UCRL-TR-113717-04

LDRD

LABORATORY DIRECTED RESEARCH AND DEVELOPMENT

FY2004 ANNUAL REPORT

LAWRENCE LIVERMORE NATIONAL LABORATORY

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Acknowledgments

This *Annual Report* provides an overview of the FY2004 Laboratory Directed Research and Development (LDRD) Program at Lawrence Livermore National Laboratory (LLNL) and presents a summary of the results achieved by each LDRD project. At LLNL, Laboratory Director Michael Anastasio and Deputy Director for Science and Technology Cherry Murray are responsible for the LDRD Program and delegate responsibility for the operation of the Program to the Associate Deputy Director for Science and Technology and the Director of the Laboratory Science and Technology Office (LSTO), Rokaya Al-Ayat. The LDRD Program at LLNL is in compliance with Department of Energy (DOE) Order 413.2 and other relevant DOE orders and guidelines.

The LDRD Program extends its sincere appreciation to the principal investigators of the FY2004 projects for providing the content of the *Annual Report* and to the publications team. The Program also thanks the following members of the LSTO team for their many contributions to this publication: Mary Callesen, administrator; Nancy Campos, database manager; Andrew Hurst, computer specialist; and Cathleen Sayre, resource manager.

Scientific Editors
Kenneth Jackson, Karl van Bibber

Publication Editors
Paul Kotta, Karen Kline

Publication Designer
Kitty Madison

Compositor
Louisa Cardoza

Michael R. Anastasio, Director

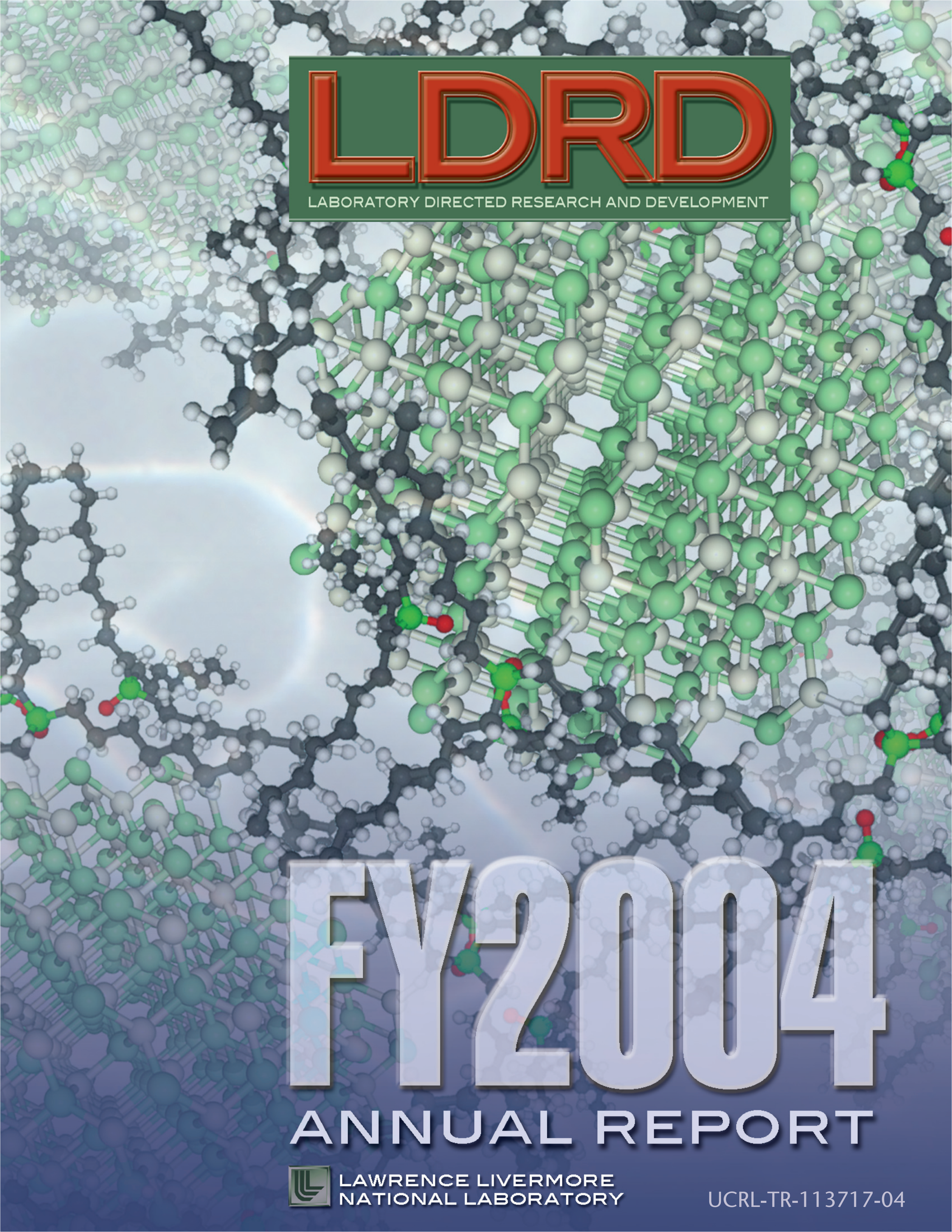
Director's Statement

The Laboratory Directed Research and Development (LDRD) Program is our primary means for pursuing innovative, long-term, high-risk, and potentially high-payoff research that supports the missions of the Laboratory, the Department of Energy, and the National Nuclear Security Administration in national security, homeland security, energy security, environmental management, bioscience and healthcare technology, and breakthroughs in fundamental science and technology. The LDRD Program was authorized by Congress in 1991 and is administered by the Laboratory Science and Technology Office. The accomplishments described in this *Annual Report* demonstrate how the LDRD portfolio is strongly aligned with these missions and contributes to the Laboratory's success in meeting its goals.

The LDRD budget of $69.8 million for FY2004 sponsored 220 projects. These projects were selected through an extensive peer-review process to ensure the highest scientific and technical quality and mission relevance. Each year, the number of meritorious proposals far exceeds the funding available, making the selection a challenging 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 and homeland security missions.



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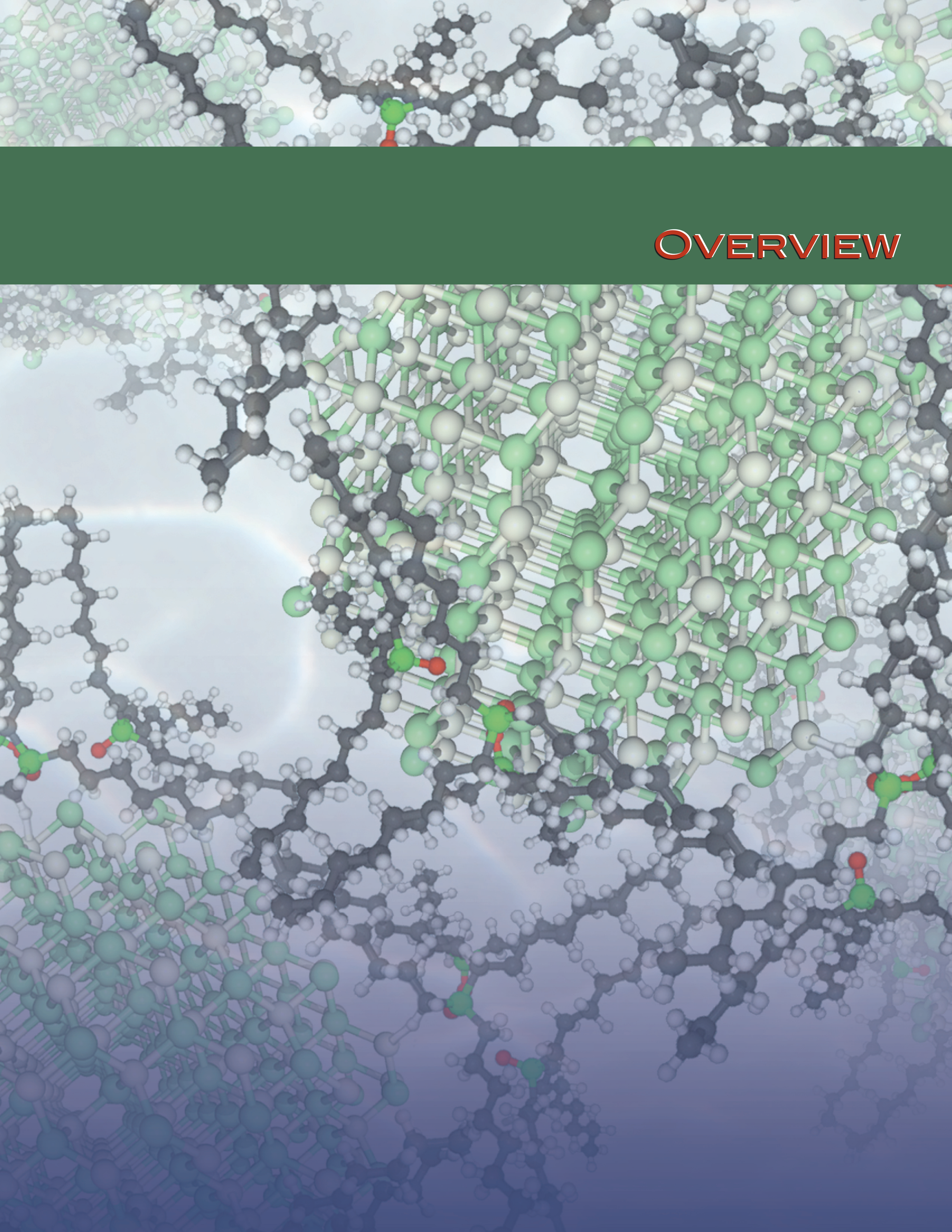
Overview



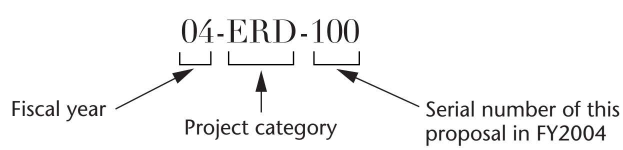
About the FY2004 LDRD Annual Report

The *FY2004 Laboratory Directed Research and Development (LDRD) Annual Report* provides a summary of LDRD-funded projects for the fiscal year and consists of two parts:

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

Project Summaries: A summary of each project, submitted by the principal investigator. Project summaries include the scope, motivation, goals, relevance to DOE/NNSA and LLNL mission areas, the technical progress achieved in FY2004, and a list of publications that resulted from the research in FY2004.

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:



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Program Overview

Investing in our nation's future

About Lawrence Livermore National Laboratory

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

The Laboratory leverages the scientific and engineering expertise and facilities developed for its primary mission to pursue advanced technologies to meet other important national security needs—homeland security, 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 healthcare technology, and breakthroughs in fundamental science and technology. With this multidisciplinary expertise, the Laboratory serves as a science and technology resource to the U.S. government and as a partner with industry and academia.

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

Laboratory Directed Research and Development Program

To fulfill its missions, LLNL must continually invest in the science and technology that form the foundation of its signature capabilities. The Laboratory Directed Research and Development (LDRD) Program, which was established by Congress at all DOE national laboratories in 1991, is LLNL's most important single resource for supporting 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 funding creative basic and applied research activities in areas aligned with its missions, the LDRD Program develops and extends the Laboratory's intellectual foundations and

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

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maintains its vitality as a premier research and applied science institution. Many of the present scientific and technical strengths of LLNL are, in large part, a product of LDRD investment choices in the past.

The LDRD Portfolio Management Process

The FY2004 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 FY2004, the top-level LDRD strategic planning process was guided by the DOE Strategic Plan[2] for the next 25 years, and by the Laboratory's own Long-Range Science and Technology (S&T) Plan that will define the scientific and technical strategy for the coming decade.

The 2004 DOE Strategic Plan articulates four strategic goals for achieving the DOE mission of advancing the national, economic, and energy security of the U.S., promoting scientific and technological innovation in support of that mission, and ensuring the environmental cleanup of the national nuclear weapons complex. In FY2004, the LLNL LDRD Program strongly supported all four DOE strategic goals:

1. Defense Strategic Goal—To protect our national security by applying advanced science and nuclear technology to the Nation's defense.
2. Energy Strategic Goal—To protect our national and economic security by promoting a diverse supply and delivery of reliable, affordable, and environmentally sound energy.
3. Science Strategic Goal—To protect our national and economic security by providing world-class scientific research capacity and advancing scientific knowledge.
4. Environment Strategic Goal—To protect the environment by providing a responsible resolution to the environmental legacy of the Cold War and by providing for the permanent disposal of the Nation's high-level radioactive waste.

The Laboratory's Long-Range S&T Plan continues to inform the LDRD portfolio planning process. Broadly inclusive and continuously evolving to meet changing circumstances, the 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 Long-Range 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
- Information, simulations, and systems
- Energy and environmental science and technology

[2] U. S. Department of Energy. (2003). *Strategic Plan*. http://strategicplan.doe.gov/ (retrieved January 28, 2005).

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The NNSA oversees LLNL's LDRD Program to ensure that it accomplishes its objectives, is administered in full compliance with DOE Orders and pertinent Congressional requirements, and is consistent with NNSA's Strategic Plan.[3] This oversight includes field and headquarters reviews of both the technical content and management processes. All LDRD proposals at LLNL are coordinated with the NNSA Livermore Site Office, and approvals are granted on a case-by-case basis. Furthermore, NNSA Headquarters approves the LDRD Program Plan and authorizes annual funding. At the end of each financial year, DOE also reports on LDRD programs at all DOE national laboratories in the DOE LDRD Annual Report to Congress.

As demonstrated in a memorandum (April 30, 2002) from the Secretary of Energy and the NNSA Administrator, the DOE/NNSA actively supports the LDRD Program. In the memo, Secretary of Energy Spencer Abraham writes: "I believe that all sponsors, including other Federal agencies, benefit from the strong science and technology base provided by the Department's [LDRD] programs. [LDRD] is at the core of our ability to develop research capabilities and apply advanced technologies to effectively meet the Department's and the Nation's needs."[4]

Structure of the LDRD Program

Project Categories

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

Strategic Initiative

The SI category focuses on innovative R&D activities that are likely to set new directions for existing programs, help develop new programmatic areas within LLNL's mission responsibilities, or enhance the Laboratory's science and technology base. Projects in this category are usually larger and more technically challenging than projects funded in other categories. An SI project must be aligned with the strategic R&D priorities of at least one of the six thematic areas of LLNL's Long-Range 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 FY2004, LLNL's Long-Range 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

[3] National Nuclear Security Administration. (2004). *Strategic Plan.* http://www.nnsa.doe.gov/stratplan_toc.htm (retrieved February 15, 2005).

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

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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 explore the feasibility of a new concept and help define and develop potential projects in the other three categories. To increase its responsiveness to Laboratory scientists and engineers, the LDRD Program funds FSs and PDs throughout the year.

Research Categories

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, specified by DOE/NNSA, 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

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The LDRD FY2004 Portfolio

Investing in our nation's future

Overview of the FY2004 Portfolio

The FY2004 LDRD portfolio was carefully structured to continue the Program's vigorous support for the strategic vision and long-term goals of DOE/NNSA and LLNL. The FY2004 projects described in this *Annual Report* underwent a stringent selection process and received ongoing management oversight. Each project was reviewed and approved by the NNSA Livermore Site Office, and NNSA Headquarters approved the LDRD FY04 Program Plan and funding on September 30, 2003.

In FY2004 the Program funded 220 projects with a total budget of $69.8 million. Figure 1 shows the number of projects in each of the four categories. Figure 2 shows the distribution of funding among the four LDRD project categories.

Strategic Initiative

In FY2004, the LDRD Program funded six SI projects. Although the SI category represented only 3% of the total number of LDRD projects for FY2004, it accounted for 16% of the budget. Strategic Initiative projects ranged in funding levels from $607K to $3.9M.

