys." *Appl. Phys. Lett.* **89**, 121902. UCRL-JRNL-225773.

Caro, A., et al., 2007. "The computational modeling of alloys at the atomic scale: from ab initio and thermodynamics to radiation-induced heterogeneous precipitation." *J. Miner. Met. Mater. Soc.* **52**, 8. UCRL-JRNL-227806.

Marian, J., and A. Caro, 2007. "Moving dislocations in disordered alloys: connecting continuum and discrete models with atomistic simulations." *Phys. Rev. B* **74**, 1. UCRL-JRNL-226405.

Fundamental Investigation of Laser-Induced Surface Damage in Optical Materials

Jeffrey D. Bude 06-ERD-035

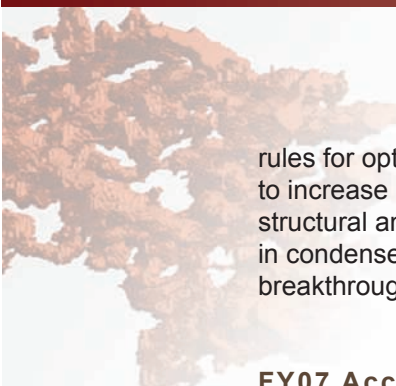
Abstract

The objective of this research is to develop a fundamental understanding of laser-induced surface damage in fused silica. Despite its excellent ultraviolet (UV) transparency, silica undergoes damage under high UV fluence. Although intrinsic bulk damage is relatively well understood, extrinsic surface damage, which is dominant in high quality silica, is poorly understood. This work will develop a suite of high-resolution optical techniques and advanced computational models to help clarify the processes behind surface damage in fused silica.

We expect to (1) identify a reliable experimental signature unique to the defects that absorb sub-bandgap light and lead to damage initiation and growth; (2) use this diagnostic to help identify the physical nature and origin of these defects, focusing on experiments to correlate photoluminescence (PL) signatures to damage susceptibility; and (3) use experiment and simulation to develop a consistent model of the dynamics of damage, including methods to directly measure the rate and location of laser-induced heating in the near-surface region for fluences below the damage threshold, and models and experiments that test the hypothesis of runaway absorption.

Mission Relevance

This work directly addresses stockpile stewardship and laser fusion challenges by optimizing the utilization of large fusion-class laser systems. It will also serve to establish science-based



rules for optics reliability predictions, improve damage diagnostics, and suggest pathways to increase damage resistance in optical materials. By furthering the understanding of structural and electronic transitions in glasses and disordered materials—a frontier problem in condensed-matter physics—this project contributes to the Laboratory’s mission in breakthroughs in fundamental science and technology.

FY07 Accomplishments and Results

In FY07, we (1) completed the setup and calibration of the PL imaging system, (2) imaged a variety of damage sites and mechanically engineered initiation sites, (3) found very short (<100-ps) PL lifetimes, (4) correlated short PL lifetimes with damage measurements, (5) explored the effects of annealing on PL response and damage susceptibility, (6) completed Raman temperature measurements and performed two-color pump-probe absorption measurements on damage sites, (7) added hybrid functional techniques to improve energy accuracy, (8) performed ab initio simulations of high-temperature absorption in silica, (9) performed measurements of temperature-dependent damage thresholds and found evidence of runaway absorption in intrinsic silica, and (10) simulated pulse-length effects on damage size due to runaway absorption.

Proposed Work for FY08

We will (1) perform direct correlations between fast PL lifetimes and damage to test the accuracy of this diagnostic, (2) improve the signal-to-noise ratio in absorption measurements, (3) develop a microdamage test capability in support of the first objective, (4) continue simulations of absorption at high temperature to develop links to defect-absorption models and simulate precursor absorption, (5) provide better temperature estimates for high-temperature damage experiments, and (6) design experiments to isolate the mechanisms behind high-temperature surface conditioning seen in high-temperature damage experiments.

Publications

Bude, J., et al., 2007. *The effect of lattice temperature on surface damage in fused silica optics*. 39th Ann. Symp. Optical Materials for High-Power Lasers, Boulder, CO, Sept. 24–26, 2007. UCRL-PRES-234883.

Trave, A., et al., 2007. *Temperature-induced changes in optical properties of silica*. 2007 APS Spring Mtg., Jacksonville, FL, Apr. 14–17, 2007. UCRL-ABS-226625.

Large-Aperture Optics Performance

Thomas G. Parham 06-ERD-054

Abstract

The performance of large-aperture optics has traditionally limited the maximum output of fusion-class lasers. Prior work has not employed realistic testbeds that allow large-aperture

optics to be studied as they actually will be used in large laser systems. Especially important is the effect of unconverted 1ω and 2ω light on 3ω damage. This project will evaluate large optics at full aperture, with unconverted light present and with the statistical fluctuations that are typical of large-aperture lasers. Specialized optomechanical hardware that edge-illuminates optics will be coupled with extensive laser diagnostics in an existing large-aperture laser system called the Precision Diagnostic System, to allow in situ analysis of damage initiation and growth.

Materials research that enhances our understanding of complex mechanisms involving the response of optical materials to intense laser irradiation will benefit the development of 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 allow operators of advanced laser systems to predict effects of experiments on the lifetime of optics and optimize experiments to minimize costs of replacing optical components. In addition, this project will help 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 of fusion-class lasers systems, and allow us to validate our theoretical and stochastic models. Our research supports the Laboratory's mission in the national security area of stockpile stewardship, because safety and reliability of the nuclear stockpile in the absence of testing will rely heavily on experimental data from fusion-class laser systems to validate complex computer simulations.

FY07 Accomplishments and Results

In FY07, we eliminated fratricide caused by intensification and ejecta from neighboring optics. This was accomplished by (1) improving propagation models, (2) benchmarking the models versus a library of flaws from FY06 data to create a new set of flaw specifications, (3) creating a new optical configuration that positions the final optics to minimize these effects, and (4) developing improved screening for flaws that cause downstream intensification. In addition, new grating-specific damage caused by stimulated Brillouin scattering was observed and eliminated by increasing the main laser bandwidth. The new configuration and flaw specifications enabled proof-of-concept for operations at full fusion-class laser requirements with acceptable damage, including use of a 2ω phase plate. We conducted 130 damage-relevant shots at 3ω and 22 shots at 2ω .

Proposed Work for FY08

Primary research in FY08 will center on issues relevant to the target chamber of fusion-class lasers. A new damage inspection system will be fielded and used to monitor optics performance during commissioning shots at the National Ignition Facility. This will provide new data on optical damage in the presence of debris and target backscatter and allow evaluation of air knives as a means to reduce debris-generated damage. New software tools will be put in place to track this large volume of new data and to predict reasonable replacement sequences for optics as they are damaged.

Publications

Conder, A., et al., 2007. *Final optics damage inspection for the National Ignition Facility*. SPIE Boulder Damage Symp., Boulder, CO, Sept. 24–26, 2007. UCR-ABS-232479.

Understanding Shape Control in Nanoparticle Synthesis

Christine A. Orme 06-LW-090

Abstract

Biologically inspired approaches to materials synthesis hold promise of unprecedented atomic-level control of structure and interfaces of nanoparticles. Use of organic molecules to control the production of inorganic technological materials remains an area with enormous potential. This project combines experimental and theoretical approaches to understand shape control in nanoscale and microscale structures with the goal of determining mechanisms by which ligands direct crystal growth processes at the atomic scale. We will use in situ electrochemical atomic force microscopy and microellipsometry to study the influence of molecules on silver- and copper-oxide growth during electrochemical deposition. This approach directly monitors the transitions between shapes, providing quantitative input to simulations.

This project will provide a capability to systematically investigate organic interactions at metal and metal-oxide surfaces. It enables a more fundamental understanding of the self-assembly process with regards to the very interesting, but predominately empirical, fields of nanoparticle synthesis, electrodeposition, and corrosion. We expect to develop mechanistic models of how particular chemical moieties can be used to influence metal and metal-oxide growth and dissolution.

Mission Relevance

The controlled synthesis of novel, nanostructured technological materials contributes to the development of a scientific platform for sensing and separation technologies in support of national and homeland security, as well as target fabrication that supports stockpile stewardship. Understanding the corrosive effects of organic acids produced by microbes in nuclear waste facilities is important to energy security and environmental management. Finally, understanding how polymeric materials degrade metallic weapons components supports the Laboratory's stockpile stewardship mission.

FY07 Accomplishments and Results

In FY07, we met our milestones by (1) identifying new ways in which common surfactants modify silver nanoparticle growth, (2) quantifying and validating facet-binding mechanisms on silver using microellipsometry, (3) quantifying kinetics of transformation in copper oxide from cubes with 1-0-0 facets to octahedrons terminated with 1-1-1 facets, (4) developing a shape-evolution model based on measured velocity maps, and (5) publishing our results and giving several invited talks. Overall, we have shown that by using in situ tools, we can produce

velocity maps for a variety of additives and growth conditions. The velocity maps, when coupled with simulations, can predict and eventually design specific nanocrystal shapes.

Publications

Gray, J. J., et al., 2007. "In-situ investigation of the silver-CTAB system." *Mater. Res. Soc. Symp. Proc.* **1017E**, 1017-DD12-40. UCRL-CONF-230182.

Gray, J. J., et al., 2007. *Shaping nanoscale crystals*. Gordon Research Conf. Thin Films and Crystal Growth Mechanisms, South Hadley, MA, June 24–29, 2007. UCRL-ABS-232111.

Gray, J. J., et al., 2007. *Understanding shape control during nanoparticle synthesis*. 13th Intl. Summer School on Crystal Growth, Park City, UT, Aug. 5–11, 2007. UCRL-ABS-232112.

Gray, J. J., et al., 2007. *Understanding shape control in nanoparticle synthesis*. 58th Ann. Mtg. Intl. Society of Electrochemistry, Banff, Canada, Sept. 9–14, 2007. UCRL-ABS-228602.

Orme, C. A., and J. L. Giocondi, 2007. "The use of scanning probe microscopy to investigate crystal–fluid interfaces." *Perspectives on Inorganic, Organic and Biological Crystal Growth: From Fundamentals to Application*, vol. 916, pp. 342–362. UCRL-BOOK-230189.

Kinetics of Phase Evolution: Coupling Microstructure with Deformation

James F. Belak 07-ERD-007

Abstract

Confidence in predictions of material behavior at extreme conditions is limited by our lack of knowledge of microstructures and their rate of formation following first-order solidification and solid–solid phase transitions. Our goal is to reliably predict microstructure by extending our highly successful molecular dynamics (MD) techniques for nucleation to hydrodynamic length- and time-scales. Phase and microstructure calculated by MD will serve as initial conditions for our continuum microstructure evolution model, whose dynamics will be validated with MD and recovery experiments. This new phase-field capability will employ an adaptive mesh refinement real-space representation of the time-dependent Ginzburg–Landau equations in a scalable simulation code amenable to LLNL's world-class computing.

We expect to obtain the first reliable prediction of material microstructure following first-order solidification and solid–solid phase transformation under extreme conditions at relevant length and time scales—a prediction observable on emerging x-ray platforms such as the Linac Coherent Light Source. This prediction is directly tied to MD simulations of nucleated microstructure using potentials applicable to metals and alloys of interest. We will create the first phase-field modeling capability scalable to LLNL's high-performance computing resources. By using this microstructure to calculate material behavior, we will increase confidence in calculations at extreme conditions, for which experimental data are either obtained indirectly or entirely lacking.



Mission Relevance

Upon completion, this new capability will be available to provide a realistic prediction of microstructure for relevant alloys under extreme conditions and to directly simulate, for the first time, experiments of first-order phase transformations under highly dynamic conditions such as those observed with the dynamic transmission electron microscope and the JASPER (Joint Actinide Shock Physics Experimental Research) facility. Our new capability will both increase confidence in stockpile calculations, in support of the national security mission, and have applications in the DOE Nuclear Fuel Initiative and the Global Nuclear Energy Partnership, in support of the energy security mission.

FY07 Accomplishments and Results

During FY07, we completed construction of our baseline crystallographic-aware phase-field code. Specifically, we (1) developed a parallel adaptive mesh-phase evolution (AMPE) code based on LLNL's SAMRAI libraries, (2) created a quaternion representation of polycrystal orientation and evolution, (3) validated our coarse-grained quaternion dynamics using molecular dynamics, (4) developed efficient numerical solvers for the quaternion equations and consistent treatment of quaternion symmetries, (5) made quantitative free-energy functionals with the CALPHAD (Computer Coupling of Phase Diagrams and Thermochemistry) approach, (6) initialized our AMPE code with an MD nucleated microstructure, (7) validated the AMPE code with published results, and (8) hired a new postdoctoral researcher.

Proposed Work for FY08

In FY08, we will capture the necessary physics in our phase-field model to validate our AMPE code with concurrent MD simulations of the evolution of the microstructure. To this end, we will implement the physics of elasticity, grain-boundary interfaces, alloy diffusion, and thermal fluctuations. While the form of energy for each of these is fairly well known, the concurrent evolution of multiple-order-parameter fields is not. We anticipate the need to develop and implement new long time-integration and spatial discretization strategies and to extend scalability to large numbers of processors. We will validate our AMPE simulation with concurrent MD propagation of the same order-parameter field.

Publications

Belak, J., et al., 2007. *Molecular dynamics and phase field modeling of pressure-driven solidification*. Minerals, Metals and Materials Soc. 136th Ann. Mtg. and Exhibition, Orlando, FL, Feb. 25–Mar. 1, 2007. UCRL-ABS-223276.

Belak, J., et al., 2006. *The linkage of molecular dynamics to phase field modeling of solidification*. Fall Mtg. of the Materials Research Society, Boston, MA, Nov. 27–Dec. 1, 2006. UCRL-ABS-222330.

Turchi, P. E. A., et al., 2007. "Interface between quantum mechanical-based approaches, experiments, and CALPHAD methodology." *CALPHAD* **31**, 4. UCRL-JRNL-217787.

Wickett, M. E., et al., 2007. *Phase field modeling with adaptive mesh refinement*. UCRL-ABS-228274.

Development of a First-Principles Computational Toolkit for Predicting the Structural, Electronic, and Transport Properties of Semiconductor Radiation-Detection Materials

Vincenzo Lordi 07-ERD-013

Abstract

The nature of carrier dynamics and lifetimes in materials used for gamma-ray detection are largely unknown yet critical for high-performance detection systems. This project's objective is to use first-principles simulations to guide and accelerate the development of new semiconductor materials for room-temperature gamma-ray detectors, by predicting the structural, electronic, and transport properties of candidate materials. For a given material, our computational toolkit evaluates the structures and densities of impurities and defects, then predicts how transport properties are affected by them. This fundamental understanding of the sensitivity of transport properties to material structure and composition will help identify promising materials and evaluate strategies for maximizing their performance.

This project will provide the first detailed, atomic-level, mechanistic description of the nature of electron and hole transport in gamma-ray-detecting semiconductor materials and their dependence on materials defects. This information is not readily accessible by experiment yet will provide tremendous insight for improving material performance. Particularly, for radiation detectors, the concentrations of impurities and defects are generally below the detection limits of experimental techniques. We anticipate that the results of this project's first-principles studies will have a large impact not only on radiation detectors but also on the larger semiconductor physics, in applications ranging from solar cells to microelectronic devices.

Mission Relevance

By furthering the development of gamma-radiation detectors, this project supports LLNL's national security mission area of nonproliferation and the homeland security mission area of detecting weapons of mass destruction.

FY07 Accomplishments and Results

In FY07, we (1) implemented routines using density functional theory and quasiparticle calculations to accurately calculate structures and formation energies for defects and impurities in semiconductor materials; (2) implemented the first version of scattering-rate codes to predict defect-limited transport properties; (3) analyzed the structural and electronic properties of all native defects (vacancies, antisites, and interstitials) in aluminum–antimony, as well as the impurities in carbon, silicon, germanium, tin, phosphorus, oxygen, sulfur, selenium, and tellurium; (4) calculated the relative carrier-scattering strengths of these defects and impurities; (5) compared the calculations to measurement results from aluminum–antimony crystals grown at LLNL; and (6) completed analysis of several dominant native defects in cadmium–tellurium and began analysis of impurities in that material.

Proposed Work for FY08

In FY08, we will (1) complete development of scattering rate codes to accurately predict the effects of defects on carrier transport in semiconductors, (2) implement codes to investigate the phonon-limited transport in these materials, and (3) continue to apply and test our techniques on materials of technological interest, particularly cadmium–zinc–tellurium and gallium–tellurium. We expect the results of our calculations to directly impact experimental materials-optimization efforts. Furthermore, the experience gained from simulations on specific materials will help advance computational materials discovery—the use of automated first-principles simulations to search for optimal materials using input from large structure databases.

Publications

Lordi, V., D. Aberg, and A. J. Williamson, 2007. *First-principles calculation of point defect scattering and carrier transport in bulk semiconductors*. 2007 American Physical Society March Mtg., Denver, CO, Mar. 5–9, 2007. UCRL-PRES-235681.

Lordi, V., et al., 2007. *First principles calculation of point defects and mobility degradation in bulk AlSb for radiation detection application*. 2007 SPIE Optics and Photonics Exhibition, San Diego, CA, Aug. 26–30, 2007. UCRL-PRES-233291.

Lordi, V., et al., 2007. “First principles calculation of point defects and mobility degradation in bulk AlSb for radiation detection application.” *Proc. SPIE* **6706**, 67060O. UCRL-PROC-233251.

Deformation of Low-Symmetry and Multiphase Materials

Nathan R. Barton 07-ERD-024

Abstract

Materials composed of low-symmetry crystals or of multiple solid phases exhibit heterogeneous deformation at the microstructural scale, presenting significant challenges to constructing macroscale constitutive models. We will develop an approach that allows for the explicit incorporation of microstructure and deformational heterogeneity in a framework appropriate to use in analysis of engineering-scale components. In multiphase materials, microstructure influences yield stress, ductility, high- and low-cycle fatigue, creep, and fracture toughness. Given advances in computing resources, constitutive models, and software infrastructure, we are in a unique position to significantly advance constitutive modeling of complex materials.

We are targeting applications involving fully developed plastic flow in which the microstructure is known. Explicit inclusion of the microstructure allows for treatment of deformational heterogeneities at the microstructural scale, and we build on emerging technologies for effectively combining microscale plasticity simulations with macroscale models. The goal is

to homogenize the microscale response to obtain effective macroscale models for materials whose macroscopic behavior is difficult to both characterize and model using conventional approaches. In addition to multiphase alloys, the approach is applicable to low-symmetry crystalline materials. New capabilities will allow for effective assessment of the impact of microstructure on performance of engineering-scale components.

Mission Relevance

In the search for robust and cost-effective systems, there is a need for tools that predict material behavior both during processing and while in service. Such behavior is central to the success of many research and development programs at the Laboratory that rely on multiphase alloys and low-symmetry crystalline materials, particularly those that support both stockpile stewardship and homeland security. Behavior prediction of these materials is also critical to the Responsive Infrastructure Initiative at NNSA and industrial production of a vast range of consumer goods. Finally, it contributes to the Laboratory's mission in advancing basic materials and computational science.

FY07 Accomplishments and Results

In FY07 we (1) assessed important microstructural features and experimental data to aid development of numerical methods for predicting multiphase microstructures, (2) determined a fine-scale implementation strategy and tested the ALE3D fine-scale model, (3) developed capabilities for obtaining steady plastic-flow solutions in ALE3D, (4) introduced new physics into software for coupling coarse and fine scales, (5) developed collaborations with researchers at the Air Force Research Laboratory and Los Alamos National Laboratory, and (6) hired a postdoctoral researcher and graduate-level summer student.

Proposed Work for FY08

Work during FY08 will focus on (1) testing parameterized plastic-flow algorithms and increasing robustness as necessary, (2) developing a technique for extracting fine-scale strain energy data, (3) comparing model predictions against results from appropriate experiments, and (4) demonstrating extension of our method to another low-symmetry or multiphase material such as beryllium or uranium.

Publications

Barton, N. R., et al., 2007. *Adaptive sampling in hierarchical simulation: embedded polycrystal plasticity*. 13th Intl. Symp. Plasticity and Its Current Applications, Alyeska, AK, June 2–6, 2007. UCRL-PRES-231402.

Barton, N. R., et al., 2007. *Efficient embedded multi-scale constitutive modeling*. 5th Intl. Conf. Materials Processing Defects, Ithaca, NY, July 18–20, 2007. UCRL-PRES-232790.

Plasticity at High Pressures and Strain Rates Using Oblique-Impact Isentropic-Compression Experiments

Jeffrey N. Florando 07-ERD-034

Abstract

Various aspects of Lawrence Livermore's national security mission depend on accurate computer code simulations of high-strain-rate plastic flow (i.e., nonreversible deformation) under conditions of high hydrostatic pressures. While progress has been made in recent years there is still an uncertainty in understanding the strength of materials under high strain rate (10^4 – 10^6 s⁻¹) and high pressure (1–100 GPa). We intend to develop an oblique-impact isentropic-compression experiment to measure the strength of materials under these conditions, which will improve the Laboratory's ability to develop predictive strength models for use in computer code simulations.

We will use an oblique-impact, isentropic-compression experiment to subject materials to the appropriate conditions. An understanding of the underlying deformation mechanisms at work under combined high pressure and strain rate will lead to development of more accurate strength models. Initial experimental and computational results have shown that the transverse wave, which is generated by the oblique impact and trails the longitudinal wave, is very sensitive to the strength of the material under pressure. Analysis of this transverse wave can, therefore, be used to assess material strength. In addition, these experiments could potentially be modified to be fielded at large gas-gun or fusion-class laser facilities.

Mission Relevance

The ability to accurately predict the strength of materials under high pressure and dynamic loading conditions (high strain rate) is a main component of the assessment and certification portion of LLNL's stockpile stewardship efforts. This project supports the stockpile stewardship mission by providing previously unobtainable experimental data on materials strength under these conditions.

FY07 Accomplishments and Results

In FY07 we met our major milestones by fielding the oblique-impact isentropic-compression experiment at low pressures. Specifically, we (1) completed sample preparation and diagnostic issues, including fabricating the grating system to measure transverse and longitudinal velocities, and measuring the strength and wave profile of the graded-density impactor; (2) performed simulations to help guide the experimental design; and (3) conducted the first experiments on copper at low pressures at Brown University.

Proposed Work for FY08

In FY08, additional low- and intermediate-pressure experiments (150–300 kbar) on copper, tantalum, and vanadium are planned at Brown University. The results from the tests will give

us the necessary data to begin development and refinement of strength models. Based on these results, designs for a 2-inch graded-density impactor and a soft-recovery experiment will be explored. Simulations will continue to be conducted to aid in the experimental design.

Investigation of the “Double-C” Behavior in the Plutonium–Gallium Time–Temperature Transformation Diagram

Kerri Jayne M. Blobaum 07-ERD-047

Abstract

The goal of this project is to verify and explain the unusual “double C” behavior in the time–temperature transformation (TTT) diagram for the delta-to-alpha-prime transformation of plutonium–gallium alloys. The double-C is an over 30-year-old mystery in plutonium science, and an understanding of this behavior will help connect 5f electronic structure to observed structural phase transformations. Furthermore, the results of this work are essential for modeling and predicting characteristics of the transformation in aging plutonium and under extreme conditions. We will use differential scanning calorimetry, optical microscopy, and x-ray diffraction to confirm the purported double-C curve behavior and to determine the mechanisms responsible for the observed kinetics.

We expect to confirm or disprove that the delta-to-alpha-prime transformation in a plutonium–gallium alloy has double-C curve kinetics. We will provide a comparison of the microstructures and morphologies of the alpha-prime products formed in the upper- and lower-C curves. This work will lead to a kinetic model and explanation of the mechanisms responsible for this unusual behavior. Because many plutonium–gallium research alloys have gallium concentrated in the centers of the grains, we will determine if this inhomogeneity contributes to the double-C curve behavior. This project will lay the foundation for studying the delta-to-alpha-prime phase transformation in aging plutonium and under pressure.

Mission Relevance

This work directly supports the stockpile stewardship mission by forming a basis for modeling plutonium–gallium phase transformations under dynamic conditions and by enhancing the Laboratory’s small-scale plutonium metallurgy capabilities.

FY07 Accomplishments and Results

To produce an alternative TTT diagram with at least three contours of constant time, we collected three sets of data in FY07, one of which suggested a confirmation of the double-C curve behavior at longer times than previously published. Additional experiments resulted in a hypothesis that an ambient-temperature conditioning treatment enables the upper-C curve. Optical microscopy suggests that the morphology of the alpha-prime particles differs depending on the conditioning and that the delta-to-alpha-prime transformation has both athermal and isothermal components. Delta-to-alpha-prime martensitic transformation was shown to apparently have more unusual features than simply the double-C behavior.

Proposed Work for FY08

In FY08, we will (1) continue optical microscopy characterization and initiate x-ray diffraction of alpha-prime formed in the upper- and lower-C curves, (2) compare these morphologies, (3) formulate an experimental design for experiments aimed at elucidating the mechanism responsible for the double-C curve kinetics, and (4) commence experimental work to understand this mechanism.

Publications

Oudot, B., et al., 2007. "Supporting evidence for double-C curve kinetics in the isothermal delta to alpha-prime transformation in a Pu-Ga alloy." *J. Alloy Comp.* **444**, 230. UCRL-JRNL-223221.

Controlling the Structure of a Quantum Solid: Hydrogen

George H. Gilmer 07-ERD-049

Abstract

Achieving inertial-confinement fusion is dependent upon our ability to create solid films of the hydrogen isotopes deuterium and tritium, both quantum solids with high homogeneity and low surface roughness. Recent studies have shown that patterned substrates can be used to direct the orientation, morphology, polymorph, and size of nucleating crystals. The purpose of this project is to explore two approaches, using experiments and atomistic simulations, to control the growth and defect population of these crystals. The first is to develop templates that will provide preferential nucleation sites and set the orientation of the crystalline material. The second is to make the hydrogen lattice amorphous or fine-grained using particle bombardment or rapid cooling.

Hydrogen isotopes provide a challenging but rich system for better understanding of templating mechanisms and crystalline-to-amorphous transitions. For example, the lattice-constant difference between isotopes provides a method to test how templating mechanisms are affected by film stress. Similarly, the different atomic diameters of the isotopes favor amorphization. Isotope molecules can be accurately modeled as computationally simple, point-interacting van der Waals molecules. Simulations by molecular dynamics use pseudopotentials to account for quantum zero-point vibrations. Thus, this project is likely to provide a physical understanding of basic crystal-growth phenomena, templating mechanisms, and defect properties of these quantum solids.

Mission Relevance

This project will couple computational material science to nanoscale research, which supports the Laboratory mission in national security. Scientific findings will aid in developing strategies for ignition targets at future large fusion-class lasers.

FY07 Accomplishments and Results

In FY07, we (1) designed and implemented a cryogenic stage with an optical microscope for the in situ imaging of crystals, (2) grew the first-ever deuterium crystals from vapor onto functionalized diamond substrates, (3) developed pseudopotentials for molecular dynamics simulations of quantum materials on fast parallel computer networks, (4) simulated the crystallization of hydrogen on several templates, (5) observed large template-dependent variations in defect densities and crystalline order, and (6) applied our modeling approach to examine the solidification of amorphous hydrogen and the process of helium bubble nucleation and growth in solid deuterium and tritium.

Proposed Work for FY08

In FY08, we will (1) use modeling and experiments to understand and mitigate surface defects such as grooves and bubbles; (2) combine modeling and experiments to examine stress and cracks in the crystalline hydrogen layer and the relationship to temperature changes; (3) simulate particle-bombardment damage to determine optimum particle flux, energy, and isotope–impurity mix for crystal growth anisotropy reduction and amorphization; (4) develop an alpha radiation system for testing deuterium films on flat substrates; (5) characterize surface damage and its effect on basal plane facets; and (6) simulate and test templating with a deuterium–tritium fuel mixture, which is subject to radiation damage because of tritium beta decay.

Publications


Gilmer, G. H., et al., 2007. *Atomistic modeling of polycrystalline surface structures and growth morphologies: applications to PVD of molecular hydrogen*. 15th Intl. Conf. Crystal Growth, Salt Lake City, UT, Aug. 12–17, 2007. UCRL-PRES-233651.

Fabrication Science for Thick Beryllium Films

Alex V. Hamza 07-ERD-060

Abstract

Design of applications for fusion ignition is at its infancy. The objective of this project is to investigate the fabrication science required for uses of fusion ignition in high-energy-density (HED) physics experiments. We propose two-dimensional hydrodynamic simulations based on existing inertial-confinement fusion ignition designs. We intend to produce low-stress, thick beryllium films on flat substrates to be used for heat shields and tampers in HED experiments. Our goal is to produce a 10- to 50- μm -thick beryllium film on a flat substrate with a film stress less than the yield strength of beryllium (300 MPa). In situ and ex situ film stress measurements will be developed.



We expect to lay the groundwork for fabrication science that will eventually enable use of fusion ignition for HED physics platforms. If successful, we will produce a viable experimental design and define issues affecting materials and fabrication. By producing low-stress, thick beryllium films, we will establish a new field of research, controlling stresses in thick films, with application in thermal barrier coatings for energy generation, corrosion barriers, and HED physics targets. The novel techniques developed for stress measurement will also spur progress in the field of thick-film growth.

Mission Relevance

Advancing the uses of fusion ignition concepts supports Lawrence Livermore's national and energy security missions. In addition, creating the capability to fabricate low-stress, thick beryllium films for heat shields and tampers for HED experiments directly supports the Stockpile Stewardship Program.

FY07 Accomplishments and Results

In FY07, we (1) investigated the two-dimensional implosion characteristics of an HED single-shell target; (2) examined foam doping levels that affect yield and symmetry of integrated implosions; (3) investigated the synthesis of low-stress, thick beryllium films experimentally by coupling an in situ optical stress-monitoring capability to a beryllium growth system on flat substrates; and (4) performed both in situ and ex situ measurements on substrates as a function of lattice geometry, power, temperature, argon pressure, and external energy.

Magnetism in Semiconductor Nanocrystals: New Physics at the Nanoscale

Robert W. Meulenberg **07-LW-041**

Abstract

The novel magnetic effects that we recently discovered in cadmium selenide nanocrystals represent a new field of physics in nanoscience. This project aims to understand how these magnetic properties arise and how they can be manipulated. We will systematically tune particle size and surface functionality and explore their effect on the magnetic properties. In turn, we hope to formulate ways to optimize and control the strength of the magnetic effect. This work will lead to a model for how the surfaces of cadmium selenide nanocrystals influence their properties, and will have applications in a range of sensor applications.

We will look for a systematic relationship between surface termination, particle size, and magnetism in cadmium selenide nanocrystals. Measurements of x-ray magnetic circular dichroism (XMCD) at a synchrotron facility will enable us to identify the chemical source of the magnetism. In addition, more detailed magnetic measurements will provide insight into the strength of magnetism in cadmium selenide samples. These studies represent very significant cutting-edge physics experiments. If initial measurements are confirmed, publication of these results in high-profile journals is very likely.

Mission Relevance

Cadmium selenide nanocrystals have potential applications in chemical sensors and bioassays based on magneto-optical effects. By establishing a knowledge of how the surfaces influence the properties of these particles, this project supports the Laboratory's missions in homeland and national security through development of new sensors.

FY07 Accomplishments and Results

In FY07, we (1) investigated whether the magnetism of cadmium selenide nanocrystals was dependent on surface termination, finding that our ability to induce paramagnetic behavior was not restricted to a specific functionality and therefore concluding that the phenomenon can be extended to ligands that coordinate with cadmium via a nitrogen atom in any aromatic system; (2) confirmed our x-ray absorption spectroscopy measurements with magnetometry measurements; (3) worked to complete an analysis of the magnetometry experiments; and (4) submitted a record of invention, "Magnetization of Undoped CdSe Nanocrystals". In addition, we published, in *Physical Review Letters*, a manuscript detailing the size-dependent shift with particle size in the L3 edge.

Proposed Work for FY08

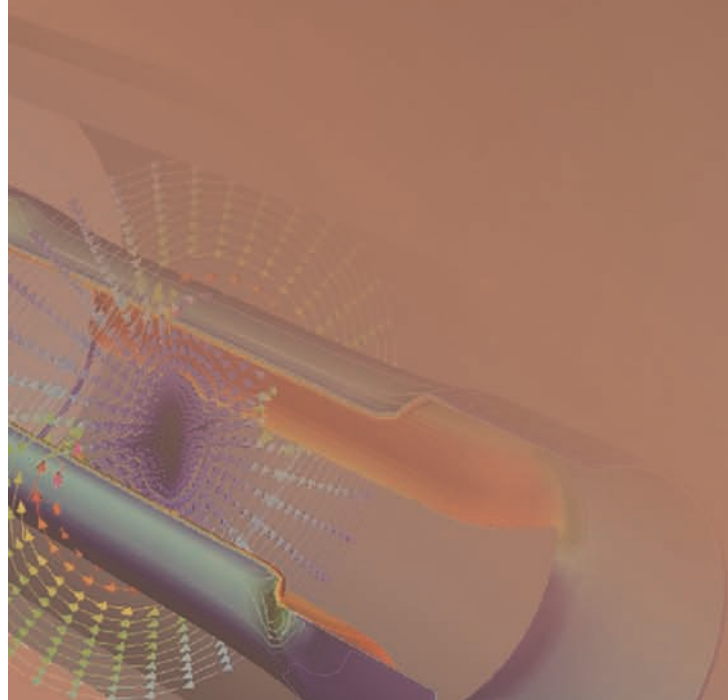
For FY08, we will (1) perform XMCD measurements of cadmium selenide nanoparticles with varying surface termination and particle size using the Advanced Photon Source at Argonne National Laboratory; (2) analyze our data and compile our results for publication; and (3) examine other semiconductor systems (e.g., zinc selenide) to determine whether the effects we discovered are present in all semiconductor quantum-dots systems.

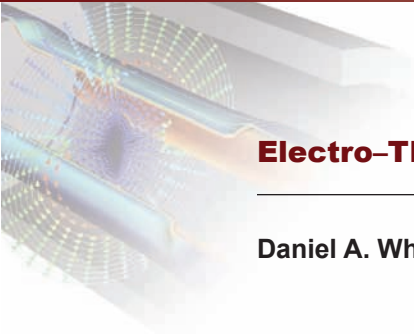
Publications

Lee, J. R. I., et al., 2007. "Experimental observation of quantum confinement in the conduction band of Cd–Se quantum dots." *Phys. Rev. Lett.* **98**, 146803. UCRL-JRNL-226853.

Laboratory Directed Research and Development

Mathematics and Computing Sciences





Electro–Thermal–Mechanical Simulation Capability

Daniel A. White 04-ERD-086

Abstract

For this project, we will research and develop numerical algorithms for three-dimensional (3D) electro–thermal–mechanical (ETM) simulations and incorporate them into Lawrence Livermore’s computational mechanics codes ALE3D and Diablo. A coupled, 3D ETM simulation solves, in a self-consistent manner, the equations of electromagnetics (EM), heat transfer, and nonlinear mechanics. Research will include advection of EM quantities in an ALE setting, algorithms for electrical contact and slide surfaces, and EM 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.

We expect to develop accurate and efficient algorithms for solving coupled ETM problems on 3D unstructured, finite-element meshes. A robust, accurate ETM simulation capability developed in this project will enable Livermore physicists and engineers to better support applications such as explosively driven magnetic-flux compressors, EM launchers, inductive heating and mixing of metals, and microelectromechanical systems.

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 and in chemical and biological detection for counterterrorism, counterproliferation, and homeland security missions. In addition, ETM simulation tools are needed for designing the EM launchers that have been proposed for DOE equation-of-state research and Department of Defense missile defense and artillery systems.

FY07 Accomplishments and Results

In FY07, we (1) developed a primary code enhancement for a realistic resonant circuit model, which is used to model capacitor banks and associated cables and pulse-shaping hardware; (2) developed a second code enhancement by incorporating a novel multigrid solver for the curl–curl equations, which resulted in a solver that is approximately ten times faster for typical problems of interest; and (3) published several conference papers and peer-reviewed journal articles on our results. The success of our research will enable us to continue development of the ETM simulation capability and to investigate novel materials and diagnostics, with support from the Office of Naval Research.

Publications

Rieben, R., et al., 2006. *Lagrangian Eulerian electromechanics in 3D*. Progress in Electromagnetics Research Symp., Cambridge, MA, Mar. 26–29, 2006. UCRL-PRES-219923.

Solberg, J., D. White, and R. A. Rieben, 2006. *A Mortar-based method for zero-conductivity regions in a coupled electro-thermal-mechanical simulation code*. 7th World Congress Computational Mechanics, Los Angeles, CA, July 16–22, 2006. UCRL-ABS-219804.

White, D., 2006. *Coupling magnetic fields and ALE hydrodynamics for 3D simulations of MFCG's*. 2006 IEEE Intl. Conf. Megagauss Magnetic Field Generation, Santa Fe, NM, Nov. 5–10, 2006. UCRL-JRNL-225074.

White, D., and B. Fasebtest, 2007. "Performance of low-rank QR approximation of the finite element Biot–Savart Law." *IEEE Trans. Mag.* **43**, 1485. UCRL-JRNL-225446.

White, D., J. Koning, and R. Rieben, 2007. *Development and application of compatible discretizations of Maxwell's Equations*. Springer Verlag, New York. UCRL-BOOK-212729.

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 analysis approach based on "natural neighbor" techniques to model extreme deformation and failure for analyses of earth penetrator weapons or dam failure. The method could overcome instabilities, insufficient treatment of boundaries, and inefficiencies of other meshless approaches by using a natural neighbor interpolation scheme that will eliminate tensile instabilities and provide improved efficiency. Finite-element approaches are often unsuccessful for these applications because of "mesh tangling." Lagrangian methods allow tracking of particles that makes handling of sophisticated material models and effects from debris and fragmentation more straightforward and natural as compared to Eulerian and ALE-type methods.

We intend to overcome many of the inherent problems associated with meshless methods. This involves the development of several neighbor-based approximation methods, stable time-step calculation, and techniques for improving efficiency. Verification and validation and material modeling will also be a key component of our effort.

Mission Relevance

The proposed approach treats both solid and fluid mechanics and is therefore applicable to several important LLNL national security and homeland security mission areas. Analyses of earth penetration such as for the Robust Nuclear Earth Penetrator and terrorist vulnerability evaluation of infrastructure elements such as dams are possible applications for this new approach. As-built x-ray tomography of laser targets and in vivo magnetic resonance imaging in biomechanics that create "point clouds" would benefit from this meshless method of expediting stress analysis.



FY07 Accomplishments and Results

In FY07, we (1) completed verification and validation of our approach for an extended-range demonstration penetrator after enhancements to our concrete models, and also performed verification and validation for blunt penetrators in reinforced concrete; (2) implemented on-the-fly updates in DYNA3D—a fully production-ready version of meshless DYNA3D is now available; (3) developed more efficient integration method and better shape functions, settled on the most efficient stabilization method, and worked to develop integration correction for patch tests; (4) implemented neighbor-based maximum entropy shape function for better treatment of boundary conditions; and (5) published several journal and conference articles. Overall, we succeeded in developing more robust, accurate, and efficient meshless methods.

Publications

Chen, J. S., W. Hu, and M. A. Puso, 2007. “Orbital HP-clouds for solving Schrödinger equation in quantum mechanics.” *Comput. Meth. Appl. Mech. Eng.* **196**, 3693. UCRL-JRNL-225453.

Puso, M. A., 2007. “An improved linear tetrahedral element for plasticity.” *Proc. 2007 ASME Applied Mechanics and Materials Conf.* UCRL-CONF-211697.

Puso, M. A., and J. Solberg, 2007. “A stabilized nodally integrated tetrahedral.” *Intl. J. Numer. Meth. Eng.* **67**, 841. UCRL-JRNL-212610.

Puso, M. A., E. Zywicz, and J. S. Chen, 2007. “A new stabilized nodal integration approach.” *Meshfree Methods for Partial Differential Equations III*. Springer, Berlin, vol. 57, pp. 207–217. UCRL-PROC-218895.

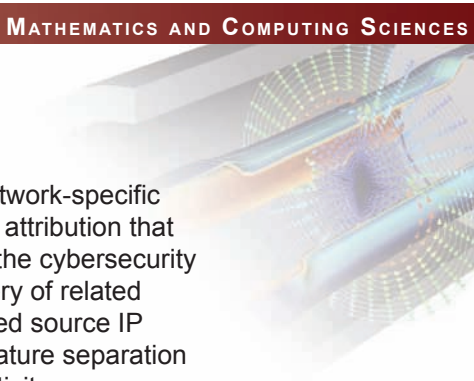
Puso, M. A., et al., in press. “Meshfree and finite element nodal integration methods.” *Int. J. Numer. Meth. Eng.* UCRL-JRNL-229168.

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 timing characteristics imposed unwittingly by the attacker’s choice of platform, software, algorithm settings, and network locations, with focus upon hostile scan activity. Most network security efforts require adversary identification and characterization but rely primarily upon easily spoofed or illicit source IP addresses. This research seeks to supplant IP address with other evidence that is characteristic of the adversary. We will measure the influence upon packet arrival patterns because of varied attack software, platform performance, and intervening network conditions by employing controlled experiments with reference platforms. Mathematical characterization will employ wavelets.



The project serves to separate adversary-specific traffic signatures from network-specific traffic signatures and 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. We will identify a precise regimen of measures well suited to feature separation and detection, resulting in improved detection of distributed surveillance activity, more-accurate attribution of distributed denial-of-service attacks, and directions for further research on improving underlying metrics.

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.

FY07 Accomplishments and Results

In FY07, we completed and characterized 700 class B network scans, employing three contemporary scan tools with parameter variation, involving both non-routed and Internet routed pathways. Each scan was analyzed using 110 separate datamode–wavelet signature combinations and ranked for effectiveness in separability of the known tool, parameter, and location groupings. In summary, this project demonstrated significant ability to distinguish tools and parameters in the presence of packet-routing unknowns and to distinguish scan routing given accurate tool and parameter identification. Collaboration with University of California at Davis produced an analytic tool employing these techniques to enhance hostile scan discrimination and providing a foundation for behavior-based identification of adversary activity.

Publications

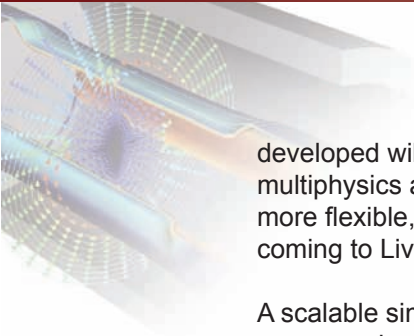
Bartoletti, T., et al., 2007. *Intelligent classification and visualization of network scans*. 13th Intl. Conf. Knowledge Discovery and Data Mining, San Jose, CA, Aug. 12–15, 2007. UCRL-CONF-228878.

Petascale Simulation Initiative

John M. May **04-ERD-102**

Abstract

This project will dramatically increase Lawrence Livermore's capability in scalable multiphysics and multiscale simulations for stockpile stewardship and other applications. Our focus will be the development of algorithms and software for efficient coupling of multiphysics calculations in a multiple program, multiple data (MPMD) environment with massively parallel computer clusters, and on adaptive algorithms for multiscale modeling. The architecture



developed will facilitate the coupling of existing, independently written codes and lead to new multiphysics and analysis capabilities. Advances to the programming environment will provide more flexible, and potentially much more efficient, use of massively parallel computers coming to Livermore. Adaptive sampling will enable larger and faster multiscale simulations.

A scalable simulation component architecture implementing our cooperative parallel (Coop) programming model will be developed and implemented for multiphysics codes, and the core capabilities will be demonstrated with a full-scale simulation on a high-performance computer. 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 for 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 missions that require the multiphysics, multiscale synthesis of existing models. It will facilitate the coupling of existing codes, thereby shortening development time for complex applications. MPMD capabilities exercised in the demonstration problem will help achieve greater machine efficiency, and adaptive sampling applied to multiscale material modeling will be useful for many applications requiring subscale physics. Our model greatly simplifies the process of orchestrating multiple instances of a simulation running concurrently.

FY07 Accomplishments and Results

In FY07, we (1) finished a prototype of the Coop runtime system; (2) implemented nonblocking Remote Method Invocation; (3) studied the tradeoff between efficiency and accuracy in adaptive sampling; (4) demonstrated an approximate 40-fold increase in speed of a material science multiscale simulation, with no significant loss of accuracy; (5) demonstrated a parameter study application using Coop; and (6) submitted several publications on cooperative parallelism and adaptive sampling. Overall, we developed, implemented, and demonstrated a new MPMD programming model for large-scale parallel applications. We showed its value in two different application areas, and intend to continue our work with users to adopt this model.

Publications

Ashby, S., and J. May, 2007. "Multiphysics simulations and petascale computing." *Petascale Computing: Algorithms and Applications*. D. Bader (Ed.), Chapman and Hall/CRC Press, London, 2007. UCRL-BOOK-227348.

Barton, N., et al., in press. "Embedded polycrystal plasticity and in situ adaptive tabulation." *Intl. J. Plasticity*. UCRL-JRNL-226715.

Kumfert, G., J. Leek, and T. Epperly, 2007. *Babel remote method invocation*. 21st IEEE Intl. Parallel and Distributed Processing Symp., Long Beach, CA, Mar. 26–30, 2007. UCRL-CONF-220411.

Catalyzing the Adoption of Software Components

Thomas Epperly 05-ERD-012

Abstract

Software component technology has redefined state-of-the-art software engineering practices in industry, enabling codes to be larger and more modular, scalable, robust, and flexible, all of which similarly benefit Lawrence Livermore computation efforts. The purpose of this project is to develop the technology to automate componentization of existing computer source code. This technology will allow code developers to define unique software components according to existing functions in the code, and to visualize and manually tailor the componentization process. By providing a lower-cost path towards componentization, we will improve the productivity of code developers and help Livermore maintain the lead in delivering large advanced simulation codes in the pursuit of science.

This project will provide (1) technology to automate construction of components by automatically partitioning existing functions into components and generating for Babel application interfaces, (2) multiple alternative componentizations, (3) ability to visualize large-scale applications and their decomposition into components, (4) means to manually tailor componentization, (5) metrics to evaluate interface suitability, (6) static analysis to determine correct use 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—including stockpile stewardship, information processing, and chemical and biological security—depend on scientific computation to further science in the national interest. This project supports LLNL missions by providing a concrete approach to the development of components within existing Laboratory applications. This work will make development and maintenance of large-scale applications less expensive and more robust and flexible.

FY07 Accomplishments and Results

In FY07 we (1) made our software analysis and visualization technology available to a new software security project; (2) researched, implemented, and tested several feature-based and topology-based clustering algorithms to discover the proper grouping of functions as components; (3) created state-of-the-art program visualizations to display software metrics and clustering results; and (4) prepared conference papers on our results. Our prototype automatic components-generation technology produces component interface definitions in the Scientific Interface Definition Language. Pattern recognition technology we developed has been successfully used at Livermore to locate specific programming constructs in source code.



Publications

Panas, T., et al. 2007. *Communicating software architecture using a unified single-view visualization*. 12th IEEE Intl. Conf. Engineering Complex Computer Systems (ICECCS 2007), Auckland, New Zealand, July 10–14, 2007. UCRL-CONF-227293.

Panas, T., D. J. Quinlan, and R. Vuduc, 2007. *Visualization of program architecture using whole-program analysis*. 1st Intl. Workshop Aerospace Software Engineering (AeroSE 07) in conjunction with the 29th Intl. Conf. Software Engineering (ICSE 2007), Minneapolis, MN, May 21–22, 2007. UCRL-PROC-231453.

Locality-Optimizing Caching Algorithms and Layouts

Peter G. Lindstrom 05-ERD-018

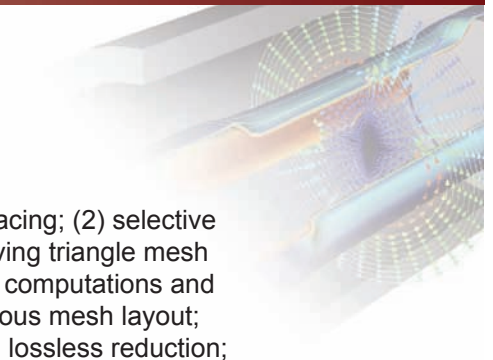
Abstract

This project proposes to investigate layout and compression techniques for large, unstructured computer simulation data to reduce bandwidth requirements and latency for visualization and analysis. Our goal is to eliminate the data-transfer bottleneck—for example, from disk to memory and from the central processing unit to the graphics processing unit—through cache-coherent data access and by trading underutilized computer power for bandwidth and storage. We are achieving this by designing algorithms that both enforce and exploit compactness and locality in unstructured data and by adapting offline computations to a novel stream-processing framework that supports pipelining of processes and low-latency sequential access to compressed data. The scalable algorithms developed in this project will run on both end-user desktops and dedicated visualization clusters.

We expect to achieve significant improvements in computer 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 computers as the gap in processor speed and input/output performance keeps widening.

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 on high-performance computers for the stockpile stewardship mission.



FY07 Accomplishments and Results

In FY07, we developed (1) cache-coherent layouts of hierarchies for ray tracing; (2) selective spectral compression of two-dimensional scalar fields; (3) an order-preserving triangle mesh compression that supports random access, with 20:1 reduction in required computations and 6:1 input/output speedup; (4) a multilevel ordering scheme for cache-oblivious mesh layout; (5) streaming hexahedral mesh connectivity compression, with up to 200:1 lossless reduction; (6) streaming, lossless compression of unstructured point sets that is more than two orders of magnitude faster than the current state of the art; (7) mesh partitioning via cache-oblivious layouts that supports instant repartitioning and load balancing; and (8) a new space-filling curve with higher locality than the conventional continuous fractal space-filling curve.

Proposed Work for FY08

In FY08, we will (1) design a compression scheme for coding fields and geometry in hexahedral meshes, (2) parallelize implementation of cache-oblivious layout and mesh partitioning for high scalability and potential replacement of the ParMETIS parallel library for graph partitioning, and (3) develop representations and techniques for distributed streaming on parallel machines by exploiting both task and data parallelism.

Publications

Callahan, S. P., et al., 2007. "Progressive volume rendering of large unstructured grids." *IEEE Trans. Visual. Comput. Graph.* **12**, 1307. UCRL-PROC-208680.

Ibarria, L., P. Lindstrom, and J. Rossignac, in press. "Spectral interpolation on 3×3 stencils for prediction and compression." *J. Comput.* UCRL-JRNL-232201.

Ibarria, L., P. Lindstrom, and J. Rossignac, 2007. "Spectral predictors." *Proc. IEEE Data Compression Conf. 2007*, pp. 163–172. UCRL-CONF-226261.

Lindstrom, P., and M. Isenburg, 2007. "Fast and efficient compression of floating-point data." *IEEE Trans. Visual. Comput. Graph.* **12**, 1245. UCRL-JRNL-220406.

Pascucci, V., et al., 2007. "Robust on-line computation of Reeb graphs: simplicity and speed." *ACM Trans. Graph.* **26**, 58. UCRL-JRNL-218501.

Vo, H., et al., 2007. "Streaming simplification of tetrahedral meshes." *IEEE Trans. Visual. Comput. Graph.* **13**, 145. UCRL-JRNL-208710.

Yoon, S.-E., and P. Lindstrom, 2007. "Mesh layouts for block-based caches." *IEEE Trans. Visual. Comput. Graph.* **12**, 1213. UCRL-JRNL-220368.

Yoon, S.-E., and P. Lindstrom, in press. "Random-accessible compressed triangle meshes." *IEEE Trans. Visual. Comput. Graph.* UCRL-JRNL-229656.

Yoon, S.-E., C. Lauterbach, and D. Manocha, 2007. "R-LODs: Fast LOD-based ray tracing of massive models." *Vis. Comput.* **22**, 772. UCRL-JRNL-219092.



A New Method for Wave Propagation in Elastic Media

Anders Petersson 05-ERD-079

Abstract

Simulation of elastic wave propagation is essential for many Lawrence Livermore applications, including monitoring underground nuclear explosions and other seismic events and the nondestructive evaluation of complex parts. We propose to develop significant improvements of the traditional finite difference technique that allow a fully second-order-accurate treatment of boundary conditions in complex domains to handle stress-free boundary conditions on topography, and use local mesh refinement to mitigate over-sampling 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-dimensional (2D) and 3D media. This open-source code will be used for many applications at LLNL and in the scientific community. The software will support applications ranging from 2D simulations in nondestructive testing to 3D earthquake modeling using computer power ranging from desktop workstations (e.g., SUN, LINUX, and OSX platforms) to massively parallel high-performance machines (e.g., Zeus, Thunder, and Frost). We plan to validate the code against benchmark problems relevant to LLNL applications and 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 Livermore's national security missions. This includes strong ground-motion prediction for the Enhanced Test Site Readiness Program and the Yucca Mountain Program, nuclear explosion monitoring and underground facilities characterization, and nondestructive testing for locating imperfections in critical components relevant to the Transformational Materials Initiative.

FY07 Accomplishments and Results

In FY07 we (1) completed theoretical development of the embedded boundary technique for enforcing stress-free boundary conditions on voids and topography with complex shapes, (2) developed patch-based local mesh refinement to allow the mesh size to approximately follow the velocity structure in the computational domain, (3) developed and implemented local mesh refinement into the Wave Propagation Project (WPP) code, (4) validated simulations with free-surface topography against a spectral element method code (SPECFEM2D), and (5) ported the WPP code to the Macintosh OS X platform to broaden the user base in the seismic modeling community.

Proposed Work for FY08

In FY08 we will enhance the WPP code by implementing (1) an embedded boundary technique for enforcing stress-free boundary conditions on topography with complex shape,

(2) a smoothing technique for removing unresolvable details in a realistic topography, (3) anelastic damping for modeling motion attenuation in dissipative materials, and (4) an improved far-field boundary condition that is stable for materials with a high ratio between compressional and shear velocities.

Publications

Appelo, D., et al., 2007. *User's guide to the Wave Propagation Program (WPP) version 1.0*. UCRL-SM-230257.

Nilsson, S., et al., 2007. "Stable difference approximations for the elastic wave equation in second-order formulation." *SIAM J. Numer. Anal.* **45**, 1902. UCRL-JRNL-222276.

Petersson, N. A., et al., 2006. *Large-scale simulations of the great 1906 San Francisco earthquake*. American Geophysical Union Fall Mtg., San Francisco, CA, Dec. 11–16, 2006. UCRL-POST-226623.

Efficient and Reliable Data Exploration via Multiscale Morse Analysis and Combinatorial Information Visualization

Valerio Pascucci 05-ERI-002

Abstract

We propose to develop 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 will use 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 Web security graphs and counterproliferation semantic graphs. The environment will use linked views to show the graphs with context information that improves the overall data understanding.

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 multiresolution 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 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 terascale scientific data of the type generated by LLNL's stockpile stewardship program, such as large-scale simulations of hydrodynamic instabilities in shocked materials or material defect simulations with billions of atoms. We target the needs of scientists to explore such datasets with confidence that important features are not overlooked.

FY07 Accomplishments and Results

For FY07, we (1) developed a complete system for two-dimensional and three-dimensional topology computations with simplification of selected topological features and quantification of metrics of interest, (2) developed methods for space-time multiscale representation of scientific data and performed extensive scaling tests for practical usage in large data sets, and (3) worked with application scientists to demonstrate the effectiveness of our approach in the quantitative analysis and comparison of complex topological features from real-world data. The results of our research have been published extensively in refereed publications and we have received the Institute of Electrical and Electronics Engineers Visualization 2006 award for best application paper.

Publications

Bethel, E. W., et al., 2007. "SciDAC visualization and analytics center for enabling technology." *J. Phys. Conf. Ser.* **78**, 012032. UCRL-JRNL-218501.

Bremer, P.-T., et al., 2007. "Topological feature extraction and tracking." *J. Phys. Conf. Ser.* **78**, 012007. UCRL-CONF-232179.

Callahan, S. P., et al., 2006. "Progressive volume rendering of large unstructured grids." *IEEE Trans. Visual. Comput. Graph.* **12**(5). UCRL-JRNL-214075.

Dillard, S. E., et al., 2007. "Tessellation of quadratic elements." *Algorithms and Computation, 17th Intl Symp. Proc.*, pp. 722–731. UCRL-BOOK-218501.

Laney, D., et al., 2007. "Understanding the structure of the turbulent mixing layer in hydrodynamic instabilities." *IEEE Trans. Visual. Comput. Graph.* **12**, 1053. UCRL-JRNL-218500.

Pascucci, V., et al., 2007. "Streaming computation of Reeb graphs: simplicity and speed." *ACM Trans. Graph.* **26**(3), 58.1. UCRL-JRNL-218501.

Predictive Knowledge Systems for Large Complex Data Sources

James M. Brase 06-SI-006

Abstract

Nonproliferation, counterterrorism, and intelligence are primarily problems of information: Sensors and data-collection systems can provide overwhelming quantities of data. Moreover, these data are often sparse, noisy, irrelevant, disjointed, and even intentionally misleading. The objective of this project is to discover complex patterns in large-scale multisource data streams and to build predictive models based on these patterns. We will create algorithms and computations capabilities that will allow analysts to extract knowledge from such data in a meaningful and timely way. To this end, we will pursue pattern discovery, learning and prediction, and data-intensive computational architectures. The project will focus on demonstrating applications in nonproliferation and homeland security.

We expect to develop (1) a technical base of algorithms and computational methods that will extend the frontiers of pattern recognition and stochastic predictive models, (2) confidence measures and performance metrics relevant to these tools, and (3) demonstrations that apply these capabilities to important problems of nonproliferation and intelligence.

Mission Relevance

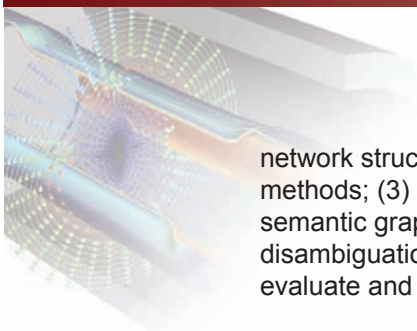
This project will enable accurate and relevant conclusions from a mix of complex data sources (e.g., imagery, communications, and text) with unprecedented levels of performance in broad-area search and site-monitoring problems relevant to nonproliferation and intelligence applications. The central core of algorithms and computational methods developed will broadly support the LLNL mission in national security, particularly nonproliferation and homeland security.

FY07 Accomplishments and Results

In FY07, we (1) completed the first phase of a network-based process simulator and transferred data sets to analysis teams; (2) completed the scenario ontology and loaded data; (3) produced initial disambiguation results; (4) developed, implemented, and published a “random tree model” search algorithm; (5) demonstrated a 100-fold reduction in time required for semantic graph searches; (6) demonstrated a new “relational random forest” node and link classifier; (7) demonstrated a new hierarchical, mixed-membership model for unsupervised group-structure discovery; (8) achieved initial-state estimation results using a sequential Monte Carlo simulation; and (9) established collaborations in role discovery with the University of Maryland and in dynamic point processes with the University of California at Santa Cruz.

Proposed Work for FY08

For FY08, we will (1) demonstrate detection of coherent spatio-temporal patterns in persistent imagery and communications data streams and analyze the detection performance limits in applications; (2) develop and analyze methods for reconstructing communication



network structure from observed event data and characterize performance limits of the methods; (3) extend algorithms for learning node, link, and subgraph labels in dynamic semantic graphs and quantify performance in controlled data sets; and (4) apply text disambiguation algorithms to large data sets from the Biodefense Knowledge Center and evaluate and document performance.

Publications

Gallagher, B., 2006. *Matching structure and semantics: a survey on graph-based pattern matching*. Association for the Advancement of Artificial Intelligence 2006 Fall Symp. Capturing and Using Patterns for Evidence Detection, Washington, DC, Oct. 13–15, 2006. UCRL-CONF-220852.

Manay, S., and D. W. Paglieroni, 2007. *Matching flexible polygons to fields of corners extracted from images*. IEEE Computer Society Conf. Computer Vision and Pattern Recognition, Minneapolis, MN, June 18–23, 2007. UCRL-CONF-226354.

Ng, B. M., 2007. *Adaptive dynamic Bayesian networks*. 2007 Joint Statistical Mtg., Salt Lake City, UT, July 29–Aug. 2, 2007. UCRL-PROC-235983.

Paglieroni, D. W., and F. Nekoogar, 2007. *Matching random tree models of spatio-temporal patterns to tables or graphs*. IEEE Symp. Computational Intelligence and Data Mining, Honolulu, HI, Apr. 1–5, 2007. UCRL-CONF-225685.

Pfeffer, A., et al., 2007. *Global/local dynamic models*. Intl. Joint Conf. Artificial Intelligence, Hyderabad, India, Jan. 6–12, 2007. UCRL-CONF-225258.

Roberts, D., and T. Eliassi-Rad, 2006. *A position paper: value of information for evidence detection*. Association for the Advancement of Artificial Intelligence 2006 Fall Symp. Capturing and Using Patterns for Evidence Detection, Washington, DC, Oct. 13–15, 2006. UCRL-CONF-223973.

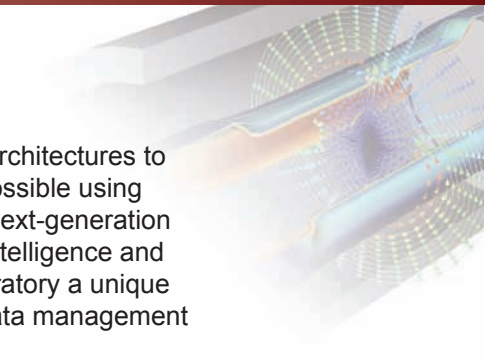
Tong, H., et al., 2007. *Fast best-effort pattern matching in large attributed graphs*. 13th ACM SIGKDD Intl. Conf. Knowledge Discovery and Data Mining, San Jose, CA, Aug. 12–15, 2007. UCRL-CONF-231426.

Scalable Data Management for Massive Semantic Graphs

Scott R. Kohn 06-ERD-009

Abstract

Semantic graphs are an important tool for the intelligence community and homeland security applications. However, current approaches do not address anticipated data sizes. We are investigating the scaling properties of semantic graphs using parallel databases running on multiple central processing units with storage arranged in an active disk architecture. We believe that such approaches can support semantic graphs that are at least 100 times larger than currently possible. Our research focuses on the algorithms, architectures, and techniques necessary to support these massive distributed semantic graphs.



The goal of this research is to understand how to use parallel active disk architectures to support semantic graphs that are orders of magnitude larger than those possible using current technology. We expect this research to guide the development of next-generation semantic graph architectures of a size and scale currently unmatched in intelligence and homeland security applications. These architectures would bring the Laboratory a unique analysis capability and help develop expertise in the area of large-scale data management and analysis.

Mission Relevance

Increasingly massive data sets collected for nonproliferation, intelligence, and military missions require new technologies to query and analyze the data. By furthering understanding of the performance characteristics of a cluster architecture on data-intensive semantic graph applications, this project supports the large-scale data management and analysis needs of the Laboratory's homeland and national security missions.

FY07 Accomplishments and Results

In FY07 we (1) created and searched a semantic graph with 290 billion edges and 11 billion vertices—the largest search to date by an order of magnitude, (2) studied the ingest and query characteristics of a parallel active-disk architecture and achieved data ingest performance 50 times faster than existing semantic graph systems, (3) compared a variety of out-of-core storage methods and developed both a new approach that was between two and seven times faster and new clustering and blocking algorithms that increased search performance by a factor of three, and (4) developed a high-performance semantic graph query language that is significantly faster than existing languages.

Proposed Work for FY08

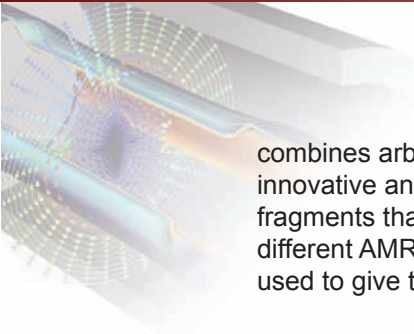
In FY08, we will (1) develop new algorithms for out-of-core semantic graph pattern matching—one of the most common analysis queries for semantic graphs—with a focus on optimizing the execution order of pattern matching to reduce query time, (2) add pattern-matching capabilities to our graph query language, and (3) continue to evaluate active-disk-style architectures, vertical databases, and other alternative out-of-core technologies such as nonvolatile memory and solid-state disks for their use in massive semantic graphs.

A Predictive Model of Fragmentation Using Adaptive Mesh Refinement and Hierarchical Material Modeling

Alice E. Koniges 06-ERD-036

Abstract

Fragmentation is a fundamental process that naturally spans microscopic to macroscopic scales and is important in many applications. Recent advances in algorithms and computer power will enable us to design and implement new algorithms for fragmentation modeling based on a parallel, multiscale adaptive-mesh-refinement (AMR) framework. The method



combines arbitrary Lagrangian–Eulerian (ALE) simulation methods with AMR and an innovative and hierarchical material model that allows voids to grow and coalesce, forming fragments that can be tracked. The model also allows for inclusion of different models on different AMR levels. Dedicated experiments will benchmark these simulations, which will be used to give the first-ever predictions of fragmentation of high-powered laser targets.

This project will bolster the use of the structured AMR application infrastructure (SAMRAI) library for multiscale simulations. Design decisions relating to fusion-class lasers rely on accurate prediction of fragmentation in the target chamber. This research will develop the first computationally based models for the fragmentation process that can be directly applied to fusion-class laser design issues. The predictive model developed with this project, along with the experimental component, are expected to yield an increased understanding of fragmentation, which will have broad impact, from fusion-class lasers to weapons systems to space shuttle re-entry vehicles.

Mission Relevance

This research has direct relevance to national security as well as to breakthroughs in fundamental science and technology. Benefits include significant enhancement and broadening of our expertise in computational solid mechanics and advancement of the application base for our AMR framework. A significantly improved capability to predict fragmentation will find application in fusion-class laser experiments in support of the stockpile stewardship mission.

FY07 Accomplishments and Results

In FY07, we (1) implemented anisotropic strength in the ALE–AMR code, (2) completed fluid multimaterial interface reconstruction in two and three dimensions (onion skin) and completed extensions to material history, (3) connected our simulations to a materials library at Lawrence Livermore, (4) began benchmarking and mixed-cell formulation, (5) began constructing additional models for brittle failure, (6) continued implementation of the laser deposition package, and (7) implemented three-dimensional onion-skin and single-crystal models.

Proposed Work for FY08

For FY08, we will (1) continue and complete our diffusion package; (2) complete implementation of laser deposition; (3) use large-scale simulations in a parallel environment to benchmark the computational strategies, including the hierarchical material modeling where useful; (4) make comparisons with experimental data to assess the effectiveness of hierarchical material modeling and scaling of failure modeling; and (5) investigate the extension of this work to other problems in multiscale modeling.

Publications

Fisher, A., et al., 2007. *Hierarchical multi-scale modeling of fragmentation in materials*. 9th U.S. National Congress Computational Mechanics, San Francisco, CA, July 23–26, 2007. UCRL-PRES-232971.

Koniges, A., et al., in press. "Development strategies for modern predictive simulation codes." *Parallel Computing: Architectures, Algorithms and Applications. Proc. Intl. Conf. ParCo 2007*. UCRL-PROC-235391.