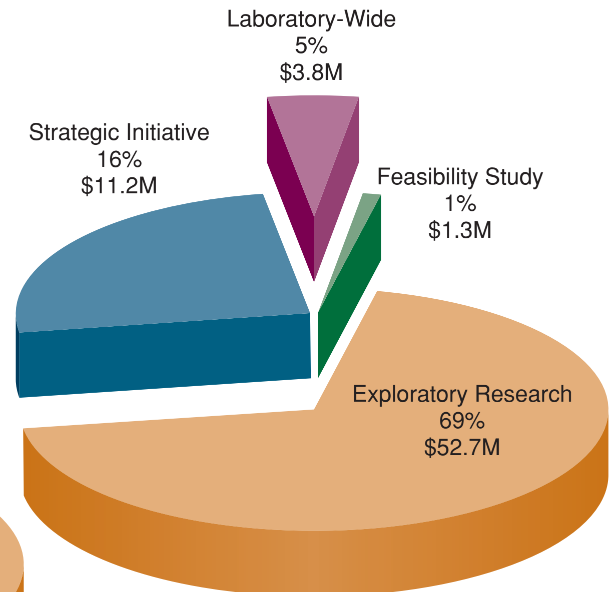

Figure 2. Distribution of funding among the four LDRD project categories. Total funding for FY2004 was $69.8M.

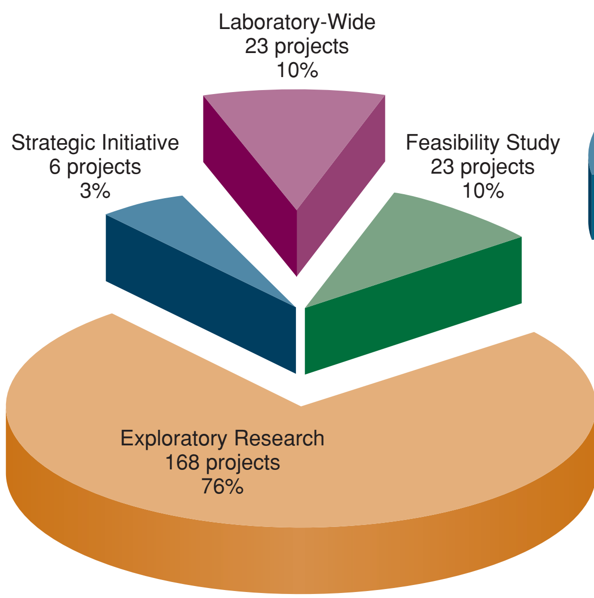

Figure 1. Number and percentage of the 220 LDRD projects in each project category in FY2004.

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Exploratory Research

In FY2004, 168 ER projects were funded. The largest project category, ERs account for 76% of LDRD projects for the fiscal year. Projects in this year's ER category ranged in budget from $44.2K to $1.2M.

Laboratory-Wide Competition

Twenty-three LW projects were funded in FY2004, which represent 10% of LDRD projects for the year and 5% of the budget. Laboratory-Wide projects are limited to $190K/yr funding, with a few exceptions. In FY2004, LW projects ranged in funding level from $73.4K to $210K.

Feasibility Study

In FY2004, the Program funded 23 FS projects, or 10% of the total. Feasibility Studies are limited to $75K and a 12-month duration.

Figure 3 shows the funding distribution by dollar amount for the 220 FY2004 projects. Sixty-six percent of the projects were in the $101K to $500K range, with 23% falling below $100K. This lowest funding level includes all the FS projects. Almost 7% of the projects were in the $501K to $1M funding range, and only 4% of the projects received more than $1M. The average funding level for the 220 projects was $317K.

Figure 4 shows the percentage of Program funding and number of projects in each reasearch category for FY2004.

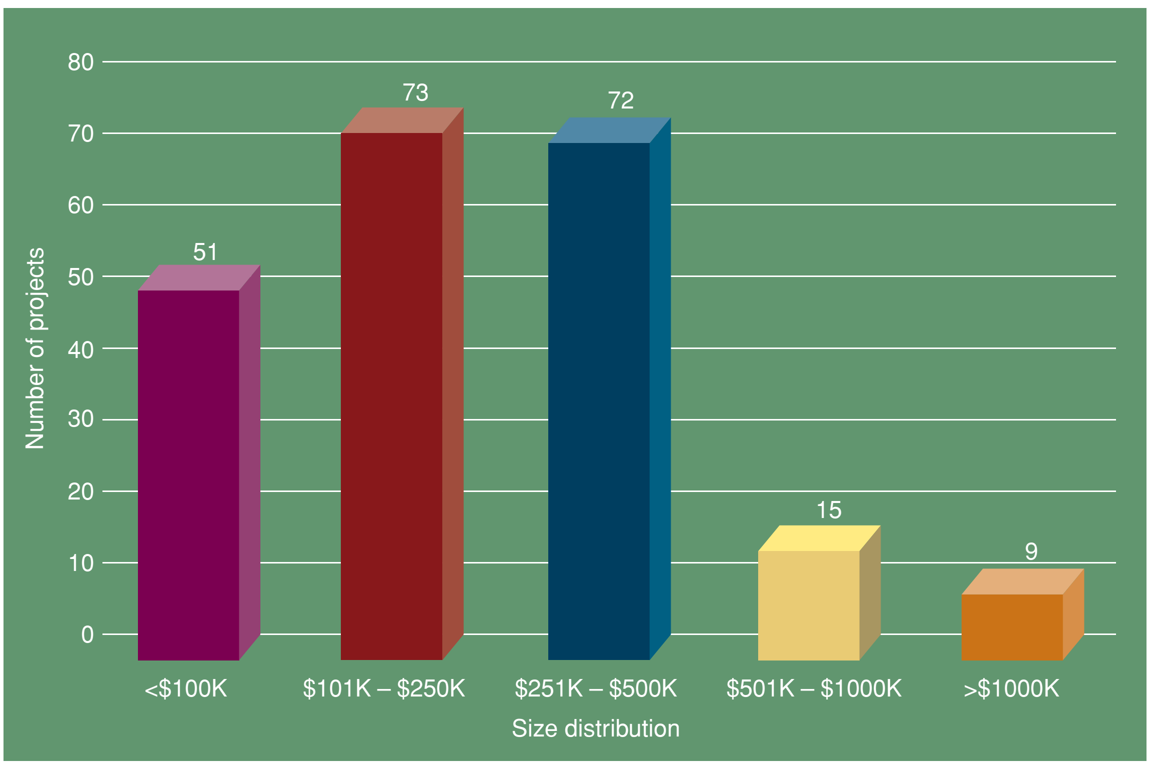

Figure 3. Number of projects and levels of funding. The average funding level for an LDRD project in FY2003 was $317K.

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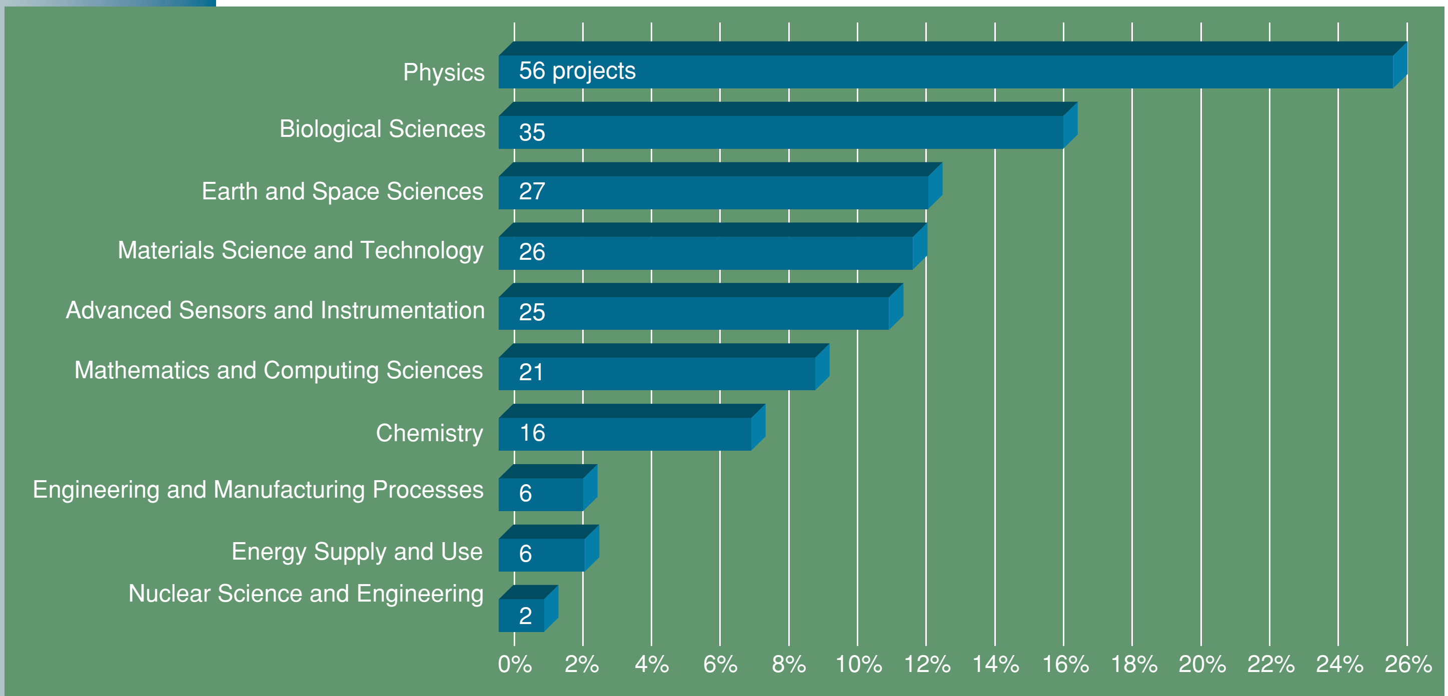

Figure 4. Percentage of LDRD funding and number of projects in each research category in FY2004.

Highlights of FY2004 LDRD Accomplishments

In FY2004, 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 FY2004 highlights that exemplify the Program's noteworthy research results, timely support for critical national needs, and external recognition of Laboratory personnel.

- ***Superheavy Elements 113 and 115 (04-ERD-085).*** A collaboration between LLNL and the Heavy Elements Group of the Joint Institute for Nuclear Research in Dubna, Russia, published, in *Physical Review C*, the discovery of elements 113 and 115. These elements are located on the "western shores" of the "island of stability"—a region where superheavy elements are predicted to be stable and comparatively long-lived, due to a double-shell closure in Z, N (proton number and neutron number). News of the discovery appeared widely in the popular press, including the front page of the *New York Times* (Sunday, February 1, 2004). In this collaboration, LLNL scientists provided the Dubna group with critical hardware, special nuclear materials, chemical and nuclear analytical techniques and expertise, and parallel data analysis. The experiments were performed at the Dubna cyclotron with a gas-filled recoil spectrometer. This was a challenging search in that the discoveries were based on only one or a few counts obtained during each month of bombardment. The discovery of these two elements and the other heaviest elements known to man (Z = 113–116), along with all of their isotopes (twelve in total), would not have been achieved without LDRD funding.
- ***Long-Range, Passive Detection of Fissile Materials (03-ERD-046).*** One of the primary needs for countering the threat of nuclear terrorism is the long-range detection of nuclear material. Detection at distances greater than 100 meters was thought impractical due to

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fluctuating levels of natural background radiation. Imaging gamma-ray detectors represent a potentially promising solution. This project prototyped a coded-aperture imager, adapting concepts from satellite-borne x-ray astronomy. In its simplest one-dimensional form, a mask consisting of a series of parallel shadow-bars, of random spacing and width, is placed a distance in front of a large-area gamma detector. The location of a point source is uniquely determined by the "bar code" shadow pattern cast on the detector. A more complicated source distribution is determined by computer inversion of the pattern. This LDRD project team achieved a major success: The detector was mounted in a truck and driven around the Laboratory site, with data accumulated and displayed in real time on a laptop computer. A hidden 1-millicurie cesium-137 radiation source was easily detected and pinpointed at a distance of 85 meters while the truck was moving 25 miles an hour. In addition to its clear applicability in homeland security, this project also provided important results for use in basic astrophysical science in the form of enhanced techniques, algorithms, and technical skills.

- ***Phonon-Dispersion Curves in Plutonium (03-ERD-017).*** Neutron scattering is the tool of choice for measuring phonon spectra in crystalline materials. However, this technique cannot be used on plutonium due to its high cross-section for neutron absorption. Consequently, measuring the phonon spectra of plutonium was long thought to be impossible. With LDRD funding, however, a Livermore group prepared thin, polycrystalline targets and measured both longitudinal and transverse phonon spectra of plutonium using high-resolution inelastic x-ray scattering at the European Synchrotron Radiation Facility in Grenoble, France. The data showed several unusual features related to phase transitions in plutonium and to strong coupling between the lattice structure and the 5f valence instabilities. These measurements promise to reveal much about the chemistry and thermal properties of plutonium and its alloys—data that are critical to the Lab's stockpile stewardship work.
- ***Uranium Nitride Fuel (03-ERD-019).*** An important aspect of nonproliferation is the development of a proliferation-resistant reactor. One such concept is the secure, transportable, autonomous reactor (STAR), which requires a fuel that would enable a compact, long-life reactor core. Such reactors would be fabricated and sealed by a trusted nation and shipped to other nations for up to 30 years of use. At the end of its life, the sealed reactor would be returned to the trusted nation for disposition. One LDRD project, "Mono-Nitride Fuel Development for STAR and Space Applications," is developing, fabricating, and irradiating modified nitride-based uranium fuels (containing zirconium nitride or hafnium nitride) for use in STARs and in space reactors. Computational methods are being used to analyze fuel performance, and the fuels are being irradiated at the McClellan Nuclear Radiation Center to verify the computational results and demonstrate fuel feasibility. In response to favorable results indicating the reactor stability and long life that would be possible with this uranium nitride fuel, NASA and the U.S. Navy recently selected this fuel technology, along with the SP100 reactor, as the starting point for the Jupiter Icy Moons Orbiter (JIMO) program. The key issue in JIMO is fuel longevity, as the program entails a voyage lasting 20 years. Because of this LDRD project, fuel is currently being produced for JIMO, and LLNL will assume a leading role in R&D for the program.
- ***A Quantum-Limited Microwave Amplifier (02-ERD-071).*** Experimental progress in many areas of basic and applied science requires ever quieter and more sensitive radio-frequency

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amplifiers. One such LDRD experiment, "Development of a Quantum-Limited Microwave Amplifier using a dc Superconducting Quantum Interference Device (SQUID)," is attempting to find dark matter in the Universe by converting the axion—a hypothetical elementary particle—to a microwave photon. Conventional microwave amplifiers have an equivalent noise temperature in the gigahertz range of around 1 kelvin. SQUIDs have long been used as extremely sensitive magnetometers, and recent progress in their design has now opened up the possibility of their use as ultrasensitive amplifiers in the gigahertz range and above. Through a combination of modeling, prototyping, and characterization, this LDRD project has been reducing the noise temperature of SQUID amplifiers towards the theoretical minimum noise temperature—the quantum limit—which at this frequency is approximately 35 millikelvins. Presently, the best devices built and measured by this project have achieved 50 miilikelvins, only about 50% higher than the quantum limit. This is thus a world record by a large factor and has tremendous implications not only for the axion dark-matter experiment, but also for research in quantum computing and quantum communication. This project's achievements have already led to the DOE Office of High Energy Physics supporting a $2M upgrade of the axion experiment, and collaborations have begun with three leading groups in the world of quantum computing. Achieving the quantum limit, expected in FY05, will constitute a major and highly visible scientific advance.