Decomposition of Large-Scale Semantic Graphs via an Efficient Communities Algorithm

Yiming Yao 06-ERD-038

Abstract

Semantic graphs have become key components in analyzing complex systems such as the Web and social networks. These types of graphs generally consist of sparsely connected clusters ("communities") whose nodes are more densely connected to each other than to other nodes in the graph. The identification of communities is invaluable in the analysis of large graphs by producing subgraphs of related data whose interrelationships can be readily characterized. A new challenge is to decompose these massive graphs into meaningful communities for effective analysis. We are addressing this issue by developing more efficient algorithms for discerning community structure that can effectively process massive graphs.

Current algorithms for detecting community structure are only capable of processing relatively small graphs with up to ten thousand nodes. Our goal for this project is to develop methodologies and corresponding algorithms capable of effectively processing graphs with up to 1 billion nodes. From a practical standpoint, we expect the developed scalable algorithms to help resolve a variety of operational issues associated with the productive use of semantic graphs at LLNL.

Mission Relevance

Algorithms to be developed in this project will lead to effective knowledge-discovery systems for the Laboratory's national security mission to counter the proliferation and use of weapons of mass destruction and will support the homeland security mission by enabling more effective knowledge extraction and inference activities. In addition, by making contributions to knowledge of graphing theoretic algorithms, the project also supports the Laboratory's mission in breakthroughs in fundamental science and technology.

FY07 Accomplishments and Results

In FY07, we (1) completed a clustering algorithm based on dynamic graph transformation and achieved significant efficiency gains over current, high-quality algorithms; (2) performed test runs that have shown computation time reductions of up to 89%; (3) implemented parallel graph clustering on a supercomputer; (4) exploited the fact that large graphs tend to be sparse, comprising loosely connected node clusters, to achieve efficiency in parallelism—our implementation performed well on several large graphs with up to one billion nodes; and (5) developed a verification model for decomposition algorithms, based upon a novel integer linear programming approach that maximizes modularity—this tool has found improved community structure in several well-studied graphs in the literature.



Publications

Yao, Y., et al., 2007. "Graph clustering evaluation via integer linear programming." *Proc. 2007 Intl. Conf. Data Mining*, p. 369. UCRL-PROC-228037.

A Novel Structure-Driven Approach to Sequence Pattern Definition for Remote Homology Detection

Carol E. Zhou 06-ERD-059

Abstract

We are developing, testing, and demonstrating an algorithm for detecting remote protein homology—an important need in protein-structure modeling, functional assignment, and the rational design of diagnostics, therapeutics, and vaccines. Current methods of homolog detection and fold assignment lack sufficient sensitivity to enable the structural modeling of a wide range of proteins of interest to biodefense applications. Our algorithm will improve sensitivity by translating key structural information into sequence patterns, which will be used to assign distantly homologous proteins to structural families and will enable the efficient structure-driven identification of domain fusion proteins. We will use laboratory methods to validate our predictions and refine the algorithm.

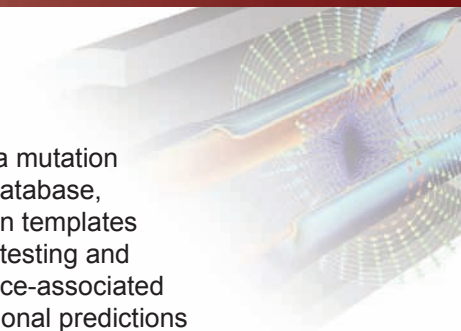
We will devise an algorithm for automatically generating sequence patterns that embody essential protein-structure information. This achievement will immediately impact the characterization of virulence proteins from one of the most important biothreat agents: *Yersinia pestis*, which causes the plague. A longer-term application of our algorithm is the three-dimensional modeling and characterization of proteins from all pathogens of interest in biodefense. Most significantly, this could lead to more effective signatures for identification of biothreat agents (including toxins), and would yield a tool for elucidating pathogen protein function and hence, mechanisms of fitness and pathogenicity. The algorithm also will be of general use for proteins that could not otherwise be characterized.

Mission Relevance

Our work supports Lawrence Livermore's mission in national security by establishing a computational capability with biodefense applications in (1) detecting and characterizing virulence proteins, (2) constructing a pathogen protein phylogeny, (3) analyzing sequence variability in rapidly evolving virus genomes, and (4) forming the rational design of diagnostics, therapeutics, and vaccines.

FY07 Accomplishments and Results

In FY07, we (1) developed an algorithm for identifying structurally conserved regions and corresponding sequential fragments in related proteins, (2) developed an algorithm for



structure-based searches for domain fusion proteins, (3) began developing a mutation mechanism for detecting remote homologs in the nonredundant sequence database, (4) wrote code to automate the process of associating putative domain fusion templates across sets of proteins within a given protein-interaction network, (5) began testing and applying this code to analyze proteins from selected sets of *Y. pestis* virulence-associated proteins, and (6) made steps toward experimentally validating our computational predictions by cloning 147 *Y. pestis* virulence-factor genes—of which 32 were expressed—and grew crystals of two factors: pesticin and ypo0408.

Proposed Work for FY08

In FY08 we will (1) develop an algorithm for pattern-based homology detection; (2) develop an algorithm for identification of interacting partners (domains) using sequence-pattern-based searches; (3) apply developed algorithms to the selected set of *Y. pestis* virulence-associated proteins, such as from an identified network associated with a virulence mechanism known as quorum sensing; (4) identify links between interacting partners from the quorum-sensing network using novel pattern-based domain-fusion search algorithm; and (5) validate computational predictions by performing experimental interaction studies using gel-shift assays and by verifying functional assignments using enzyme assays.

Publications

Chakicherla, A., et al., 2007. *Function prediction of an interacting protein pair using domain-fusion analysis: SpaR and SpaK*. 11th Ann. Intl. Conf. Research in Computational Biology (RECOMB 2007), Oakland, CA, Apr. 21–25, 2007. UCRL-POST-229464.

Zemla, A., and C. E. Zhou, in press. “Structural re-alignment in an immunogenic surface region of ricin A chain.” *Bioinformatics Biol*. UCRL-JRNL-233290.

Zemla, A., et al., 2007. *Predicting structure and function for novel proteins of the iron oxidizing bacterium, leptospirillum group II, from a natural acidophilic community*. JGI Users Mtg., Walnut Creek, CA, Mar. 28–30, 2007. UCRL-ABS-228552.

Zemla, A., et al., in press. “STRALCP—structure alignment-based clustering of proteins.” *Nucleic Acids Res*. UCRL-JRNL-220404.

Zemla, A., et al., 2006. *STRALCP—structure alignment-based clustering of proteins and its application to the detection of conserved regions in proteins*. 2006 Department of Homeland Security Conf. UCRL-POST-224222.

Zemla, A., et al., 2006. *Structure-driven domain fusion analysis and its application to the detection of functional homologues*. 2006 Department of Homeland Security Conf. UCRL-POST-224844.



Quantum Monte Carlo Assessment of the Relevance of Electronic Correlations in Defects and Equation of State in Metals

Randolph Q. Hood 06-LW-024

Abstract

We propose to develop accurate quantum Monte Carlo (QMC) capabilities to calculate defect formation energies and equations of state (EOS) for metallic systems. We will determine, for the first time in metals, the significance of electronic correlation effects in EOS and metal–insulator transitions, and the effects on formation energies of point defects, impurities, surfaces, and interfaces beyond Density Functional Theory (DFT). These parameters determine the mechanical properties and microstructural evolution of metals under extreme conditions. Technical limitations have restricted application of QMC to semiconductors, insulators, and homogeneous electron gas. We have recently overcome the largest obstacle with a new formulation based on optimized, nonorthogonal orbitals.

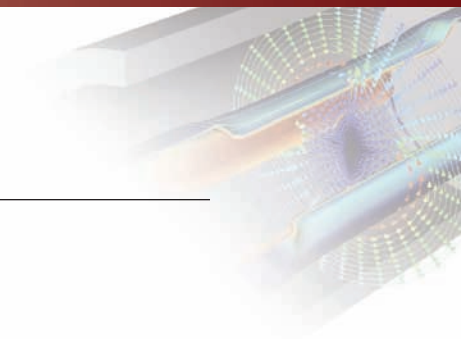
Initially, we plan to develop the method and software to generate nonorthogonal transformations for real metals as an input to the CASINO QMC code. QMC values for the total formation energies of bulk aluminum will be obtained and we will begin to calculate some simple defects such as vacancies or surfaces for the same system. In the longer term, we will generate a formation energy table for a long list of defect structures for different materials. Under extreme pressures, when accurate experimental measurements become exceedingly difficult, tables of this sort provide data for fitting empirical interatomic potentials and continuum models. Until now, less-accurate DFT calculations have been used for this purpose.

Mission Relevance

The calculations associated with this research, which take advantage of LLNL's unique supercomputing capabilities, are key to describing matter at extreme regimes for stockpile stewardship and assessment.

FY07 Accomplishments and Results

In FY07, we determined that the EOS for aluminum obtained by our QMC assessment had a nonphysical sharp feature in its form and a lattice constant 2% less than experimental values. Therefore, we developed multideterminant wavefunctions using variance minimization and obtained a smooth EOS, though the lattice constant was still 2% lower than experimental values. We optimized multideterminant wavefunctions using energy minimization to obtain a more accurate EOS before studying other metals. Through algorithmic improvements, our QMC code is now five times faster and runs on the BlueGene/L supercomputer at Livermore. Finally, we have begun preparing our QMC results on aluminum for publication.



Diffusion Monte Carlo without All the Hops

Vasily V. Bulatov 06-LW-028

Abstract

We intend to develop a new general method for diffusion Monte Carlo (DMC) simulations based on the theory of first-passage processes. This method will be applicable to a wide variety of conditions where the standard DMC simulation is ineffective—for example, when the density of diffusing particles is low and/or the distribution of their diffusion rates is wide. Our approach will boost the performance of DMC simulations by projecting the system's state directly to collisions while preserving the random walk statistics. We will implement the new method in an efficient computer code (both sequential and parallel versions) convenient for a wide variety of applications relevant to Laboratory missions and beyond.

The successful project will result in a new, efficient methodology for DMC simulations applicable to a wide variety of conditions encountered in physics, chemistry, biology, and materials sciences. The new approach will overcome an age-old limitation of the standard DMC method and open multiple new opportunities for predictive modeling of complex processes relevant to Laboratory missions. The new development will be implemented in an efficient parallel code with user-definable local rules, rate constants, and simulation geometries in one, two, and three dimensions. The code will be maintained as an open-source software and made available to computational scientists at the Laboratory and beyond.

Mission Relevance

The DMC simulations can be applied to stockpile science and technology, such as predictive modeling of plutonium aging; to energy science and security, such as materials for fusion reactors and nuclear waste management; and to breakthroughs in fundamental chemistry and material science. Development of new algorithms and parallel computing techniques also contribute to LLNL's expertise in numerical simulations, which support the Laboratory's mission in fundamental science and technology.

FY07 Accomplishments and Results

In FY07, we (1) extended our simulation method to deal with diffusion of multiple particles in confined geometries, (2) developed mathematical algorithms for three-dimensional kinetic Monte Carlo simulations using spherical protective domains, (3) examined and evaluated several algorithms for a parallel kinetic Monte Carlo code based on a spatial domain decomposition scheme, (4) examined the potential of a new algorithm for boosting performance of quantum Monte Carlo simulations, and (5) developed a user-friendly interface for setting up the geometry and creating input decks of the unit processes and their kinetic rates.



Knowledge-Based Coreference Resolution

Terence Critchlow 07-ERD-027

Abstract

Extracting knowledge buried within unstructured electronic documents is becoming an increasingly critical issue to the intelligence community. Millions of such documents are created daily; obtaining knowledge from them requires identifying relevant documents, recognizing real-world entities, determining relationships among entities, extracting events, identifying when the same event is discussed in multiple documents, and providing a summary of events contained within a collection. One critical step not being addressed by the larger community is knowledge-based, non-pronoun, non-proper-name (nominal) coreference resolution, which is required to identify each mention of relevant entities in a document and to match an entity to its relationships. Our project addresses this deficiency by leveraging the unique capabilities of LLNL, including work in graph-based entity disambiguation.

To meet our goal of significantly improving nominal coreference resolution, we are developing novel research algorithms to create concept-independent signatures for entities in documents and accurately resolve nominals using background corpora and semantic graphs as external knowledge. These algorithms will perform significantly better than state-of-the-art coreference resolution techniques and are expected to provide accuracies greater than 75% for relevant nominal coreferences.

Mission Relevance

By developing advanced text-analysis algorithms that will help analysts extract knowledge from massive volumes of text documents in counterterrorism and other homeland security applications, this project supports the Laboratory's national security mission.

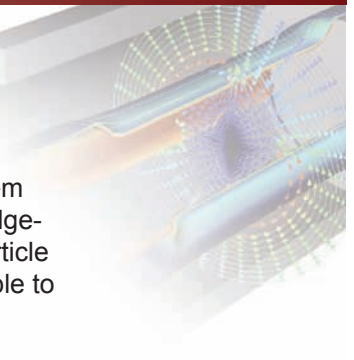
FY07 Accomplishment and Results

In FY07, we (1) analyzed and annotated documents from several corpora, including the benchmark datasets ACE2005 and MUC6/7, email messages from the ProMED Mail archive, and news reports gathered from the Open Source Center; (2) developed a baseline machine learning system for resolving coreferent phrases that performs as well as published results; (3) created a suite of tools to analyze background data to extract different types of coreference-specific relationships; and (4) implemented an enhanced scoring algorithm to assist in developing the system. A unique aspect of our system is that it classifies mention types in a way that enables us to develop specialized algorithms for each category and to focus on those coreferences that most improve information-extraction tasks.

Proposed Work for FY08

In FY08, we will conduct an in-depth exploration of the baseline prototype to catalog the contribution of each feature and experiment with different classifiers, then convert the

individual components developed in FY07 into features that can be incorporated into the baseline system, including examination of the impact of each new feature on system performance. We will specifically examine the impact of features derived from knowledge-rich external sources. We will initially investigate use of the *New York Times* historic article archive. This work entails producing a code base, in the ML language, that is deployable to any corpus of interest and available to different endeavors at LLNL.



Advanced Computational Techniques for Uncertainty Quantification

Charles H. Tong 07-ERD-028

Abstract

This project is aimed at developing advanced uncertainty quantification methods that can efficiently and accurately handle large-scale multiphysics simulations distinguished by large numbers of inputs and expensive evaluations. This research aligns well with LLNL's increased emphasis on modeling and simulation, and the technologies developed will benefit many Livermore simulation-based applications. We will focus our algorithm research and development effort on (1) derivative-based global sensitivity analysis, (2) sensitivity analysis for high-dimensional problems, (3) new response surface methods, and (4) risk analysis. Software and papers will be written to document our results.

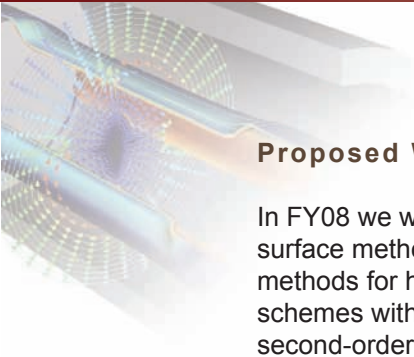
We anticipate that our research results will be published in journals and conference proceedings. The developed software will find immediate use in varied LLNL applications as well as the broad scientific community.

Mission Relevance

Our project aligns well with the LLNL goal of advancing the state of the art in large-scale simulations, on which the Stockpile Stewardship Program is based. It also will be useful in support of DOE's new energy research initiatives and help fulfill the Laboratory's mission in energy security. In addition, this research effort will leverage the Laboratory's current expertise in local and global sensitivity analyses and keep it at the forefront of high-consequence, high-fidelity, and high-dimensional simulation models.

FY07 Accomplishments and Results

In FY07, we (1) implemented a prototype for derivative-based global sensitivity analysis and demonstrated it on several problems modeled as partial differential equations with adjoints; (2) began work on "black box" test problems in which finite differences are needed; (3) developed methods for second-order sensitivity indices, incorporated refinement strategies to improve robustness for first- and second-order sensitivities, and began constructing a robust screening method; and (4) acquired and began working with a model for uncertainty quantification requirements for structures under seismic effects.



Proposed Work for FY08

In FY08 we will (1) implement, test, and document a final version of the adaptive-response surface method using local sensitivity analysis; (2) explore Bayesian sensitivity analysis methods for high-dimensional applications and compare the effectiveness of Bayesian schemes with the novel scheme we developed in FY07; (3) complete a suite of first- and second-order variance-decomposition capabilities for uncorrelated and correlated inputs; (4) investigate risk-analysis approaches and apply them to building structures; and (5) improve and document our black box test problems.

Publications

Tong, C., 2007. *Toward a more robust variance-based global sensitivity analysis of model outputs*. UCRL-TR-235561.

Tong, C., and F. Graziani, 2007. *A global sensitivity analysis methodology for multi-physics applications*. UCRL-TR-227800.

VidCharts: Real-Time Algorithms for Large-Scale Video Analysis, Compression, and Visualization

Mark A. Duchaineau 07-ERD-035

Abstract

This project aims to produce techniques for processing huge streams of imagery for national security applications. Our core technical ideas include a novel algorithm to compute dense image correspondences and a progressive, hierarchical processing framework. Leveraging large-data work performed at the Laboratory in support of massive simulations, we will produce a prototype automatic visual summary and drill-down system to allow hours of video to be analyzed accurately in minutes. This technology has the potential to accelerate, by orders of magnitude, the human-analysis tasks of categorizing, indexing, annotating, and otherwise managing the petabytes of video data being generated worldwide each day.

If successful, this project will result in a prototype visual indexing system for huge video streams, including scene segmentation, pan/zoom/mover analysis, space-time drill-down, visualization of complex movers in the summary view, and three-dimensional scene and camera parameter extractions. In our system, processing speeds will be accelerated by orders of magnitude using both novel streaming hardware optimizations and the progressive, multiresolution streaming algorithm pipelines developed in LLNL's large-data research efforts. Furthermore, our system will yield over a thousand-fold compression with enhanced quality for repeated imaging of a scene from a moving camera. The project also aims to create the first scalable video content indexing and query system.

Mission Relevance

The project supports the Laboratory's national security mission, particularly arms control and nonproliferation, which are the most urgent and critical application areas for new software

systems to accelerate the accurate analysis and management of huge streams of imagery. Numerous monitoring, tracking, discovery, and operational activities fall within this application area.

FY07 Accomplishments and Results

We (1) created an experimental test bed for performing image-pair correspondence computations, grouping these into temporal abstractions; (2) developed an initial interactive video summary and drill-down visualization prototype; (3) accelerated by more than 100 times the most computationally intense algorithm kernels using graphics processing units; (4) successfully applied streaming processing algorithm frameworks and multiple-resolution, hierarchical data organizations to massive video analysis; and (5) extracted three-dimensional information from uncalibrated, unstable aerial snapshots. Of particular note was our development of new wavelet-compression algorithms optimized for large-format video and the creation of a mosaic summary and superhigh-resolution drill-down application using experimental aerial video.

Proposed Work for FY08

In FY08, we will (1) pursue three-dimensional extraction from multiview correspondences; (2) provide robust mover and background segmentation and mixing estimation on mover and silhouette boundaries; (3) devise view-dependent interaction acceleration with motion compensation for large-format video; (4) continue to expand the core selective-refinement infrastructure upon which the experimental test bed is based; (5) study sensor, camera, and environmental artifacts and noise to improve the performance of the correspondence algorithm in the most challenging conditions; and (6) improve the graphics processor unit's iterative optimization of mosaic stitching and resolution enhancement.

Publications

Duchaineau, M. A., J. D. Cohen, and S. Vaidya, 2007. *Toward fast computation of dense image correspondence on the GPU*. 11th Annual Workshop on High-Performance Embedded Computing, Lexington, MA, Sept. 18–20, 2007. UCRL-CONF-233816.

Tsap, L. V., M. A. Duchaineau, and D. B. Goldgof, 2007. "Data-driven feature modeling, recognition, and analysis in a discovery of supersonic cracks in multimillion-atom simulations." *Pattern Recogn.* **40**, 2400. UCRL-JC-143820.

Software Security Analysis

Daniel J. Quinlan 07-ERD-057

Abstract

Lawrence Livermore, as well as the DOE, obtains software from a wide variety of sources, both as binaries and source code. Currently, LLNL has almost no ability to determine whether such software is free from intentional or unintentional security defects. Furthermore, existing



security-analysis algorithms and techniques do not exploit large-scale parallelism. Leveraging LLNL's unique parallel-computing technology to address large-scale program analysis problems, we are doing research on the security analysis of software in both binary and source-code form. This project is defining mechanisms for the analysis of binaries and the automated source-to-source transformation of vulnerable code to secure code, something not possible with any existing security analysis capability of which we are aware.

We expect this project to greatly increase LLNL's ability to construct and maintain highly secure and reliable software systems. We plan to release tools developed in this project through the open-source channel so that they also benefit other researchers and tool developers in building their own binary and source-code analysis tools. At a more fundamental level, this project also will advance our understanding in building practical and effective analysis tools for binary and source-code applications. Our work builds on existing collaborations with Argonne National Laboratory and several universities to support a program-analysis framework able to handle the compilation and analysis of LLNL's largest-scale Advanced Simulation and Computing applications.

Mission Relevance

The project supports LLNL's national security mission by improving the Laboratory's cybersecurity infrastructure with tools to support the general analysis and reverse engineering of outside software. Our work also will build a significant level of internal expertise in software security analysis.

FY07 Accomplishments and Results

In FY07, we (1) developed a global program-analysis capability to support more-complex analysis of source code and support side-by-side analysis with binaries, (2) developed a mechanism to specify security flaws in source code and search for such flaws in applications, (3) demonstrated attribution of source information in binaries and construction of a control flow graph from a Linux binary without symbol information, and (4) published a paper on C++ security analysis and four other papers on the recognition of general security analysis flaws.

Proposed Work for FY08

In FY08, we will focus on binary analysis and the development of tools for analyzing source code. Specifically, we will (1) define additional detection algorithms for Software Assurance Metrics and Tool Evaluation Program security bugs and implement these using the ROSE code, (2) identify classes of security bugs that can be automatically rendered secure, (3) develop the security bug specification mechanism into a general tool for the analysis of large-scale applications, (4) target a specific analysis that is not well handled at the scale of million-line applications and define the techniques and parallel algorithms to solve using massively parallel computation, and (5) validate the approach by applying these techniques and algorithms to LLNL application codes such as VisIt or ALE3D.

Publications

Panas, T., D. Quinlan, and R. Vuduc, 2007. "Tool support for inspecting the code quality of HPC applications." *Proc. 29th Intl. Conf. Software Engineering*. UCRL-PROC-229217.

Panas, T., D. Quinlan, and R. Vuduc, 2007. "Visualization of program architecture using whole-program analysis." *Proc. 29th Intl. Conf. Software Engineering*. UCRL-PROC-231453.

Panas, T., et al., 2007. *Communicating software architecture using a unified single-view visualization*. 12th IEEE Intl. Conf. Engineering Complex Computer Systems, Auckland, New Zealand, July 11–14, 2007. UCRL-CONF-227293.

Quinlan, D., and R. Vuduc, 2007. *Techniques for specifying bug patterns*. Intl. Symp. Software Testing and Analysis, London, UK, July 9–12, 2007. UCRL-CONF-230552.

Quinlan, D., et al., 2006. *Support for whole-program analysis and the verification of the one-definition rule in C++*. Static Analysis Summit, Gaithersburg, MA, June 29, 2006. UCRL-CONF-222122.

Verification and Validation of Radiation Hydrodynamics for Astrophysical Applications

Louis H. Howel 07-ERD-061

Abstract

This project's aim is to verify and validate an adaptive-mesh radiation hydrodynamics code with applications in astrophysics and high-energy-density physics. Verification tests will include uncomplicated problems with known or analytic solutions that will be used to measure the accuracy and resolution convergence of numerical solutions. These tests will be applied to code units (e.g., hydrodynamics) and, as possible, to integrated radiation hydrodynamics simulations. Verification will also include more complex tests (e.g., the crooked-pipe test). Validation will use data from actual experiments. This project will be conducted in collaboration with Stony Brook University and the Computational Astrophysics Consortium.

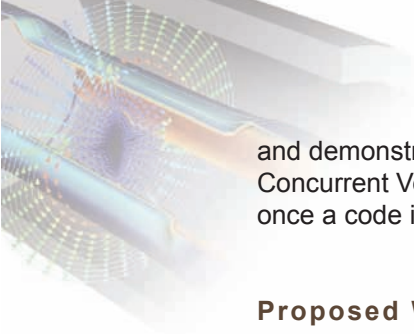
The immediate result of this work will be increased understanding of the simulation accuracy of the adaptive-mesh radiation hydrodynamics code for particular astrophysics problems, including supernova explosions, with a focus on radiating shock instabilities. These instabilities are of great interest to the Stockpile Stewardship Program. Another expected result is an advancement in methodology for validating such code.

Mission Relevance

This project supports stockpile stewardship by developing and applying new verification and validation methods for astrophysical radiation hydrodynamics to quantify the uncertainties in large-scale simulations for NNSA applications.

FY07 Accomplishments and Results

After a midyear start, we (1) began assembling the tools needed to perform verification of the Castro code, (2) completed development of a general test suite for Boxlib-based codes and some analysis routines, (3) used these tools to perform shock-tube hydrodynamics tests



and demonstrate the Sedov problem, and (4) registered all of these tools into the project Concurrent Versions System repository and our test suite, which enabled us to ensure that once a code is verified, it is the verified version that is preserved.

Proposed Work for FY08

We will (1) continue our verification work on the radiation hydrodynamics solver, using standard test problems from the literature; (2) explore validation opportunities with experimentalists, to find metrics for radiation and hydrodynamic simulation–experiment comparison; and (3) continue to document all radiation test problems in the project Concurrent Versions System repository and place into the test suite.

Storage-Intensive Supercomputing

Maya B. Gokhale 07-ERD-063

Abstract

This project addresses efficient computation of data-intensive problems in national security and basic science by advancing storage-intensive supercomputing (SISC) capabilities. We propose to (1) develop new algorithms and applications to solve large-scale data analytics problems on this class of architectures; (2) explore new programming models, tools, and libraries to address the difficulty in developing software applications for storage-intensive architectures; and (3) develop new system architectures for SISC in partnership with industry collaborators. These efforts will enable an order-of-magnitude improvement in price/performance ratios over today's data-intensive architectures for a broad range of data-intensive problems.

Across the laboratory, and in the scientific and national security communities at large, scientists and analysts are searching for techniques, tools, and computing architectures to manage and analyze large datasets. Such data-intensive problems are particularly common in scientific simulation, defense applications, and sensor-related activities. For applications that require frequent access to storage, the traditional technology is inadequate. Our goal is to enable applications that simply cannot run on current systems and to deliver an order-of-magnitude improvement in performance and productivity over current systems.

Mission Relevance

This project will deliver a new capability to solve data-intensive problems in nonproliferation and homeland security, defense applications, and analysis of scientific simulation data. Storage-intensive architectures offer an advantage over computation-intensive architectures by optimizing access to large data sets. Example problems include analysis of stockpile stewardship simulations, large-scale graphs used to identify terrorist networks, massive astronomy datasets, and fusion-class laser optics imagery to assess damage.

FY07 Accomplishments and Results

In FY07 we created a set of data-intensive application benchmarks. The benchmarks were drawn from a wide range of domains and consist of (1) level-set expansion of scale-free graphs, (2) entity extraction in unstructured text, (3) language classification using n -gram profiles, and (4) image resampling. The first two types of benchmarks used data stored on a prototype solid-state disk array, whereas the last two types of benchmarks included versions accelerated by field-programmable gate arrays and graphics processing units, respectively. The accelerated versions showed a speedup of a factor of 5 to 85 over their sequential counterparts. We also developed an input–output tracing tool to record the actual read and write operations of the benchmarks and wrote a technical report to document the benchmarks and their input–output characteristics.

Proposed Work for FY08

In FY08 we will (1) collaborate with industrial partners to develop SISC system architectures that are flexible, scalable, robust, and cost-effective; (2) address the difficulty of programming scalable applications software with file systems and programming models focused on data-intensive problems; (3) create software tools for performance measurement; and (4) develop new approaches for mapping algorithms and applications onto SISC architectures.

Publications

Jacob, A., and M. Gokhale, 2007. *Language classification using N-grams accelerated by FPGA-based Bloom filters*. 1st Intl. Workshop High-Performance Reconfigurable Computing Technology and Applications, Reno, NV, Nov. 11, 2007. UCRL-CONF-234633.

Laboratory Directed Research and Development

Nuclear Science and Engineering





Decontamination of Terrorist-Dispersed Radionuclides from Surfaces in Urban Environments

Robert P. Fischer 05-ERD-029

Abstract

This project will develop radionuclide-specific decontamination agents and optimize them for use in the mass transit infrastructure in urban environments. The work will address recognized data gaps by advancing the basic scientific knowledge of radionuclide substrate interactions in urban environments following a terrorist attack with a radiological dispersal device (RDD). The final deliverable will be threefold: prototype decontamination agents for americium and cesium in urban environments to take to technology transfer, a better understanding of surface decontamination in a series of reports, and a model for radionuclide surface interactions.

The project will identify and optimize a new generation of radionuclide-specific decontamination agents and improved fate transport modeling for responding to an RDD event. This research will result in a substantial improvement over existing response capability. Results will be published in peer-reviewed journals.

Mission Relevance

This project enhances U.S. capability to respond effectively to an RDD. It supports national security, homeland security, and environmental-management missions by developing effective decontamination agents for use in urban settings and by advancing the basic scientific knowledge of radionuclide substrate interactions in the urban environment.

FY07 Accomplishments and Results

In FY07 we (1) fielded two outdoor explosives tests; (2) tested prospective decontamination agents through a series of sorption studies, establishing provisional efficacy information; and (3) studied the interaction of cesium with concrete surfaces under a number of experimental parameters. In summary, this project showed that penetration of cesium in real-world materials may significantly differ from standard laboratory test articles. For instance, cesium penetration on dry materials was found to be significant at depths of several millimeters to several centimeters. Such data will be of great use in the further development of decontamination agents. Finally, this project developed four chelators optimized for use in urban environments.

Publications

Fischer, R. P., et al., 2007. *Decontamination of terrorist dispersed radionuclides from surfaces in the urban environment*. UCRL-PRES-231780.

Fischer, R. P., et al., 2007. *Decontamination of terrorist-dispersed radionuclides from surfaces in urban environments*. Am. Nuc. Soc. Workshop Decommissioning, Decontamination, and Reutilization. Chattanooga, TN, Sept. 16–19, 2007. UCRL-ABS-227694.

Gates-Anderson, D. D., et al., 2007. "Dirty bomb fallout." *Nucl. Eng. Int.* **52**, 28. UCRL-PROC-226374.

Gates-Anderson, D. D., et al., 2007. *Field evaluation of the explosive deposition of cesium on concrete surfaces following the detonation of a mock radiological dispersal device*. Waste Management 2007, Tuscon, AZ, Feb. 26–Mar. 2, 2007. UCRL-POST-227962.

Sutton, M., et al., 2007. *Simulating Cs radiological dispersal devices for deposition, dose, and decontamination studies*. 52th Ann. Mtg. Health Phys. Soc. Portland, OR, July 8–12, 2007. UCRL-ABS-227787.

A Coupled, Multiphysics Code for Accurate Modeling of Nuclear Reactors

Richard Procassini 06-ERD-069

Abstract

We are performing algorithmic research and development necessary to produce a prototype coupled, multiphysics code that is applicable to the accurate modeling of nuclear reactors. This code—Osiris—will permit reactor designers to overcome many of the limitations they currently face, including the use of lumped-parameter models, and the use of multiple codes within a multistep modeling methodology. Our approach to producing this new code leverages our proven expertise in code development and our suite of existing code modules. This project will pave the way for LLNL to contribute to the future needs of DOE's Global Nuclear Energy Partnership (GNEP) initiative.

We are leveraging our expertise in code development and suite of existing codes to produce a prototype high-performance, coupled, multiphysics code that is applicable to the design of reactor systems. The intent is to replace an existing set of legacy codes, which require significant approximations and assumptions, with an integrated, coupled code that permits the design of a reactor core using a first-principles approach to model the physics. This would greatly simplify the reactor-core design process. The high-performance capabilities of this code would enable routine calculations at scales that are impossible for the current modeling methodology.

Mission Relevance

This project takes advantage of the Laboratory's expertise in code development and nuclear reactor modeling to provide tools that will respond to future needs of the DOE's GNEP initiative, which will contribute to the future of nuclear power options for the United States and help to realize the DOE's goal of enhanced energy security. Additional benefits may be obtained from this work in the area of code validation supporting LLNL's stockpile stewardship efforts.



FY07 Accomplishments and Results

Our accomplishments in FY07 include the development of an incompressible Navier–Stokes flow solver in the Vulcan thermal-hydraulics module, which was combined with an existing thermal-transport solver. In addition, we investigated algorithms for the parallelization of a Vulcan thermal-hydraulics module within the Overture framework code, using a parallel, multigrid elliptic equation solver.

Gallium Telluride Semiconductor Radiation Detector

Stephen A. Payne 07-ERD-008

Abstract

The objective of our proposed work is to perform a feasibility study to determine if a gallium telluride (GaTe) semiconducting crystal can serve as a high-performance, room-temperature, gamma-radiation detector. Determining if a GaTe detector offers compelling advantages over the state of the art will help decide if an additional material development campaign is required. Our partner, the historically black university Fisk, is providing the needed expertise in the growth of semiconductor crystals, while the materials characterization and device fabrication will be performed at Lawrence Livermore.

We expect to determine the feasibility of using GaTe semiconducting crystals for an advanced radiation detector. Overall, GaTe has many compelling features: it is a simple binary material, has a high Z of 52 for good gamma stopping power, congruently melts at 836°C , offers an ideal bandgap of 1.7 eV for room-temperature operation, and is comprised of readily available, high-purity starting materials (elemental gallium and telluride). We will initially develop 0.5-cm thick crystals to determine their material properties (carrier mobility, lifetime, etc.) relevant to performance as a high-resolution radiation detector.

Mission Relevance

An inexpensive, sensitive, high-resolution, room-temperature radiation detector is urgently needed for widespread deployment at ports of entry, throughout cities, and for use in military operations. Semiconductor radiation detectors serve a crucial role in detecting nuclear weapons and radiological dispersal devices by virtue of their ability to distinguish isotopes. Thus, advanced radiation detector materials support critical Laboratory missions in national and homeland security.

FY07 Accomplishments and Results

In FY07, we (1) grew millimeter-sized GaTe crystals and measured a mobility of about $100 \text{ cm}^2/\text{Vs}$, carrier lifetime of $2.6 \mu\text{s}$, and resistivity of $2 \times 10^7 \text{ Ohm-cm}$; (2) used x-ray diffraction to verify that we had the correct chemical phase of the material; and (3) determined, on the basis of these preliminary results, that GaTe offers promise as a radiation detector. The initial success of this study has resulted in continued support from the Department of Homeland Security.

Publications

Conway, A., et al., 2007. *Exploration of GaTe for gamma detectors*. IEEE Nuclear Science Symp., Honolulu, HI, Oct. 27–Nov. 3, 2007. UCRL-ABS-230896.

Detection, Classification, and Estimation of Radioactive Contraband from Uncertain, Low-Count Measurements

James V. Candy 07-ERD-019

Abstract

The detection of smuggled radioactive materials is a serious issue in national security. The objective of this project is to investigate fast, reliable radiation detection methods capable of making a more rapid decision with higher confidence, along with the ability to quantify performance. We are focused on the detection, classification, and estimation of illicit radioactive material from highly uncertain, low-count radionuclide measurements using a statistical approach based on Bayesian inference and physics-based signal processing for container scanning. Modern computers enable the development of physics-based statistical models that capture essential signatures of radionuclides and incorporate them into a Bayesian sequential scheme capable of online, near-real-time operation.

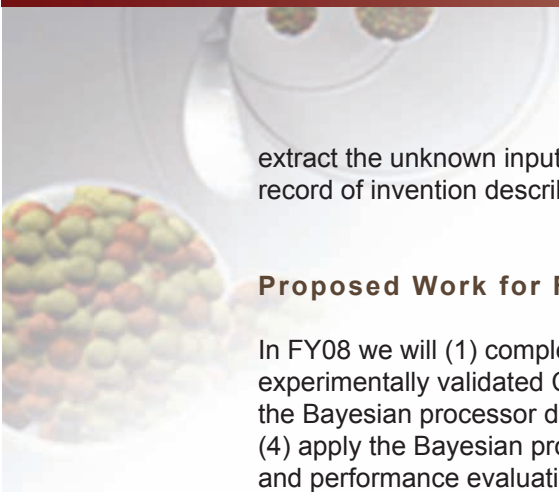
We expect to develop solutions for the detection, classification, and estimation of a moving radionuclide source and detector. Our goal is to reliably detect several kilograms of shielded plutonium in less than a minute, with a 95% detection probability and a 5% false alarm rate. The Bayesian approach enables development of a sequential framework that establishes the foundation for future problems that are both time and space varying or equivalently statistically nonstationary. This approach is applicable, in principle, to a large variety of model-based problems in many other critical areas of Laboratory work, including defect detection for the Stockpile Stewardship Program. Advanced signal- and image-processing techniques for the next generation of processors will evolve from this project.

Mission Relevance

The detection of illicit radionuclides is a top priority in furthering LLNL's national security mission. Radionuclide detection, classification, and estimation are critical for detecting the transportation of radiological materials by terrorists, an important goal in both national and international security. This technology also supports the life extension program because of its potential application in defect detection.

FY07 Accomplishments and Results

After a midyear start, we (1) developed a radionuclide signal-processing model based on decomposition into monoenergetic sources; (2) incorporated these embedded physics-based models into a sequential Bayesian processor; (3) validated these models with sophisticated simulations (using the x-ray transmission radiography code COG) and controlled experiments; (4) applied the processor to known, controlled experimental data; (5) published our results in conference papers that illustrate the capability of the Bayesian processor to



extract the unknown input (deconvolution) from measured radiation data; and (6) developed a record of invention describing the initial detection–classification system design.

Proposed Work for FY08

In FY08 we will (1) complete the sodium iodide experiments, (2) initiate a set of experimentally validated COG simulations to incorporate more of the transport physics into the Bayesian processor design, (3) begin to validate and refine out initial design concepts, (4) apply the Bayesian processor design to an equivalent experimental setup for validation and performance evaluation, (5) begin implementing the full system described in our record of invention, and (6) design sequential methods capable of classifying various radionuclides of interest to terrorists.

Publications

Candy, J. V., 2007. *Bayesian processing for SNM enhancement from uncertain radionuclide measurements*. UCRL-AR-234614.

Candy, J. V., 2007. “Bootstrap particle filtering.” *IEEE Sig. Proc. Mag.* **24(4)**, 73. UCRL-JRNL-227731.

Candy, J. V., 2007. “Nonlinear statistical signal processing: a particle filtering approach.” *Imaging for Detection and Identification*. J. Byrnes (Ed.), Springer, New York, chap. 9, pp. 129-150. UCRL-JRNL-234770.

Candy, J. V., et al., 2007. “Bayesian processing for the detection of radioactive contraband from uncertain measurements.” *Proc. 2nd Intl. Workshop Computational Advances in Multi-Sensor Adaptive Processing*. UCRL-JRNL-232317.

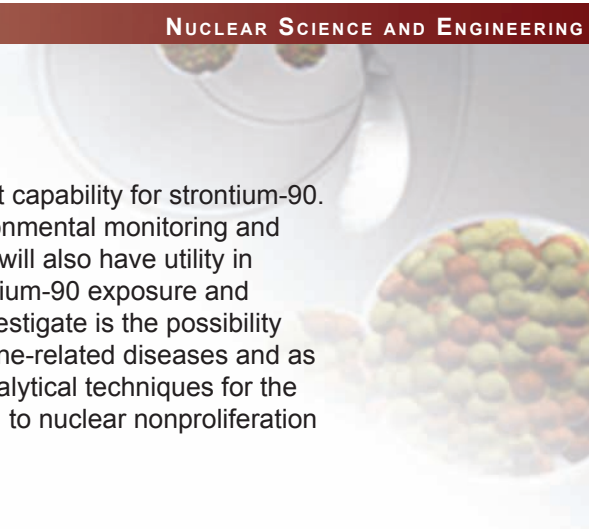
Sale, K., et al., 2007. “A Bayesian signal processor approach to spectroscopic portal system measurements.” *Proc. SPIE Imaging Conf. 2007*. UCRL-CONF-233728.

Accelerator Mass Spectrometry of Strontium-90 for Biomonitoring and Human Health

Scott J. Tumey 07-ERI-002

Abstract

A high-yield fission product, strontium-90 is one of the most hazardous constituents of nuclear waste. Being a pure beta emitter, strontium-90 is difficult to measure accurately in environmental samples because it generally occurs in the presence of other beta emitters—that is, fission products. An alternative method to measure strontium-90, with potentially significant advantages over radiation counting, is accelerator mass spectrometry (AMS), which offers enhanced sensitivity, throughput, and expediency. Therefore, we propose to develop a method to quantify strontium-90 by AMS, which combined with the high-throughput design of the Center for Accelerator Mass Spectrometry facility at Livermore, would result in a state-of-the-art measurement system for this radionuclide.



The primary outcome of this project will be a robust measurement capability for strontium-90. This capability will have immediate application to improved environmental monitoring and dose assessment for the Marshall Islands Project. The capability will also have utility in human health studies focusing on the relationship between strontium-90 exposure and cancer rates. A potentially high-impact application that we will investigate is the possibility for strontium-90 to compliment calcium-41 as a bioindicator of bone-related diseases and as a tool for evaluating the efficacy of various treatments. Finally, analytical techniques for the detection of strontium-90 have an obvious and natural application to nuclear nonproliferation and homeland security.

Mission Relevance

This project is well aligned with the national security mission of the Laboratory because of the role of strontium-90 in homeland security and nuclear nonproliferation—almost all of this isotope is produced in the nuclear fuel cycle or past nuclear tests. Therefore, its detection in environmental samples could serve as a diagnostic for clandestine reprocessing of nuclear fuel for weapons use. Additionally, the application of strontium-90 to human health studies and its potential use in detection and treatment of bone-related diseases are important contributions to science in the public interest.

FY07 Accomplishments and Results

In FY07, we (1) selected strontium fluoride as the most suitable target material since it yields robust beam currents, can be easily precipitated from solution and collected by centrifugation, and is stable in air and therefore requires no special handling; (2) measured charge-state distributions of strontium at various energies that have not been previously published in the literature and will comprise an important component of a journal article; (3) observed a pervasive interference from the isobar zirconium-90 that appears to originate from the aluminum target holder, and so began investigating alternative materials to minimize this interference; and (4) begun designing an improved detector that will more effectively resolve strontium-90 from zirconium-90.

Proposed Work for FY08

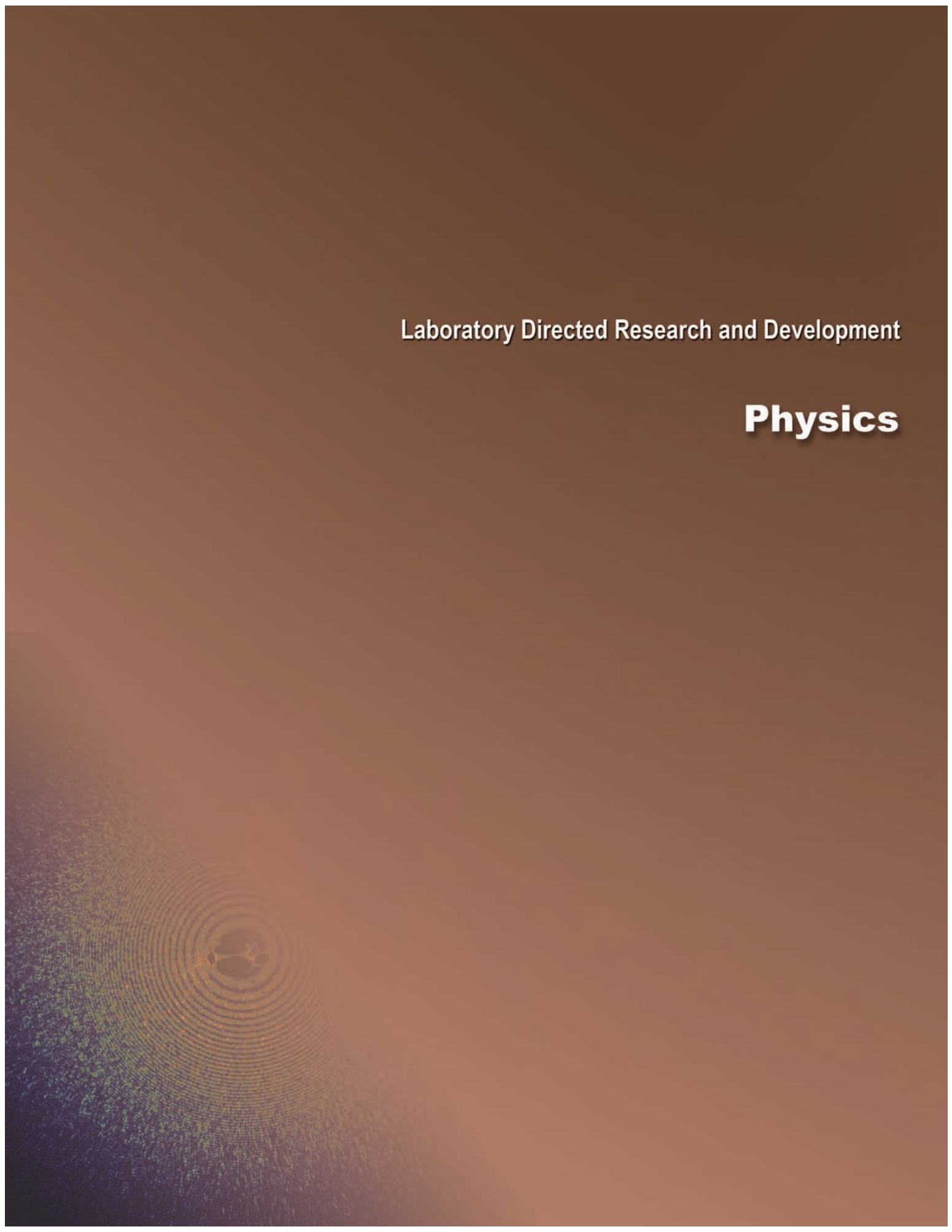
Work in FY08 will focus on the development of appropriate sample preparation methods for real matrices. Given the potential applications of this work, treatment of soil, water, and urine will be our main priority. Specifically, we will (1) assess our ability to accurately measure strontium-90 in these matrices by using existing standard reference materials; (2) begin to participate in the DOE Laboratory Accreditation Program, which will be important in establishing our involvement in the Federal Radiological Monitoring and Assessment Center in the event of a radiological incident; and (3) participate in exercises for the measurement of strontium-90 in various matrices.

Publications

Tumey, S. J., et al., 2007. *Accelerator mass spectrometry of strontium-90 for homeland security, environmental monitoring, and human health*. 9th European Conf. Accelerators in Applied Research and Technology, Florence, Italy, Sept. 3–7, 2007. UCRL-ABS-231565.

Laboratory Directed Research and Development

Physics





Nonequilibrium Phase Transitions

Andrew Ng 04-ERD-108

Abstract

The exploration of nonequilibrium phase transitions is a scientific frontier that holds promise for discovering new phases, metastable states, chemical reaction pathways, and biological functioning processes. We will conduct the first systematic study of phase transitions in an extreme, nonequilibrium regime to examine lattice disordering and melting, quantify the role of electronic excitation on phase-transition kinetics, and develop approaches in finite-temperature condensed matter for constructing an equation of state. The project will use measurements to correlate optical and structural properties under ultrafast laser excitation to help develop density functional theory approaches, and work will be conducted in collaboration with the University of Toronto.

We seek 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 also will 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, as well as breakthroughs in fundamental science.

FY07 Accomplishments and Results

In FY07, we (1) completed development of the 600-fs, 30-keV electron gun and demonstrated the viability of single-shot, ultrafast electron diffraction measurement at 400 nm on gold nanofoils; (2) obtained new findings that elucidated ionization dynamics and nonequilibrium effects on phonon modes; and (3) presented work on ultrafast electron diffraction and broadband dielectric function measurements in a keynote conference address as well as several invited talks. Overall, the project's most notable achievement is the discovery of quasi-steady states of superheated lattice with highly excited nonequilibrium electrons. This fundamentally changes the existing understanding of warm dense matter produced by ultrafast excitation of a solid using laser x-ray charged particles.

Publications

Murray, E. D., et al., 2007. "Phonon dispersion relations and softening in photoexcited bismuth from first principles." *Phys. Rev. B*. **75**, 184301. UCRL-JRNL-224921.

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 compare experiments with high-fidelity modeling to understand the new abilities that the XFEL will bring to biological imaging. These capabilities will allow the determination of the atomic structure of any protein.

Each of our experiments, which will be a world first and a major new result in x-ray science in every case, will (1) determine the duration and fluence of XFEL pulses required for single-molecule imaging, (2) demonstrate reconstruction methods, and (3) perform ultrahigh-resolution three-dimensional 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) beyond the baseline design, a high-priority project of the DOE Office of Science, in support of LLNL's mission in breakthrough science and technology.

FY07 Accomplishments and Results

We (1) performed the first-ever demonstration of the ultrafast diffractive x-ray imaging of free injected particles, a major milestone of the project; (2) characterized our injection system in experiments at the FLASH free-electron laser facility in Hamburg, Germany, and found it capable of carrying out experiments at higher resolution at the upcoming LCLS; (3) reconstructed images of injected living hydrated biological cells and test particles; (4) improved the accuracy of our method of time-delay holography and published results in *Nature*; (5) conducted ultrafast time-resolved imaging of laser-ablation of materials at a time resolution of 1 ps and a spatial resolution of 40 nm; and (6) conducted diffraction simulations on systems of proteins in evaporating droplets, utilizing analytical models to determine droplet conditions.

Proposed Work for FY08

We will (1) take advantage of the increased x-ray penetration of FLASH—which has been upgraded to a wavelength of 6 nm—to obtain high-resolution images of biological cells beyond radiation-damage limits; (2) increase the efficiency of the particle injection system and diagnostics for use in the three-dimensional imaging of reproducible structures, which will be done with gold rod nanoparticles and asymmetric virus particles; (3) further develop time-resolved imaging methods, including the tomographic time-resolved imaging of ablation, and measure the effect of a tamper on XFEL-induced particle explosion; and (4) develop a plan for the first experiments at the LCLS based on FLASH experiments and modeling.

Publications

Bajt, S., 2007. *Multilayers for next generation x-ray sources*. SPIE Optics 2007, Prague, Czech Republic, Apr. 16–20 2007. UCRL-ABS-226344.

Bajt, S., and H. N. Chapman, 2007. “Multilayers are enabling new science with x-ray free electron lasers.” *SPIE Newsroom* **200**, 7. UCRL-JRNL-232981.

Bajt, S., et al., in press. “A camera for coherent diffractive imaging and holography with a soft-x-ray free electron laser.” *Appl. Optic*. UCRL-JRNL-235064.

Bajt, S. et al., 2007. *Development of multilayer-based optical elements for FLASH experiments*. ALS Users’ Mtg., Berkeley, CA, Oct. 4–6, 2007. UCRL-POST-224923.

Bajt, S., et al., 2007. “Multilayers for next generation x-ray sources.” *Proc. SPIE* **6586**, 65860J. UCRL-CONF-230785.

Barty, A., et al., in press. “Ultrafast imaging of nanoscale dynamics with x-ray free electron laser pulses.” *Nano Lett*. UCRL-JRNL-235384.

Bogan, M., 2007. *Femtosecond diffractive imaging of nanoparticles*. 52nd Intl. Union Vacuum Science, Technique, and Applications Workshop, Erice, Italy, July 26, 2007. UCRL-PRES-233737.

Bogan, M., 2007. *The role of mass spectrometry in ultrafast x-ray laser diffractive imaging of electrosprayed biomolecules, nanoparticles, and cells*. 55th Am. Soc. Mass Spectrometry Conf., Indianapolis, IN, June 3–8, 2007. UCRL-ABS-231478.

Bogan, M., et al., 2007. *Shotgun femtosecond diffractive imaging of free nanoscale biomaterials*. Coherence 2007, Asilomar, CA, June 25–28 2007. UCRL-POST-231879.

Bogan, M. J., et al., in press. “Single particle diffraction imaging.” *Nano Lett*. UCRL-JRNL-235405.

Chalupsky, J., et al., 2007. “Characteristics of focused soft x-ray free-electron laser beam determined by ablation of organic molecular solids.” *Optic. Express* **15**, 6036. UCRL-JRNL-228426.

Chapman, H. N., 2007. *Diffractive imaging at LCLS, FLASH, and ALS*. Gulliver Multiscale Bioimaging Workshop, Berkeley, CA, May 17–19, 2007. UCRL-PRES-218769.

Chapman, H. N., 2007. *Diffraction imaging of exploding particles*. FLASH Collaboration Mtg., Hamburg, Germany, Jan. 27, 2007. UCRL-PRES-224915.

Chapman, H. N., 2007. *Ultrafast coherent diffractive x-ray imaging*. Frontiers in Optics 2007, San Jose, CA, Sept. 2007. UCRL-ABS-230661.

Chapman, H. N., 2007. *Ultrafast coherent diffractive x-ray imaging with x-ray free-electron lasers*. Coherence 2007, Asilomar, CA, June 25–28, 2007. UCRL-PRES-230661.

Chapman, H. N., 2007. *Ultrafast coherent diffractive x-ray imaging with x-ray free-electron lasers*. Gordon Research Conf. Electron Distribution and Bonding, Hadley, MA, July 1–6, 2007. UCRL-PRES-230661.

Chapman, H. N., 2007. *Ultrafast diffractive imaging with the FLASH FEL*. 2007 Synchrotron Radiation Instrumentation Conference, Baton Rouge, LA, Apr. 23–28, 2007. UCRL-ABS-229743.

Chapman, H. N., et al., 2007. “Femtosecond diffractive imaging with a soft-x-ray free-electron laser.” *Nat. Phys.* **2**, 839. UCRL-JRNL-219848.

Chapman, H. N., et al., 2007. *Femtosecond pump-probe x-ray diffraction imaging*. Coherence 2007, Asilomar, CA, June 25–28, 2007. UCRL-POST-233016.

Chapman, H. N., et al., 2007. “Femtosecond time-delay x-ray holography.” *Nature* **448**, 676. UCRL-JRNL-229436.

Gaffney, K. J., and H. N. Chapman, 2007. “Imaging atomic structure and dynamics with ultrafast x-ray scattering.” *Science* **316**, 1444. UCRL-JRNL-228540.

Hau-Riege, S. P., 2007. “Damage-resistant single-pulse optics for x-ray free electron lasers.” *SPIE Newsroom* <<http://spie.org/newsroom>> (Dec. 14, 2007). UCRL-JRNL-231965.

Hau-Riege, S. P., 2007. *Damage-resistant single-pulse optics for x-ray free electron lasers*. SPIE Optics 2007, Prague, Czech Republic, Apr. 16–20 2007. UCRL-ABS-226238.

Hau-Riege, S. P., et al., 2007. “Damage threshold of inorganic solids under free-electron-laser irradiation at 32.5 nm wavelength.” *Appl. Phys. Lett.* **90**, 173128. UCRL-JRNL-223837.

Hau-Riege, S. P., 2007. “X-ray atomic scattering factors of low-Z ions with a core hole.” *Phys. Rev.* **76**, 042511. UCRL-JRNL-231967.

Hau-Riege, S. P., and H. N. Chapman, 2007. “Reflection of attosecond x-ray free electron laser pulses.” *Rev. Sci. Instrum.* **78**, 013104. UCRL-JRNL-226114.

Hau-Riege, S. P., et al. 2007. “Soft x-ray free-electron laser interaction with materials.” *Phys. Rev.* **76**, 046403. UCRL-JRNL-229456.

Hau-Riege, S. P., et al., 2007. “Damage-resistant single-pulse optics for x-ray free electron lasers.” *Proc. SPIE* **6586**, 65860T. UCRL-CONF-230792.

Hau-Riege, S. P., et al., 2007. “Encapsulation and diffraction-pattern-correction methods to reduce the effect of damage in x-ray diffraction imaging of single biological molecules.” *Phys. Rev. Lett.* **98**, 198302. UCRL-JRNL-226873.

Hau-Riege, S. P., et al., 2007. "Subnanometer-scale measurements of the interaction of ultrafast soft x-ray free-electron-laser pulses with matter." *Phys. Rev. Lett.* **98**, 145502. UCRL-JRNL-223562.

He, Y. B., et al., 2007. "Critical parameters influencing the EUV-induced damage of Ru-capped multilayer mirrors." *J. Phys. Chem. C* **111**, 10988. UCRL-JRNL-227206.

Marchesini, S., 2007. "A unified evaluation of iterative projection algorithms for phase retrieval." *Rev. Sci. Instrum.* **78**, 011301. UCRL-JRNL-219850.

Marchesini, S., 2007. "Phase retrieval and saddle-point optimization." *J. Opt. Soc. Am. Optic. Image Sci. Vis.* **24**, 3289. UCRL-JRNL-226292.

Seibert, M., and Bogan, M., 2007. *DNA nanostructures in the gas phase*. XFEL Users' Mtg., Hamburg, Germany, Jan. 24–25, 2007. UCRL-PRES-231482.

van Tilborg, J., et al., 2007. *Experimental nonlinear studies of atoms and molecules with an intense vacuum ultraviolet photon source*. 38th Ann. Mtg. Division of Atomic, Molecular, and Optical Physics, Calgary, Canada, June 5–9, 2007. UCRL-ABS-231226.

Physics from the Main Injector Particle Production Experiment

Peter D. Barnes 05-ERD-007

Abstract

This project aims to deliver the essential physics cross sections needed for two applications: particle production and scattering cross sections relevant to future proton radiography facilities, and accurate physics models of the Neutrinos at the Main Injector (NuMI)/Main Injector Neutrino Oscillation Search (MINOS) neutrino beam at Fermilab, enabling high-accuracy neutrino measurements. The project supports analysis of the required physics data taken by the Fermilab E907 Main Injector Particle Production (MIPP) experiment, which can measure particle production and total cross sections from proton, pion, and kaon beams, from 5- to 120-GeV/c momentum, on nuclear targets from hydrogen to uranium, and on the Fermilab NuMI Beam Target for the MINOS Experiment.

The project will determine the cross sections with 2% relative and 5% absolute error. For NuMI/MINOS this will allow a reduction of the neutrino spectrum systematic error (the dominant systematic error) from 10% to approximately 2%. The charged-particle cross sections obtained for radiography will allow high-accuracy modeling and analysis of future experiments at Advanced Hydro Facility– (AHF-) class facilities. The project will result in publication of the cross sections, which will be put into LLNL databases and made available to simulation codes, and will enhance strong collaborations with DOE Office of Science–supported national laboratory and university groups.

Mission Relevance

The data analysis 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.

FY07 Accomplishments and Results

In FY07 we (1) developed the reconstruction algorithms, (2) completed particle identification in the detectors, and (3) wrote a paper describing the experiment and performance. Because algorithm reconstruction was more time-consuming than anticipated, we were unable to perform final cross section analysis or insert the data into LLNL data tables.

Publications

Abrams, R. J. et al. (2007). *The Fermilab MIPP experiment (FNAL-E907)*. UCRL-JRNL-235281.

Neutron Capture Cross-Section Measurements at the Detector for Advanced Neutron Capture Experiments

Winifred E. Parker 05-ERD-011

Abstract

Many important nuclear capture cross sections are not well known and are very difficult to model. Isotopes important to the slow neutron capture process have no experimental measurements at all, whereas some isotopes of interest to stockpile stewardship have existing measurements that are in marked disagreement with each other. We are measuring several neutron-capture cross sections using stable and radioactive targets—a novel and high-risk technique—with the Detector for Advanced Neutron Capture Experiments (DANCE) detector 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 for the DANCE array.

We expect to increase knowledge of the properties of unstable nuclei, including level-density information and statistical theory of gamma-ray decay. The discrepancy in existing europium data is expected to be resolved, so problems of europium and gadolinium cross sections for stockpile stewardship calculations may be solved. No measurements had been done on the americium nuclide that we propose to study. Furthermore, modern approaches involving quantification of margins and uncertainties require improved physics input. If this new technique involving unstable nuclei as targets is successful, the impact of this work will potentially be significant to astrophysics because current models give results that differ from global isotopic abundances by an order of magnitude.

Mission Relevance

This project supports the national security mission. If the project is successful, the unstable target approach will directly affect 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 production radionuclides, cross sections for their destruction are unknown.

FY07 Accomplishments and Results

In FY07 we (1) finished europium cross-section measurements and analysis of scissors-mode resonance, achieving an accurate europium-151,153 (n,gamma) cross sections for use in stockpile stewardship; (2) updated the gadolinium model, which is based on the europium model; (3) measured gadolinium-160, which was the final target for our gadolinium data set; (4) conducted gadolinium analysis in collaboration with North Carolina State University; (5) analyzed preliminary americium-242m data; and (6) fabricated a thicker americium-242m target. In summary, this project was a success as we provided capture cross sections for basic science and stockpile stewardship, and developed the ability to make radioactive targets for the DANCE array.

Publications

Agvaanluvsan, U. et al., 2007. *Neutron capture cross section on $^{151,153}\text{Eu}$* . UCRL-TR-234006.

Agvaanluvsan, U., et al., 2006. *Radiative neutron-capture studies using DANCE*. UCRL-POST-226629.

Agvaanluvsan, U., et al., in press. "The radiative strength function using the neutron-capture reaction on $^{151,153}\text{Eu}$." *Nucl. Instr. Meth. Phys. Res. B*. UCRL-PROC 215984.

Agvaanluvsan, U., et al., in press. "Progress on the europium neutron-capture study using DANCE." *Proc. 19th Intl. Conf. Application of Accelerators in Research and Industry*. UCRL-PROC-225343.

Dashdorj, D., et al., 2007. *Updates of gadolinium neutron-capture measurements at DANCE*. Ann. Mtg. Division of Nuclear Physics, Newport News, VA, Oct. 10–13, 2007. UCRL-PRES-235595.

Mitchell, G., et al., in press. *Spin and parity assignments for $^{94,95}\text{Mo}$ neutron resonances measured with the DANCE array*. Workshop on Photon Strength Function and Related Topics, Prague, Czech Republic, June 17–20, 2007. UCRL-PROC-233859.

Parker, W. E., et al., 2007. *Review of Livermore-led neutron capture studies using DANCE*. Los Alamos Neutron Science Center (LANSCE) User Group 8 Mtg., Santa Fe, NM, June 10–12, 2007. UCRL-CONF-231210.

Parker, W. E., et al., 2007. *Review of Livermore-led neutron capture studies using DANCE*. Intl. Conf. Nuclear Data for Science and Technology, Nice, France, April 22–27, 2007. UCRL-ABS-224774.

Parker, W. E., et al., 2007. *Neutron capture reactions for stockpile stewardship and basic science*. UCRL-TR-234820.

Parker, W. E., et al., 2007. *New measurement of the ^{242m}Am (n,γ) cross section using DANCE*. *Proc. DANCE Collaboration Workshop*. UCRL-PRES-229569.

Parker, W. E., et al., in press. "Review of Livermore-led neutron capture studies using DANCE." Los Alamos Neutron Science Center (LANSCE) User Group 8 Mtg., Santa Fe, NM, June 10–12, 2007. UCRL-PRES-231589.

Walker, C., et al., 2007. "Testing the possibility of using gadolinium for neutron capture therapy." Los Alamos Neutron Science Center (LANSCE) User Group 8 Mtg., Santa Fe, NM, June 10–12, 2007. UCRL-POST-231656.

Optical Properties as a Real-Time, In Situ Materials Diagnostic at Extreme Conditions

Jeffrey H. Nguyen 05-ERD-030

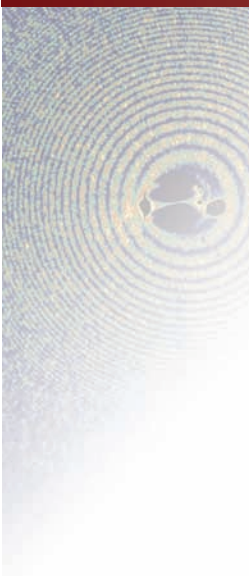
Abstract

Although characterizing the state of materials at subnanosecond time resolution is critical in experiments for 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 the Laboratory's Advanced Simulations and Computing (ASC) calculations of these same quantities. Direct comparison between theory and experiment will enable association of the observed optical properties with other materials properties by establishing spectroscopic fingerprints for each crystalline phase as well as the liquid.

We expect to develop a diagnostic capable of measuring in situ the structure of a material in real time as it is subjected to extreme temperatures and pressures in dynamic experiments at gas guns (at Los Alamos National Laboratory and at the Joint Actinide Shock Physics Experimental Facility at Livermore), high-power lasers, and Sandia's Z machine. Moreover, the real-time nature of the diagnostic will enable us to infer structural quantities while they are changing. 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 capable of characterizing material properties such as crystal structures and kinetics of phase transitions, 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 Sandia's Z machine to support basic science research. The project also leverages ASC theoretical



effort on optical properties for stockpile stewardship. The physical data obtained from this diagnostic is directly related to experimental milestones and can be used for validation of hydrodynamic codes.

FY07 Accomplishments and Results

In FY07, we measured the broadband emission of shocked and quasi-isentropically compressed tantalum at high temperatures and pressures. From the data, we were able for the first time to determine the temperature of tantalum along a high temperature quasi-isentrope. In correlation with the theoretical calculation of tantalum, emissivity and optical properties of tantalum can be deduced.

Publications

Nguyen, J. H., et al., 2007. *Kinetics of phase transition under tailored dynamic compression*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 23–29, 2007. UCRL-ABS-228667.

Patterson, J. R., 2007. *Making and characterizing off-Hugoniot states in gas gun experiments*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 23–29, 2007. UCRL-PRES-232157.

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 (thousands of degrees kelvin). Properties of high-pressure hydrogen in this regime are important to a range of basic and applied sciences, including condensed-matter theory, modeling efforts, planetary science, and hydrogen energy storage. High-pressure hydrogen is the subject of intense theoretical and experimental studies, both static and dynamic. Our proposed high-pressure and high-temperature experiments bridge the gap between static and dynamic experiments. We propose to apply state-of-the-art high-pressure, x-ray, laser, and spectroscopic capabilities. These studies address important issues such as the equation of state at high pressures and temperatures, phase lines, and novel phase transitions.

The rich physics in the regime between dynamic and static studies are targeted by our project. Hydrogen shockwave studies (150 GPa, 3000 K) reveal 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 properties to 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 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 high-temperature hydrogen directly address current and future needs in stockpile stewardship (specifically, the extreme dynamics of materials) and hydrogen energy storage, in support of LLNL's national and energy security missions.