- ***ICE: The Image Content Engine (03-SI-003).*** Advancements in imaging-sensor technologies, particularly for remotely sensed images, are resulting in volumes of data that overwhelm human analysts. The goal of this project is to develop a framework—the Image Content Engine (ICE)—that will allow human analysts to interpret massive volumes of imagery in a timely fashion. The ICE approach is to focus the attention of humans on relatively few small areas with specified attributes. This will be achieved by capturing content extracted from images as nodes in semantic graphs whose links define relationships between pieces of image content and information obtained from non-image sources. The work supports LLNL's mission in national security by developing the technology to allow content extracted from images to be seamlessly integrated with information from other sources, enabling analysts to submit sophisticated queries relevant to defense, intelligence, counterterrorism, deployment strategies, determine the functions of enigma facilities, and detect the production of weapons of mass destruction.

Patents

Projects sponsored by LDRD consistently account for a large percentage of the patents issued for LLNL research, especially considering that the LDRD Program funding represents 6% of the total LLNL operating budget (with the exception of FY2000, when the Program received only 4% of the operating budget). Table 1 shows the number of patents resulting from LDRD-funded research since 1997.

	1997	1998	1999	2000	2001	2002	2003	2004
All LLNL patents	64	78	84	93	89	97	71	95
LDRD patents	29	39	45	35	42	27	29	55

Table 1. Patents resulting from LDRD-funded research as a percentage of all LLNL patents from 1997 to 2004.

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Awards

- ***American Physical Society Award for Excellence in Plasma Physics.*** LLNL physicist Siegfried Glenzer received the American Physical Society's 2003 Award for Excellence in Plasma Physics Research. Much of the work recognized by the APS was funded by LDRD (LDRD project 01-ERD-107).
- ***American Physical Society Fellows.*** Of the six Livermore scientists who were elected as Fellows of the American Physical Society in 2004, two were elected on the basis of work that was largely supported by LDRD.
  - Giulia Galli—For work in ab initio molecular dynamics and amorphous and liquid semiconductors and quantum systems (e.g., LDRD project 03-SI-001)
  - Erich Ormand—For work in nuclear structure physics and nuclear physics as applied to stockpile stewardship (e.g., LDRD project 04-ERD-058)
- ***Chemistry Abstract Service's "Most Intriguing Documents."*** In the third quarter of 2003, Chemistry Abstract Service (CAS), a division of the American Chemical Society, chose six documents, out of over 200,000 reviewed by CAS scientists, as the most intriguing. Of these six articles, two were related to work funded by LDRD.
  - "Autonomous Detection of Aerosolized *Bacillus anthracis* and *Yersinia pestis*" (***Analytical Chemistry***, Oct. 15, 2003) (Fred Milanovich, LDRD project 99-SI-016)
  - "Single-Molecule Measurement of Protein Folding Kinetics using a Microfabricated Mixer" (***Science***, Aug. 29, 2003) (Olgica Bakajin, LDRD project 02-ERD-040)
- ***Fellow of the Acoustical Society of America.*** Livermore scientist David Chambers was named a Fellow by the Acoustical Society of America in recognition for work supplied by LDRD funding (LDRD projects 99-LW-045 and 00-LW-049).
- ***Presidential Early Career Awards.*** LLNL scientist Brian Wirth received a 2003 Presidential Early Career Award for Scientists and Engineers. LDRD has funded important work by Brian (e.g., LDRD project 01-ERD-042).
- ***R&D 100 Awards.*** In 2004, Laboratory technologies won five "R&D 100" awards from *R&D Magazine*. Of these, LDRD support directly contributed to three awards:
  - APDS: Autonomous Pathogen Detection System (Fred Milanovich, LDRD project 99-SI-016)
  - Gene Silencing with SiHybrids (Allen Christian, LDRD project 03-ERD-038)
  - Inductrack Magnetic Levitation System (Richard Post, LDRD project 95-LW-065; others)
- ***UC Davis Marr Award.*** In 2004, Laboratory postdoctoral researcher Richard Snavely received the UC Davis 2004 Marr Award for best Ph.D. dissertation at the university. This work in inertial confinement fusion was supported largely by LDRD (e.g., LDRD project 02-SI-004).

LDRD researchers were also recognized by two major LLNL award programs. Although these are internal awards and fellowships, they are based on national and international recognition for these scientists and engineers.

- ***LLNL 2003 Science & Technology Awards.*** One of the two FY2003 Science and Technology Awards went to a team, led by LLNL scientist Joe Wong, whose research was conducted with substantial LDRD support: Complete Phonon Dispersions in delta-Phase Pu–Ga Alloys by Inelastic X-Ray Scattering (LDRD project 03-ERD-017).
- ***LLNL 2003 Teller Fellow.*** Kenneth G. Caldeira was selected as a 2003 Teller Fellow for his work on climate change and carbon dioxide sequestration, much of which was performed with LDRD funding (e.g., LDRD project 01-ERD-091).

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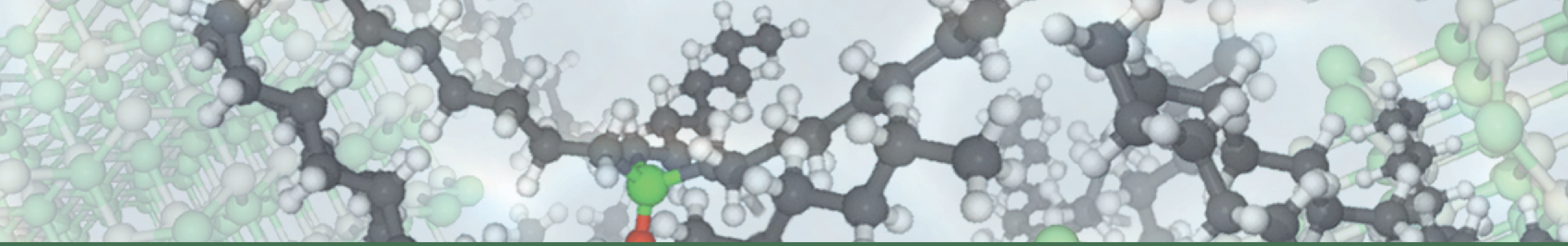
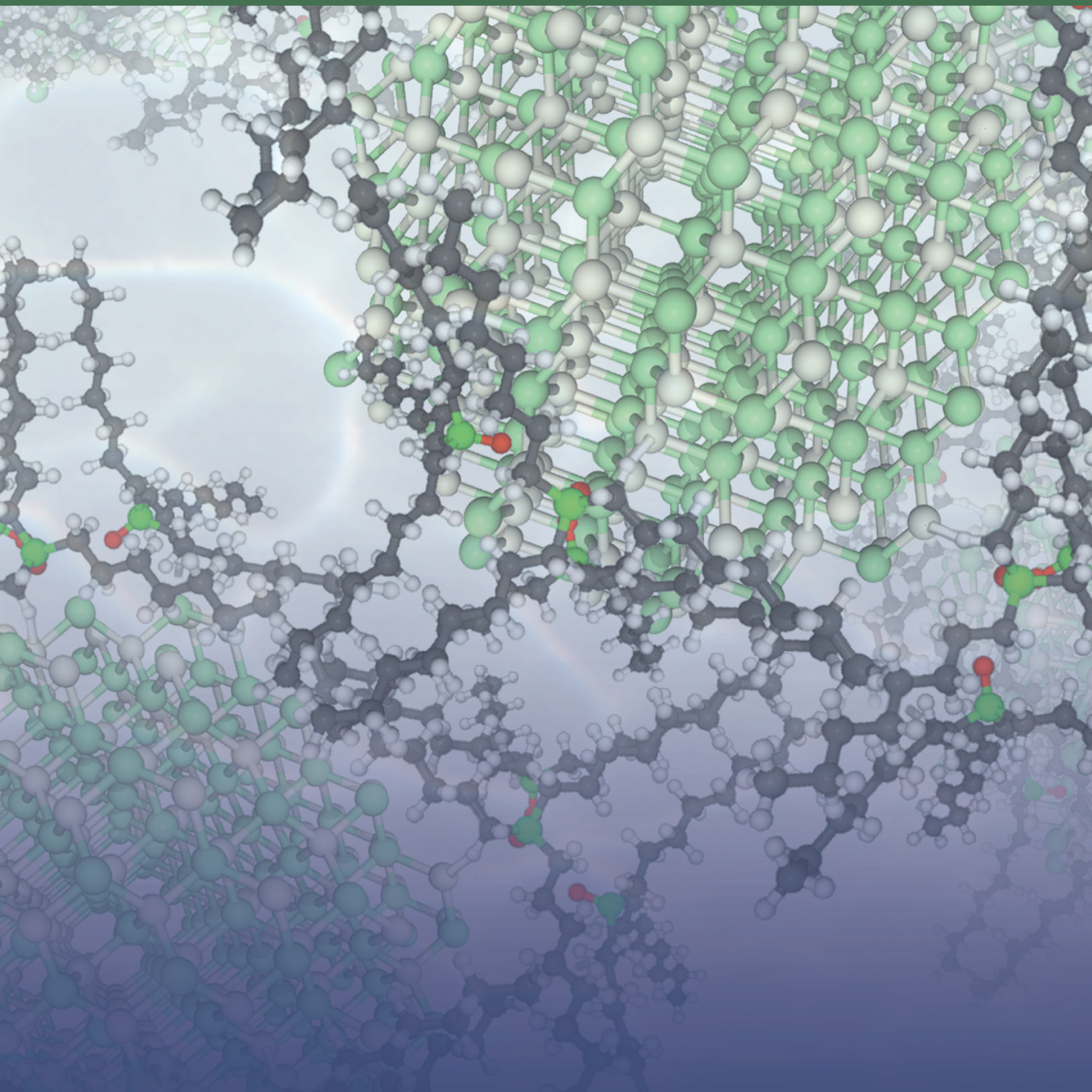


Advanced Sensors and Instrumentation



Advanced Sensors and Instrumentation

Stroke Sensor Development using Microdot Array Sensors

Jerry C. Carter

01-ERD-101

Abstract

In the U.S., over 700,000 people a year suffer a stroke, which is a major cause of mortality and disability. Currently, the only approved therapy for treatment of acute ischemic stroke is intravenous thrombolytic drugs, which must be administered within 3 h of the onset of symptoms; drugs administered after this may result in a fatal intracranial hemorrhage. Consequently, only 2 to 3% of people with acute stroke are treated with these drugs. We propose to develop and demonstrate a minimally invasive optical-fiber-based sensor for rapid *in vivo* measurements of multiple stroke biomarkers, e.g., blood gas and enzymes. Such a tool could extend the use of thrombolytic drug treatment to acute stroke victims outside the current limitation of 3 h.

Successful completion of this project will result in a unique multianalyte biosensor platform. Specifically, we expect to (1) demonstrate world-class science, the first reproducible fiber-based sensors and the first fiber-based enzyme biosensor; (2) develop a portfolio of intellectual property; (3) hire new postdocs and provide research topics for graduate students; (4) develop a science and technology base in multianalyte fiber-based sensors; (5) develop new biosensors for disease diagnostics; and (6) assess this technology for potential applications to stockpile surveillance.

Mission Relevance

In support of LLNL's bioscience mission, the microdot-array sensor platform will complement existing stroke-related efforts, in which mechanical devices are being designed for removing endovascular obstructions in the brain. In addition, the multianalyte fiber-based sensors developed by this project are potentially useful for stockpile surveillance applications.

FY04 Accomplishments and Results

In FY2004, we improved our surface-functionalization technique, which allowed us to reproducibly print microdot arrays. We demonstrated the multianalyte printing capabilities of our custom, in-house fabrication system and the proof-of-concept for detecting enzymes on fibers. However, consistently reproducing these results turned out to be more difficult than we anticipated and prevented us from producing a multianalyte sensor for pH, 0_2, and matrix metalloproteinase. A UC graduate student will continue to pursue this issue under separate funding. A prototype sensor was developed, which led to follow-on funding for bioagent detection in groundwater; another follow-on project has been proposed for Department of Defense funding. We filed a patent application describing the fabrication technology developed during this project and expect to produce two additional publication submissions by year's end.

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Publications

Carter, J. C. et al. (in press). "Fabricating optical fiber imaging biosensors using inkjet printing technology." *Biosensors & Bioelectronics*. UCRL-JC-150484.

Carter, J. C. et al. (2004). *Fabricating optical fiber imaging biosensors using inkjet printing technology*. Presented at the 2004 LLNL Sensor Workshop, Livermore, CA, April 6-7, 2004. UCRL-ABS-203723.

Single-Cell Proteomics with Ultrahigh-Sensitivity Mass Spectrometry

Matthias Frank 02-ERD-002

Abstract

This project will measure, for the first time, the expression of biomolecules in single cells by ultrahigh-sensitivity mass spectrometry to probe individual cells on a molecular level. The sensitivity and selectivity of time-of-flight mass spectrometry (TOF-MS) will be improved by a factor of 1000 using a combination of single-particle aerosol TOF-MS and advanced laser desorption and ionization techniques. These techniques will be used to study molecular changes in individual cells and detect and identify single biological aerosol particles in real time. Characteristic mass signatures will be obtained from biological-warfare simulants, and the sensitivity and specificity of detection technique will be optimized.

The capability developed in this project will enable studies that probe individual cells at the molecular level to (1) understand molecular changes that occur during cell development; (2) investigate subtle differences between individual normal and cancerous cells; and (3) detect and identify single, biological aerosol particles in real time. This technology is expected to have significant impact on the field of autonomous biodetection and rapid disease diagnostics. Single-cell mass spectrometry could detect and help understand changes in protein expression or cell composition occurring in the early stages of a disease. Accurate measurement of protein expression in cancer cells could also be used to identify metastatic cancer and determine the progression of the disease.

Mission Relevance

The ability to study biochemical processes at the cellular level will lead to improved bioanalytical tools to support LLNL's missions in countering the threat of bioterrorism and in bioscience and technology to improve human health.

FY04 Accomplishments and Results

In FY04, we continued to measure the mass spectral signatures from several bacterial species, viruses, and toxin simulants and study their dependence on desorption and ionization laser parameters and on growth conditions. Marker peaks critical for the identification of bacterial spores were investigated using carefully designed isotope-labeling experiments. We also performed our first measurements on a close (but harmless) relative of *Mycobacterium tuberculosis*. We began exploring the use of matrix-assisted laser desorption and ionization in combination with an improved aerosol TOF mass spectrometer to facilitate the

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measurement of protein expressions in single cells for studies of cancer and other diseases. This research resulted in three papers published in peer-reviewed journals.

Publications

Steele, P. T. et al., “Laser power dependence of mass spectral signatures from individual bacterial spores in bioaerosol mass spectrometry.” *Analytical Chemistry* **75**, 5480. UCRL-JC-153095.

Fergenson, D. P. et al., “Reagentless detection and classification of individual bioaerosol particles in seconds.” *Analytical Chemistry* **76**, 373. UCRL-JC-151115.

Ultrasonic Nondestructive Evaluation of Multilayered Structures

Michael J. Quarry 02-ERD-010

Abstract

This project developed ultrasonic nondestructive evaluation techniques based on guided and bulk waves in multilayered structures. First, a guided wave technique was developed to preferentially excite dominant modes with energy in the layer of interest via an ultrasonic array. These modes were excited by designing arrays with proper element spacing and frequency. The guided-wave technique enables the inspection of inaccessible areas without disassembly. Second, a bulk wave technique used Fermat’s principle of least time and wave-based properties to reconstruct array data. The algorithms overcame the difficulties of multiple reflections and refractions at interfaces to accurately detect and localize flaws through multiple layers.

The project produced a prototype of an array system for inspecting inaccessible areas of multilayered structures using guided waves. Applicability of the technique to various materials and curvatures was demonstrated. Potential inspection applications include detecting inaccessible layers of weapons systems, tubing, and fusion-class laser targets. Also, bulk-wave-reconstruction algorithms were developed to utilize array technology for imaging multilayered structures. The algorithms enable imaging of explosive materials for defense programs and near-earth tunnels for nonproliferation applications.