FY07 Accomplishments and Results

In FY07, we measured the indirect and direct band gaps of hydrogen to be 4.67 eV at 165 GPa (indirect) and 223 Pa (direct). This measurement is the first-ever robust determination of the band gap of hydrogen at ultrahigh pressure, which is a fundamental parameter for benchmarking theoretical predictions of metallization. We also discovered evidence for a new high-temperature phase transition in solid deuterium. This evidence is supported by theoretical predictions and is the first-ever evidence of this new phase transition. These results achieved the very ambitious proposed goal of discovering new phase transitions in solid hydrogen, and our work has been published in the high-profile journal *Physical Review Letters*.

Proposed Work for FY08

In FY08 we will extend our studies to the fluid phase of hydrogen. Specifically, we will apply resistive and laser heating techniques to high-pressure diamond anvil cells to generate the necessary pressure and temperature conditions for achieving the liquid phase. Using laser spectroscopy and x-ray diffraction techniques, we will measure the hydrogen melt curve and structure of the liquid phase. Because hydrogen under these conditions is highly reactive, we will use novel techniques such as “tailored” and pulsed laser heating to preserve sample integrity. Our results will have broad, far-reaching importance to basic and applied sciences and will resolve controversies regarding hydrogen's melt curve.

Publications

Baer, B. J., W. J. Evans, and C. S. Yoo, 2007. *Coherent anti-Stokes Raman spectroscopy of deuterium at 298 K up to 190 GPa*. Joint 21st AIRAPT and 45th EHPRG Intl. Conf. High Pressure Science and Technology, Catania, Italy, Sept. 17–21, 2007. UCRL-ABS-231208.

Baer, B. J., W. J. Evans, and C. S. Yoo, 2007. *Coherent anti-Stokes Raman spectroscopy study of highly compressed deuterium*. American Physical Society Mtg., Denver, CO, Mar. 5–9, 2007. UCRL-PRES-228436.

Baer, B. J., W. J. Evans, and C. S. Yoo, 2007. “Coherent anti-Stokes Raman spectroscopy of highly compressed solid deuterium at 300 K: evidence for a new phase and implications for the band gap.” *Phys. Rev. Lett.* 235503. UCRL-JRNL-226865.

Baer, B. J., W. J. Evans, and C. S. Yoo, 2007. *High pressure deuterium studied using coherent anti-Stokes Raman spectroscopy*. American Physical Society Mtg., Denver, CO, Mar. 5–9, 2007. UCRL-ABS-226285.



Hydrodynamic, Atomic Kinetic, and Monte Carlo Radiation-Transfer Models of the X-Ray Spectra of Compact Binaries

Christopher W. Mauche 05-ERD-044

Abstract

This project models the x-ray spectra of compact binaries (i.e., white dwarf, neutron star, and black hole binaries) by constructing highly detailed, three-dimensional (3D) 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 are being used to interpret the existing Chandra and Newton X-Ray Multimirror Mission and future Constellation-X and X-Ray Evolving-Universe Spectroscopy high-resolution x-ray spectra of compact binaries.

The project will produce highly detailed, 3D time-dependent hydrodynamic models of the flow of plasma in compact binaries. These models are being used to produce the first realistic x-ray spectral models of compact binaries with sufficient detail to predict relative and absolute line strengths and shapes as a function of binary phase. Although we focused initially on high-mass x-ray binaries (HMXBs), the capabilities we will develop will be applicable to all types of x-ray sources dominated by photoionized 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 radiation-hydrodynamic codes that simulate nuclear explosives for the stockpile stewardship mission.

FY07 Accomplishments and Results

During FY07, we (1) hired a postdoctoral candidate to assist with our research, (2) performed 3D FLASH hydrodynamic simulations of HMXBs, (3) modified our Monte Carlo radiation-transfer code to accept FLASH output files, (4) used this code to conduct Monte Carlo calculations of the x-ray radiation transfer, (5) made comparisons between these model spectra and Chandra x-ray spectra of the HMXB Vela X-1, and (6) presented our results at two astrophysics conferences. We close this project having built all the tools needed to bring our research to fruition and with excellent prospects for additional Chandra x-ray grating observations of HMXBs with support from the National Aeronautics and Space Administration.

Publications

Akiyama, S., et al., 2007. "X-ray spectral formation in high-mass x-ray binaries: the case of Vela X-1." *Bull. Am. Astron. Soc.* **210**. UCRL-ABS-229350.

Mauche, C. W., D. A. Liedahl, and T. Plewa, 2007. "Hydrodynamical and spectral simulations of HMXB winds." *Bull. Am. Astron. Soc.* **331**. UCRL-ABS-223074.

Mauche, C. W., et al., in press. "Hydrodynamic and spectral simulations of HMXB winds." *Progr. Theor. Phys. Suppl.* UCRL-CONF-229626.

The Opacity of the Solar Interior

Pravesh K. Patel 05-ERD-045

Abstract

This project will make experimental measurements of the opacity of materials in the high-density, high-temperature regime of stellar physics. Opacity, which governs the transport of radiation 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. This project 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/c³), 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.

FY07 Accomplishments and Results

In FY07 we performed the most detailed plasma characterization experiments to date of a solid-density proton-heated target. These experiments, performed on the new Titan laser, used new techniques (optical and x-ray) to measure and optimize plasma temperature and uniformity. Fast time and spectrally resolved optical measurements enabled us to distinguish fast-electron heating from proton heating and to correlate the time history of self-emission to the incident proton energy.

Publications

Akli, K., et al., 2006. *Temperature sensitivity of Cu K-alpha imaging efficiency using a spherical Bragg reflecting crystal*. Conference on High-Temperature Plasma Diagnostics, Williamsburg, VA, May 7–11, 2006. UCRL-ABS-219295.

Beg, F., et al. 2007. *Recent work on production of hot plasmas and transport of hot electrons*. U.S.-Japan Fast Ignition Workshop, Otsu, Japan, Jan. 9, 2007. UCRL-PROC-228354.

Hey, D., et al., 2006. *Measurement of conversion efficiency of laser energy into accelerated protons*. 9th Intl. Fast Ignition Workshop, Cambridge, MA, Nov. 3–5, 2006. UCRL-ABS-222630.

Mackinnon, A. J., et al. 2006. *Studies of electron and proton isochoric heating for fast ignition*. 21st IAEA Fusion Energy Conference, Chengdu, China, Oct. 16, 2006. UCRL-ABS-218402.

Moon, S. J., et al., 2007. *Modeling the dynamics of reduced mass targets heated by ultra-intense lasers*. 6th Intl. Conf. High-Energy-Density Laboratory Astrophysics, Houston, TX, Mar. 11–14, 2006. UCRL-ABS-218529.

Optics Characterization for High-Energy, Split-Beam, Short-Pulse Lasers

Craig W. Siders 05-ERD-060

Abstract

With this project, we intend to provide critical technology for high-energy, split-beam, short-pulse (HESBSP) laser systems. These systems—with chirped-pulse amplification and a compact, folded compressor in a split-beam geometry—could significantly enhance high-energy-density (HED) science and inertial fusion facilities by enabling high-energy x-ray radiographs of laser-driven experiments. The goals of this project are evaluation of final focus and alignment of short pulses, as well as development of precision metrology for determining the alignment, in both space and time, of a focused split-beam pulse for fusion-class laser systems.

We will develop and test several key technology components for the back end of an HESBSP laser system. This project should solve most of the outstanding technology challenges associated with focusing and characterizing such lasers. The split-beam concept represents a new method for achieving very-high-power, short-pulse laser operations that will enable deployment of both a multiple time-frame and a multiple-view, high-energy K_α radiographic diagnostic capability. This work is also expected to result in numerous publications and new intellectual property.

Mission Relevance

New technology components for back-end and precision laser-pulse diagnostics are critical to the generation and compression of multikilojoule pulses in chirped-pulse amplification

systems. These systems support LLNL's national and energy security missions by advancing many areas of HED and inertial confinement fusion. These include (1) compression of multikilojoule pulses in chirped-pulse amplification systems for use in x-ray radiography; (2) scaling of high-energy, short-pulse systems to about 100 kJ using split-beam configurations with application to fast ignition; and (3) development of a novel pulse-metrology HESBSP technology with applications to future fusion-class lasers.

FY07 Accomplishments and Results

In FY07, we (1) completed construction and characterization of all spatial and spectral diagnostics; (2) integrated the diagnostics package into the HESBSP system and injected and characterized a surrogate laser pulse; (3) constructed an ultralow-energy inter-split-beam timing device using scanning cross-correlation; (4) demonstrated a single-shot, 200-ps window, third-order cross-correlator for pre-pulse contrast measurement with 106 dynamic range; and (5) and commissioned a device that can measure the near-transform-limited HESBSP using frequency-resolved optical gating. Our efforts for this project have set the stage to successfully conduct short-pulse laser-driven HED and inertial-confinement fusion experiments on advanced, high-power laser systems.

Publications

Barty, C. P. J., et al., 2007. *Advanced petawatt laser capabilities on the National Ignition Facility*. 7th Japan–U.S. Fast Ignition Workshop, Otsu, Japan, Jan. 9, 2007. UCRL-ABS-230163.

Crane, J., et al., 2007. *Alignment and diagnostics for high-energy petawatt systems*. 7th Japan–U.S. Fast Ignition Workshop, Otsu, Japan, Jan. 9, 2007. UCRL-PRES-227170.

Haefner, C., et al., 2007. *Development of diagnostics for high-energy petawatt*. Joint Conf. Ultrafast Optics and High Field Short Wavelength Phenomena, Santa Fe, NM, Sept. 2–7, 2007. UCRL-ABS-232484.

Jovanovic, I., et al., 2007. *High-dynamic-range, 200-ps window, single-shot cross-correlator for ultrahigh intensity laser characterization*. Conf. Laser and Electrooptics, Baltimore, MD, May 6–11, 2007. UCRL-POST-230745.

Rushford, M. C., et al., 2007. *Coherent addition of pulse for energy (CAPE) instrument and data fitting model study*. 13th Ann. Signal and Imaging Sciences Workshop, CASIS, Livermore, CA, Nov. 15–16, 2007. UCRL-PRES-225807.

Siders, C. W., et al., 2007. *10-kJ status and 100-kJ future for NIF petawatt technology*. Joint Conf. Ultrafast Optics and High Field Short Wavelength Phenomena, Santa Fe, NM, Sept. 2–7, 2007. UCRL-PRES-234491.

Precision Split-Beam, Chirped-Pulse, Seed Laser Technology

Jay W. Dawson 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:glass laser systems.

This project will develop seed laser technology for scaling high-energy, short-pulse laser beams on 40-cm-aperture neodymium: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.

FY07 Accomplishments and Results

In FY07 we successfully constructed a fiber laser system suitable for high-energy glass laser systems. This mode-locked fiber laser was phase-locked to an external clock and shown to be robust in a relatively harsh environment. Pulses were chirped using a chirped-fiber Bragg grating, amplified to 100 μJ , and then recompressed, upon which good pulse quality was verified. We measured pre-pulse contrast from the laser system and showed it to exceed 10^8 for picosecond pulses. Our system, which was packaged for mounting in standard 19-inch racks, can also run for long periods of time without operator interaction. Publications describing these achievements appeared in refereed journals.

Publications

An, J., et al., 2007. "Grating-less, fiber-based oscillator that generates 25-nJ pulses at 80 MHz, compressible to 150 fs." *Optic. Lett.* **32**, 2010. UCRL-JRNL-227086.

Dawson, J. W., et al., 2007. *Comparison of amplified spontaneous emission pulse cleaners for use in chirped-pulse amplification front-end lasers*. Ultrafast Optics Conf., Santa Fe, NM, Sept. 2–7, 2007. UCRL-CONF-232702.

Dawson, J. W., et al., 2007. *Fiber laser front ends for high-energy, short-pulse lasers*. IEEE Lasers and Electro-Optics Society Conf., Orlando, FL, Oct. 25, 2007. UCRL-CONF-232200.

Development of Hot, Tunable Radiation Sources for Material Science Studies and Simulating Radiation Transport in Dense Astrophysical Plasmas

Marilyn B. Schneider 05-ERD-068

Abstract

A high-temperature radiation source would enable studies of materials under extreme conditions to be performed at the OMEGA laser facility and other large-scale laser facilities. An appropriate source produces hot radiation, maintains a heated material sample in local thermodynamic equilibrium (LTE), and allows good (optically thin) experimental access to the heated sample. This project is developing such a hot radiation source and the spectroscopic techniques to characterize it. The source will be incorporated into the thin back wall of a reduced-scale hohlraum. Our research leverages previous efforts that developed radiation sources capable of heating targets to about 50 eV.

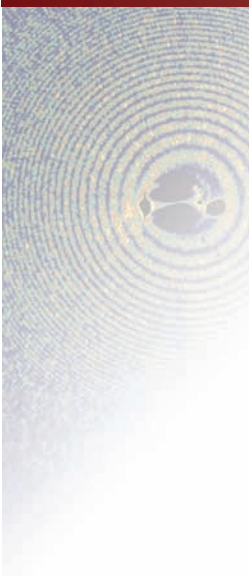
This project is producing a well-characterized hot radiation platform for studies of materials under extreme conditions. The platform enables the researchers to put a physics package in the location of the witness plate and, with the appropriate spectrometers and backlighters, to perform opacity or atomic physics studies.

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.

FY07 Accomplishments and Results

All FY07 milestones to complete the project were met. Specifically, we (1) performed a scaling study of the source temperature as a function of back wall thickness, hohlraum size and shape, and back wall composition; (2) used this source to heat a sample (chromium or titanium) and measured the soft x-ray emission spectra of the sample; (3) added a hydrodynamics model to an atomic kinetics code to model the experiment—this code uses the measured radiation drive (as a function of time) to predict the sample spectrum; (4) performed a secondary experiment that determined the electron temperature in the gold plasma of the laser deposition region from high-resolution L-shell spectra; and (5) developed



a hybrid atomic structure collisional–radiative model and benchmarked it against data from Super-EBIT, Livermore’s electron-beam ion trap facility.

Publications

Austheim-Smith, C., et al., 2006. *Calibration of a laser plasma diagnostic variable spaced reflection grating spectrometer using EBIT*. Twenty Years of Spectroscopy with EBIT, Berkeley, CA, Nov. 12–16, 2006. UCRL-POST-226044.

Cone, K. V., et al., 2006. *Calibration of a laser plasma diagnostic crystal spectrometer using LLNL’s EBIT facility*. Twenty Years of Spectroscopy with EBIT, Berkeley, CA, Nov. 12–16, 2006. UCRL-POST-226042.

Schneider, D. E., et al., 2007. “Development of a thermal x-radiation source using high energy density.” *Physics* **3**, 256. UCRL-JRNL-227227.

Schneider, M. B., et al., 2007. *Development of a thermal x-radiation source using “hot” hohlraums*. American Physical Society 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-POST-225415.

Schneider, M. B., et. al., 2006. “Plasma filling in reduced-scale hohlraums irradiated with multiple beam cones.” *Phys. Plasma* **13**, 112701. UCRL-JRNL-223616.

Widmann, K., et al. 2006. *Determination of the electron temperature of a hot gold plasma utilizing high-resolution x-ray spectroscopy of the L-band emission*. American Physical Society 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-ABS-223466.

Developing the Physics Basis of Fast-Ignition Experiments at Future Large Fusion-Class Lasers

Andrew J. Mackinnon 05-ERI-001

Abstract

This project seeks to establish the physics basis, measurement techniques, and numerical designs for future integrated fast ignition (FI) experiments, an innovative approach to achieving fusion. This research is being conducted on U.S. and foreign laser facilities, in collaboration with U.S. universities. The team comprises experts in FI modeling and experiments. Advanced modeling of the short-pulse ignition process with particle-in-code (PIC) and hybrid PIC codes have been benchmarked against experiments and used to assess short-pulse heating in the design of experiments at future fusion-class lasers. Experiments are investigating electron transport, developing diagnostics of electron and proton isochoric heating, and studying optimization of focused 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. The 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.

FY07 Accomplishments and Results

We (1) conducted experiments exploring electron transport, proton conversion from hydrogen-rich targets, and optimized proton focusing; (2) developed a prototype target design for fusion-class fast-pulse lasers using K fluorescence and neutron diagnostics to determine short-pulse particle energy delivery to the target; and (3) studied electron transport on slabs and with cone wire surrogates on the Vulcan laser. Over the course of this project we investigated hydrodynamics and electron and proton FI sources, published 21 papers (including articles in *Physical Review Letters* and *Physics of Plasmas*), and developed the point design of experiments to be conducted at the National Ignition Facility after it comes online. Follow-on work includes refining our experimental designs for future experiments at Omega and the National Ignition Facility and refining the optimum level of fluorescent doping for future experiments on Titan and Omega.

Publications

Akli, K., et al., 2007. "Temperature sensitivity of Cu K-alpha imaging efficiency using a spherical Bragg reflecting crystal." *Phys. Plasma*. **14**, 023102. UCRL-JRNL-219295.

Hatchett, S. P., 2007. "Hydrodynamics of conically guided fast-ignition targets." *Fusion. Sci. Tech*. **49**, 327. UCRL-JRNL-215884.

Mackinnon, A. J., et al., 2007. "Proton radiography of a laser-driven implosion." *Phys. Rev. Lett*. **97**, 045001. UCRL-JRNL-210529.

Notely, M., et al., 2007. "Development of time-resolved x-ray spectroscopy in high-intensity laser-plasma interactions." *Rev. Sci. Instrum*. **10**, 10f322. UCRL-CONF-221180.

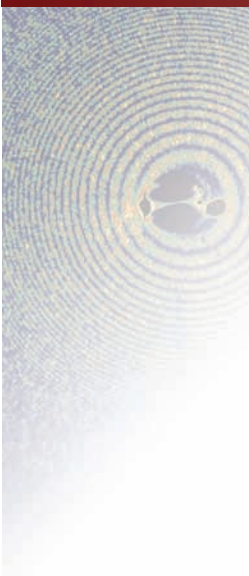
Key, M. H., 2007. "Status and prospects of the fast-ignition inertial fusion concept." *Phys. Plasma*. **5**, 055502. UCRL-TR-226302.

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. Dense matter at solid density and above will be probed 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, material science, and high-energy-density experiments. The



experiments have been conducted at multiple laser and synchrotron facilities, in collaboration with research teams at University of California campuses 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 plasmon density of states in cold matter and in dense plasmas.

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 also 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.

FY07 Accomplishments and Results

Using the benchmark data on the plasmon spectra that we demonstrated in FY06, we measured plasmons as a function of scattering angle and plasma density. The measurements, made at the Omega laser, succeeded in validating plasmon dispersion by comparing results from forward scatter at 25° with side scatter at 90°. These results demonstrated that density inferred from the plasmon shift is within 10% of the measurement made with Compton scattering. Overall, our experiments delivered data on all five proposed milestones and was of sufficient quality to test theoretical models in regimes of high interest to the Laboratory. The project was successful in observing plasmons and validating plasmon energy shift, and our results were published in several prominent journals.

Publications

Glenzer, S. H., 2007. "Observations of plasmons in warm dense matter." *Phys. Rev. Lett.* **98**, 065002. UCRL-JRNL-224979.

Glenzer, S. H., 2007. "X-ray Thomson scattering in dense plasmas." *AIP Conf. Proc.* **926**, 8. UCRL-PROC-231029.

Gregori, G., 2007. "Analytical derivation of static structure factor in strongly-coupled non-equilibrium plasmas for x-ray scattering studies." *High Energ. Dens. Phys.* **3**, 99. UCRL-JRNL-226394.

Hoell, A., 2007. "Thomson scattering at near-solid-density plasmas." *High Energ. Dens. Phys.* **3**, 120. UCRL-JRNL-226389.

Kritcher, A. L., 2007. "K-alpha conversion efficiency measurements for x-ray scattering in inertial confinement fusion plasmas." *High Energ. Dens. Phys.* **3**, 156. UCRL-JRNL-227913.

Reighard, A., in press. "Measurements of the cooling layer in a collapsed radiative shock in argon via Thomson scattering." *Phys. Rev. Lett.* UCRL-JRNL-235243.

Schwarz, V., 2007. "Hypernetted chain calculations for two-component plasmas." *Contrib. Plasma Phys.* **47**, 324. UCRL-JRNL-226395.

Sengebusch, A., 2007. "Shift of Cl K α and K β lines in laser-produced dense plasmas." *Contrib. Plasma Phys.* **47**, 1. UCRL-JRNL-227696.

Understanding the Nuclear Magnetic Fields

Peter Beiersdorfer 05-LW-006

Abstract

There is reasonable doubt if recently developed models of magnetic field generation in nuclei are correct, because a spectral line predicted by these models could not be found in a laser-excitation experiment. Correct models are needed for interpretation of parity nonconservation (PNC) experiments, as well as for nuclear materials and energy research. We propose to conduct high-precision measurements of the hyperfine structure in praseodymium, thallium, and bismuth to provide data for developing correct nuclear models and distinguishing between current models for $n = 2$ electron energy splittings in lithium-like ions. The experiment, carried out at the Livermore high-energy electron beam ion trap (SuperEBIT), utilizes passive emission spectroscopy with a high-resolution, soft-x-ray spectrometer.

Standard Model uncertainties stem largely from the uncertainties of various nuclear parameters. An improved knowledge of these parameters will be essential for further progress with PNC experiments. This project provides the necessary data to evaluate and distinguish among different models for generating magnetic fields in the nucleus, both near to and far from the doubly magic (both proton and neutron closed shells) nucleus praseodymium-208. The results will guide improved descriptions of the nuclear-atomic interaction and help select those needed for the most accurate evaluation of PNC experiments. Our measurements will be the first to use antisymmetric wave functions, and thus probe the nuclear magnetic fields differently.

Mission Relevance

The proposed work, at the interface of atomic and nuclear physics, supports the Laboratory's stockpile stewardship mission, insofar as it extends and enhances the Laboratory's competency in precision quantum electrodynamics of highly ionized atoms, and in understanding nuclear structure. It furthermore 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.

FY07 Accomplishments and Results

In FY07, we have completed our project as planned by measuring the hyperfine splitting effects of antisymmetric wave functions. The SuperEBIT has been used to measure the 2p excited-level hyperfine splitting in beryllium-like ions of bismuth, reaching an even higher precision (0.3%) than for the symmetric wave function splitting in the ground state of lithium-like ions of bismuth (0.6%). A first theoretical estimate is close to the experimental finding for beryllium-like bismuth ions, but theory is now struggling to match experimental accuracy. The experimental findings on lithium- and beryllium-like ions of praseodymium and bismuth provide the most accurate and complete data for understanding nuclear magnetic-field generation and are already being used in the planning of laser-excitation experiments in Germany.

Publications

Beiersdorfer, P., et al., in press. "Hyperfine splitting of the $2s_{1/2}$ and $2p_{1/2}$ levels in Li-like $^{209}\text{Bi}^{80+}$." *Phys. Rev. Lett.* UCRL-JRNL-234445.

Beiersdorfer, P., et al., 2007. *Measurement of the 2s hyperfine splitting in Bi^{80+}* . XXV Intl. Conf. Photonic, Electronic and Atomic Collisions, Freiburg, Germany, July 25–31, 2007. UCRL-ABS-229857.

Novel High-Energy-Density Source

James H. Hammer 06-SI-001

Abstract

High-energy-density science, the study of matter under extreme conditions, is a key to mastery of fission and fusion science and its applications. This project's objective is to develop a novel high-energy-density source of higher quality than achievable with other pulsed-power technologies. To this end, we will closely couple multidimensional code simulations and experiments. We expect to achieve a high-energy-density source that will be of broad utility to the Stockpile Stewardship Program and that will provide a capability complementary to future fusion-class lasers.

Mission Relevance

This work directly supports stockpile stewardship efforts at Lawrence Livermore by providing a new high-energy-density source for experiments, as well as by validating codes used for high-energy-density modeling.

FY07 Accomplishments and Results

For FY07, we (1) achieved significant advances in computational modeling of the core concept, bringing experiments and simulation into much better agreement than previously obtained; (2) conducted further analysis of a novel thermal x-ray source that showed even higher performance than early estimates and that has potential application to a range of high-energy-density experiments; and (3) conducted experiments to improve our understanding of a key physics issue, along with an analytic analysis of the requirements for an accurate computational model. Our analytic understanding, combined with improved agreement between models and experiment, suggests that we are converging on a validated predictive model.

Proposed Work for FY08

In FY08, we will (1) continue to extend source development, characterize output, and develop a stable, reproducible platform for high-energy-density experiments; (2) make further improvements in predictive capability by detailed comparisons of calculational models with data; (3) make innovative variations on the core concept; and (4) use the improved models to evaluate the potential of the concept for a variety of future applications.

Active Detection and Imaging of Nuclear Materials with High-Brightness Gamma Rays

Christopher P. Barty 06-SI-002

Abstract

This proposal leverages LLNL's world-leading capabilities in laser science, x-ray source development, and nuclear science to enable a new class of active interrogation techniques. We propose to demonstrate the use of new linac/laser-based gamma-ray sources for isotopic imaging of well-shielded objects, thus providing an effective means of detecting concealed highly enriched uranium, including ^{235}U . Isotopic selectivity of the detection scheme is based on nuclear resonance fluorescence from target nuclei. Specifically, these efforts include (1) finding nuclear resonance fluorescence lines in ^{235}U and ^{239}Pu ; (2) developing, modeling, and demonstrating linac/laser-based, high-brightness x-ray generation; (3) demonstrating nuclear resonance fluorescence detection of ^{238}U ; and (4) developing modeling tools for optimizing nuclear resonance fluorescence-based detection and imaging.

If successful, this project will create the world's brightest gamma-ray source and enable, for the first time, an effective detection modality for hidden highly enriched uranium. This detection capability could launch "nuclear photo science" as a new field of study and result in numerous follow-on applications. This capability represents a potential solution to a longstanding challenge of significant importance to homeland security and nonproliferation. A completely new concept, Compton scattering in thermonuclear plasmas, and simulations of weapons-relevant inverse-density radiography will result from this research.

Mission Relevance

The ability to detect and image highly enriched uranium would support LLNL's national security mission, specifically homeland security and counterproliferation. Furthermore, the picosecond, high-spatial-resolution, tunable, megaelectronvolt source capability developed as part of this proposal would also impact a wide range of applications of importance to stockpile stewardship (in support of national security) and high-energy-density science and technology (in support of national and energy security).

FY07 Accomplishments and Results

In FY07, we discovered a deeply penetrating 2.143-MeV nuclear resonance fluorescence line in ^{239}Pu , ideal for detection. Our new magnesium-based photocathode radiofrequency gun and linac upgrade yielded exceptional results: a greater than 120-MV/m gradient and 3-nC charge, 5×10^{-4} quantum efficiency (50 times greater than a conventional copper photocathode), 2- μm emittance, 50-MeV acceleration, and 40- μm focus. This was made possible by successfully completing a ultraviolet fiber laser using a highly stable fiber oscillator and coupling 1-ps, 100- μJ , 263-nm pulses to a hyper-Michelson temporal shaper.

Proposed Work for FY08

In FY08, we will (1) complete the interaction laser and achieve first light; (2) perform an in-depth characterization and optimization campaign of the x-rays and electron beam, which will result in world-record electron beam brightness and enable nuclear resonance fluorescence experiments with ^{238}U at 680 keV; (3) produce conventional, high-precision radiographs in the 500-keV range, along with uranium K-edge imaging at 116 keV; (4) perform an inverse-density radiography experiment with ^7Li at 477 keV; and (5) continue our work on nonlinear trapping and pulse recirculation, along with the study of x-ray scanning, inverse-density charge-coupled device imaging, and spectral brightness optimization, with the ultimate goal of generating extremely narrow-band x-rays for direct nuclear resonance fluorescence excitation and imaging.

Publications

Anderson, S. G., et al., 2007. *Commissioning of a high-brightness photoinjector for Compton scattering x-ray sources*. 22nd Particle Accelerator Conf., Albuquerque, NM, June 25–29, 2007. UCRL-PROC-232211.

Barty, C. P. J., F. V. Hartemann, and C. W. Siders, 2007. *Ultrabright gamma-ray beams with lasers: nuclear photoscience and applications*. Ultrafast Optics 2007, Santa Fe, NM, Sept. 2–7, 2007. UCRL-PRES-233703.

Barty, C. P. J., et al., 2006. *Possibilities for nuclear photo-science with intense lasers*. Intl. Conf. Ultrahigh Intensity Lasers, Cassis, France, Sept. 25–29, 2006. UCRL-PROC-222451.

Gibson, D. J., et al., 2006. *Electron beam optimization for a high-brightness gamma-ray source*. 48th Annual Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-ABS-223370.

Gibson, D. J., et al., 2007. *Optimal design of a tunable Thomson-scattering based gamma-ray source*. 22nd Particle Accelerator Conf., Albuquerque, NM, June 25–29, 2007. UCRL-PROC-231689.

Hartemann, F. V., C. W. Siders, and C. P. J. Barty, 2007. *Compton scattering in ignited thermonuclear plasmas*. Ultrafast Optics 2007, Santa Fe, NM, Sept. 2–7, 2007. UCRL-PRES-233911.

Hartemann, F. V., et al., 2007. “Compton scattering x-ray sources driven by laser Wakefield acceleration.” *Phys. Rev. Spec. Top. Accel. Beams* **10**, 011301. UCRL-JRNL-216408.

Hartemann, F. V., et al., 2007. *Gamma-ray Compton light source development at LLNL*. 22nd Particle Accelerator Conf., Albuquerque, NM, June 25–29, 2007. UCRL-PROC-232248.

Hartemann, F. V., et al., 2007. *Gamma-ray Compton light source development at LLNL*. American Physical Society March Meeting, Denver, CO, Mar. 5–9, 2007. UCRL-ABS-227156.

Hartemann, F. V., et al., 2007. *Gamma-ray Compton light source development at LLNL*. IEEE Pulsed Power and Plasma Science Conf., Albuquerque, NM, June 17–22, 2007. UCRL-PROC-233910.

Hartemann, F. V., et al., 2006. *Nuclear photo-science and applications with T-Rex sources*. 2006 Linear Accelerator Conf., Knoxville, TN, Aug. 24–25, 2006. UCRL-PROC-224082.

McNabb, D. P., et al., 2006. *Mapping isotopic distributions in cargo to detect SNM and its configuration*. Division of Nuclear Physics Annual Mtg., Nashville, TN, Oct. 25–28, 2006. UCRL-ABS-222552.

McNabb, D. P., et al., 2007. *Mapping isotopic distributions inside enclosures with Thomson-backscatter sources to detect SNM and its configuration*. Intl. Conf. Nuclear Data for Science and Technology, Nice, France, Apr. 22–27, 2007. UCRL-ABS-224351.

Shverdin, M. Y., et al., 2007. *Fiber-based, spatially and temporally shaped picosecond UV laser for advanced RF gun applications*. 22nd Particle Accelerator Conf., Albuquerque, NM, June 25–29, 2007. UCRL-CONF-231791.

Shverdin, M. Y., et al., 2007. *High power picosecond laser pulse recirculation*. Nonlinear Optics, Kona, HI, July 30–Aug. 3, 2007. UCRL-PROC-229440.

Shverdin, M. Y., et al., 2007. *High power picosecond laser pulse recirculation*. Posipol 2007 Workshop, Paris, France, May 23–25, 2007. UCRL-PROC-230727.

The Ultrafast Lattice Response of the Shocked Solid

Hector E. Lorenzana 06-SI-004

Abstract

Our goal is to perform the first studies exploiting the new generation of bright, ultrafast x-ray sources to investigate real-time dynamics of shocked materials at characteristic temporal and spatial scales of the controlling physical phenomena. We will measure the microscopic state of materials and their evolution under shock loading by a combination of dynamic x-ray diffraction and scattering. We will characterize, in situ, two phenomena: condensed-matter phase transformations and incipient damage. We will couple our experimental results with state-of-the-art simulations which, for the first time, will match the length and time scales of the experiments. These results will lay the foundation for next-generation physics-based models of material behavior under extreme conditions.

We expect to perform the first comprehensive real-time, in situ study of the lattice response under shock loading, a problem of great scientific importance. Lattice-level properties of solids undergoing phase transitions, plastic deformation, or void growth during a shock are largely unknown, yet this information is crucial for a predictive understanding of material response and failure. Success in this endeavor through laser and synchrotron-based experiments coupled with simulations will position LLNL at the forefront of a new field—high-energy-density materials science. The close coupling of experiment and theory will validate existing modeling at the atomistic, mesoscopic, and continuum scales, and spur development of the next generation of dynamic material models.

Mission Relevance

Success in stockpile stewardship, high-energy-density science, and other national security missions demand a fundamental understanding of dynamic materials behavior. The expertise developed in this project will further current and future research in support of stockpile stewardship facilities such as the Joint Actinide Shock Physics Experimental Research gas gun at the Nevada Test Site and high-pressure materials studies on advanced fusion-class lasers, as well as basic science facilities such as the Linac Coherent Light Source at Stanford. This work will drive development of computational tools for predicting performance, safety, and reliability of nuclear weapons as well as for high-power laser research.

FY07 Accomplishments and Results

In FY07, we continued studies of the in situ, lattice-level dynamic response of the shocked solid. Specifically, we (1) utilized both molecular dynamics simulations and experiments to investigate the one- to three-dimensional plastic transition and coexistence region of a solid–solid phase transformation, (2) demonstrated that the lattice response along the [100] direction of iron exhibits no plasticity at short (<5 ns) timescales and that the recovered microstructure after transformation and release returns to a single crystal, (3) modeled loading/unloading history in a continuum simulation using Livermore-developed crystal plasticity codes, (4) demonstrated a polycrystalline diffraction methodology for investigating shocked materials and, (5) installed and tested a laser drive system at the Advanced Photon Source for in situ damage studies.

Proposed Work for FY08

In FY08, we will continue the study of transformation phenomena by examining lattice response to shock loading along different crystallographic directions, including transformation pressures, timescales, and microstructure. We will (1) use polycrystalline diffraction techniques to investigate, for the first time, general shock-driven transformations, including melt and refreeze phenomena; (2) continue exploring pressure effects on one- to three-dimensional lattice-relaxation timescales; (3) conduct synchrotron-based work to characterize time-dependent, shock-driven void growth in a metal, using both experiment and theory; and (4) use diffuse scattering to measure defect nucleation rates and density in a single-crystal metal.

Publications

Bringa, E. M., et al., 2007. "Shock deformation of fcc metals on sub-nanosecond time scales." *Nat. Mater.* **5**, 805. UCRL-JRNL-210478.

El-Dasher, B., et al., 2007. *Recovery studies of shocked iron single crystals*. 15th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 23–29, 2007. UCRL-ABS-226301.

Garcia-Baonza, V. G., et al., in press. "Temperature dependence of phonons from an anharmonic potential." *J. Phys. Chem.* UCRL-JRNL-230701.

Hawreliak, J., 2007. *In-situ probing of lattice response in shock compressed materials using x-ray diffraction*. 15th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 23–29, 2007. UCRL-PROC-233046.

Hawreliak, J., et al., 2007. "An analysis of the x-ray diffraction signal for the (alpha)–(epsilon) transition in shock-compressed iron: simulation and experiment." *Phys. Rev. B* **74**, 184107. UCRL-JRNL-220707.

Hawreliak, J., et al., 2006. *Modeling planetary interiors in laser based experiments using shockless compression*. 6th Intl. Conf. High Energy Density Laboratory Astrophysics, Mar. 11–14, 2006, Houston, TX. UCRL-PROC-221172.

Hawreliak, J., et al., 2007. "Nanosecond diffraction from polycrystalline and amorphous materials in a pinhole camera geometry suitable for laser shock compression experiments." *Rev. Sci. Instrum.* **78**, 083908. UCRL-JRNL-227167.

Hawreliak, J., et al., 2005. *Shock induced (alpha)-(epsilon) phase change in iron: analysis of MD simulations and experiment*. 14th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 5, 2005. UCRL-PROC-215261.

Remington, B. A., et al., 2007. "Material dynamics under extreme conditions of pressure and strain rate." *Mater. Sci. Tech.* **22**, 474. UCRL-JRNL-215701.

Wark, J. S., et al., 2005. *Picosecond x-ray diffraction from laser-shocked copper and iron*. 14th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Baltimore, MD, July 31–Aug. 5, 2005. UCRL-CONF-215010.

Laser-Driven Dynamic Hohlraums

Sharon Glendinning 06-ERD-017

Abstract

This project proposes to use experiments and simulations to explore the laser-driven dynamic hohlraum (LDDH), a laser-driven analog to the dynamic hohlraum used at the Sandia Z machine, as an ignition target for future fusion-class lasers. The project will investigate the neutron production history of capsules driven with an LDDH, determine the radiation production during an implosion, use polar direct drive on the Omega laser, and optimize the LDDH as a potential bright x-ray source and general hohlraum driver. Experiments will be fielded on the Omega laser to test simulations of the LDDH addressing each of these points.

We expect to demonstrate the feasibility of the LDDH as a candidate for an ignition target, x-ray source development, and hohlraum driver for diverse high-energy-density physics experiments in regimes not otherwise achievable. A noncryogenic ignition option could allow for more ignition experiments at lower cost than currently feasible, making possible more experiments on the effects and uses of ignition.

Mission Relevance

The LDDH, once demonstrated, could become a platform for a variety of high-energy-density physics experiments that support the Laboratory's stockpile stewardship mission. Radiatively driven shocks are also of great interest in basic science, especially astrophysics, and thus this project supports the Laboratory's mission in breakthrough science.

FY07 Accomplishment and Results

In FY07, we conducted our first experiments focused on creating a bright x-ray backlighting source. Simulations predicted that an LDDH filled with krypton would give a smooth spectrum above 3 keV (essential for opacity experiments) and that a krypton-filled LDDH would produce about twice the x-ray yield of a comparable single-shell implosion using hydrogen. Two-dimensional simulations predicted that the source could be generated even with a very asymmetric implosion. These predictions were confirmed in krypton-filled LDDH experiments fielded on the Omega laser. In addition, we performed double shell (ignition-relevant) experiments studying the effect of nonuniform irradiation (polar direct drive) on shell symmetry and neutron yield for comparison with simulations.

Proposed Work for FY08

We will capitalize on our backlighter success in FY07 by designing and testing an LDDH backlighter that should have bright emission in a higher-energy spectral band (>6 keV) and by demonstrating the use of this backlighter to characterize a hot plasma by measuring the absorption spectrum of a plastic-tamped, laser-heated nonlocal-thermodynamic-equilibrium iron sample. In addition, we intend to field experiments using capsules that are a closer match to future fusion-class laser hohlraums. Simulations indicate that inner capsules with a

high-Z dopant in the shell would be more suitable for the drive (the ignition LDDH uses a gold alloy inner capsule). This will allow us to test our simulations of drive coupling and hence of scalability to future laser ignition for the LDDH.

Publications

Glendinning, S. G., et al., 2006. *Progress in laser-driven dynamic hohlraum implosions*. 48th Mtg. APS Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-PRES-225662.

Hansen, J. F., S. G. Glendinning, and R. F. Heeter, 2007. *Dynamic hohlraums as x-ray sources in high-energy-density science*. 49th Mtg. APS Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-ABS-232402.

Hansen, J. F., et al., in press. “Dynamic hohlraums as x-ray sources in high-energy-density science.” *Rev. Sci. Instr.* UCRL-JRNL-233875.

Measurements to Facilitate Advanced Tokamak Science in Burning Plasma Experiments

Steven L. Allen 06-ERD-024

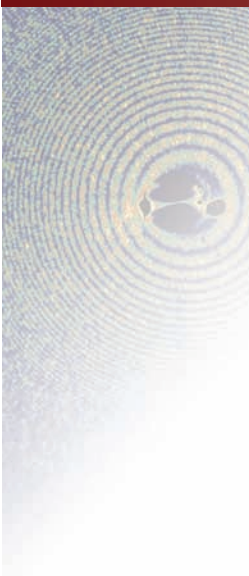
Abstract

Measuring the plasma current profile in advanced tokamaks is currently based on the motional Stark effect (MSE). When a high-energy neutral beam interacts with the plasma in a magnetic field, the MSE results in a polarized photon that can be used to measure the local plasma current and electric field. This project will investigate the possibility of making high-resolution measurements of the plasma current profile in a burning plasma environment. This will include modeling the spectrum and propagation through a polarizing-preserving optical train, detecting the full modulated-intensity spectrum, making detailed magnetohydrodynamic measurements with appropriate processing of the high-time-resolution measurements, and developing system calibration techniques.

We will carry out a conceptual study of an MSE diagnostic on several levels. On the modeling side, the plasma code CORSICA will be upgraded with synthetic diagnostics to calculate the actual MSE signals in a burning plasma. Because the measurement will be in a high-neutron environment, we will develop the capability to optimize the optical design with the constraint of neutron shielding. Adequate shielding favors multiple mirrors, but accurate measurements favor a simple, no-mirror system. We will characterize prototypical optics and model their polarization performance. Polarization-sensitive optics may be degraded by plasma exposure, and we would start these measurements in existing devices.

Mission Relevance

This project will contribute to burning plasma fusion research, an important step in achieving fusion power for the country's long-term energy needs. The combined development of a new



diagnostic tool and a new predictive code for the International Thermonuclear Experimental Reactor scenarios will advance plasma physics research, in support of LLNL and DOE missions in fundamental science and energy security.

FY07 Accomplishments and Results

In FY07, we (1) collaborated with the University of Arizona Polarization Lab to obtain Mueller matrices measurements of a precision dielectric mirror typical of that used in current tokamaks; (2) analyzed the mirror after plasma treatment that could be used to remove tritium from the vessel walls; (3) made the first Mueller matrix measurements of a rhodium mirror, a prototype for burning plasma machines; (4) developed a model of beam transport through a neutral gas, compared it with experimental data, and submitted the results for publication; and (5) developed, with the University of Arizona, a new in situ calibration technique, which we are using to characterize the polarization response of systems on existing tokamaks.

Proposed Work for FY08

For FY08, we will (1) improve our calculation of the MSE spectrum; (2) conduct Mueller matrix measurements of metal and dielectric mirrors in collaboration with the University of Arizona; (3) model a typical polarization optical train with these measured matrices; (4) process the full temporal spectrum of a typical system, enabling plasma magnetohydrodynamic and system polarization evaluation; (5) develop a low-Faraday rotation optical train (vacuum window); (6) examine system calibration techniques that are appropriate for a burning plasma environment; and (7) begin measurements of the MSE spectrum with fast digitizers.

Publications

Gu, M., in press. "Atomic models for motional Stark effects diagnostics." *J. Phys. B Atom. Mol. Opt. Phys.* UCRL-JRNL-233134.

Investigating New Regimes of Material Strength at Ultrahigh Strain Rates and Pressures

Stephen M. Pollaine 06-ERD-027

Abstract

In this project, we propose to measure material strength of metals at high pressures (>1 Mbar) and strain rates up to 10^9 /s by developing two new VISAR techniques: compression wave reverberations in thin samples and hysteresis between loading and unloading waves in thick samples. We will also develop a new hohlraum isentropic compression experiment (ICE) drive that will serve as a platform for these techniques. This new technology will test various models of material strength, such as that of Preston–Tonks–Wallace, and will provide parameters for those models. This project will be exploring new regimes never before measured, where current models of strength differ dramatically from each other.

We expect to develop two new experimental techniques for measuring strength and a new hohlraum ICE drive platform that will deliver larger, more uniform loading than is currently possible. These techniques and platform will be extended to the National Ignition Facility, Joint Actinide Shock Physics Experimental Research, Phoenix, and Sandia's ZR facility programs. In this project, we will take the first-ever time-resolved measurements of the strength of nanocrystalline materials at ultrahigh strain rates. Also, our work will contribute to understanding the effect of strength on the shock processing of interstellar dust grains, which affects the size distribution of the grains and, indirectly, the rate of star formation. Our results will be submitted for publication in peer-reviewed physics and materials science journals.

Mission Relevance

The new VISAR techniques will make it possible to measure the strength of materials important for stockpile stewardship on existing facilities, while the development of the hohlraum-ICE drive is a critical advance for stockpile stewardship experiments on fusion-class lasers. Investigating new regimes of material strength at high strain rates and pressures also contributes to the Laboratory's mission in breakthroughs in fundamental science.

FY07 Accomplishments and Results

In FY07, we (1) demonstrated that our Omega halfraums remain open past 80 ns, and simulations showed they can be scaled to fusion-class lasers, with a 195-eV drive; (2) developed a new way of measuring preheat in these hohlraums, with an unprecedented sensitivity of 100 K; (3) demonstrated the first hohlraum on the Janus laser, reaching 110 eV; (4) completed the investigation of the Rayleigh-Taylor feed-through method of measuring strength and eliminated it as a viable candidate, allowing us to concentrate our efforts on the remaining two techniques; (5) developed a new reverse-reservoir technique for delivering a pressure drive that drops sharply—needed for the hysteresis strength technique—and collected our first data with the technique using aluminum at 350 kbar; and (6) published a paper on the high-planarity halfraum drive in *Physics of Plasmas*.

Proposed Work for FY08

In FY08, we will (1) extend our strength measurements to the mini-Z and gas guns, to span strain rates from 10^5 to 10^8 /s at pressures from 0.5 to 1.0 Mbar; (2) continue our strength experiments on Janus and Omega using our halfraum platforms; (3) analyze all data to show how material strength varies with strain rate in the transition from the thermal-activation regime to the phonon-drag regime; and (4) measure, for the first time ever, the dynamic strength of nanocrystalline materials and the material of interstellar grains.

Publications

Pollaine, S., et al., 2007. *Material strength hohlraum development*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 24–29, 2007. UCRL-PRES-231876.

Smith, R. F., et al., 2007. "High-planarity x-ray drive for ultra-fast shockless-compression experiments." *Phys. Plasma* **14**, 057105. UCRL-JRNL-229932.

The Properties of Confined Water and Fluid Flow at the Nanoscale

Eric R. Schwegler 06-ERD-039

Abstract

The properties of confined water affect a wide range of scientific and technological areas of interest, including protein folding, cell-membrane flow, materials properties in confined media, and nanofluidic devices. We propose to develop accurate computational tools to study fluids in confined, nanoscale geometries, and to apply these techniques to probe the structural and electronic properties of water confined between hydrophilic and hydrophobic substrates, including the presence of simple ions at the interfaces. In particular, we will use a series of ab initio molecular dynamics simulations to build an understanding of how hydrogen bonding and solvation are modified at the nanoscale.

The primary results of this research project will be threefold: (1) a first-principles-based computational framework for investigating and characterizing liquids that are confined at the nanoscale, (2) a better understanding of how the properties of water change upon nanoscale confinement by hydrophobic and hydrophilic surfaces, and (3) a suitable empirical model that can be used to describe the flow of water in confined media.

Mission Relevance

This project will contribute to a detailed understanding of how the structural, electronic, and dynamical properties of water are modified upon nanoscale confinement, which is relevant to the development of nanoscale-material sensor technologies for national security missions in nonproliferation and counterproliferation, as well as for homeland security missions in counterterrorism.

FY07 Accomplishments and Results

In FY07, we have (1) completed a series of first-principles simulations of water confined by representative hydrophobic (graphitic sheets and carbon nanotubes) and hydrophilic (silicon carbide) surfaces, and the solvation of small hydrophobic solutes (benzene and hexafluorobenzene); (2) completed an initial investigation of ion solvation in carbon nanotubes; (3) performed calculations of the x-ray absorption spectra of confined water; and (4) prepared two manuscripts describing our work on hydrophobic surface confinement, which includes a comparison of first-principles and classical molecular dynamics results and our work on hydrophobic solutes.

Proposed Work for FY08

In FY08 we will (1) conduct ab initio molecular dynamics simulations of ion solvation in a carbon nanotube, then directly compare our results to experimental measurements; (2) evaluate the use of empirical potentials that explicitly account for polarization effects to reproduce the properties of confined water; (3) develop and apply a computational framework for carrying out fluid-flow simulations with classical molecular dynamics; and (4) determine

the spectroscopic signatures of confined water based on nuclear magnetic resonance and x-ray absorption spectra.

Publications

Allesch, M., E. Schwegler, and G. Galli, 2007. "Structure of hydrophobic hydration of benzene and hexafluorobenzene from first principles." *J. Phys. Chem. B* **111**, 1081. UCRL-JRNL-225529.

Schwegler, E., 2007. *Ab-initio simulations of interfacial systems*. 2007 Defense Threat Reduction Agency Computational Chemistry Workshop, Maui, HI, Aug. 13, 2007. UCRL-PRES-235665.

Schwegler, E., 2007. *Ab-initio simulations of materials and molecular systems*. SciDAC 2007, Boston, MA, June 24–28, 2007. UCRL-PRES-233319.

Schwegler, E., 2007. "First principles simulations of aqueous solutions." *J. Phys.* **78**, 012065. UCRL-JRNL-232936.

Schwegler, E., 2007. *Ion solvation in confined water: a first-principles molecular dynamics investigation*. 2007 Am. Phys. Soc. March Mtg., Denver, CO, Mar. 5–9, 2007. UCRL-ABS-226642.

Spheromak Energy Transport Studies via Neutral Beam Injection

Harry S. McLean 06-ERD-042

Abstract

Promising results from the Sustained Spheromak Physics Experiment (SSPX) at Livermore provide impetus for adding neutral beam injection (NBI) heating to advance the physics of energy transport and pressure limits for the spheromak. Our project will represent the first-time application of NBI auxiliary heating to a spheromak fusion reactor. Design studies will determine optimum injection angles, desired target plasma conditions, timing, and spheromak operating conditions and diagnostics needed to produce the desired transport data.

Successful application of NBI heating to a spheromak plasma would be a significant step forward for this magnetic fusion energy design—past experience with other experiments have shown NBI to be a powerful driver for significantly enhancing the quality of scientific output. We expect to (1) develop and apply simulation tools for studying auxiliary heating in spheromak plasmas that could be extended to other magnetic fusion concepts; (2) increase visibility for the SSPX experiment, both nationally and internationally, by showing the feasibility of using neutral beams to heat spheromak plasmas; and (3) position the DOE-funded SSPX for strong scientific contributions during the next three years. A stronger SSPX program will enhance the Laboratory's ability to attract top-quality students, postdoctoral researchers, and scientists.

Mission Relevance

Plasma physics is at the core of the Laboratory's national security mission. The proposed work would provide a testbed for computational simulations from the NIMROD three-dimensional magnetohydrodynamics code. Research in magnetohydrodynamics theory and magnetic-field fluctuation theory also supports the Laboratory's mission in breakthroughs in fundamental science and technology.

FY07 Accomplishments and Results

In FY07, we (1) simulated beam heating with the Corsica integrated magnetic-fusion code using new data from long-pulse plasmas produced by experiments; (2) developed target plasmas suitable for application of NBI auxiliary heating and measurement of the time history of electron temperature; (3) explored the pressure limit for predicted spheromak target-plasma conditions; (4) performed initial measurements of ion temperature, plasma rotation, and plasma fluctuations using a newly developed, compact neutral-particle analyzer and ion-Doppler spectrometer; and (5) completed installation and performed measurements with a double-pulse Thomson scattering system. Results of our studies have been prepared for submittal to *Nuclear Fusion*.

Publications

Cohen, B. I., et al., 2007. *NIMROD simulations of the SSPX spheromak studying viscosity and artificial density diffusivity, and linear MHD stability analysis of SSPX*. Intl. Sherwood Fusion Theory Conf. Annapolis, MD, Apr. 23–25, 2007. UCRL-ABS-228288.

Hill, D. N., et al., 2006. *Confinement studies in high temperature spheromak plasmas*. 21st IAEA Fusion Energy Conf., Chengdu, China, Oct. 16–21, 2006. UCRL-CONF-225553.

Jayakumar, R., et al., 2007. *Studies on neutral beam injection into the SSPX spheromak plasma*. UCRL-TR-235704.

King, J., et al., 2007. *Ion Doppler spectroscopy measurements on SSPX*. 49th Ann. Mtg. Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-ABS-232994.

LoDestro, L. L., et al., 2007. *Linear MHD stability analysis of the SSPX spheromak*. Innovative Confinement Concepts Workshop, College Park, MD, Feb. 12, 2007. UCRL-ABS-226899.

Ludington, A. R., et al., 2007. *Time-resolved temperature measurements in SSPX*. UCRL-TR-223809.

McLean, H. S., et al., 2007. *SSPX achievements and future directions for spheromak research*. 49th Ann. Mtg. Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-ABS-232976.

Heavy Quark Jet Tomography of Compressed Nuclear Matter

Ron Soltz 06-ERD-045

Abstract

This project is investigating the compressed nuclear medium created in high-energy nuclear collisions at the Relativistic Heavy Ion Collider (RHIC) and the Large Hadron Collider (LHC) using heavy quark jets. The azimuthal correlations of electrons with associated charged hadrons can be used to identify jets containing heavy flavor quarks, which lose energy as they traverse a quark–gluon plasma (QGP). We will use the PHENIX experiment at RHIC to compare e-h correlations in p + p, d + gold, and gold + gold collisions. At the same time, we are preparing to move to the next frontier of heavy-ion physics at the LHC, scheduled to begin operations in FY07. We will also use the ALICE experiment's electromagnetic calorimeter to study the heavy quark jet tomography of the QGP produced at the LHC.

This research is of high significance for heavy-ion physics and will contribute to our understanding of the dense matter formed at RHIC and LHC. The characterization of the mass dependence of jet quenching will help explain the underlying coupling of quarks and gluons in high-temperature nuclear matter and enable the quantitative evaluation of the properties of the QGP. We expect to publish our results in peer-reviewed journals.

Mission Relevance

Jet tomography is a promising and active area of research in relativistic heavy ion physics, which supports LLNL's missions in stockpile stewardship, energy security, and breakthroughs in fundamental science and technology. Our scientific leadership in this area will attract top postdoctoral applicants to the Laboratory. Training postdoctoral researchers in relativistic heavy ion physics has proven to be an effective means of cultivating talent and new detection concepts for the Laboratory's efforts in nonproliferation and homeland security.

FY07 Accomplishments and Results

In FY07 we (1) completed the simulations needed for corrections and theoretical comparisons of the e-h correlations at RHIC and published a paper on results from the gold + gold collisions, (2) hired a postdoc to work on PHENIX and ALICE, (3) collected data during our seventh RHIC run, (4) continued development of the offline software for the electromagnetic calorimeter, (5) participated in the first LHC run on p + p collisions, and (6) helped install and commission the first electromagnetic calorimeter module. This work will be continued by the DOE Office of Science as part of the ALICE collaboration, in which LLNL has established a leadership role. In addition, theoretical calculations that we conducted on BluGene/L using the techniques of lattice quantum chromodynamics were instrumental in a team with LLNL researchers winning the Gordon Bell Special Achievement Award at SuperComputing 2006.

Publications

Adare, A., 2007. "Energy loss and flow of heavy quarks in Au+Au collisions at $\sqrt{s}=200\text{GeV}$." *Phys. Rev. Lett.* **98**, 172301. UCRL-JRNL-228385.

Blyth, S. L., et al., 2007. "A cone jet-finding algorithm for heavy ion collision at LHC energies." *J. Phys. G Nucl. Phys.* **344**, 271. UCRL-JRNL-228100.

Klay, J., 2007. "Jet physics with ALICE." *Nucl. Phys. A* **787**, 52. UCRL-PROC-225837.

Foam-Walled Hohlräume for Increased X-Ray Conversion Efficiency

Peter E. Young 06-ERD-053

Abstract

Recent analytic and computational analysis of the radiation heat front in the high-Z wall of laser-irradiated hohlraum targets suggests that an optimum wall density exists at which radiation energy is maximum. Since the increased radiation energy is the result of decreased energy transferred into hydrodynamic energy, reducing wall density has the added benefit of reduced filling of the hohlraum. The predicted increase in x-ray flux (~20%) would provide an increased margin of energy in experiments using large laser facilities. We demonstrated the effect by comparing hohlraums made with 100-mg/cm³ and 4-g/cm³ tantalum foam.

Successful demonstration of increased x-ray conversion efficiency and lower hohlraum filling using foam-walled hohlraums will have an important impact on target designs for both internal-confinement fusion and high-energy-density experiments planned for large laser facilities. Reduction of the required laser energy for a given x-ray flux will increase the energy margin for such experiments and will reduce the operating costs of those facilities by reducing damage to final optics. Once the physics of foam-walled hohlraums has been demonstrated, such hohlraums will likely become a standard experimental platform for large lasers. The experimental demonstration of the density-dependent x-ray flux has already motivated the development of approximately 400-mg/cm³ gold foams.

Mission Relevance

Target designs that increase the x-ray conversion and decrease the amount of hohlraum filling because of ablated plasma are very important for the internal-confinement fusion and high-energy-density experiments planned for large laser facilities, which support LLNL's national and energy security missions.

FY07 Accomplishments and Results

We demonstrated an enhancement of hohlraum radiation temperature using tantalum foam. By using a reverse-ramp laser pulse, we achieved nearly constant radiation temperature over the duration of the pulse, allowing quantitative comparison with the predictions of analytic theory. The analytic predictions and two-dimensional simulations agreed well with both the

absolute value of the observed radiation temperature and the dependence on the hohlraum wall density. An approximately 20% enhancement in the radiation flux was observed with the lower density foam.

Mitigation of Electromagnetic Pulse Effects from Short-Pulse Lasers and Fusion Neutrons

David C. Eder 06-ERD-055

Abstract

The objective of this project is to mitigate the impact of electromagnetic pulses (EMP) at petawatt-class laser facilities. The primary source of EMP at such facilities is high-energy (>1 -MeV) electrons generated by the short (<1 -ps) laser pulses. At the Titan petawatt-class laser facility, we are measuring the number and spatial distribution of electrons escaping from the target and the resulting transient currents and EMP. The electron properties are used in three-dimensional electromagnetic simulations, and the calculated fields are compared to measurements. The frequency dependence of the EMP is important because different shielding techniques are required for high-frequency (1- to 5-GHz) EMP as compared to more conventional EMP in the 10- to 500-MHz range. The relative importance of system-generated EMP is also evaluated.


This project will provide the first quantitative understanding of EMP, including system-generated EMP, in petawatt-class laser facilities. This information can be used to mitigate the impact of EMP through improved shielding and reduced source generation. The predictive simulation capability developed in this project can be applied to future systems to mitigate EMP and greatly reduce the occurrence of diagnostic damage and data loss. The experimental and computational techniques we develop will be applicable to EMP at longer-pulse laser facilities, as well. In addition, the data obtained on the dependence of escaping electron number and spatial distribution on laser and target conditions is of interest to a wide range of research, including fast ignition.

Mission Relevance

The work supports stockpile stewardship in two ways. First, knowledge of EMP and system response is critical for the reliable operation of facilities used to validate ignition simulations and simulate weapons effects. Second, possible future underground experiments require knowledge of SGEMP on components that did not exist during earlier testing. This work on EMP mitigation techniques also will apply to short-pulse lasers for fast ignition, in support of Lawrence Livermore's energy security mission.

FY07 Accomplishments and Results

For FY07, we (1) explored EMP reduction experimentally and with simulations using different beam dump designs; (2) studied the effect of equipment in the Titan chamber on probe signals; (3) improved our EMSolve model, which is used to calculate EMP given properties of electrons leaving the target; (4) added chamber windows to the simulations and measured



EMP outside of the windows; (5) compared computed and measured EMP data with particular attention to the frequency dependence; (6) measured the relative importance of system-generated EMP; and (7) obtained a very large amount of data from dedicated shots on Titan, allowing us to study the effect of pulse duration, target size and thickness, and laser energy on the properties of escaping electrons and EMP.

Proposed Work for FY08

In FY08, we will (1) analyze Titan data obtained in FY07 to determine the impact of varying laser and target parameters; (2) compare calculated EMP properties, such as frequency, with observed values for our data set using the measured properties of the escaping electrons as input into the simulations; (3) develop a predictive capability for EMP and mitigation approaches based on an improved understanding of the underlying mechanisms; (4) test our predictive capability with additional Titan experiments; and (5) use these techniques to predict and validate EMP levels for fusion-class laser facilities.

Publications

Brown, C. G., et al., in press. "Electromagnetic pulses at short-pulse laser facilities." *J. Phys. Conf. Ser.* UCRL-CONF-234483.

Eder, D., et al., 2007. *Facility impacts from neutron and gamma radiation, electromagnetic pulses, debris/shrapnel, and tritium.* Intl. Workshop ITER-LMJ-NIF Components in Harsh Environments, Cadarache, France, June 27–29, 2007. UCRL-PRES-232107.

Laser Beam Propagation in High-Temperature Plasmas

Dustin H. Froula 06-ERD-056

Abstract

The primary goal of this research is to develop an understanding of laser beam propagation through high-temperature plasmas. This research is of interest to numerous Laboratory programs in material strength, radiation transport, and hydrodynamics. Experiments and numerical modeling will focus on blue (3ω) high-intensity laser beams interacting with the high-temperature plasmas that will occur in high-energy-density (HED) physics experiments such as those at fusion-class lasers. Determining the limits of efficient laser beam propagation will strongly influence design of a wide variety of experiments that scale strongly with laser intensity, and will allow us to access higher regimes of pressure, temperature, and strain rate in matter.

In this work, we will resolve important scientific questions relating to laser–plasma interactions in high-temperature plasmas, which are important for HED science applications. We will determine temperature and beam intensity limits for laser beam propagation, which will serve as input parameters for the design of critical stockpile stewardship–related experiments. These results will provide the first predictive laser–plasma modeling.

Mission Relevance

Five key areas within the Stockpile Stewardship Program would significantly benefit from the ability to predict the design space limits for laser-driven HED experiments: (1) fusion ignition, (2) material dynamics, (3) experiments using special nuclear materials, (4) nuclear weapons effects testing, and (5) hydrodynamics. This project will also enable cutting-edge research in other LLNL mission areas such as inertial confinement fusion, in support of the energy security mission.

FY07 Accomplishments and Results

In FY07, we (1) performed new experiments at the Omega Laser Facility to measure laser beam coupling through plasmas that have been previously characterized, (2) scaled the electron temperature to investigate laser beam propagation in plasmas, (3) extended our target platform to produce the first experiments in a 4-mm-long transparent plasma, (4) explored fundamental properties of filamentation when a laser beam is defocused, and (5) validated our laser–plasma interaction modeling using our propagation results.

Publications

Froula, D. H., et al., 2007. “0.351-nm transmitted beam diagnostic at the Omega Laser Facility.” *Rev. Sci. Instrum.* **77**, 10E507. UCRL-JRNL-221120.

Froula, D. H., et al., 2007. “Ideal laser beam propagation through high temperature ignition hohlraum plasmas.” *Phys. Rev. Lett.* **98**, 085001. UCRL-JRNL-225079.

Froula, D. H., et al., 2007. “Laser beam propagation through inertial confinement fusion hohlraum plasmas.” *Phys. Plasma* **14**, 055705. UCRL-CONF-225802.

Froula, D. H., et al., in press. “Pushing the limits of plasma length in inertial fusion laser–plasma interaction studies.” *Phys. Rev. Lett.* UCRL-JRNL-233624.

Froula, D. H., et al., 2007. “Thomson scattering techniques in a laser produced plasma.” *Rev. Sci. Instrum.* **77**, 10E522. UCRL-CONF-221161.

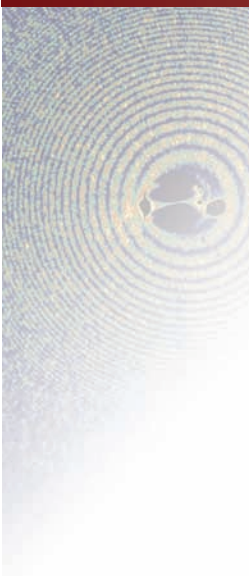
Meezan, N. B., et al., 2007. “Role of hydrodynamic simulations in laser–plasma interaction predictive capability.” *Phys. Plasma* **14**, 056304. UCRL-JRNL-225530.

Compact, High-Intensity Neutron Source Driven By Pyroelectric Crystals

Jeffrey D. Morse 06-ERD-065

Abstract

The objective of this effort is to establish a new paradigm for active neutron-interrogation systems. We will explore the potential for achieving an extremely compact, high-intensity



neutron source exploiting nuclear fusion reactions driven by pyroelectric crystals. The concept being investigated represents a revolutionary approach for accelerator-induced nuclear fusion reactions in a compact platform. Pyroelectric-crystal-driven neutron sources would potentially eliminate the need for large, high-voltage power supplies and radically change the size and configuration of the ion accelerator, enabling a palm-sized neutron source. Thus, this project could have broad impact on weapons science, nuclear physics, and homeland security applications.

We will quantitatively determine the potential for scaling pyroelectric-crystal-driven ion and neutron sources to fluxes of greater than 10^6 n/sec. Our technical approach is to (1) complete a modeling study of the crystal-based neutron source; (2) demonstrate experimental scaling—that is, neutron output up to three orders of magnitude greater than the initial results; and (3) test and evaluate the neutron source in actual applications. We plan to investigate a new source approach that will further intensify the ion beam, enabling both pulsed and continuous operation.

Mission Relevance

Neutron interrogation provides a noninvasive method of screening cargo and shipping containers for special nuclear materials smuggled through ports. This project supports Lawrence Livermore's national security mission by investigating a promising new technique that may enable a field version or even a handheld neutron source with the ability to interrogate targets anywhere, not just at ports. This approach offers the further possibility of a remote, autonomous neutron probe for the covert interrogation of targets and threat identification.

FY07 Accomplishments and Results

In FY07, we established the experimental capability to generate neutrons via the pyrofusion effect. Our unique approach is to control the thermal cycle of a pyroelectric crystal assembly using a lithium tantalite crystal with a tungsten needle affixed to the charge surface. The onset of ionization of a 3- to 5-microTorr background of deuterium gas was observed—as our Monte Carlo models had predicted—followed by persistent deuteron beams as measured with a Faraday cup positioned in front of the deuterated target. Ion beam currents on the order of 8 to 10 nA were measured, for cycle times on the order of 400 to 600 s, yielding greater than 1.9×10^5 neutrons per cycle—more than a factor of two greater than previously reported for the $d(D,n)$ reaction driven by pyroelectric crystals.

Proposed Work for FY08

Proposed work for FY08 includes (1) continuing to optimize and integrate a gated ion source with the pyroelectric crystal high-voltage transformer; (2) demonstrating efficient generation of ion beams with energies in excess of 100 kV for effective fusion reactions with a deuterated or tritiated target; (3) modeling target reactions to predict the expected performance and to confirm experimental results for the neutron source; (4) combining ion trajectory models and ion optics designs to optimize the beam profile impinging upon the target, thereby increasing overall neutron yield; and (5) conducting further optimization of thermal ramping approaches for crystal actuation to provide a robust neutron source design.

Nuclear Physics the Monte Carlo Way

William E. Ormand 06-LW-013

Abstract

This research effort will investigate the application of Monte Carlo methods to the nuclear many-body problem. In this project, we will examine improvements and applications of the auxiliary-field Monte Carlo (AFMC) method, which can provide exact results for systems with extraordinarily large dimensions (e.g., 10^{20} and beyond) where brute-force matrix diagonalization is hopelessly overmatched to problems of interest in nuclear structure and nuclear astrophysics. In the past, applications were limited because of the notorious “sign problem,” which recently has been mitigated considerably and will be resolved more generally with this project.