Mission Relevance

The guided- and bulk-wave inspection techniques developed in this work will advance stockpile stewardship, particularly weapons inspection without disassembly. Experiments also showed the scalability of guided waves to the smaller curvatures in future fusion-class laser targets. In addition, the planar, multilayer-diffraction tomography reconstruction algorithms may image density distributions in explosives and shallow, near-earth environments to detect buried hazardous waste and covert trans-border tunnels, in support of nonproliferation and counterterrorism.

FY04 Accomplishments and Results

We evaluated the quantitative detection abilities of both techniques by constructing multilayer plates of Al, epoxy, and steel with notches of various depths in the bottom layer.

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The guided-wave technique showed sensitivity to the smallest (10%) flaw. A guided wave was used to detect a notch machined by electrical discharge in a curved multilayer structure. Additional experiments showed applicability to smaller curvatures by detecting microcracking in a 6-mm-diam stainless-steel tube. For bulk waves, an Al-Cu planar sample was constructed with side-drilled holes of various diameters, and data were collected with a 5-MHz ultrasonic array. The bent-ray algorithm imaged a 0.75 mm flaw, whereas the wave-based algorithm showed resolution of 1.5 mm.

Publications

Quarry, M. J. "Guided wave inspection of multilayered structures." *Review of Prog. in Quant. Nondestructive Eval.* **23A** 246–253. UCRL-JC-152109.

Development of Ultrasensitive High-Speed Biological Assays Based on Two-Dimensional Flow-Cell Detection of Single Molecules

Christopher W. Hollars 02-ERD-018

Abstract

This project is focused on developing a practical device for rapid assay of biological species such as proteins and DNA at ultralow levels. Such a device will have applications in the detection of biological warfare (BW) agents and would provide novel assays for medical applications, drug discovery, biotechnology, and basic biology research. Our objective is to detect and identify single, fluorescently labeled molecules using total internal reflection and an electron multiplied-charge-coupled device camera. The analyte solution will be analyzed in a microfluidics device. Our goal is throughput 1,000 to 10,000 times greater than that of typical microstream devices, which will allow biological assays to be performed in approximately 10 min.

The outcome of this research will be an assay capability for high-speed analysis of biological molecules at ultralow concentrations. This will represent a significant breakthrough in the detection sensitivity (100 to 1000 times better) and will enable analysis of small biological systems, ranging from large molecules, proteins, and DNA to toxins, viruses, and bacteria.

Mission Relevance

This device will have applications in the detection of potential BW agents in support of the Laboratory's missions of nonproliferation and counterterrorism, and will provide novel assays for medicine, drug discovery, biotechnology, and basic biology research in support of LLNL's mission to improve human health.

FY04 Accomplishments and Results

In FY04, we focused on overcoming several technical challenges to enable the application of this novel analytical technique. These efforts resulted in a two-color analysis of dual-labeled target molecules in a flowing solution. After we carefully determined the optimal analysis conditions, these measurements demonstrated 50-picomolar detection limits within a 2-min analysis. This result supports the claims that single-molecule detection in flowing solutions can

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be applied to the ultrasensitive analysis of biological molecules. We have prepared several publications describing the results of this project.

Development of a Fast Microfluidic Mixer for Studies of Protein Folding Kinetics

Olgica Bakajin 02-ERD-040

Abstract

The goal of this project is to develop a robust, microfluidic laminar-flow mixer with a considerably lower consumption rate, shorter dead time, and better time resolution than currently available mixers. The mixer will be used to study the kinetics of fast protein-folding reactions using ultraviolet- (UV-) light-induced fluorescence and fluorescence resonant energy transfer (FRET). The project involves using computational modeling to develop the mixer design, developing fabrication processes and techniques for assembly and characterization of the microfluidic mixer, and using the mixer to measure protein-folding kinetics. The mixers will be created with photolithographic patterning and wet and reactive ion etching of fused silica wafers.

The result of this project will be a robust microfluidic mixer that will offer a 500-fold improvement in mixing times and sample consumption compared to currently available mixers and will be compatible with most spectroscopic methods. This new capability will be useful for studies of DNA interactions with proteins, for single-molecule detection in low concentrations of toxins, and for studies of aggregation of prions or of the amyloid-beta peptide implicated in Alzheimer's disease.

Mission Relevance

By developing new microfluidic devices with a wide range of potential applications, from detecting biological warfare agents to health care, this project supports the LLNL mission of counterterrorism and bioscience to improve human health.

FY04 Accomplishments and Results

We built the fastest reported microfluidic mixer with mixing times as short as 8 μs and sample consumption in femtomoles. This device allows us to access protein folding kinetics under conditions far from equilibrium and at the previously inaccessible timescales at which important structural events occur. We designed, optimized, and characterized the instrument, and, in collaboration with UCLA, demonstrated the feasibility of protein-folding measurements. Our mixer is fast, reliable, repeatable, easy to batch-fabricate, and consumes very small amounts of precious sample, all of which will enable studies of many proteins in the future. We also fabricated mixers in which UV-induced fluorescence can be observed and which will enable future studies.

Publications

Hertzog, D. E. et al. (2004). "Femtomole mixer for microsecond kinetic studies of protein folding." *Analytical Chem* **76**, 7169. UCRL-JRNL-206676.

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Hertzog, D. E., J. Santiago, and O. Bakajin. (2004). Microfluidic Mixers for UV Studies of Unlabeled Proteins. Micro Total Analysis Systems 2004. UCRL-POST-207476.

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

S. Darin Kinion 02-ERD-071

Abstract

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

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

Mission Relevance

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

FY04 Accomplishments and Results

Our major accomplishment in FY04 was measuring a noise temperature within 50% of the standard quantum limit at 700 MHz; this result was presented at the American Physical Society March meeting this year. A second accomplishment was improving the noise temperature measurement using a shot-noise thermometer; this allows us to measure noise temperature as a function of frequency, which will be very important to finding the minimum. Finally, significant progress was made both in establishing collaborations with groups using SETs as well making the first measurement of charge sensitivity using a SQUID readout. Preliminary results already showed about a factor-of-3 improvement over a traditional heterojunction field effect transistor readout.

Proposed Work for FY05

Work will focus on migrating the SQUID fabrication technology to LLNL. A thin-film deposition at LLNL will be used to perform the SQUID fabrication at the wafer-level. The result should be hundreds of working amplifiers ready to go into experiments around the world.

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Collaboration will continue with SET groups to demonstrate the quantum limit for charge amplification, resulting in multiple high-visibility publications.

Advancing the Technology of Tabletop, Mesoscale Nondestructive Characterization

Harry E. Martz

03-SI-004

Abstract

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

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

Mission Relevance

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

FY04 Accomplishments and Results

We performed several types of simulation studies to better understand x-ray imaging of mesoscale objects. We chose the solid deuterium–tritium fuel layer in an inertial-confinement fusion capsule with a Be ablator as one example for our research. We simulated x-ray projection systems with a coherent parallel beam and a point source, and a large-size source with a Wölter x-ray imaging optic. These studies showed that imaging was possible with either approach. We developed a Wölter optic simulation capability for 3-D objects and used it to study laminographic and tomographic approaches to object recovery for the Wölter microscope.

Proposed Work for FY05

We expect to (1) determine definitively whether multislice modeling is needed for the objects of interest; (2) finalize the design of a multiple-materials step wedge; (3) fabricate the multiple-materials step wedge; (4) complete validation of point-projection HADES models, including phase contrast for various phantoms; (5) validate exit-wave recovery for uniform materials; and (6) characterize artifacts that could be problematic when using existing tomography algorithms in this kind of imaging.

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Publications

Aufderheide, M. B., A. Barty, and H. E. Martz. (2004). *Simulation of phase effects in imaging for mesoscale NDE*. 31st Ann. Rev. of Progress in Quantitative Nondestructive Evaluation, Golden, CO, July 25–30, 2004. UCRL-CONF-206263.

Jackson, J. A. (2004). *Wölter x-ray microscope computed tomography ray-trace model with preliminary simulation results*. UCRL-TR-206864.

Kozioziemski, B. J., et al. *Quantitative characterization of inertial confinement fusion capsules using phase contract enhanced x-ray imaging*. UCRL-JRNL-205025.

Martz, H. E. et al. (2004). *X-ray nondestructive characterization of mesoscale objects*. UCRL-ABS-206253.

Schneberk, D. J., H. E., Martz, and W. D. Brown. (2004). *Dimensional measurements of three tubes by computed tomography*. UCRL-TR-207195.

Schneberk, D. J., J. A. Jackson, and H. E. Martz. (2004). *Possible laminographic and tomosynthesis applications for Wölter microscope scan geometries*. UCRL-TR-207196.

Ultrafast Radiation Detection by Modulation of an Optical Probe Beam

Mark E. Lowry 03-ERD-007

Abstract

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

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

Mission Relevance

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

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FY04 Accomplishments and Results

In FY04, we developed a radiation optic (radoptic) effect model by comparing optical excitation theory to FY03 x-ray excitation experimental results, allowing us to quantitatively predict x-ray sensitivity. We developed the first single-pixel cavity-based radiation sensor (RadSensor) device. To guide device optimization, we developed a cavity device model that incorporates the basic cavity physics, the appropriate materials physics, and the experimentally validated radoptic effect model. Also developed and implemented were the mirror deposition technologies for cavity formation and the single-pixel x-ray instrumentation system. We fielded two distinct versions of these single-pixel cavities on the LLNL Ultra-Short Pulse Laser facility and successfully demonstrated an improvement in sensitivity that scales with the cavity finesse.

Proposed Work for FY05

We will focus on evaluating the temporal response of the detector to x-ray illumination. There are two major tasks: (1) procure fast-response materials and (2) develop and implement the necessary measurement techniques. On the materials development side, we will procure low-temperature-grown GaAs and pursue either the neutron irradiation or proton bombardment of single-crystal GaAs. On the fast measurement side, subpicosecond response can be obtained using cross-correlation techniques. However, this will be challenging in terms of x-rays and optics. Our backup plan will be to use streak camera recording. Our goal will be to demonstrate near picosecond or subpicosecond response.

Publications

Lowry, M. E. et al. (in press). "X-ray detection by direct modulation of an optical probe beam—RadSensor: Progress on development for imaging applications." *Rev. Sci. Instruments*. UCRL-CONF-203607.

DNA Detection through Designed Apertures

Sonia E. Letant 03-ERD-013

Abstract

Our objective is to demonstrate a selective Coulter counter for bio-organisms by using a rigid silicon aperture which will mimic an ion channel. The project involves three main steps: (1) design and fabrication of single apertures with a tunable diameter from the nanometer to the micron range on silicon platforms; (2) functionalization of the aperture with chemical probes to provide selectivity; and finally, (3) measurement of bio-organism transport through the functionalized apertures by the current blockade technique at the single bio-organism detection level. The challenges are to design single apertures in silicon with tunable diameter, length, and shape and to control their chemical functionalization. This will be achieved by combining nanomachining and self-assembly.

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Although efforts have focused on building a single nanopore on various rigid substrates, no one has addressed either of the two key issues: finding a fast, reliable, and inexpensive technique to prepare single nanoapertures, and controlling the chemical functionality in these confined apertures. Our new approach, at the cutting edge of nanotechnology and self-assembly, should accomplish both and transform synthetic nanopores into extremely selective sensors able to detect and identify single organisms in real time without requiring polymerase chain reaction amplification. Such a result would open the door to a new class of biosensors for military and civilian applications.

Mission Relevance

Our project supports LLNL's national security mission by developing a new class of biosensors able to identify bio-organisms with single-organism sensitivity. This technology could enable fast and sensitive sensors to improve early response to terrorist or accidental biological contamination of air or water. This technique has potential applications for homeland defense and several DOE programs in counterterrorism, preventing the proliferation and use of biological weapons, and attribution.

FY04 Accomplishments and Results

In FY04 we accomplished the following: prepared apertures with diameters down to 30 nm by electrochemistry; drilled apertures with controlled diameters ranging from microns to tens of nanometers with a focused ion beam; measured conductivity through unfunctionalized apertures to demonstrate that the channels were open; calibrated the oxide growth rate on flat surfaces by ellipsometry and transfered it to the aperture walls; synthesized partially ordered top coats on flat surfaces and studied them by ellipsometry, Fourier transform infrared, and x-ray photoemission spectroscopy; functionalized a single aperture from bare silicon, to silicon oxide, to self-assembled monolayer and finally to DNA; and demonstrated control of each functionalization step at the nanometer scale by measuring the I-V curve through the device.

Proposed Work for FY05

Our goals for FY05 are to: (1) show that we can control aperture fabrication, oxidation and surface functionalization in order to achieve precise aperture diameters; (2) test DNA functionalized apertures with DNA functionalized beads to demonstrate the first selective Coulter counter working in the virus-size range; and (3) test the apertures with T7 viruses engineered by the phage display technique.

Publications

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

Nilsson, J., L. Hope-Weeks, and S. E. Letant. (2004). *Single aperture functionalization for bio-organism sensing applications*. Presented at the MRS Fall Meeting, Boston, MA, Nov. 29–Dec. 3, 2004. UCRL-ABS-207152.

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Microfluidic System for Solution Array-Based Bioassays

George M. Dougherty 03-ERD-024

Abstract

We are developing a new method for performing multi-target biodetection assays using nanobarcode (NBC) particles—short, metallic nanowires bearing patterns of light and dark stripes, analogous to the stripes in a supermarket barcode. This approach offers advantages in speed, cost, and flexibility over existing systems. The assay results can be read using standard light microscopy, avoiding the need for complicated flow cytometry techniques. Furthermore, the properties of the particles are such that they can be manipulated, transported, and trapped in microfluidic systems using magnetic and electric fields. Methods based on NBCs promise advancements in biodetection for homeland and national security applications, as well as new capabilities for medical diagnostics and screening.

The goal of the project is to demonstrate a prototype bioassay system based on NBC particles. This system will be capable of assaying several biowarfare (BW) agent simulants simultaneously. We also expect to achieve a number of important scientific goals, advancing the state of the art in particle-based biochemical assays and in the manipulation and control of metallic nanoparticles within aqueous solutions. We are also investigating methods for using NBCs as covert tags for material identification and forensics. Finally, the basic-science advancements from this project will contribute to nanotechnology and nanoparticle science in general.

Mission Relevance

This project supports LLNL's national-security mission areas of homeland security and counterterrorism by developing technology to counter weapons of mass destruction. The resulting instruments will be capable of detecting BW agents in the field and therefore have applications in surveillance, detection, and treaty verification. The project also supports LLNL's mission in science and technology to improve human health.

FY04 Accomplishments and Results

In FY04, we (1) completed the development of a multiplex biodetection panel based on NBC particles and obtained test results showing performance equal to state-of-the-art immunoassays; (2) carried out extensive studies of metallic nanoparticle transport in aqueous solutions, including studies of electromagnetic manipulation effects; (3) developed a microfluidic method for creating self-assembled, two-dimensional arrays of particles for readout and analysis; and (4) completed the development of our benchtop optical readout system. The results we obtained this year led to three journal articles submitted, a conference presentation, and a record of invention (ROI).

Proposed Work for FY05

In FY05, we plan to build a prototype system that will perform in situ biodetection assays, something not possible with any other fielded real-time, solution-array pathogen-detection system in existence. This system will also demonstrate our ability to isolate NBCs that are used as covert material tags for purposes of forensics and attribution. Ongoing work in

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biochemical assay development, optical readout techniques, and particle transport will be conducted to optimize system performance and meet real-world operating challenges. We anticipate additional publications and intellectual property.