With successful implementation of these Monte Carlo methods, the range of topics in nuclear theory that we can explore is far reaching. We will be able to (1) perform the first truly detailed studies of intermediate-mass nuclei at the limits of stability with realistic interactions; (2) compute weak transition rates important for theories of supernova core collapse; (3) compute level densities, electro-weak rates, and binding energies important for nucleosynthesis in the r-process; and (4) study the evolution of the giant-dipole resonance in nuclei at high excitation energy. Our work will provide a tool for future theoretical studies of nuclear structure. Results will be published in physics journals, and we hope to provide data that is important to the astrophysics community.

Mission Relevance

This project supports LLNL’s stockpile stewardship and homeland security missions by providing a scientific basis and a new tool for theoretical studies in nuclear structure, which impacts the nuclear data needs of these missions. The project utilizes high-performance computing to address important problems in nuclear theory, thus supporting the Laboratory’s mission in breakthroughs in fundamental science, and also will provide critical underpinning for the Rare-Isotope Accelerator, a proposed DOE nuclear physics facility.

FY07 Accomplishments and Results

In FY07 we (1) achieved a breakthrough by demonstrating, in critical test cases, that we defeated the sign problem for general nuclear physics applications—and these test cases showed excellent agreement between the AFMC calculations and those obtained with diagonalization; (2) completed calculations for simple sd-shell nuclei as well as the much more complicated fp-shell nuclei iron-54 and iron-60; and (3) demonstrated the ability to compute the density of states, which is key for nuclear-reaction studies important for understanding nucleosynthesis. This project’s results are significant as this is the first successful application of its kind, and it has the potential to revolutionize our approach to the nuclear many-body problem.

Publications

Ormand, W. E., and G. Stoitcheva, 2007. "Defeating the sign problem in the auxiliary-field Monte Carlo method for nuclear structure: level densities and other properties." *Bull. Am. Phys. Soc.* **52**, 25. UCRL-ABS-232452.

Stoitcheva, G., and W. E. Ormand, 2007. "Defeating the sign problem in the auxiliary-field Monte Carlo method for nuclear structure: the shifted-contour method." *Bull. Am. Phys. Soc.* **52**, 24. UCRL-ABS-232453.

Observation of Coherent Terahertz-Frequency Emission from Shocked Polarizable Materials

Evan J. Reed 06-LW-063

Abstract

We recently predicted that coherent electromagnetic radiation (<100 THz) can be generated in crystalline materials subjected to a shock wave. This phenomenon represents a fundamentally new form of coherent optical radiation. The predicted coherent emission contains information regarding shock speed, lattice and atomic structure of the crystal, and shock-front rise time, suggesting this effect represents a new experimental probe. In this project, we propose to perform the first-ever experimental measurements of terahertz emission from shocked materials and determine the ultrafast dynamical processes responsible for this emission. The experiments will utilize ultrafast laser-driven shock waves in ionic crystals, coupled with a theoretical effort for guidance and interpretation.

We expect to make the first observation of terahertz radiation from a shocked crystal in search of our predicted emission of coherent photons. We will extract the information contained in observed coherent or other emission regarding shock-front rise time, picosecond-timescale shock-speed history, and crystal lattice constant. If successful, this will result in a paper reporting the observation of a fundamentally new source of coherent optical radiation or a measurement of the rise time of an elastic shock wave. We will assess the utility of terahertz emission as a fundamentally new general probe into dynamical processes in shock waves. Our results will be submitted for publication in peer-reviewed journals.

Mission Relevance

New tools for understanding shock wave properties and materials phenomena are central to mission needs in stockpile stewardship. This work will provide fundamentally new insight into shock wave properties and phenomena in materials. It also provides a new source of terahertz radiation with unique properties, which can be used for applications in defending against explosives and chemical and biological threats, in support of the Laboratory's missions in national security and homeland security.

FY07 Accomplishments and Results

For FY07, we successfully performed the first observation of terahertz radiation emission from a shocked material and determined the multiple ultrafast processes for the emission, including shock rise time. This work is yielding a fundamentally new, ultrafast-timescale diagnostic for shock wave experiments with capabilities unlike existing probes. We also performed simulations that indicate the time resolution is potentially subpicosecond—an unprecedented value. This accomplishment represents a key step toward observation of our predicted coherent radiation and indicates that, under some circumstances, such radiation can potentially be observed with our experimental setup. We filed a provisional patent and published several papers this year.

Proposed Work for FY08

In FY08, we plan to search for temporally coherent radiation generated by strain waves in gallium–nitrogen superlattices and ionic crystals. We will attempt to maximize frequency, coherence length, and amplitude of this radiation. A clear observation will yield a high-profile paper reporting a fundamentally new source of coherent radiation. We also will observe and characterize terahertz emission associated with other ultrafast dynamical processes that occur within shocks. We will assess the utility of terahertz emission spectra as a fundamentally new general probe into dynamical processes in shock waves. Accomplishment of this objective also may yield a high-visibility paper reporting direct experimental observation of previously unobservable ultrafast phenomena.

Publications

Reed, E. J., M. Soljacic, and J. D. Joannopoulos, 2007. “Maxwell equation simulations of coherent optical photon emission from shock waves in crystals.” *Phys. Rev. E* **75**, 056611. UCRL-JRNL-226463.

Reed, E. J., et al., 2007. *Coherent THz frequency radiation from shock waves: a new ultrafast strain wave detection mechanism*. 15th APS Topical Conf. Shock Compression of Condensed Matter, Kohala Coast, HI, June 24–29, 2007. UCRL-CONF-232143.

Reed, E. J., et al., 2007. “Molecular dynamics simulations of coherent optical photon emission from shock waves in crystals.” *Phys. Rev. B* **75**, 174302. UCRL-JRNL-233178-DRAFT.

Reed, E. J., et al., 2007. “Nonlinear strain and shock wave sources of THz and optical frequency radiation.” *Mater. Today* **10**, 44. UCRL-JRNL-228830-DRAFT.



Multipulse, High-Energy Backlighting for a Compton-Radiography Ignition Diagnostic for High-Power Lasers

Riccardo Tommasini 07-ERD-004

Abstract

We propose to develop a multiframe radiography capability to obtain ultrahigh-speed movies of an imploding target capsule as it evolves through ignition time for single high-power laser shots. Transmission radiography of ignition capsules requires high-photon energies because imploded cores are opaque below about 15 keV and the anticipated self-emission would overwhelm backlighter brightness below some threshold greater than 70 keV. Therefore, we will exploit the capabilities of the Advanced Radiographic Capability laser facility at Livermore to develop multipulsed backlighting sources that emit in the spectral region above 75 keV. At these photon energies, the main source of opacity for the dense cold deuterium–tritium fuel is Compton scattering, hence the designation “Compton radiography.”

We expect to achieve a high-energy radiography diagnostic able to give several images of the dense cold fuel surrounding the capsule hot spot, having about 10-ps time resolution, over a time span of a few hundred picoseconds. This capability will help provide the time history of the compressed fuel shape leading up to the implosion’s maximum density. Obtaining multiple time-spaced radiographs in a single fusion-class laser shot will yield constant conditions for the ignition-driving laser and a reduced number of shots to gain the actual time history of the imploding target. This novel diagnostic will allow us to observe fuel asymmetries near peak compression and could be an invaluable aid in achieving ignition.

Mission Relevance

Our proposal is exceptionally well aligned with the stockpile stewardship and energy security missions of the Laboratory, by developing a unique Compton radiography ignition diagnostic for fusion-class lasers and a multipulse, high-energy radiography capability to increase the accuracy of measurements and reduce the number of laser shots needed to reach a scientific result. The Compton radiography diagnostic will greatly improve our ability to achieve ignition on fusion-class lasers, giving a time sequence of two-dimensional images of the compressed deuterium–tritium fuel.

FY07 Accomplishments and Results

In FY07, we (1) estimated the target motion to be marginal on a 10-ps time scale, (2) designed a collimator near the hohlraum to prevent backlighters from adversely affecting images from other backlighters, (3) separated the backlighters sufficiently to eliminate timing conflicts between backlighters imaged on a single detector, (4) developed a point-projection radiography geometry having a separate target for each backlighter, (5) performed dedicated experiments demonstrating backlighter performances at the Titan laser facility and measured conversion efficiencies to line emissions up to 75 keV and to 75- to 100-keV bremsstrahlung, and (6) maximized signal-to-noise by choosing point-projection radiography using targets made of high-Z wire sandwiched between lower Z materials.

Proposed Work for FY08

Based on the results obtained so far, we estimate that a 75- to 100-keV bremsstrahlung backlighter in a point-projection geometry will be a useful diagnostic for low-yield (~100 kJ) failures of fusion-class lasers. High-yield (~1 MJ) failures would require extension of our results to the 150- to 200-keV energy range. Therefore, in FY08, we will (1) extend the conversion efficiency measurements to bremsstrahlung emission above 100 keV, (2) demonstrate sufficient two-dimensional spatial resolution at and above 100 keV, (3) demonstrate the multipulse capabilities for short interframe delays, and (4) test a prototype of the x-ray radiography detector to be used on advanced, high-power lasers.

Publications

Tommasini, R., et al., 2007. *Development of Compton radiography using high-Z backlighters produced by ultra-intense lasers*. 15th Intl. Conf. Atomic Processes in Plasma, Gaithersburg, MD, Mar. 19–22, 2007. UCRL-ABS-230812.

Tommasini, R., et al., 2007. *Ultra-intense-laser produced high-Z backlighters for Compton radiography*. SPIE Optics and Photonics Conf., San Diego, CA, Aug. 10–14, 2007. UCRL-CONF-233900.

Cladding-Pumped Raman Fiber Lasers

Jay W. Dawson 07-ERD-005

Abstract

We propose to develop a cladding-pumped Raman fiber amplifier. This device will enable us to generate extremely high-energy pulses in multimode, rare-earth-doped fiber amplifiers and efficiently convert that light to a diffraction-limited laser beam with tens of millijoule pulse energies and hundreds of watts of power. This new capability will enable applications of interest to the Laboratory such as x-ray generation; very high-energy, short-pulse laser front ends; and high-speed cutting and welding in compact, robust all-fiber formats. We believe this technology will scale several orders of magnitude beyond the current state of the art in fiber-pulse energy with diffraction-limited beam quality.

We expect to demonstrate a cladding-pumped Raman laser with greater than 200-kW peak power pulses in a short length of optical fiber. This will show that it is possible to construct short and efficient cladding-pumped Raman lasers with good beam quality. This will result in one or more publications and possibly some significant intellectual property. We further expect to investigate theoretically the scaling properties of such a laser. The laser proposed here would be capable of accessing new regimes of pulse energy and average power in a compact and robust format not available in other types of systems. The inherent bandwidth of the Raman transition would also enable short-pulse applications of the system.

Mission Relevance

High-average-power, high-energy laser systems support our national security mission along a broad front of applications. These include materials processing such as laser peening to strengthen aircraft engines and airframes, x-ray generation such as backlighters for future fusion-class laser experiments and for pulse-probe shock experiments, and remote sensing of chemical-absorption signatures to determine the presence and distance of chemicals relevant to nonproliferation and homeland security.

FY07 Accomplishments and Results

For FY07, we (1) performed a theoretical evaluation of the scalability of Raman fiber amplifiers, validating our assumptions about the performance of these types of systems; (2) obtained a specialized fiber optic to allow preliminary experimental study of our concept for producing high-average-power systems; and (3) successfully tested the basic physics of our concept using a neodymium–yttrium lithium fluoride Q-switched laser followed by a ytterbium fiber amplifier as a pump laser.

Proposed Work for FY08

Our goals for FY08 are to (1) develop a design for a large-core-area, single-mode cladding-pumped Raman gain medium; (2) build the designed medium in collaboration with a specialty fiber vendor; (3) demonstrate 10-mJ pulse energies with high beam quality; and (4) produce a publication for a peer-reviewed journal and obtain at least one patent.

Maximizing the Science from Astrophysical, Time-Domain Surveys: Targeted Follow-up

Kem H. Cook 07-ERD-014

Abstract

We intend to maximize data from astrophysical, time-domain surveys that will produce significant science on issues ranging from new exoplanets to a better understanding of dark energy. Using time-domain data either in place at Lawrence Livermore and/or publicly available (for example, the Optical Gravitational Lensing Experiment and Microlensing Observations in Astrophysics data sets) with data-management experience from the Large Synoptic Survey Telescope precursor projects, we will perform new studies and identify follow-up observations.

By building new computational capabilities, we expect to produce new results in several fields of astrophysics, including (1) finding extrasolar planets representative of those in our solar system; (2) reducing uncertainties in the dark energy equation-of-state; (3) characterizing the historical rate of Type Ia supernovae through light echo spectroscopy; (4) obtaining galactic halo structure and formation history from Lowell Observatory Near-Earth Object Search (LONEOS) RR Lyr stars; (5) determining the delta Scuti distance to the Large Magellanic Cloud, a key rung in the cosmic distance ladder; (6) exploring stellar physics (e.g., limb

darkening via microlensing); and (7) determining the time evolution of stellar pulsations and their effect on mass loss. We intend to leverage our work and the significant computational and modeling capabilities at LLNL to develop data mining tools that we will make available to the scientific community.

Mission Relevance

The project is related to Laboratory research efforts in exploration and use of space, which will benefit LLNL missions in national security and fundamental science. Determining the abundance of planets in the galaxy will showcase Livermore's scientific and computing capabilities, which helps retain and recruit the best scientific and technical minds. This project will demonstrate how coordinated follow-up to a wide-field, time-domain survey such as the Large Synoptic Survey Telescope (a part of the Laboratory's science and technology plan) can greatly enhance its scientific payoff.

FY07 Accomplishments and Results

In FY07, we (1) parallelized a planetary microlensing modeling code, leading to the discovery of two possible planet systems; (2) discovered candidate light echos from historical supernovae in the Milky Way Galaxy; (3) discovered an eruptive variable with light echoes from our mining of the Super Massively Compact Halo Object (SuperMACHO) data set; (4) conducted spectroscopic follow-up observations of light echoes in the Large Magellanic Cloud; (5) started follow-up observations to characterize two Halo RR Lyrae populations; and (6) continued data reductions on archival data at LLNL for additional follow-up studies and science extraction.

Proposed Work for FY08

In FY08, we will continue follow-up programs with the Equation of State: Supernovae Trace Cosmic Expansion (ESSENCE), Probing Lensing Anomalies Network (PLANET), and LONEOS surveys, as well as the survey for galactic light echoes. Specifically, we will (1) conduct full analysis of the ESSENCE five-year data set, which will promote additional follow-up studies on other topics; (2) analyze LONEOS image data, with results available for follow-up spectroscopy (five nights are already scheduled) and modeling; (3) complete the SuperMACHO efficiency analysis; (4) continue Hubble Space Telescope follow-up; (5) continue PLANET observations, with real-time analysis to discover new extrasolar planets; (6) continue our imaging survey for light echoes in the galaxy and begin spectroscopic characterization of those found; and (7) complete our follow-up spectroscopy of ancient supernovae light echoes in the Large Magellanic Cloud.

Publications

Cook, K. H., S. Nikolaev, and M. E. Huber, 2007. *The Large Synoptic Survey Telescope and foundations for data exploitation of petabyte data sets*. UCRL-TR-228468.

Cook, K., et al., 2007. *Science results from the MACHO and superMACHO surveys*. American Astronomical Society 207th Mtg., Washington, DC, Jan. 8–12, 2006. UCRL-PRES-229881.

Garg, A., et al., 2007. "Lightcurves of Type Ia supernovae from near the time of explosion." *Astronom. J.* **133**, 403. UCRL-JRNL-224121.

Huber, M., et al., 2007. "Time-series data and the virtual observatory: the faint sky variability survey dataset." *Bull. Am. Astron. Soc.* **38**, 1041. UCRL-POST-227082.

Lehner, M. J., et al., 2007. "TAOS—The Taiwanese-American Occultation Survey." *Astron. Nachr.* **327**, 814. UCRL-PROC-225991.

Miceli, A., et al., in press. "Evidence for distinct components of the galactic stellar halo from 838 RR Lyrae stars discovered in the LONESO-I survey." *Astrophys. J.* UCRL-JRNL-228444.

Discovery of a Light Higgs Boson with b Quarks

David J. Lange 07-ERD-015

Abstract

A new era in particle physics will begin when the Large Hadron Collider (LHC) comes fully online in 2008 and advances the high-energy frontier to a center-of-mass energy of 14 TeV. We are developing a technique that may discover the Higgs boson by enhancing detection of b quarks in the first data samples produced from experiments at the LHC. We will develop the required particle-identification techniques, trigger algorithms, and analysis codes to search for the Higgs boson in its dominant decay mode.

Discovery of the Higgs boson would represent the beginning of a new era in particle physics—understanding new phenomena at the teraelectronvolt scale. Understanding the light Higgs sector will be one of the first physics goals of the LHC, and we will play a leading role in the search for the Higgs boson with b quarks. We will also ready a high-level trigger (HLT) system, which will be critical for Higgs analysis.

Mission Relevance

The data-mining, analysis, and computing capabilities developed in this project for collider physics have direct application in homeland security applications for detecting subtle patterns in massive sets of disparate data. This work also supports the Laboratory's mission in breakthrough science by enabling LLNL to play an important role in discoveries that will have a profound impact on particle physics. This research will also be an exceptional recruiting tool for outstanding young scientists with both radiation-detection and computer-simulations capability required for the Laboratory's nonproliferation and homeland security work.

FY07 Accomplishments and Results

In FY07 we (1) deployed the HLT configuration management system in collaboration with researchers at the European Organization for Nuclear Research, (2) assessed HLT system timing with the requirement to unpack raw data into the format used by HLT algorithms, (3) provided and validated the code to convert simulation into raw data, and (4) assessed

the potential of Higgs production channels, revealing two new mechanisms with potential for discovery using the first LHC data. The HLT algorithms required for these channels are now in place for first data taking.

Proposed Work for FY08

With the first physics data expected from LHC in FY08, we will participate in HLT operation, benchmark our Higgs analysis, and validate the detector systems with these data. In particular, we will complete development and commissioning of the HLT configuration database as well as the raw data conversion to a common reduced format in the HLT. We will continue to develop the Higgs production mechanisms from our FY07 work. With a trigger in place, we will determine how to distinguish these modes from background to isolate the Higgs signal. We will also use the first LHC data samples and our analysis code to study the production of Z bosons to validate the detector and current experimental observations.

A New Approach to Simulating Inhomogeneous Plasmas for Inertial-Fusion Energy and Other Applications

David P. Grote 07-ERD-016

Abstract

We propose to develop a novel approach to simulating inhomogeneous magnetized plasmas, and to begin applying this new method to problems of importance to Lawrence Livermore. Numerical simulations of such plasmas, which arise in fast ignition, magnetic-fusion energy, and space sciences, are challenging. The newly developed drift-Lorentz particle mover has proven very effective for heavy-ion fusion applications by enabling a significant reduction in computational time requirements. The goal of our research is to extend this invention, combining it with implicitness and collisions, so that it can be applied to fast ignition for inertial fusion and a broad range of other significant problems.

If the project is successful, we expect to demonstrate an advanced plasma simulation capability that substantially reduces the computational effort needed for simulations of inhomogeneous plasmas, based on a novel method that differs qualitatively from anything now available. For fast ignition, this should enable more robust calculations, with a more accurate treatment of interparticle collisions. This approach may also prove a simpler alternative to the gyrokinetic formulation widely used in magnetic-fusion energy core-turbulence studies, which becomes complex when applied to edge plasmas. While development of this method for the latter application is beyond the scope of this proposal, this work would place LLNL in a better position to assess the promise of the idea.

Mission Relevance

This project is designed to strengthen LLNL's capabilities in fast ignition for inertial-fusion energy, a long-standing Laboratory strategic goal. With a strong foundation created for magnetic-fusion energy and heavy-ion fusion, we seek to build capabilities as interest in inertial-fusion energy increases. This capability therefore supports the Laboratory's missions

in energy security and breakthroughs in fundamental science and technology, as well as advancing the supercomputing environment on which stockpile stewardship is based.

FY07 Accomplishments and Results

In FY07, we generalized a grid-based Langevin collision operator to the case of unlike-particle scattering, conducted tests that showed good agreement with theoretical results, and ported the operator to the WARP time-dependent plasma code. We continue to explore approaches that may offer greater computational efficiency. In addition, we implemented partial implicitness into the new drift-Lorentz particle mover and used the new capability to simulate the ion-temperature gradient instability, obtaining the correct linear growth rate and saturation level with a large-time-step simulation. Finally, we developed and began implementing an implicit electrostatic version of the drift-Lorentz mover.

Proposed Work for FY08

In FY08, we will (1) implement and begin testing an implicit electromagnetic or Darwin version of the drift-Lorentz mover in the user-steerable WARP code, which will be used to understand the properties of the algorithm; (2) test our grid-based Langevin collision operator, in conjunction with the drift-Lorentz mover, on the collisional ion-temperature gradient problem; and (3) implement the collision operator in the Large Scale Plasma code for fast-ignition studies.

Publications

Cohen, R. H., et al. 2007. "Large-timestep mover for particle simulations of arbitrarily magnetized species." *Nucl. Instr. Meth. A* **577**, 52. UCRL-CONF-222185.

A Novel Method for Extracting Signals from Noisy Broadband Data Using Poynting Vector Measurements

Charles R. Carrigan 07-ERD-018

Abstract

We propose to explore and develop techniques for exploiting electric and magnetic field measurements in noisy electromagnetic environments using the Poynting vector (electric \times magnetic field) to extract low-level signals. Because facilities of interest radiate electromagnetic signals from electric equipment and instrumentation, the proposed work is particularly relevant to proliferation detection, as well as other information-gathering objectives. We will develop a method to identify unknown signals and increase the signal-to-noise ratio in an omnidirectional signal-acquisition system by tagging the frequency spectrum of a broadband signal. A combination of experiments and simulations will be used to accomplish this project.

If successful, we will develop a new method that allows for reconstructing an electromagnetic signal of interest from a noisy broadband data stream acquired from an omnidirectional acquisition system. With minimal information about a signal of interest, we also will be able to determine the direction of its source. In principle, this allows us to reconstruct signals from facilities and other electromagnetic sources of interest or to determine the direction to such facilities and sources if we have knowledge about the signal. Analysis applications that associate a collection of signals or harmonics with activities at predetermined standoff locations will benefit considerably from this technique.

Mission Relevance

This project is consistent with the Laboratory's commitment to exploring innovative system concepts to address challenges and develop novel approaches for national security issues including homeland security, nonproliferation, and counterterrorism. Specific relevant challenges include information processing to yield enhanced detection and characterization requiring recovery of sparse, noisy measurements relating to proliferation detection; tagging, tracking, and locating high-value objects; and predictive simulation and modeling approaches for optimization of sensor and data-collection system performance.

FY07 Accomplishments and Results

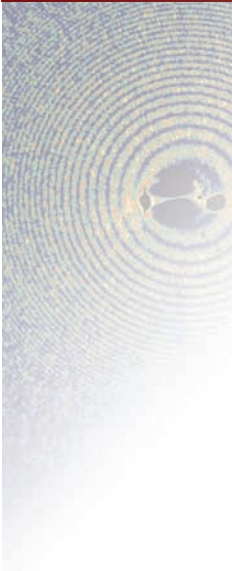
In FY07, we (1) performed an experiment tracking a moving source, demonstrating correspondence with the Poynting vector direction in a complex and noisy electromagnetic environment; (2) evaluated calibration of electric and magnetic field sensors with the goal of refining our directionality calculations; (3) introduced significant, state-of-art enhancements to a MATLAB signal-analysis program used for obtaining direction from raw electric and magnetic field data; (4) began laboratory experiments to evaluate the Poynting vector resulting from single electromagnetic sources; and (5) performed a series of computer simulations of electromagnetic sources in realistic environments that included ground clutter, combined noise and signal sources, and ground planes, to provide well-defined source input for testing the MATLAB analysis program.

Techniques for Supernova Cosmology with the Large Synoptic Survey Telescope

Scot S. Olivier 07-ERD-023

Abstract

We propose to develop and test techniques for precisely quantifying the properties of cosmic dark energy using observations of supernovae made with the Large Synoptic Survey Telescope (LSST), a large-aperture, wide-field optical imaging facility that will be the most powerful astronomical survey instrument in the next decade. The LSST is designed to address fundamental questions concerning the structure and evolution of the universe. Leveraging LLNL's leadership in several related research areas, we intend to establish the Laboratory as a leader in the use of the LSST for supernovae cosmology to elucidate the nature of the mysterious dark energy that is driving the acceleration of cosmic expansion.



The discovery of accelerated expansion of the universe is based on less than 100 type Ia supernovae, indicating their importance as distance indicators. In contrast, the LSST survey is planned to measure the multicolor light curves of hundreds of thousands of type Ia supernovae out to redshifts of up to approximately 1.2. This survey should enable precise investigations of cosmic expansion, yielding unprecedented constraints on the equation of state of dark energy. Our goal is to develop tools to simulate the capability of LSST in discovering distant supernovae and to optimize strategies in controlling systematic errors.

Mission Relevance

This project will help make important advances in astrophysics and space science with national security applications by leveraging the Laboratory's expertise in high-energy-density physics, nuclear fusion, instrumentation and diagnostics, and scientific computing and data management. In particular, large-scale data management and image analysis techniques have specific relevance to similar efforts in nonproliferation and homeland security.

FY07 Accomplishments and Results

In FY07, we (1) developed techniques to analyze the accuracy of redshift estimates from supernova light curves (photometric redshifts) and supported the cadence simulation for the LSST 10-year baseline survey strategy, (2) analyzed the performance of an LLNL-developed intensity transport (curvature) algorithm for LSST wavefront sensing, (3) developed a cadence simulator for supernovae observations with the University of California's Kast spectrograph on the Lick Observatory's 3-m Shane telescope and developed a proposal (which was accepted) to conduct multi-epoch spectral observations of supernovae to quantify the intrinsic variability in their spectral evolution, and (4) began planning to utilize Thunder simulations of cosmological structure as a starting point for comprehensive LSST simulation.

Proposed Work for FY08

In FY08, we plan to (1) continue investigations into the accuracy of redshift estimates from supernova light curves and the effects of these on measurements of cosmological parameters, (2) complete the performance analysis of the LSST wavefront sensing and reconstruction system, (3) analyze observations of supernovae to quantify intrinsic variability in spectral evolution, and (4) incorporate models of supernovae measurements into the comprehensive LSST simulation framework and begin simulation runs.

Publications

Asztalos, S. J., et al., 2007. "Properties of ellipticity correlation with atmospheric structure from Gemini South." *Astrophys. J.* **659**, 69. UCRL-JRNL-226988.

de Vries, W. H., et al., 2007. "Image ellipticity from atmospheric aberrations." *Astrophys. J.* **662**, 744. UCRL-JRNL-228829.

Garg, A., et al., 2007. "Light curves of type Ia supernovae from near the time of explosion." *Astronom. J.* **133**, 404. UCRL-JRNL-224121.

Electronic Anomalies in Ordered and Disordered Cerium at High Pressures and Temperatures

Magnus J. Lipp 07-ERD-029

Abstract

Cerium exhibits fascinating anomalous behavior under varying conditions of pressure and temperature that are the result of strong electron correlation—it exhibits an isostructural, large volume collapse (VC) ending in a critical point and shows a melt line with a pronounced minimum. However, the phase diagram is not well understood, and only a few data exist above the critical point. Moreover, hypothesized VC mechanisms are controversial, and the magnetic susceptibility appears to switch from Curie–Weiss to Pauli paramagnetism across the VC. Our study addresses these intensely debated issues by determining the structure of solid and liquid cerium with high precision using high-energy x-ray scattering and by measuring susceptibility in a large-volume diamond-anvil cell.

This project will increase our understanding of electron correlation effects as they are displayed in the behavior of cerium, including improved values for the critical point and determining (1) if its anomalous transition is indeed isostructural, (2) if a second-order transition extends from the critical point to the melt-line minimum, (3) if a remnant of this transition continues into the melt and if there are separate liquids of different density, and (4) if the change in entropy across the transition is caused only by electrons, an assumption recently challenged on the basis of neutron scattering of phonons. Our experiments will interface heavily with theoretical efforts centering on the different mechanisms proposed to cause the anomalous transition.

Mission Relevance

The physics of f-electron correlation in the lanthanides and actinides is central to stockpile stewardship. Such research will provide crucial data in support of the Laboratory's national security and stockpile stewardship missions.

FY07 Accomplishments and Results

We (1) constrained the location of the VC critical point to approximately 1.5 GPa and 480 K; (2) observed that the critical behavior of the bulk modulus continues above the critical point and exhibits a clear minimum, meaning that a remnant of the VC continues into the melt without a change in lattice symmetry; (3) conducted x-ray emission spectroscopy—we found that the bare moments remain across the VC, which can only be reconciled with old susceptibility data by screening of the moments, as is precisely predicted by the Kondo VC model; (4) conducted x-ray scattering on liquid bismuth, revealing suppression of the solidification deep into the solid part of the phase diagram; and (5) modified designer anvil technology to increase the pressure range available for our experiments.

Proposed Work for FY08

In FY08, we plan to (1) perform x-ray diffraction measurements above the critical point to follow the remnants of VC up to the melt, (2) employ x-ray scattering above the melt to provide equation-of-state data on the liquid, (3) perform traditional susceptibility measurements to address screening of the magnetic moments, and (4) implement the large-volume diamond-anvil cell, which will allow for a larger pressure range (up to 10 GPa) and provide the necessary signal-to-noise ratios for susceptibility measurements and x-ray scattering from the liquid.

Publications

Lipp, M. J., et al., 2007. *The volume collapse in cerium revisited*. Predictive Capability for Strongly Correlated Systems Fall 2007 Coordination Meeting, Davis, CA, Sept. 15–16, 2007. UCRL-PRES-234630.

Exploring Phase Transition Mechanisms Using Ramp Compression

Jon H. Eggert 07-ERD-032

Abstract

We propose to use ramp compression, coupled with emerging experimental probes developed at Lawrence Livermore, to examine phase-transition mechanisms. While phase transitions occur in many materials when they are compressed or heated, a detailed understanding of transition mechanisms including pathways, nucleation and growth, metastable or transitional phases, and the influence of grain size and texture is still missing. Without detailed mechanism information, we cannot have a true predictive capability of material response to important dynamically driven phase transitions. We will experimentally determine nucleation and growth kinetics, strain-rate driven metastability, grain size, and orientation effects. All requisite diagnostics have been demonstrated or are being built for other applications.

We will study solid–solid transitions in iron and bismuth because of their novel phase diagrams and extensive existing data under static conditions. By comparing our results with those from gas-gun ramp compression and dynamic diamond anvil cells, we will identify the contribution of varying strain rates. We will develop a kinetic formalism that includes both nucleation and growth, and free-energy considerations that can be applied consistently at various strain rates. This will provide the basis for a predictive capability for simulations of dynamic phase transitions. Many of these experiments will yield high-quality research publications.

Mission Relevance

The development of a predictive capability for simulations of dynamic phase transitions will benefit the Stockpile Stewardship Program, as well as missions in energy security, environmental management, and breakthroughs in fundamental science and technology.

Development of effective fusion-class laser capsules and drives will benefit greatly from these predictive simulations. Phase-transition mechanisms have long been an important, but elusive goal of condensed-matter physics—the unique capabilities found at LLNL will enable great strides toward this goal.

FY07 Accomplishments and Results

In FY07, we performed waveform analysis of solid–solid transitions in bismuth, iron, and silicon on the Janus and Omega lasers. In all of these materials we have observed an over-pressurization of the phase transition. We have found for bismuth and iron that the over-pressurization versus strain rate shows a strong correlation. We believe that this over-pressurization is a much more robust and material-dependent effect of kinetics than previous observations have yielded. We have submitted a paper on our results on bismuth and are preparing an article on the silicon and iron results.

Publications

Celliers, P. M., et al., 2007. *Dynamic compression of diamond*. Joint 21st AIRAPT and 45th EHPRG Intl. Conf. High Pressure Science and Technology, Catania, Italy, Sept. 17–21, 2007. UCRL-PRES-234708.

Smith, F., et al., 2007. *Ramp compression of diamond to over 1000 GPa*. 15th American Physical Society Topical Conf. Shock Compression of Condensed Matter, Waikoloa, HI, June 24–29, 2007. UCRL-PRES-232153.

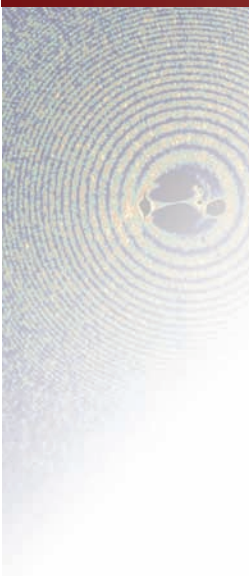
The Development of Scaled Astrophysical Experiments for Current and Future Lasers

Richard I. Klein 07-ERD-038

Abstract

We intend to design and field scaled astrophysical experiments for the Laboratory's intense-short-pulse Titan laser in the collisionless plasma regime to investigate plasma-scaling properties with regard to powerful astrophysical phenomena such as gamma-ray bursts and supernovae. In addition, we propose to design an experiment using high-gain thermonuclear ignition coupled with petawatt lasers to produce conditions relevant to magnetized, radiation-dominated accretion disks surrounding stellar black holes. This work will enable development of a high-energy-density (HED) platform for scaled astrophysical science on fusion-class lasers. We will implement proton deflectometry to characterize magnetic fields associated with the interaction of short-pulse, high-energy lasers with solids.