Publications

Dougherty, G. M., et al. (2004). *Multiplex biodetection using solution arrays based on encoded nanowire particles*. Presented at the Matls. Research Soc. Spring Mtg., San Francisco, CA, Apr. 12–16 2004. UCRL-PRES-203512.

Photochromic Radiation Dosimetry

Nerine J. Cherepy 03-ERD-040

Abstract

This project demonstrated fluorescence-based radiation dosimetry systems and established several appropriate fields of use. We are developing systems in which radiation converts photochromic molecules into fluorescers, which are then laser interrogated. The first approach is a detector based on remote laser interrogation of gradient index millispheres with a scintillator–photochrome coating sensitive to alphas and betas, which permanently accumulates fluorescence signal in proportion to dose. Another approach involves a reversible photochromic radiation sensor that also utilizes fluorescence readout. It may be implemented as a small photochrome-embedded plastic transducer coupled to a large plastic scintillator panel, or as a liquid dispersed in a liquid scintillation cocktail.

This project has explored new concepts in radiation detection. We determined performance potential for several of these concepts, which can now be developed into projects for the intelligence and defense community. The gradient-index sphere dosimeter offers a truly remote radiation-detection capability for the first time. The Defense Advanced Research Projects Agency has opted to continue the research and development of this remote dosimetry system. Its successful implementation will provide an enabling technology for important problems in homeland defense and nuclear nonproliferation. In addition, photochromic materials and expertise developed during this project will be used to demonstrate charged-particle detection and tracking via photochromic fluorescer detection.

Mission Relevance

This project supports LLNL's national-security mission by furthering the development of advanced technical surveillance capabilities to provide early warning and assessment of the location of nuclear materials. The radiation sensor systems we seek to develop have unique features to enable detection of proliferation activities and remote sites. It will contribute directly to national and international security, counterterrorism, nonproliferation, homeland security, and military intelligence.

FY04 Accomplishments and Results

In FY04, we continued materials development and evaluation and determined

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performance characteristics, such as sensitivity, linearity with exposure, and stability of a stored signal over time. We synthesized and characterized the photophysics of many different photochromic materials that permanently convert to a fluoresent form upon exposure to scintillation; synthesized, purified, and characterized a reversible photochrome (naphthothioindigo; one publication will result); and prepared crystalline BaF_2 nanoparticles using a micro-emulsion synthetic technique for use in low-cost BaF_2 scintillating coatings.

Long-Range, Passive Detection of Fissile Material

Lorenzo Fabris

03-ERD-048

Abstract

One of the key needs for counteracting the nuclear terrorist threat is the long-range detection of nuclear material. While it is theoretically possible to detect gamma-ray emissions from these materials at distances of greater than 100 m, detection at 100 m has long been thought impractical due to fluctuating levels of natural background radiation. This problem can be overcome through the use of imaging gamma-ray detectors. We propose to design and build a prototype large-size (6400-cm^2) gamma-ray imager and demonstrate its capability to detect nuclear materials from a distance of ~100 m in a search scenario.

We will demonstrate the possibility of detecting nuclear material to distances of 100 m, while driving past it at 25 mph. In addition to this, a complete set of electronics for medium to high channel counts and high photon rates will be made available to the whole community. This set of electronics will be easily adaptable to read out a broad class of detectors, process their signals, and move the results into personal computers through a high-speed interface. A successful demonstration of this technology will lead to a full-size, fieldable unit.

Mission Relevance

The successful demonstration of this technology will respond to a major national security challenge, i.e., remote detection of radioactive materials that could be used in a nuclear or radiological weapon, and will support NNSA and Laboratory missions in nonproliferation and homeland security.

FY04 Accomplishments and Results

During FY04 we successfully completed the detector system, the electronics readout, and the image-reconstruction software. The system was operated for several months and used to perform several measurements in standing source–standing detector and moving source–standing detector scenarios. This work demonstrated the ability of our system to detect a stationary 1-mCi source at distances exceeding 80 m when the instrument is moving at speeds of up to 10 mph. We also achieved the capability of generating real-time images of a source moving at variable velocity in front of the detector. This system could be deployed for case studies useful to the design of a next-generation, commercial version of this prototype. Papers were published at two international conferences.

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Publications

Ziock, K. P. et al. (2004). "Large-area imaging detector for long-range, passive detection of fissile material." *IEEE Transactions on Nuclear Science* **51**, 2238. UCRL-JRNL-206121.

Ziock, K. P. et al. (2004). *Source-search sensitivity of a large-area, coded-aperture, gamma-ray imager*. IEEE Nuclear Science Symposium, October 16–22, 2004, Rome, Italy. UCRL-ABS-204377.

Carbon-Nanotube Permeable Membranes

David J. Eaglesham 03-ERD-050

Abstract

This project will measure the motion of molecules down the axis of a carbon nanotube (CNT). This will be accomplished by embedding ordered nanotubes in a thin film to make a semipermeable membrane with ideal pores. We will observe diffusion both by permeation and direct observation of single fluorescent molecules.

The motion of molecules through pores of molecular dimensions is a crucial problem in nanoscience. Diffusion down a nanotube has been frequently simulated but never measured, so these experiments will have a significant impact on the field and enhance our understanding of molecularly confined motion—critical for integrated sensors and separation systems. Finally, these semipermeable membranes are chemically selective and of molecular size and could have broad application.

This project will build a deeper understanding of the physics of permeation separation through fabrication of a series of media with precisely controlled pore geometry and surface chemistry. If successful, this work could have applications in various basic- and applied-science challenges. This could include applications such as micro gas chromatography (GC), microcapillary protein-separation, and micro-isotope purification.

Mission Relevance

A semipermeable membrane with molecular size selectivity would further the Laboratory's national-security mission by contributing to the development of more compact, highly integrated sensors. LLNL faces the challenge of interfacing a sensitive molecular detection and identification system to a complex environment. This microseparation capability would also enable analytical science at volumes needed for single-cell analysis, which could benefit the Laboratory's mission in science and technology to improve human health.

FY04 Accomplishments and Results

In FY04, we built a capability for embedding CNTs into an inorganic material and building a free-standing membrane. This capability has made us the hub of a broad collaboration. We also achieved the first permeation measurements for noble gases. Our results were published in a conference presentation and a proceeding. Our fabrication capability will enable future work in CNT-based GC separation.

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Proposed Work for FY05

Because the slow rate of diffusion through the CNTs observed in FY04 probably indicates that the majority of these tubes are bamboo-like in structure, we will pursue three approaches to achieve faster diffusion rates in FY05: (1) working with shorter tubes, (2) using single-walled CNT "forests," and (3) using lateral transport through a single-walled nanotube lying horizontally on a substrate. We will also complete a crucial water-transport experiment and demonstrate how this scales with molecular-size nanotube material, thereby building a conceptual framework for understanding the bulk, surface, and molecular aspects of the transport process.

Publications

Holk, J. K. et al. (2004). "Carbon nanotube-based permeable membranes." *Proc. Mat. Res. Soc.* **820**, 3.1. UCRL-CONF-203753.

Holt, J. K. et al. (2004). "Fabrication of a carbon nanotube-embedded silicon nitride membrane for studies of nanometer-scale mass transport." *Nanoletters* **4**, 2245. UCRL-CONF-204540.

Space-Time Secure Communications for Hostile Environments

James V. Candy 03-LW-005

Abstract

Communicating in a complex environment is a daunting problem. Such an environment can be a hostile urban setting populated with a multitude of buildings and vehicles, military operations in an environment with obstructive topographic features, or even a maze of buried pipes within a municipal water supply. The obstructions inherent in these environments cause transmitted signals to reflect, refract, and disperse in a multitude of directions, resulting in signal distortions at network receiver locations. In this project, which incorporates theory, simulation, and experiments, we are developing secure time-reversal (TR) communications methods to improve communications in noisy, distorted environments.

Successful development and demonstration of TR receiver performance will lead to the next generation advance in improved wireless communication systems for both military and civilian (commercial) applications in complex or hostile environments, including caves, urban environments, water pipes, nuclear power plant systems, and border-protection systems.

Mission Relevance

Development of the TR receiver will greatly benefit national-security and military applications. Channel reliability in a hostile environment is a prime concern of security forces, especially in counterterrorism activities. The TR receiver technology also promotes protection of communication channels against intercept and thus supports LLNL's national-security and homeland security missions.

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FY04 Accomplishments and Results

In FY04, we (1) completed theoretical development of a receiver design for multichannel communications systems; (2) developed and deployed an eight-element acoustic array; (3) used simulations to perform multichannel TR array design and conducted controlled laboratory experiments on acoustics using the TR array; (4) demonstrated a critical theoretical development for future electromagnetic (EM) experiments using 1-bit analog/digital (A/D) conversion for TR receivers; (5) performed controlled experiments demonstrating TR receiver performance; (6) designed a highly scattering medium on an aluminum plate and used simulations to perform proof-of-principle TR receiver designs; and (7) initiated several spin-off projects using related TR concepts.

Proposed Work for FY05

We will continue developing a fundamental theoretical basis for TR communications by pursuing both simulations and laboratory evaluations. Evaluation of the techniques in a hostile environment will lead to (1) theoretical development, (2) simulation-based evaluation of the benefits of TR, (3) controlled laboratory validation of the simulation results, (4) an operational brass-board design of an actual TR receiver for EM applications using the 1-bit A/D concept, (5) development of new TR principles using 1-bit A/D converters, and (6) participation in TR communications experiments in wideband EM and acoustic-pipe experiments.

Publications

Candy, J. et al. (2004). “Performance of a multichannel time-reversal receiver design in a highly reverberant environment.” *J. Acoust. Soc. Amer.* **116**, 2574. UCRL-ABS-201966.

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Chambers, D., C. Kent, and A. Meyer. (2004). Time-reversal communication through a highly scattering medium. *J. Acoust. Soc. Amer.* **115**, 2526. UCRL-ABS-201967.

Chambers, D. et al. (2004). “Time reversal and the spatio-temporal matched filter.” *J. Acoust. Soc. Amer.* **116**, 1348, UCRL-JRNL-202959.

Guidry, B. et al. (2003). “Experimental design and processing for time-reversal communications in a highly reverberative environment.” *J. Acoust. Soc. Amer.* **114**, 2367. UCRL-ABS-20073.

Coherent Anti-Stokes Raman Microscopy: Specific Molecular In Vivo Imaging at Superresolution without Fluorescence Labels

Thomas R. Huser 03-LW-056

Abstract

Coherent anti-Stokes Raman scattering (CARS) microscopy images cells without the need for extrinsic markers such as fluorescent labels. The CARS technique provides three-dimensional (3-D) sectioning capability in vibrational marker modes with very low background, and can study subcellular processes with picosecond time resolution. This technique also has

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applications in materials science for the investigation of stress and corrosion, the analysis of composite systems (e.g., alloys and high explosives), and in forensics applications. This project proposes to develop a unique CARS microscope, extend its spatial resolution beyond the diffraction limit, increase its sensitivity to the single-molecule level, and apply this technique to study the interaction pathway of viruses with cells.

The project will develop a CARS microscope system that will provide imaging capabilities based on intrinsic markers in an entirely non-invasive fashion and with spatial resolution that no other technique can provide.

Mission Relevance

The CARS microscopy technique has application to the study of cell–virus interactions in support of the Laboratory's mission in bioscience to improve human health. The capability is also directly relevant to the imaging component of DOE's Genomics:GtL Program.

FY04 Accomplishments and Results

We successfully demonstrated biological imaging with the CARS microscope by imaging and identifying individual bacterial endospores. We studied the effect of virus invasion in single living cells and determined its main spectroscopic markers, and investigated the interaction of lipoprotein particles with endothelial cells in collaboration with the University of California at Davis, determining changes in these particles during the hydrolyzation process. This work has enabled follow-on studies requiring high-resolution and high-sensitivity imaging capabilities.

Publications

Chan, J. W. et al. (2004). "Reagentless identification of single bacterial spores in aqueous solution by confocal laser tweezers Raman spectroscopy." *Analytical Chemistry* **76**, 599 UCRL-JC-155491.

A Novel Antimatter Detector with Application to Dark Matter Searches

William W. Craig 03-LW-059

Abstract

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

The GAPS approach will provide an entirely new and very effective technique for detecting antimatter in cosmic rays, one the key questions of cosmology. As such, it will produce novel

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and important publications as a direct result of beamline testing and enable a new class of space-borne instrumentation to observe cosmic antimatter.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we completed the construction, laboratory testing, and calibration of the GAPS prototype. A primary task was to generate the software for real-time data processing; the first version was completed this year. The prototype was readied for shipping to the Japanese High-Energy Accelerator Research Organization (KEK), where 20 shifts of beam time were used to perform initial system validation. Testing using laboratory sources and simulated triggers and vetoes to validate end-to-end instrument performance was completed prior to a successful preship review. The initial beamline characterization was fully successful and set the stage for our science data runs in FY05.

Proposed Work for FY05

We will complete characterization of the performance of the GAPS prototype instrument using the KEK accelerator facility and will obtain additional beamline data and perform data analysis to determine absolute efficiency and background rejection capability. We will also derive the energy and time resolution requirements for space and balloon-borne implementations of the GAPS concept. Assuming that the beam tests validate our calculations, we will then develop a design for a balloon-borne instrument to detect the low-energy antiproton component in the cosmic ray flux. The beamline results from FY04 and FY05 will be written up for publication; at least two significant publications are expected.

Low-Voltage, High-Precision Spatial Light Modulator

Alexandros P. Papavasiliou 04-ERD-014

Abstract

This project aims to demonstrate a technology—a spatial light modulator (SLM)—that will make it possible to co-locate electronics components with microelectromechanical systems (MEMS) sensors. This SLM technology will enable large arrays of devices that would otherwise require an excessively large number of wires (e.g., devices that are sensitive to the parasitic capacitance of macroscopic wires) and devices (such as autonomous devices) that must fit into an extremely small package, precluding a multiple-chip approach. The SLM will

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demonstrate the technology's (1) scalability by requiring far fewer than one wire per actuator and (2) increased sensitivity by using the electronics to sense actuator position.

Expected Results

This project will produce SLM-based technology to integrate MEMS sensors and actuators with electronics. In addition, this work will advance the capabilities of SLMs by substantially reducing the required voltage and increasing positioning accuracy. Once demonstrated, this technology will help develop new sensors and optical systems.

Mission Relevance

This work will support the Laboratory's national-security mission by advancing adaptive optics for surveillance in nonproliferation applications. Numerous additional national security applications include large sensor arrays, low-noise sensors, and autonomous devices.

FY04 Accomplishments and Results

Our accomplishments in FY04 include (1) first-principles modeling of three-electrode and lever-arm MEMS actuators; (2) optimal design of three-electrode and lever-arm MEMS actuators; (3) design and layout of MEMS actuators, lateral resonators, and cantilever devices for MEMS foundry fabrication; (4) fabrication of the designed devices through a MEMS foundry; and (5) design of an interconnect chip for MEMS bonding. In preparation for designing the integrated circuits that will act as a voltage driver for the MEMS devices, we began determining the appropriate complementary metal oxide semiconductor foundry technology.

Ultrafast Transient Recording Enhancements for Optical Streak Cameras

Corey V. Bennett 04-ERD-025

Abstract

Several high-energy-density physics (HEDP) and inertial-confinement fusion experiments on current and future fusion-class laser systems will require hard x-ray and neutron diagnostics with resolution of ~ 1 ps or less and a high dynamic range. The Linac Coherent Light Source at the Stanford Linear Accelerator (SLAC) will need to measure timing and pulse shapes of its 100-fs FWHM x-ray pulse. These measurement requirements are far beyond existing solutions. We will investigate ultrafast optical recorder technologies to develop a rugged, fiber-based temporal imaging system for optical-streak cameras that can meet these measurement requirements. This system will (1) improve system resolution and dynamic range by "stretching" an input signal in time, allowing ultrafast signal recording with slower, higher-dynamic-range instruments, and (2) convert light from 1550 to 775 nm, allowing streak cameras to record signals from detectors using 1550-nm technology.

If successful, the project will demonstrate single-shot operation of a robust temporal imaging system and streak camera recorder having less than 300-fs resolution, a dynamic range greater than 100, and compatiblity with current and future hard x-ray detectors that

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produce a modulated optical carrier at 1550 nm. Arrays of microlenses and fiber optic components will be investigated for spatial imaging applications, including demonstrating a low channel-count system and developing a complete ultrafast imaging system plan.

Mission Relevance

Our goal is to ensure delivery of the next-generation diagnostics needed for stockpile-stewardship experiments at current and future large laser systems. These diagnostics will enable HEDP and ICF experiments, including measuring reaction history, dynamic holhraum temperature, dynamic opacity, and detecting subpicosecond backscatter bursts.

FY04 Accomplishments and Results

A preliminary temporal imaging system design was completed using an older modeling code. We then developed more-detailed models of the system using the codes LinkSim and MATLAB to incorporate all possible physics effects in this system. We also designed and assembled the time lens pump laser and pump dispersion system, which produces the chirped optical pulse in the nonlinear crystal. We completed the design and construction of a precision dispersion measurement system, which will enable proper focusing of the temporal imaging system, as well as the characterization and correction of possible distortions due to higher order dispersive aberrations.

Proposed Work for FY05

We will (1) finish construction of the input dispersion and the measurement and correction of higher-order phase terms in the input and pump dispersive delay lines, which are the primary aberrations that affect the systems impulse response; (2) design and implement high-energy-laser-compatible timing and triggering systems; (3) design and construct the nonlinear crystal sum-frequency-generation device; and (4) complete measurements that will allow us to predict the final system's impulse response shape, the system's temporal field of view, signal up-conversion efficiency, any added timing jitter, and the dynamic range of the temporal imaging system. These measurements will allow us to compare theoretical and experimentally determined maximum allowed output dispersion loss.

Resolution Boosting for Wide-Field and Compact Snapshot Spectrographs

David J. Erskine 04-ERD-067

Abstract

This project develops a technique for boosting the spectral resolution and stability of any existing grating spectrograph by a factor of 5 to 10, by combining it with a small interferometer and processing the data with a custom algorithm. The interferometer creates a periodic grid overlaying the input spectrum, which causes moiré patterns that depend on the narrow spectral features. The moiré patterns are recorded for several interferometer delay values. The patterns are processed to reconstruct the narrow spectral features that

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originally produced them, but which are normally unresolvable without the interferometer. Hence the resolution of the grating spectrograph is increased. This same algorithm and moiré principle can be used to boost the time resolution of streak camera recordings.

The goal is to demonstrate a low cost technique for boosting the resolution performance of existing optical grating spectrographs by 5 to 10×, and increasing the low-light signal-to-noise ratio of Fourier transform spectrometers by 10 to 100×. This performance boost can be exchanged for a much wider field of view, or a much more compact, low-cost, and lightweight spectrograph for a given resolution, thereby dramatically reducing the cost of airborne or spaceborne spectrographs for reconnaissance, aerospace, or planetary exploration applications. Single-shot spectral data (such as from shockwave experiments) can be recorded at wider field of view. The algorithms developed here can boost time resolution for streak cameras in shock experiments using an analogous moiré principle.

Mission Relevance

The spectroscopic techniques developed in this project will offer improved diagnostics for fusion-class lasers and gas gun experiments in support of LLNL's stockpile stewardship mission, and improve the resolution and reduce the cost of spectrographs for remote sensing, in support of LLNL's missions in nonproliferation, advanced military applications, and breakthroughs in basic science.

FY04 Accomplishments and Results

In FY04, we demonstrated a factor-of-6 spectral resolution boosting, from a native 25,000 to final 140,000, by adding a small interferometer to a commercial 0.6-m-long spectrograph. We measured the iodine spectrum, which has many narrow spectral features. Repeating the experiment with a 2× wider input slit would produce a 12× resolution boost (12,500 boosted to 140,000). Achieving 140,000 in a portable tabletop instrument is remarkable when compared to the 50,000-resolution Lick Observatory spectrograph, which is 7-m long and weighs many tons. We demonstrated that multiple spectral moiré data can be combined to form a composite spectrum having almost unlimited spectral resolution.

Publications

Erskine, D. J. and J. Edelstein. (2004). "Interferometric resolution boosting for spectrographs." *Proc. SPIE Conf. on Astron. Instrumentation*, Glasgow UK, June 21, 2004. UCRL-PROC-204704.

Erskine, D. J. and J. Edelstein. (2004). *Multiple-delay externally dispersed interferometry.* Presented at the OSA Conf. on Fourier Transform Spectroscopy, Alexandria, VA, Jan. 31, 2005. UCRL-PROC-206872.

An Integrated Laboratory for the Study of Interventional Device Dynamics

Duncan J. Maitland 04-ERD-093

Abstract

We propose to develop an integrated capability 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 our 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 believe 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. 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 in support of the Laboratory's national security and homeland security missions.

FY04 Accomplishments and Results

Prototype and beta test versions of the foam SMP device for treating aneurysms were successfully demonstrated. We performed computational modeling of the fluid flow within healthy and diseased basilar arteries and validated the new simulation code, StarCD. All components of the process for creating simulation/physical models of vascular anatomies were exercised. Finally, we began PIV experiments on the laser-activated shape memory polymer foam for treating aneurysms.

Proposed Work for FY05

Research in FY05 will focus on four areas: (1) conducting detailed studies of flow and temperature for the SMP foam device; (2) building and using the PIV system at LLNL; (3) expanding the CFD code to include temperature, device dynamics and laser transport; and (4) fabricating and testing a bifurcated stent.

Explosive Particle Imager for Standoff Detection

Gregory L. Klunder 04-ERD-097

Abstract

Increased security measures due to terrorist threats have created a strong demand for new technologies to detect explosive materials. During the assembly of improvised explosive

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devices small particulates can be left on the packages and/or persons involved with the activities. Current airport explosive detection methods require that a particle is collected on the swipe and introduced into the instrument. This project will develop an imaging system that can screen remotely for explosive particles based on their diffuse reflectance spectral signatures. A near-infrared (NIR) camera with a tunable filter will acquire spectral image cubes in the 1000- to 1700-nm range, where signature CH, OH, NH vibrational overtones and combination bands are observed. Many energetic materials have unique spectra in the NIR and may be discriminated from background and differentiated from other particles. A particle array of explosive and nonexplosive materials of various sizes will be studied to determine the limits of detection and the ability to differentiate different particles. Principal component analysis will be applied to image cubes to discriminate explosives from common background materials.

Limits of detection will be determined experimentally and experimental parameters will be evaluated to estimate improvements. Limits of detection will depend on a number of parameters, including reflectivity of the material, background, field of view, distance to the object, collection efficiency of the optics, lighting, and spectral response of the camera.

Mission Relevance

Developing a system for rapid screening to detect the presence of explosives at a standoff distance would contribute to the Laboratory's missions in counterproliferation and counterterrorism and find applications in the field, including support to first responders, that support LLNL's national security and homeland security missions.

FY04 Accomplishments and Results

During FY04, an NIR camera system with a tunable filter capable of chemical imaging was purchased and assembled. Particles of TNT (~500-μm diameter) were discriminated from inert materials based on their diffuse reflectance NIR spectra. Principal component analysis on the image cubes demonstrated the ability to highlight suspect particles. For the operating conditions tested, we estimate a particle detection limit of 100-μm diameter for a material detected from distance of 1 m. A similar limit of detection could be extrapolated to a distance of 2 to 3 m, with improved optics and lighting conditions. Thus, from these initial results, standoff detection of explosive particles is feasible as a screening technique.

Proposed Work for FY05

Work proposed for FY05 will expand on FY04 results. Particles will be screened on different surfaces to determine the sensitivity and selectivity of detection under more realistic conditions. In addition, we will begin modeling the diffuse reflectance measurements based on Mie scattering theory and Kubelka-Munk reflectance theory. Results from the model will be compared to experimental results and predictions will be made to determine a detection limit based on fundamental parameters of light absorption and scattering. These results will provide a better understanding of reflectance spectroscopic detection of particles on surfaces.

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Volume Radoptic Detectors

Mark E. Lowry 04-ERD-099

Abstract

This project performed a preliminary investigation of a novel, solid-state, uncooled radiation-detection approach. The underlying principle of this detector is the radiation-induced change in the index of refraction of a semiconductor, dubbed the radoptic effect, which enables a monolithic transparent semiconductor to be used as a kind of optoelectronic bubble chamber to image tracks (e.g. of recoil electrons, or protons). This approach promises very high performance in quantum efficiency, temporal response, and imaging (without collimation) for the difficult problems of neutrons, gammas, and charged-particle detection. Our approach is unique in that it uses optical readout techniques to interrogate the required high volume of material.

This novel concept represents a significant departure from other radiation-detection approaches. As such, it is very high risk, but also has the potential for very high payoff. With this project we expect to resolve many of the critical technical questions. If we are successful and these answers are favorable, we will have enabled a clear path to the future research directions and development paths for this technology applied to large-volume radiation detectors, including the development of a small-scale prototype.

Mission Relevance

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

FY04 Accomplishments and Results

In FY2004, we successfully (1) concluded initial analytical and numerical studies of optical scattering from radiation-induced phase objects (i.e., the radoptic effect), (2) estimated the expected scattered light as a function of neutron and gamma energies using various particle electron-hole pair conversion schemes, (3) studied the literature of background scattering in various semiconducting media, and (4) investigated various optical readout schemes and image reconstruction algorithms. These results lead us to conclude that it is probably possible to detect single-particle induced radoptic phase objects in semiconductors.

Prototype Chip Fabrication for Electrochemical Pathogen Detection

Paul T. Henderson 04-FS-010

Abstract

We propose to develop a prototype that will demonstrate the feasibility of a low-cost, rapid, pathogen-DNA detection system that does not require expensive reagents and can be

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automated. The method involves (1) capturing a pathogenic DNA target sequence between two microelectrodes on a silicon chip, (2) metallating the immobilized DNA duplex using a novel process for the photochemically mediated deposition of silver metal onto the DNA duplex, and (3) detecting the duplex using an electrical current across the newly-formed nanowire junction. This project leverages a recently developed LLNL process for laser-mediated deposition of silver onto DNA. Application of this technology to surface-bound DNA requires determining the best combination of chemical and light source and optimizing the microelectrode geometry on the chip.

If successful, this project will produce a very sensitive prototype device, because a substantial current can originate from the capture of a single DNA target sequence. Once the chip performance is evaluated, the data can be used to develop devices to monitor environmental and forensic samples and for health-care-related diagnostics and monitoring. The ability to manufacture a portable, electrochemistry-based pathogen detector has the potential to benefit medical and genomics efforts.

Mission Relevance

A low-cost, field-deployable pathogen-DNA detection system would enable civilian and military applications that require the rapid detection of elevated levels of pathogens. The prototype system developed in this project will find myriad uses for biodetection in support of LLNL missions in national and homeland security and in biotechnology to improve human health.

FY04 Accomplishments and Results

We designed and fabricated silicon chips containing microelectrodes spanned by DNA bridges to determine the feasibility of photochemically plating metal onto the DNA bridge, thereby completing the circuit. We used this method to build a prototype device for measuring the changes in DNA conductivity. The method apparently produces small metal particles across the DNA, particularly in the presence on a photoactive compound that was chosen from among many candidates. We found that conductivity is enhanced by depositing additional metal using a conventional chemistry technique known as development. We also set the stage for future work to test the selectivity of this method using pathogenic DNA from a variety of anthrax strains.

Detecting, Locating, and Characterizing Remote Power Sources

Philip E. Harben 04-FS-017

Abstract

This project evaluated long-range electromagnetic (EM) detection, location, and characterization of isolated power sources using B-field gradiometers, E-field sensors, and Poynting vector and harmonic ratio analysis. The research consisted of a field experiment and experimental data analysis to determine if this approach is feasible for long-range standoff detection. The project attempted to answer the following questions: (1) Can B-field

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gradiometer measurements enhance detection ranges? (2) Can new sensors effectively measure E-fields? (3) Can Poynting vectors be used to point to the source? (4) Do spatial highs and nulls exist in the EM field? (5) Can harmonics help in characterization? and (6) Are stand-off ranges large enough to be useful for practical applications?

If successful this project will determine the limitations of employing various EM-field-detection techniques to the search for isolated power sources. A successful study should highlight those detection, location, and characterization techniques that have merit and the application space best suited for further research and development. Conclusions will include analyses involving fundamental power frequencies and higher harmonics.

Mission Relevance

With sufficient standoff detection range and an ability to calculate a back azimuth to an isolated power source, an operational system could be developed to provide a new remote-sensing capability. This method would help detect, locate, and characterize concealed weapons-of-mass-destruction facilities and activities in support of the Laboratory's nonproliferation and counterproliferation missions.

FY04 Accomplishments and Results

A field experiment was conducted at the Carrizo Plain in Southern California to measure EM fields from a generator powering three different loads: a 300-ft string of lights, an inverter-powered electric motor, and a resistive heater bank. The site was chosen for overall low background EM noise. Full E- and B-field vector measurements were made at 100,000 samples/s on scripted load variations at distances up to 4 km from the source(s). The data show measurable E and B fields within 100 m of the source and unexpectedly large broadband background noise variations. The data taken are of sufficient quality to evaluate Poynting vector back-azimuth calculations, higher harmonic detectability, and stacking methods to enhance detectability at even larger standoffs.

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

Todd A. Sulchek

04-FS-034

Abstract

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

First, we expect to create an AFM liquid cell with greatly expanded capabilities. By reducing the volume contained, we enable in situ AFM experiments for a broad range of proteins. We seek to demonstrate this capability with the effect on calcium oxylate of osteopontin, a major

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influence of kidney stone formation. Second, the liquid cell will enable imaging at physiological conditions (at both elevated temperatures and precisely controlled solution environments). Third, this small-volume technique will enable cantilever-based sensing experiments by maintaining high concentrations of the biological or chemical signal of interest.

Mission Relevance

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

FY04 Accomplishments and Results

After a late-year start, we completed the design and fabrication of prototype microfluidic cells, including (1) completing the mask design to pattern the channel layer; (2) devising a method to reduce the hysteresis caused by force between the channel layer and the sample; (3) fabricating a mold that will form the bulk of the fluid cell that allows precise positioning of the cantilever die to the inlet and outlet ports; (4) testing the fabrication of most of the individual steps of the fluid cell; and (5) finishing the process for submitting the Record of Invention.

Proposed Work for FY05

After verifying the functioning of the liquid cell prototype, including perfecting the transfer of the channel layer to the cell and aligning the channel to the inlet and outlet ports, we will test the prototype by imaging under microfluidic flow, then apply the liquid cell to study the effect of osteopontin on calcium oxylate at physiological conditions. We plan to make minor improvements to our device after our initial studies, including maximizing the scan size by optimizing the channel aspect ratio.