We expect to experimentally demonstrate scaling in collisionless plasmas on the Titan laser for possible astrophysics experiments. We will design and field a set of scaled experiments to study the Weibel instability relevant to gamma-ray burst shocks and also design an experiment using the concept of high-gain thermonuclear ignition to develop astrophysics experiments for fusion-class lasers. The success of this project will give us the ability to successfully scale



Laboratory experiments to astrophysical phenomena, and test scaling methodology relevant to the science-based Stockpile Stewardship Program.

Mission Relevance

The experiments we will perform on Titan and the integral experiment proposed for fusion-class lasers are unique, and our methodology will give us insight into how to perform scaled multiphysics experiments in support of the Laboratory's stockpile stewardship efforts as well as fusion-energy research. This is a new paradigm for HED astrophysics, and will enable the study of magnetized radiative dynamics and radiation pressure-dominated flows, in support of the Laboratory's mission in breakthrough science.

FY07 Accomplishments and Results

In FY07, we (1) participated in laser experiments to determine the range of plasma parameters and observables accessible on Titan (e.g., laser intensities, density-scale lengths, electron temperatures, magnetic fields, electric fields, harmonics, and pressures), which required experimental scans and simulations with both radiation-hydrodynamics and plasma simulation codes; (2) conducted an experiment on Titan to study magnetic fields generated by the interaction of an ultra-intense laser with a thin metallic foil; (3) investigated magnetic fields as a function of target thickness and laser-pulse energy; and (4) performed simulations using the Large Scale Plasma simulation code to determine the influence of magnetic and electric fields on proton trajectory.

Short-Pulse Laser Applications Design

Richard P. Town 07-ERD-040

Abstract

The achievement of laser fusion ignition in the laboratory is a high priority for Lawrence Livermore. We intend to optimize fast ignition to enable high yield for fusion-class lasers, and apply these optimized designs to stockpile-stewardship-relevant experiments. We will use our recently developed, integrated computational tool to accomplish these tasks. In addition, we will develop and design a number of short-pulse, high-intensity laser applications that will be of utility to the stockpile program.

This project will optimize a variety of short-pulse laser applications, and specifically, fast ignition. In conventional inertial-confinement fusion, long-pulse (nanosecond-scale) laser beams both compress and heat the fuel until it ignites. In fast ignition, the roles of compression and ignition are separated, with long pulses compressing the fuel at low temperature and a short-pulse (picosecond-scale) laser providing the "spark plug." We expect to optimize fast ignition for fusion-class lasers to enable high yield. This will enable new experiments relevant to stockpile stewardship. If successful, the designs we develop with this project will be adopted by high-energy-density physics and inertial-confinement fusion programs.

Mission Relevance

One of the major goals of the Laboratory's current science and technology strategic initiative is to explore the scientific opportunities in high-energy-density science with particular focus on short-pulse, high-energy lasers. Our project will design a range of short-pulse laser, high-energy-density applications of relevance to the Stockpile Stewardship Program, as well as help fulfill LLNL's mission in breakthrough fundamental science and technology. This project also contributes towards the Laboratory's energy security mission, insofar as fast ignition, if experimentally confirmed, is an attractive option as a practical inertial-confinement fusion concept.

FY07 Accomplishments and Results

In FY07, we have (1) examined hot-electron conversion efficiency and energy distribution by performing two-dimensional particle-in-code simulations—absorption efficiency has been improved using grooved compared to flat surfaces; (2) performed indirect-drive integrated fast-ignition modeling that showed a dense and cold fuel assembly about five core radii away from the laser deposition region; (3) developed direct and indirect drive, one-dimensional optimized quasi-isochoric hydrodynamic implosions on the fusion-class-laser scale—two-dimensional simulations of these designs are underway with a reentrant gold cone that has shown reduced transport distances; and (4) identified an important energy-loss mechanism (explosive hydrodynamic expansion) that brings our calculations into closer agreement with experiments for hot hohlraum design optimization. This project has been incorporated into a new strategic initiative on “Fast-Ignition Proof-of-Principle Experiments,” which will expand upon this new approach to ignition in advanced, high-power lasers.

Publications

Clark, D. S., and M. Tabak, 2007. “A self-similar isochoric implosion for fast ignition.” *Nucl. Fusion* **47**, 1147. UCRL-JRNL-229868.

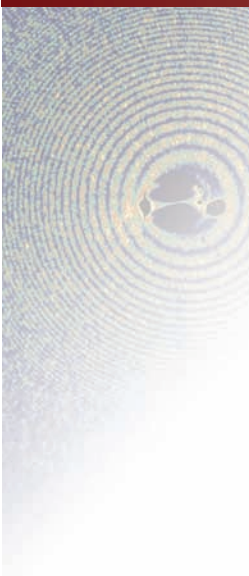
Clark, D. S., and M. Tabak, 2006. *Isochoric implosions for fast ignition*. 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-PRES-225372.

Kemp, A. J., et al., 2006. *Collisional relaxation of super thermal electrons generated by relativistic laser pulses in dense plasma*. Fast Ignition Workshop, Cambridge, MA, Nov. 3–5, 2006. UCRL-PRES-225465.

Kemp, A. J., et al., 2007. “Emittance growth mechanisms for laser-accelerated proton beams.” *Phys. Rev. E* **75**, 56401. UCRL-JRNL-226983.

Kemp, A. J., et al., 2007. *Laser heating of solid matter by light pressure-driven shocks at ultra-relativistic intensities*. Anomalous Absorption Conf., Maui, HI, Aug. 27–31, 2007. UCRL-PRES-233896.

Lasinski, B. F., et al., 2006. *PIC simulations of short-pulse, high-intensity light impinging on structured targets*. 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-PRES-225535.



Lasinski, B. F., et al., 2006. *PIC simulations of short-pulse, high-intensity light impinging on structured targets*. Fast Ignition Workshop, Cambridge, MA, Nov. 3–5, 2006. UCRL-PRES-225535.

Lasinski, B. F., et al., 2007. *PIC simulations of short-pulse, high-intensity light interacting with cone targets*. 49th Ann. Mtg. Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-ABS-232688.

Tabak, M., 2007. *Introduction to fast ignition*. Workshop on Nuclear Astrophysics, Livermore, CA, Aug. 27–30, 2007. UCRL-PRES-234061.

Tabak, M., 2007. *Suprathermal electrons and K-alpha production*. 5th Conf. Inertial Fusion Sciences and Applications, Kobe, Japan, Sept. 9–14, 2007. UCRL-POST-234120.

Tabak, M., 2007. *Suprathermal pressure in low mass short-pulse laser irradiated targets*. 49th Ann. Mtg. Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-POST-230122.

Tabak, M., et al., 2007. *Hydrodynamic assembly for fast ignition*. 49th Ann. Mtg. Division of Plasma Physics, Orlando, FL, Nov. 12–16, 2007. UCRL-PRES-230121.

Tabak, M., et al., 2006. *Hydrodynamic issues for fast ignition*. 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-POST-225428.

Tabak, M., et al., 2007. *Target assembly for fast ignition*. 5th Conf. Inertial Fusion Sciences and Applications, Kobe, Japan. Sept. 9–14, 2007. UCRL-POST-234228.

Town, R. P. J., et al., 2006. *Integrated modeling of fast ignition experiments*. Fast Ignition Workshop, Cambridge, MA, Nov. 3–5, 2006. UCRL-ABS-222479.

Town, R. P. J., et al., 2007. *Proton deflectometry of electric and magnetic fields*. 5th Conf. Inertial Fusion Sciences and Applications, Kobe, Japan. Sept. 9–14, 2007. UCRL-ABS-230120.

Town, R. P. J., et al., 2006. *Proton deflectometry of electric and magnetic fields*. 48th Ann. Mtg. Division of Plasma Physics, Philadelphia, PA, Oct. 30–Nov. 3, 2006. UCRL-ABS-222480.

Town, R. P. J., et al., 2007. *Status of fast ignition point designs for the NIF*. 5th Conf. Inertial Fusion Sciences and Applications, Kobe, Japan, Sept. 9–14, 2007. UCRL-ABS-232204.

Molecular Dynamics Simulations of Hot, Radiative Plasmas

Frank R. Graziani 07-ERD-044

Abstract

Applications as varied as inertial-confinement fusion and the physics of stars involve understanding the complex processes present in hot, dense radiative plasmas. Processes

interact with each other in highly nonlinear ways, and hence uncertainties in the physics can be amplified exponentially. Recent theoretical investigations of dense plasmas have been performed but are controversial because of the approximations made in those analyses. Direct numerical simulation of the complex interactions in plasmas offers an alternative, but they either rely on the collisionless approximation or ignore radiation. We propose to develop a new numerical simulation capability that addresses a currently unsolved problem—the extension of molecular dynamics to collisional plasmas where radiation is present.

This new code will be applied to the problem of thermalization. That is, given two species of plasma, each with their own temperature, what is the rate at which they relax to a common temperature because of the exchange of momentum and energy? This is the classic problem solved by Landau and Spitzer for Coulombic systems. We will initially consider single-species hot plasmas with varying ion atomic numbers, and investigate the effects of radiation on thermalization rates. Our final applications will be to multispecies, hot, dense radiative plasmas. Learning how the Landau–Spitzer model behaves in radiative plasmas could have significant impacts on Livermore’s supercomputing codes.

Mission Relevance

This project supports the Laboratory’s work in advanced computer simulations, especially as they relate to stockpile stewardship and energy security. This simulation capability will be the first time a microphysical (molecular dynamics) description of a plasma with radiation present will be done. Learning how Landau–Spitzer behaves in radiative plasmas could have large impacts on Laboratory supercomputer codes.

FY07 Accomplishments and Results

In FY07, we (1) implemented Coulomb-like semi-classical potentials in a domain-decomposition molecular dynamics code; (2) used the Ewald technique to verify expected power-law scaling; (3) validated our code against the molecular dynamics code used at Los Alamos National Laboratory; (4) ran simulations for all four of the Hansen–McDonald temperature equilibration problems, which showed an error in their analysis that proves the Landau–Spitzer model is not valid for moderately coupled Coulomb systems; (5) benchmarked the code against weakly coupled cases and confirmed that Landau–Spitzer works in the ideal plasma regime; and (6) wrote a stand-alone molecular dynamics code that includes radiation and verified that the bremsstrahlung emission is dipole.

Proposed Work for FY08

For FY08, we will explore the inclusion of radiation into the domain-decomposition molecular dynamics code. Issues to be addressed include scalability, boundary conditions, and the effect of radiation on energy exchange rates. The atomic physics of partially ionized plasmas will be explored in our stand-alone molecular dynamics code.

Quantum Properties of Plutonium and Plutonium Compounds

Michael J. Fluss 07-ERD-048

Abstract

The anomalous properties of plutonium and plutonium compounds arise from electronic correlation effects, but the nature, organizing principle, and order parameter of these correlations is unknown. In addition to ongoing negative-pressure tuning with americium, we will exploit physical property measurements of plutonium at very low temperatures using low-specific-activity plutonium-244 and characterizing plutonium superconductivity to understand the element's ground state. We are using the 300-keV transmission electron microscope for atomic-scale characterization, which should lead to an understanding of structure property aspects of the superconducting compound PuCoGa_5 . The goal is to understand how plutonium's quantum properties are connected to its unusual physical properties.

This project will result in an improved physical picture of the electronic structure of plutonium and a concomitant understanding of the consequences of real-world effects such as impurities, radiation damage, radiogenic products, pressure, and temperature. Measurement of the spin-pairing mechanism will identify the underlying basis for many of plutonium's properties. In a scientifically broader sense, significant progress in this area will result in an improved physical picture for many other complex materials such as copper-oxide superconductors. Successful processing and use of plutonium-244 metal in scientific studies creates important new skills and knowledge for LLNL that opens the door to a completely new class of experimentation on plutonium.

Mission Relevance

This project sustains and nurtures the Laboratory's expertise in fundamental plutonium science by focusing on a 21st century solid-state challenge—understanding of the 4f and 5f elements in the vicinity of the f-electron localization. The project will result in new skills and usher in a new class of plutonium research using low-activity plutonium-244 to perform experiments impossible any other way. The research proposed here, if successful, has the potential to profoundly change our understanding of the fundamental properties of plutonium and plutonium compounds, an important effort that supports the Laboratory's mission in stockpile stewardship and the ability to create breakthroughs in fundamental science and technology.

FY07 Accomplishments and Results

In FY07, we (1) made measurements to determine the spin pairing believed to be extant in plutonium and its compounds, (2) prepared and characterized transmission electron microscope specimens of aged and un-aged (annealed) PuCoGa_5 to determine consequences of radiation defects, (3) identified a "critical" plutonium–americium composition by observing a new metastable disordered phase of plutonium–americium at about 30 K, (4) observed a nonlinear increase in the magnetic susceptibility of plutonium with

americium concentration, (5) designed new instrumentation to measure physical properties of plutonium-244, (6) constructed an apparatus to measure magnetoresistivity and the Hall effect on plutonium foils, and (7) published several papers and gave several invited and conference presentations.

Proposed Work for FY08

For FY08, we will (1) complete negative-pressure studies using americium chemical tuning and a combination of disorder studies as a function of composition, with determination of the magnetization, resistivity, and Hall effect; (2) complete electron transmission microscopy of the PuCoGa_5 superconductor to identify the physical structure property basis for critical temperature reduction from radiation and to correlate our results with earlier muon and nuclear magnetic-resonance studies; (3) continue preparations to determine the ultralow-temperature physical property measurements of alpha- and delta-plutonium using low-specific-activity plutonium-244; and (4) investigate the electron-pairing mechanism in plutonium and its alloys using tunneling experiments such as Andreev scattering.

Publications

Chapline, G. F., M. J. Fluss, and S. K. McCall, 2007. "Plutonium and quantum criticality." *J. Alloys Comp.* **444–445**, 142. UCRL-PROC-226225.

Heffner, R. H., et al., 2007. "The search for magnetic order in (delta)-Pu metal using muon spin relaxation." *J. Alloys Comp.* **444–445**, 80. UCRL-PROC-225444.

Jackson, D. D., et al., 2007. "Cubic laves ferromagnet TbNi_2Mn investigated through ambient-pressure magnetization and specific heat and high pressure AC magnetic susceptibility." *Phys. Rev. B* **75**, 224422. UCRL-JRNL-226777.

Jackson, D. D., et al., 2007. "Unconventional ferromagnetic ordering of Ce in the anisotropic metal CeCrSb_3 ." *Phys. Rev. B* **76**, 64408. UCRL-JRNL-224810.

McCall, S. K., et al., in press. "Influence of self damage on $\text{Pu}(\text{Am})$ observed through magnetization measurements." *Physica B*. UCRL-CONF-229633.

McCall, S. K., et al., 2007. "Isochronal annealing studies in Pu and Pu alloys using magnetic susceptibility." *J. Alloys Comp.* **444–445**, 168. UCRL-PROC-225490.

McCall, S. K., et al., 2007. "Radiation damage effects on the magnetic properties of $\text{Pu}_{(1-x)}\text{Am}_x$ ($x=0.224$)." *Actinides 2006 — Basic Science, Applications and Technology, MRS Symp. Proc.* **986**, 0986-0003-03. UCRL-PROC-226893.

Finding and Characterizing Rare Events in Two Next-Generation Particle Astrophysics Experiments

Adam Bernstein 07-ERD-056

Abstract

We propose to directly detect dark matter as well as measure the neutrino-oscillation parameter θ_{13} —both endeavors that are widely acknowledged as among the most important in particle astrophysics for the coming decades. We will build detectors with unprecedented levels of sensitivity for the dark matter search, and with record low levels of systematic error for oscillation measurement.

If successful, we will be key collaborators in first-ever direct detection of dark-matter particles and the first-ever measurement of the neutrino-oscillation parameter θ_{13} . The detection of dark matter is one of the most important scientific questions in the 21st century. Measurement of the neutrino-oscillation parameter has nearly the same level of scientific significance. This project will attract top scientists to the Laboratory, with technical skills that are directly relevant to immediate needs in LLNL radiation-detection programs.

Mission Relevance

This project will help fulfill LLNL's mission of breakthrough fundamental science and technology. The skills being developed here have direct relevance to Laboratory global nuclear security missions. In particular, the advanced simulation capabilities, event-selection algorithms, and advanced detection concepts we pursue will have immediate relevance to ongoing reactor monitoring and special nuclear material detection efforts by the DOE and Department of Homeland Security. Additionally, it has been recognized that antineutrinos are a potentially valuable signature for reactor monitoring and treaty verification. This project increases our already significant competency in antineutrino detection, and will be concurrently applied to nonproliferation efforts and basic science research.

FY07 Accomplishments and Results

In FY07, we (1) established a world-record limit on allowable properties of dark matter with our 10-kg dual-phase xenon detector and submitted our results to *Physical Review Letters*, (2) formed a collaboration to build a 300-kg xenon detector for the Homestake Deep Underground Science and Engineering Laboratory in South Dakota, (3) initiated electronics design and performance simulation for the 300-kg detector, and (4) implemented a custom Monte Carlo capability for detailed calibration studies of the Double Chooz detector at the Chooz nuclear power station in France.

Proposed Work for FY08

In FY08, we will (1) simulate, assemble, and deploy a prototype 300-kg dark-matter detector; (2) perform simulation and design work for a 2-m-diameter gadolinium-doped water shield to screen backgrounds from this highly sensitive detector; and (3) expand our simulation capability for the Double Chooz antineutrino detector.

A Plasma Amplifier toward Zettawatt Laser Powers

Robert Kirkwood 07-ERI-004

Abstract

We propose to develop a technique to increase laser power with pulse compression in a plasma, which builds on recent successes with the Janus laser. The goal is to demonstrate pulse compression that will allow laser facilities to operate with as much as 1000-times higher output powers than obtained with conventional approaches. This proposed technology is possible because of the much higher power-handling limits of a plasma ($>10^{14}$ W/cm²) than of conventional solid-state optics ($<10^{11}$ W/cm²). The project is a critical step in enhancing the high-power operating regime for fusion-class lasers to the zettawatt realm, and has broad application in inertial fusion, radiation sources, and particle accelerators, as well as providing critical data on saturation of stimulated Raman scattering.

We expect that experiments with the Janus and Compact Multipulse Terawatt (COMET) laser systems in the Jupiter Laser Facility at LLNL will demonstrate depletion of the Janus beam by Raman amplification of the 1-ps COMET beam for more than 10 ps, thereby demonstrating pulse compression of a 1-ns beam in a plasma. We also will establish the laser beam intensity and plasma conditions necessary to compress an entire 1-ns pump in future experiments. In addition, we will obtain measurements of the saturated levels of seeded stimulated Raman scattering to verify models of wave saturation, on a time scale too rapid for ion wave motions. Finally, we will perform experiments combining pump beams for pulse compression of multiple beams. We expect our results will lead to publications in peer-reviewed journals.

Mission Relevance

The success of this research will support Laboratory missions in both stockpile stewardship and energy security, and open up new opportunities in high-energy-density science by substantially increasing the power of many existing lasers. The success of these experiments would leverage the existing petawatt beam program aimed at opening new, high-power operating regimes, and would enable study of laser–plasma interactions on the short time scale necessary to identify electron and ion saturation processes.

FY07 Accomplishments and Results

In FY07, we planned and set up experiments to demonstrate the focal quality of an amplified ultrashort-pulse seed beam and study its dependence on the plasma length, to optimize plans for depleting the long-pulse pump beam that amplifies the seed. This additional objective was adopted because of favorable results in similar experiments recently carried out at Princeton, and will allow an optimized configuration for our upgraded frequency shifter. We also designed and began construction of a frequency shifter having increased output energy, which is comprised of a two-stage Raman gas cell—this will facilitate experiments in FY08.

Proposed Work for FY08

In FY08, we will perform a series of experiments to demonstrate the compression of a 10- to 15-ps portion of a nanosecond pulse to greater than 1-ps duration by using plasma conditions optimized for good focal spot quality and minimum seed energy, which we will initially identify. When successful, this will be the first demonstration of pulse compression in a plasma relevant to 1-ns pulses. The results will build on our experiments with large plasma length at various values of seed- and pump-beam intensity.

Publications

Kirkwood, R. K., et al. 2007. "Amplification of an ultrashort pulse laser by stimulated Raman scattering of a 1 ns pulse in a low density plasma." *Phys. Plasmas* **14**, 113109. UCRL-JRNL-235750.

Helium Burning in Steady State and Explosive Nucleosynthesis

Jason T. Burke 07-LW-006

Abstract

We propose a project involving helium burning in stellar environments using the Silicon Telescope Array for Reaction Studies (STARS) and Livermore Berkeley Array for Collaborative Experiments (LIBERACE) combined detector system. We will perform a triple-alpha experiment to measure the radiative width of the Hoyle resonance to a precision better than 5% in a single experiment, which makes use of a highly segmented large-area silicon detector setup in the STARS system. A second experiment measures a key cross section that produces titanium-44 in supernova from the reaction $^{40}\text{Ca}(\alpha, \gamma)^{44}\text{Ti}$. Decay of titanium-44 is a signature of the dynamics of explosive helium burning in supernova remnants such as Cassiopeia A. This experiment will use two methods to determine the cross section below 4 MeV—an in-beam measurement followed by a low-background count of the activation product—which will be performed as a collaborative effort with the University of Washington.

We expect to produce, for the first time, a high-precision single measurement of the triple-alpha radiative width. The result will be used as a critical input to stellar modeling codes used by the astrophysics community. The cross section for the production of titanium-44 from the reaction of $^{40}\text{Ca}(\alpha, \gamma)^{44}\text{Ti}$ currently has an uncertainty factor of two or more. Our self-consistent, independent determinations of the cross section should provide an absolute value with an uncertainty on the order of 5%. This would have direct impact on the interpretation of supernova remnant Cassiopeia A. Both results will be submitted to highly regarded journals such as *Nature* and *Physical Review Letters*.

Mission Relevance

Exploring stellar thermonuclear phenomena similar to those in man-made nuclear explosions benefits fundamental research used in stockpile assessment and in thermonuclear experiments at future, fusion-class lasers, in support of the Laboratory's national security

mission. Specifically, this project explores the ability to measure low-yield cross sections through direct (in-beam) and activation methods, which is of high scientific value to the nuclear and astrophysics community. Our experiments in cross-section measurements, particle detection, gamma-ray spectroscopy, nuclear physics, and astrophysics will benefit Laboratory efforts in fundamental science and technology breakthroughs, as well as attract talented student and postdoctoral researchers.

FY07 Accomplishments and Results

In FY07 we (1) completed our preliminary analysis of the results of the successful triple-alpha experiment, (2) fabricated and set up the data-acquisition system and designed and assembled the calcium-40 target production apparatus at Lawrence Livermore, (3) performed calculations to establish count rates and developed target fabrication techniques to minimize contaminant backgrounds for the in-beam portion of the $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$ experiment, and (4) gave several invited talks on this work at nuclear physics and chemical society meetings.

Proposed Work for FY08

For FY08, we will (1) perform the $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$ cross-section in-beam production run, (2) perform low-background counting of the titanium-44 to confirm the in-beam results, and (3) prepare papers for publication summarizing the triple-alpha results as well as both results for the $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$ cross-section experiments.

Uncovering Supersymmetric Leptons at the Large Hadron Collider

Jeffrey B. Gronberg 07-LW-037

Abstract

A new era in particle physics will begin when the Large Hadron Collider (LHC) comes fully online in 2008. The LHC will be the first collider to reach the energy regime where current theory predicts a completely new realm of particle physics. Although expected to discover new fundamental particles, the LHC will be blind to an entire class of new particles—supersymmetric leptons. We propose to develop a novel triggering technique—based on photon–photon interactions of beam protons—that will allow these particles to be discovered. We will study the analogous process of dimuon production in photon–photon events from actual data to determine whether a supersymmetric lepton trigger would be successful.

The LHC experiments, as currently designed, will miss a significant piece of the puzzle necessary to discover the fundamental physics occurring at the teraelectronvolt scale. To complete that picture, experiments will need to have the capability to observe and study supersymmetric leptons. In this project we will analyze dimuon production and, using these results, determine the feasibility of a supersymmetric lepton trigger. A trigger upgrade allowing supersymmetric leptons to be discovered would be a major enhancement to the physics program of the LHC, leading to major physics results, carving out a scientific leadership role for LLNL, and resulting in papers published in high-profile, peer-reviewed journals.

Mission Relevance

By providing access to the latest developments in detector technology, data processing, and data mining, this project furthers LLNL's missions in nonproliferation and homeland security, which heavily utilize all three of these capabilities. This project also will help recruit scientific talent in high-energy physics, whose expertise is applicable to radiation detection and other cutting-edge national security work.

FY07 Accomplishments and Results

In FY07, we (1) analyzed the production cross section for sleptons and determined that we should produce about 100 events per slepton type over the course of a year's data taking; (2) established the Monte Carlo simulation and detector modeling of slepton production and decay, demonstrating that the major slepton decay mode has a reasonable efficiency in the level-1 trigger of the Compact Muon Solenoid particle detector at LHC; (3) added forward-proton tracking code to the simulation and began analyzing detector efficiency; (4) developed an initial dimuon trigger algorithm; (5) added Standard Model backgrounds to the simulations to determine signal efficiency and required background rejection; and (6) hired a postdoctoral researcher.

Proposed Work for FY08


In FY08, with access to the first data sets from LHC, we will implement a dilepton trigger to analyze the production of gamma–gamma events, to measure and benchmark backgrounds to the slepton trigger, and to observe counterfeit rates in the trigger. This information will be fed back into the analysis of the slepton trigger algorithm to determine its final efficiency. The slepton trigger design and the analysis of gamma–gamma to dilepton events will be published in a peer-reviewed journal.

Molecular-to-Extended-Solid Transformations in Compressed Carbon Dioxide: Six-Fold-Coordinated Carbon Dioxide

Valentin Iota-Herbei 07-LW-049

Abstract

This project will determine the stability and other properties of six-fold-coordinated extended-solid carbon dioxide (CO_2) VI, believed to be similar to the ultrahard silicon dioxide– (SiO_2) phase stishovite. We will determine (1) the phase–temperature domain of stability for six-fold CO_2 , (2) its atomic structure and physical properties, and (3) the local coordination of carbon in the new phase. Our experiments will establish the connection between the presumed CO_2 intermediate bonding phases (associated II and bent molecular IV) and their extended-solid counterparts. We will use x-ray spectroscopy and diamond anvil cells to measure the degree of intermolecular electron sharing in the intermediate phases, and to establish whether electron delocalization occurs gradually via intermediate phases or is an abrupt process.



This project will address two important scientific issues. The first is determining the existence, stability, and physical properties of a six-fold-coordinated phase of CO₂ at high pressures and temperatures. If proven, this phase would be the first example of six-fold coordination in carbon found in nature. The discovery of this presumably superhard, energy-dense material would have implications for volcanic activity in terrestrial planets. The second issue is mapping, using ab initio calculations, the evolution of intermolecular and intramolecular interactions during the transition of CO₂ from molecular to extended solid. This accomplishment would have implications for a wide range of materials.

Mission Relevance

Six-fold-coordinated CO₂ VI is likely a superhard, energy-dense material, with potential technological applications. In addition, this work will provide a comprehensive understanding of the role of electron delocalization in the phase stability of compressed molecular systems. The experimental methods and physical understanding developed herein will help other projects on molecular systems (e.g., dihydrogen and dideuterium) of interest to stockpile stewardship.

FY07 Accomplishments and Results

We studied the phase diagram of CO₂ at pressures up to 100 GPa and temperatures to 2000 K. Using externally heated membrane-actuated diamond anvil cells, we successfully reduced the influence of transition kinetics effects and discovered two new extended solids. One of the two new phases discovered, CO₂ VI, likely consists of a six-fold-coordinated carbon network similar to SiO₂ polymorph stishovite. The discovery and characterization of this new phase extends the similarity between SiO₂ and CO₂, with major implications for Earth interior models and the physics of superhard materials.

A Proposal for First-Ever Measurement of Coherent Neutrino–Nucleus Scattering

Celeste D. Winant 07-LW-062

Abstract

We propose to build and deploy a 10-kg dual-phase argon ionization detector to measure coherent neutrino–nucleus scattering, which is described by the reaction (neutrino) + (Z,N) → (neutrino) + (Z,N). Our group would be the first to make this measurement, and its detection would validate a central tenet of the Standard Model. The existence of this process is also relevant to astrophysics, where coherent neutrino scattering is assumed to moderate energy transport within neutron stars. We will first use a neutron beam at Lawrence Livermore to demonstrate that our detector can achieve the sensitivity required for coherent scattering detection. We will then search for coherent scattering with reactor antineutrinos by deploying our detector at the San Onofre Nuclear Generating Station.

We expect to field a detector with the sensitivity needed to detect coherent neutrino–nucleus scattering at the San Onofre Nuclear Generating Station. By achieving this goal, we also will

have (1) developed a detector with unprecedented sensitivity to low-energy nuclear recoil events, (2) furthered our knowledge and control of backgrounds in such experiments, (3) measured the spectrum of medium-energy (100-keV to 10-MeV) neutrons, and (4) established a dual-phase research program at the Laboratory. These intermediate goals have far-reaching consequences, and will benefit other research pursuits at LLNL (e.g., dark matter, neutron studies, and general radiation detection).

Mission Relevance

The fundamental science and applied technology missions of LLNL are supported through development of novel detectors with potential for large scientific return in astrophysics and particle physics. In addition, dual-phase detectors are expected to improve cooperative monitoring of nuclear reactors, as required by the Nuclear Nonproliferation Treaty, thereby supporting a fundamental Laboratory mission of ensuring national security.

FY07 Accomplishments and Results

In FY07, we (1) studied and optimized the gas mixture and pressure of our gas-phase detector, (2) designed a dual-phase detector for deployment at a reactor, and (3) measured the low-energy quench factor of argon by exposing the detector to a pulsed beam of 80-keV neutrons, resulting in an approximately flat argon recoil spectrum with an endpoint of 8 keV. The electron-equivalent energy calibration was performed with a built-in iron-55 source.

Fourier Transform Holography with Coded Apertures

Stefan Hau-Riege 07-LW-086

Abstract

We will develop a new materials and biological imaging technique that will boost holographic signals by two orders of magnitude and improve resolution ten times. In Fourier holography, resolution is limited by the reference object size, whereas a small object only weakly scatters the incident wave. Using a specially designed reference object with point-like autocorrelation—a uniformly redundant array (URA), also known as a coded aperture—we can improve the signal and resolution. Material science and biological applications include imaging high-power laser target aerogel foams, copper inclusions in beryllium–copper alloys, voids in plutonium, organelles of cells, and even macromolecules. We plan to put our technique into practice by fabricating reference objects and collecting holograms at the University of California Berkeley’s Advanced Light Source (ALS) and the free-electron laser FLASH in Hamburg, Germany.

We expect to perform an experimental demonstration of a new imaging technique with improved signal and resolution, which has been simulated at LLNL. We intend to obtain a high-resolution lithographic URA and image samples of interest to Livermore. In parallel, we propose to use known reference macromolecules to obtain images at atomic resolution. Eventually we expect to produce a high-resolution, three-dimensional hologram, and publish our results in peer-reviewed journals.

Mission Relevance

Our advanced imaging technique will enable analysis of the mechanical properties of materials of interest to the Stockpile Stewardship Program. When ultimately applied to bioimaging, this technique will also benefit the homeland security mission by providing an extremely powerful tool for determining pathogen structure. In addition, this technique will place LLNL at the forefront of imaging science, and advance collaborations at two major x-ray facilities: Berkeley's ALS and the Linac Coherent Light Source at Stanford.

FY07 Accomplishments and Results

In FY07 we pioneered Fourier–Hadamard transform x-ray holography to explore the nanoworld and the ultrafast. Specifically, we (1) used focused ion beams to nanofabricate URAs for use as binary reference next to test objects, coccolith structures, and bacterial cells; (2) imaged these structures with 44-nm spatial resolution at the ALS and 15-fs temporal resolution at the free-electron laser FLASH, which is among the best-ever reported for holography of micron-sized objects; (3) further identified DNA nanostructures as potential complex references, and submitted a record of invention; (4) made significant progress in linking and aligning rod-shaped tobacco mosaic viruses and the bacteriophage MS2 and cowpea mosaic virus; and (5) presented our results at numerous conferences, which have been submitted for publication.

Proposed Work for FY08

In FY08 we will embark on three-dimensional imaging. We will first image test objects and then focus our attention on objects of programmatic interest to LLNL, such as ultralow density nanofoams (aerogels). Because our URA-holographic imaging technique allows us to obtain images more rapidly and of higher quality than existing methods, we will be able to study different types of aerogels made of germanium, aluminum, silicon, or tantalum and reliably determine their three-dimensional morphology. We will further fabricate two-dimensional arrays of aligned macromolecular hybrid complexes or weave DNA strands into desired shapes ("DNA origami") and image them.

Publications

Boutet, S., et al., 2007. *Femtosecond diffraction imaging and coded aperture holography of a single cell*. Coherence 2007—Intl. Workshop Phase Retrieval and Coherent Scattering, Pacific Grove, CA, June 25–28, 2007. UCRL-POST-235322.

Chung, S.-W., et al., 2007. *Directed assembly of virus particles at nanoscale chemical templates*. 2007 Materials Research Society Spring Mtg., San Francisco, CA, Apr. 9–13, 2007. UCRL-PRES-229811.

Marchesini, S., and S. Hau-Riege, 2007. *Computational lenses*. Coherence 2007—Intl. Workshop Phase Retrieval and Coherent Scattering, Pacific Grove, CA, June 25–28, 2007. UCRL-PRES-235339.

Marchesini, S., et al., 2007. *Ultrafast coherent diffractive imaging with x-ray free-electron lasers*. III Reunión Nacional de Usuarios de Radiación de Sincrotrón (Spanish Synchrotron Users Network), Jaca, Spain, July 9–11, 2007. UCRL-PRES-235353.

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