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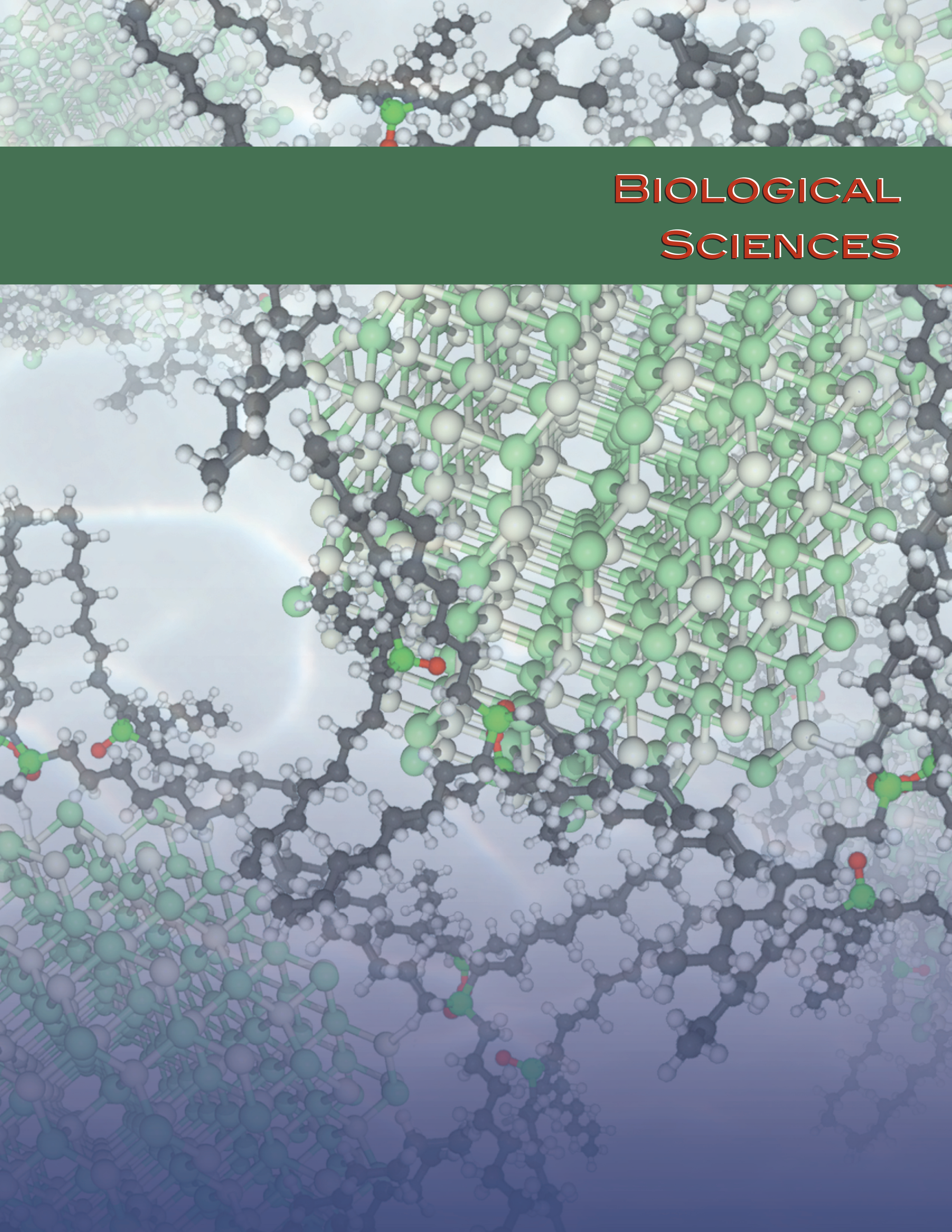
Biological Sciences



Biological Sciences

A Three-Dimensional Model of Signaling and Transport Pathways in Epithelial Cells

Andrew A. Quong

02-ERD-016

Abstract

Epithelial cells, which line all of the cavities and free surfaces of the body, act as a barrier to the environment and prevent the entry of pathogens into the body. This project is developing a fully three-dimensional (3-D) model of epithelial cells that includes ion transport across cell membranes. We are modeling epithelial cells using experimental data to identify the dominant components, develop a mesoscale model as a system of partial differential equations, and determine requisite parameters for the model. Results of the model can test hypotheses regarding the relationship between defective ion transport and disease, and it will be the starting point for studying the response of epithelial cells to various pathogens.

A 3-D model of the epithelial cell will be completed that includes passive diffusion of ions within the cell and across membranes, the active transport of ions across the cell membrane, and structural predictions and molecular simulations of the proteins responsible for metal-ion chelation and transport processes. A validated model will allow us to understand how a single change in a complex network can change the phenotypical behavior of a system.

Mission Relevance

A 3-D cellular model implemented within the ALE3D code will contribute to both LLNL's biosecurity mission by helping protect the civilian population against potential biological terrorism, and its mission of bioscience to improve human health by helping to determine the pathogenic pathways of genetic diseases.

FY04 Accomplishments and Results

In FY04, we developed a 3-D model of the epithelial cell that uses experimental data to determine diffusion parameters and for validation. We completed a trafficking model for the Ca^{2+} ion, including major compartments in the cell, such as the nucleus and endoplasmic reticulum. The model accounts for nonlinear calcium-induced calcium release instigated by the small molecule messenger 1-4,5-inositol-triphosphate (IP3) and explicitly includes mobile buffers. Computed results compared well with dye experiments in actual cells.

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

David S. Clague

02-ERD-066

Abstract

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

The project will deliver validated computational analysis tools to characterize particulate transport in viscous and gaseous flows. In addition, the newly developed capability will be useful for optimizing filtration, collection, and sample-preparation systems for chemical and biological early-detection systems.

Mission Relevance

This effort will directly contribute to the Laboratory's missions in homeland defense and counterterrorism by providing simulation tools for sample collection, filtration, and preparation systems for mitigating or preventing warfare or terrorist activities involving chemical and biological agents. In addition, this work is directly relevant to the characterization of fluid transport in porous media, which is relevant to environmental remediation and other work in support of the Laboratory's environmental management mission.

FY04 Accomplishments and Results

In FY04, we integrated the colloidal interactions module, which accounts for electrostatic, van der Waals, and hydrophobic/hydrophilic forces between surfaces, and we incorporated Brownian and buoyancy forces. Furthermore, we extended the viscous-flow LB capability to handle particulate flow in the gas phase by adding a new particle-tracking algorithm, including a capability to handle Reynolds-number-dependent inertial and buoyancy effects, and introducing the ability to handle larger time scales.

Proposed Work for FY05

We will finalize the hybridization of the LB and Fast QR (for determining the eigenvalues of a matrix) algorithm electromagnetism capabilities and compare results with existing theory and experiment, analyze relevant filter and collector efficiencies with and without environmental temperature effects, finalize inclusion of the shear-thinning model, and explore the implementation of unstructured grids to optimize physical resolution in desired domains. In addition, we will incorporate newly developed parallel optimizations, develop underlying scripts for structured data input and output for rapid flow visualization, apply Brownian

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motion capability to characterize particle petitioning and transport in nanoporous membranes with colloidal interactions, and publish our results.

Pathomics

Kenneth W. Turteltaub

03-SI-005

Abstract

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

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

Mission Relevance

This research, needed to meet gaps in our ability to detect and mitigate the effects of infectious disease whether natural or from acts of bioterrorism, supports the Laboratory's national security and homeland security missions as well as missions of the National Institutes of Health.

FY04 Accomplishments and Results

In FY04 we (1) enhanced the team with experts in real-time polymerase chain reaction (RT-PCR) and high-throughput assay technologies, statistics, bioinformatics and immunology; (2) completed studies of normal gene (mRNA) and protein expression in humans and mice; (3) completed wet-lab work [microarray, RT-PCR and protein profiles, two-dimensional fluorescence difference gel electrophoresis (2D-DIGE), liquid chromatography/mass spectrometry (LC/MS)] on humans with vaccinia; statistical and bioinformatic analyses were begun; (4) completed four pilot studies of mice with cowpox virus (CPV); results show down- and up-regulation of mRNA and proteins prior to CPV syndrome; (5) conducted genetic immunization of mice with host response markers produced in response to *Yersinia pestis* infection of human cells; (6) completed wet-lab work on a 2D-DIGE variablility study; and (7) established sample prep and LC/MS methods for quantification of proteins in mouse serum matrix.

Proposed Work for FY05

Work planned for FY05 includes (1) publish results from human and mouse normal range studies; (2) complete analysis of human vaccinia study; (3) validate CPV findings (mRNA and proteins) and extend to human studies of bacteremia and pneumonia (viral and bacterial) infections and animal studies of LPS, anthrax and SARS exposure; (4) develop host-response

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marker panel, yielding signatures that differentiate normal, sick (virus) and sick (bacteria) samples; (5) complete a human genetic variability study using 2D-DIGE; (6) complete a variability study of proteins in serum using LC/MS and apply to infected serum; and (7) develop genetic immunization and aptamers for molecular recognition.

Publications

Langlois, R.G. et al. (2004). "Protein profile alterations in hemodialysis patients." *Amer. J. Nephrology* **24**, 269-274. UCRL-JRNL-201081.

Langlois, R.G. et al. (2004). *Serum antigen profile alterations in hemodialysis patients.* Presented at the 22nd Cong. Intl. Soc. AnalyticalCytology, May 22–27, 2004. *Cytometry Part A* **59A** (1). UCRL-ABS-200268.

An Agent that Can Prohibit Microbial Development and Infection

Allen T. Christian 03-ERD-038

Abstract

We have developed a process that functions as RNA interference (RNAi), but is extremely long lasting. The agent that produces this effect—short, interfering DNA–RNA hybrids (siHybrids)—can be inserted into a cell and remain quiescent, without affecting cellular processes, until the gene against which it is targeted is induced. The agent then silences the genetic response without affecting the host cell in any other way. This project was meant to help develop the widely-used methods of RNA interference into therapeutically viable systems to combat infectious agents and other diseases.

A successful completion to this project will advance basic research by allowing simple gene knockouts to determine infectious pathways. It will impact applied research by serving as a broad-spectrum antiviral and antibacterial agent that is capable of attacking antibiotic-resistant and genetically engineered organisms. Preliminary results presented at a series of national meetings indicate that these results are of seminal importance to the field.

Mission Relevance

These molecules relate directly to the LLNL mission of combating weapons of mass destruction by rendering biowarfare agents null before or after infecting subjects. Other applications directly support LLNL's mission in bioscience and technology to improve human health.

FY04 Accomplishments and Results

Two major accomplishments in FY04 were the acceptance of a paper in Cancer Gene Therapy on the unaided uptake of siHybrids in human cells, and receiving an R&D 100 Award for this work. The paper detailed results related to the potential therapeutic use of the siHybrids. We also established a collaboration with the California State Department of Health to take advantage of these findings for human immunodeficiency virus (HIV) therapy. We demonstrated the effectiveness of siHybrids against bacteria but have not yet published the

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corresponding results without RNA data to match our protein data. However, collaborations with several microbiology groups helped us make progress in overcoming technical difficulties, and future work should establish the potential use of siHybrids against bacteria. We also collaborated with several cancer-therapy groups to transition our technology to Food and Drug Administration clinical trials.

Publications

Chen, A. A., L. C. Dugan, and A. T. Christian. (in press). "Unaided delivery of siHybrids mediates post-transcriptional gene silencing in human cells." *Cancer Gene Therapy*. UCRL-MI-202299.

Identifying Gene-Regulation Mechanisms using Rule-Based Classifiers

Krzysztof A. Fidelis 03-ERD-049

Abstract

The project is developing a new approach to identifying gene-regulation mechanisms based on supervised machine learning. Although the components and mechanisms of gene regulatory circuitry by which genes are switched on and off in different cell types, in different stages of development, and under different metabolic and environmental conditions are poorly understood, they are central to a more complete understanding of human disease, genetic susceptibility, and genome function. Our aim is to bring a new level of mathematical sophistication to the interpretation of these data. Using the formalism of rough sets, our work addresses this important problem and offers enabling technology for building tools and expertise for future efforts in gene-circuitry modeling.

Cataloging the associations between structural features of regulatory elements and the expression patterns of the corresponding genes provides the basis for modeling the mechanisms by which gene activity is controlled in living cells. The techniques developed in the project can be expanded to many applications, including study of the mechanisms of cellular homeostasis and environmental response in a wide variety of living systems.

Mission Relevance

Analysis of gene-regulation mechanisms can yield significant insight into several topics relevant to the Laboratory's missions within the area of homeland security (i.e., countering the threat of bioterrorism), environmental management (e.g., bioremediation), and human health (e.g., controlling and treating disease).

FY04 Accomplishments and Results

In the final year of the project we developed two new ways of testing our approach, and we conducted multiple performance analyses. The results of these tests allow us to compare our approach with other work in this area and the results are encouraging. We were also successful in developing a modified, iterative approach to the derivation of rules governing gene regulation. So far we have only used yeast for our work, but the methods we

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developed should allow analysis of gene-regulation mechanisms in any species. We presented our work at four conferences and submitted a paper to Genome Research.

Publications

Wilczynski, B. et al. (2004). *An iterative approach to gene regulation pathways discovery*. UCRL-ABS-201512.

Wilczynski, B. et al. (2004). *A rule-based framework for gene regulation pathway discovery*. UCRL-JC-154487.

Microbial Pathways

Harry R. Beller 03-ERD-062

Abstract

This project makes use of emerging genome-enabled techniques [such as comparative genomics and whole-genome complementary DNA (cDNA) microarrays] to explore novel, bacterial metabolic pathways and bacteria–mineral interactions that can mediate the behavior of uranium and other environmentally relevant elements in the subsurface. This is particularly pertinent to a remediation process that is of great interest to DOE: the in situ, reductive immobilization of metals, including radionuclides. Understanding microbial processes that mediate radionuclide contamination in the environment also has relevance to national security. The project is expected to encompass collaborations between the Laboratory and the DOE Joint Genome Institute.

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

Mission Relevance

This project supports LLNL's mission in environmental management and restoration. Microbial processes active in the near-surface environment are also important considerations in national security, especially issues concerning radionuclide and toxic metal contamination of groundwater.

FY04 Accomplishments and Results

Accomplishments to date include (1) the first-ever demonstration of anaerobic U(IV) oxidation in an autotrophic bacterium, (2) completion of the genome of *T. denitrificans* (we have resolved all misassemblies and have closed all gaps in the draft DNA sequence), (3) isolation of RNA from *T. denitrificans* for use in whole-genome cDNA microarrays (RNA was harvested from cells exposed to eight sets of conditions that tested differential gene expression from exposure to different electron donors), and (4) hiring of a postdoctoral researcher for this project.

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Proposed Work for FY05

Proposed work in FY05 includes (1) performing a comparative whole-genomic analysis of *T. denitrificans* against three bacterial species selected for their metabolic and biogeochemical capabilities, (2) preprocessing, normalizing, and computationally analyzing the cDNA microarray data generated in FY04, (3) initiating development of a genetic system in *T. denitrificans* for knockout-mutation (i.e., hypothesis-testing) studies, and (4) developing in vivo and in vitro assays for anaerobic Fe(II) oxidation.

Publications

Beller, H. R. (in press). “Anaerobic, nitrate-dependent oxidation of U(IV) oxide minerals by the chemolithoautotrophic bacterium *Thiobacillus denitrificans*.” *Applied and Environmental Microbiology*. UCRL-JRNL-204909.

Protein Model Database

Krzysztof A. Fidelis 03-ERD-063

Abstract

The project’s objective is to develop a global database of theoretically derived protein structures, the Protein Model Database (PMD). In the data describing the chain of biological events leading from sequence to function, protein models still remain the only primary data without a central public database. The motivation of this project is to provide such a repository, along with a means to assess model quality. In addition, model-generating capability will be afforded by (1) high-throughput genome-scale automated modeling and structure prediction, and (2) high-quality, supervised collaborative modeling by the best research groups worldwide. The implementation includes development of a dedicated relational database and tools for model analysis and assessment.

Presently, more than 99% of all sequenced proteins are left without the corresponding 3-D structure data. The PMD, with its capacity to promote high-quality, well-annotated models, will help to both close this gap and make theoretical models more accessible to biologists. Availability of structure data helps interpret results of biological experiments and guides the design of new ones. It is also critical to building models of biological processes, including insights to corresponding molecular interactions and the underlying physicochemical phenomena.

Mission Relevance

Establishing the PMD would enable the development of protein structure models and facilitate the ways they can benefit other projects, ranging from studies of human genome and research on human disease, to bio-defense and environmental studies. The basic nature and general applicability of protein-structure data is relevant to several Laboratory and DOE missions, from homeland defense to bioscience to improve human health.

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FY04 Accomplishments and Results

In FY04, we completed a relational database to store and query the data and developed new methods to analyze, compare, and classify protein structure models. These new methods included development and implementation of the model consensus algorithm, development of the distant-similarity detection software, and implementation of the automated classification of structure. The results of this work allow us to accept first models and to perform initial model analysis.

Publications

Daniluk, P. et al. (2004). *Identifying structural similarity of proteins using local descriptors of protein structures*. Pacific Symposium on Biocomputing, Kona, HI, 1/5-1/10, 2004. UCRL-ABS-201511.

The Instrumented Cell

Allen T. Christian 03-ERD-068

Abstract

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

This project aims to develop models of chemical interactions within and among cells, following perturbations made within the cell and to the cell's environment. The concept is that all of the pathways of a cell could be computationally modeled, allowing us to create predictive models of cellular responses to stimuli. Accomplishing this goal at the statistically relevant level of the individual cell would revolutionize biology, from basic research to industrial pharmacology.

Mission Relevance

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

FY04 Accomplishments and Results

For the IC platform, we developed the engineering component of a flow-through, single-cell analytical tool and filed several records of invention. We also investigated the ability of the siHybrids to enter cells without transfection, which is tremendously relevant for clinical

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applications, and studied the bacterial effects of the siHybrids. We have also began two collaborations. One, with Lawrence Berkeley National Laboratory, is to help use infrared microscopy in high-throughput applications relevant to Genomics:GtL. In this collaboration, we designed and built a custom IC system. We also began collaborating with the University of California at Merced in stem-cell research, using the IC system to study stem-cell proliferation.

Proposed Work for FY05

In FY05, we will elucidate the mechanisms of siHybrid delivery and function to improve the efficacy of the gene-silencing technology for therapeutic and research applications. Our primary goal is to understand how siHybrids function during unaided delivery into cells. Our research demonstrates that the siHybrids always enter cells without active delivery but do not always induce gene silencing. Our goal is to learn whether the molecules are intact when they enter the cell, and whether they are encapsulated in vesicles that prevent the interfering RNA enzymes from reaching them. Our IC system is ideal for this work, because it will allow real-time imaging of the molecules as they enter the cell.

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

Lawrence C. Dugan 03-ERI-004

Abstract

The objective of this proposal is to answer a number of important questions concerning a new gene-silencing technology known as small interfering RNA–DNA hybrids (siHybrids). This project is motivated by the improvements siHybrids have shown over small, interfering RNA (siRNA), a powerful tool for silencing gene function that is being considered for use in clinical settings, including cancer and HIV therapy. In this project, cells are being treated with siHybrid molecules and then collected and analyzed for the presence of the siHybrids using proton-induced x-ray emission (PIXE) and nano-secondary ion mass spectroscopy (nanoSIMS). We are also using molecular biology and biochemical techniques along with chromatin immunoprecipitation technology to determine how siHybrids function (e.g., whether they function similarly to siRNA).

Successful completion of this project will provide a population-based answer to the question of how many siHybrid molecules are required to cause an effect in a cell, where the change occurs, how the siHybrids enter cells without transfection reagent, and how long the molecules are present within the cell. Furthermore, this project should identify proteins involved in the process of siHybrid construction and in the effects of siHybrids. Finally, this information will also help determine gene functions in such cellular processes as development, differentiation, cell signaling, and cell death.

Mission Relevance

This research supports LLNL mission areas of (1) national security, by providing a potential means to mitigate the effects of biological warfare agents, and (2) bioscience and biotechnology to improve human health, by providing mechanistic information for a process being tested for use in treating cancer and infectious diseases.

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FY04 Accomplishments and Results

We successfully repeated quantification studies using PIXE analysis. Cells were transfected with Br-labeled siHybrids and analyzed. We found a 2- to 4-h delay from the time the siHybrids are added to the cell culture until the Br tag is detected within the cells. Interestingly, this corresponds to the timeframe of extracellular nuclease-mediated degradation of siHybrids and siRNA. We also used the Western and Northern blot techniques and reverse transcription polymerase chain reaction to show that RNA antisense siHybrids function similarly to siRNA. We obtained, from the Integrated Molecular Analysis of Genomes and their Expression Consortium, the vector of messenger RNA for the human Lamin A/C gene and confirmed its sequence to begin biochemical studies of hybrid mechanisms.

Proposed Work for FY05

We will (1) extend our use of PIXE to measure the kinetics of siHybrid uptake and the duration of the signal; (2) use PIXE and nanoSIMS to identify the subcellular compartment in which Br-labeled siHybrids reside; (3) isolate siHybrids delivered without transfection reagent via high-performance liquid chromatography, and analyze them by mass spectrometry to determine their degree of integrity; (4) begin extensive biochemical analysis of this mechanism using cell lysates, in vitro transcribed Lamin A/C mRNA, and siHybrids; (5) compare RNA antisense and DNA antisense siHybrids against siRNA; and (6) examine chromatin modifications to explain long-term silencing effects. These experiments will determine whether siHybrids function similarly to siRNA-based RNA interference.

Publications

Dugan, L., et al. (2004). *Tracking of dual-labeled siHybrid gene-silencing molecules in individual human cells using confocal laser scanning microscopy*. 48th Biophysical Soc. Ann. Mtg., Baltimore, MD, Feb. 14–18, 2004. UCRL-ABS-200454.

Cellular Response to Heat Stress: System Stability and Epigenetic Mechanisms

Halima Amer

03-ERI-005

Abstract

We propose to study the mechanism of epigenetic stress response by monitoring single cells for the persistent activation of heat-shock proteins over many generations. This unique, single-cell approach will remove the ambiguities associated with monitoring expression in cellular populations. The nature of the distribution of the cellular-stress response and the subsequent establishment of clonal populations exhibiting a range of stress responses will provide useful insights into factors underlying such response and phenotypic heterogeneity. Developing the techniques for quantitatively monitoring such phenotypic change will provide a tool for subsequent studies and form the groundwork for systems-level modeling of cellular adaptation.

The project will further the development of a unique experimental platform in which the variation of protein-expression levels can be studied in single cells as part of a wider cellular population. In addition to examining the variation in cellular response, a deeper

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understanding of the mechanism of heat-shock response at the cellular level will be gained. Both of these factors will ultimately contribute to understanding phenotypic variation across an apparently genotypically identical population of cells under different environmental conditions.

Mission Relevance

By providing insight into the epigenetic activation of protective systems in cells that are exposed to stresses such as heat or radiation, this project will support LLNL's mission in bioscience to improve human health. The project draws together many disciplines and will set the groundwork for future collaborations on a wide range of biological applications, including those relevant to the National Institutes of Health and the Department of Defense.

FY04 Accomplishments and Results

A central component of monitoring single-cell response to heat shock is the development of a cell line that is stably transfected with a single copy of a fluorescent protein attached to a heat shock protein promoter region. This requires the use of a custom-synthesized plasmid containing genetic elements from retroviruses. In FY04 the DNA plasmid needed for retroviral transfection of immortalized human cells was designed and synthesized. The resulting plasmid was tested by redigestion. Gel electrophoresis analysis yielded a DNA band corresponding to the correct length for the heat shock protein promoter and red fluorescent protein. The successful completion of this plasmid will allow reliable transfection and response monitoring of single cells.

Force Spectroscopy to Study Multivalent Binding in Protein-Antibody Interactions

Todd A. Sulchek 03-ERI-009

Abstract

We propose to use atomic-force microscopy (AFM) to measure the binding force between the Muc1 protein, an indicator of some cancers, and its antibody. This binding interaction is the targeting mechanism in radioimmunotherapy, which is used to treat cancer tumors. The Muc1 protein will be embedded in a suspended lipid bilayer that will simplify measurement while maintaining a cell-like environment. The goals of this project are to determine the unbinding force and the shape of the interaction potential of one, two, and three Muc1–antibody bonds; determine the optimal number of antibodies and tether length; and study details of multivalent binding. This force-measurement technique will open the door to studying other molecular interactions that occur at the interfaces of cells.

We expect to (1) develop a versatile and controllable method to study single-molecule binding that can be applied to other protein-binding systems; (2) provide quantitative data to support an experimental medical technology that could be used to help patients by improving methods for targeted delivery of radiotherapy; and (3) understand details of multivalent binding, which will have implications to all drug development because making stronger bonds between a drug and its target and making a greater number of parallel bonds are two approaches to developing effective drugs.

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Mission Relevance

The capability to measure antibody–protein interactions, combined with expertise in suspended bilayers, will enable numerous biological applications. These include applications in characterizing cell–pathogen interactions, which support the Laboratory's homeland security mission. In addition, research on drug–cell and cell–cell interactions supports LLNL's mission in bioscience to improve human health.

FY04 Accomplishments and Results

We developed a chemistry for reliably binding both the antibody and the target protein to the cantilever tip and sample in a reliable, robust manner. This was accomplished by covalently linking the proteins to a polyethylene glycol tether. Using the Wave Metrics software, we wrote routines to batch-analyze force curves, locate all unbinding events, and calculate both the rupture force and rupture location. We fit the tether stretch to a freely jointed chain model to verify single tether stretches. Single-molecule Muc1-antibody binding strengths were measured using a competitive assay that blocks the specific binding with excess free Muc1 antigen. These measurements revealed a specific single binding strength of 145 pN.

Proposed Work for FY05

In FY05, we will determine the loading rate from theoretical fits of data and plot rupture force versus loading rate and compare this to Bell Theory to determine the characteristic bond length. We will also apply our results to the multiple bond-formation cases. Working with UC Davis collaborators, we plan to obtain new antibody clones and attach multiple antibodies to the cantilever tip. Finally, we will collaborate with Lawrence Berkeley National Laboratory colleagues to incorporate the full Muc1 protein into suspended lipid bilayers to create a model cell surface upon which the bonds form.

Publications

Sulchek, T. et al. (2004). *Cantilever-based force spectroscopy for chemical and biological detection*. UCRL-PROC-204264.

Sulchek, T. et al. (2004). "Force spectroscopy to study multivalent binding in protein-antibody interactions." *Biophys. J.* **86**, 478. UCRL-ABS-202844.

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

Ted A. Laurence 03-ERI-010

Abstract

The objective of this project is to develop methods to measure concentrations in chemical microenvironments in cells and tissues using recently developed functionalized metal nanoparticles (50–100 nm). Surface-enhanced Raman spectroscopy (SERS) allows sensitive detection of changes in the state of chemical groups attached to single nanoparticles. A nanoscale pH meter, which has already been tested in a cell-free medium, will be tested

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inside cells to determine optimal conditions and methods for these measurements. When tested and calibrated, the nanoscale pH meter will be used to image the local pH inside and outside cells in tumor tissues, which contain acidic extracellular matrices, and to study the characteristic pH drop associated with the early stages of apoptosis.

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

Mission Relevance

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

FY04 Accomplishments and Results

We worked on characterizing pH response in bulk solutions, developing a rapid assay that focuses only on the relevant lines to allow higher throughput in the number of particles analyzed. Our measurements demonstrated the dependence of SERS signal on pH. In addition, we characterized signal intensity fluctuations: By looking at correlation functions derived from the fluctuating signals, we were able to observe diffusion processes. The visibility of the rotational diffusion process was found to be polarization dependent, indicating a dipole nature to the scattering particles (or aggregates). Finally, we began studying the micro-injection of cells using the nanoscale pH meters; preliminary results indicated the presence of nanoparticles in the cells.

Proposed Work for FY05

In FY05, we will (1) test the signal intensity and spectra of the nanoparticles in buffer conditions similar to those found in cells (fluorescence-based pH indicators will be studied under the same conditions for comparison); (2) test methods of incorporating the SERS nanoparticles and fluorescent pH indicators into cells, including micro-injection, electroporation, and endocytosis (requiring incorporation into liposomes); (3) monitor cells over extended periods (hours to days) to test the long-term stability of the nanoparticles; and (4) measure the pH of the intracellular and extracellular environments of tumor cells, cells undergoing apoptosis, and cells with laser-ablated mitochondria.

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Publications

Laurence, T. et al. (2003). *Application of SERS nanoparticles for intracellular pH measurements*. 43rd Ann. Mtg. Am Soc. Cell Biology, San Francisco, CA, Dec. 13–17, 2003. UCRL-JC-154465-ABS.

Laurence, T. et al. (2004). *Application of SERS nanoparticles for intracellular pH measurements*. Biophysical Society Annual Meeting, Baltimore, MD, Feb. 14–18, 2004. UCRL-ABS-202843.

Single-Molecule Techniques for the Study of Chromatin Assembly and Remodeling

Christopher Jeans 03-ERI-011

Abstract

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

The images of chromatin and the measured forces will be used to gain insight into the nature of the interactions that hold the chromatin fiber together under different conditions. This will give us valuable new information about the structure of chromosomes and how remodeling affects this structure. Improved knowledge of the remodeling process will aid the understanding of a number of cellular processes, from DNA replication and repair to the mechanisms of gene activation and silencing.

Mission Relevance

This work will address questions of general biological interest through the development of novel techniques for the manipulation of single molecules and will provide a new capability that may provide critical data for biodefense applications in support of homeland security and for DOE's Genomics: GtL program.

FY04 Accomplishments and Results

Chromatin from chicken erythrocytes (CE) has been imaged using AFM in the presence and absence of linker histones. Linker histones are known to compact nucleosomes on the DNA into thicker fibers; this was demonstrated and quantified using image analysis. Chinese hamster ovary (CHO) chromatin is biochemically distinct from CE chromatin, containing different linker histones and having a different amount of DNA strongly associated with each nucleosome. Our preliminary AFM imaging of CHO chromatin shows individual nucleosomes to be morphologically identical to CE nucleosomes, but their packing into fibers is less ordered. Growth of CHO cells in the presence of sodium butyrate results in

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biochemical changes in the isolated chromatin, which will be studied further with AFM in FY05.

Proposed Work for FY05

Imaging of chromatin from CE, CHO and sodium butyrate-treated CHO cells will be carried out, and image analysis performed to identify any significant structural differences in DNA packing. Sodium butyrate alters the chemical modifications on the histone proteins, resulting in remodeling of the chromatin. Imaging will be extended from dry samples to samples in buffer. Once fluid imaging protocols have been developed, manipulation of the fibers with the AFM tip will be performed with the aim of measuring the forces required to elongate the different types of fiber. Chemical and compositional modifications will be performed on the chromatin in situ, and changes in the characteristics of the fibers will be measured.

Publications

Jeans, C. et al. (2004). *Single molecule studies of chromatin*. Presented at the 48th Ann. Biophys. Soc. Conf., Baltimore, MD, Feb.14–18, 2004. UCRL-CONF-202850.

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

Nan Shen 03-ERI-012

Abstract

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

We will integrate the capabilities of femtosecond laser cell surgery and high-resolution optical microscopy. The resultant system will allow us to study cell behaviors and responses in real time by probing cellular structures using femtosecond laser surgery techniques. To test this system and characterize the physical effects of laser surgery on cells, we will use atomic-force microscopy (AFM) or scanning electron microscopy to study laser-irradiated cell samples. We also plan to label mitochondria in a number of cell lines for the investigation of their roles in critical cell functions.

Mission Relevance

Data that result from this project will further our understanding cellular mechanisms and support DOE missions in bioscience to improve human health. Applications also include a variety of DOE efforts such as the Genomics:GtL Program. Ultimately, this understanding will

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prove important in helping to determine complete mechanisms for pathogenicity, in support of the Laboratory's homeland security mission.

FY04 Accomplishments and Results

We constructed an experimental setup consisting of a femtosecond laser oscillator, a cavity dumper system, an inverted microscope, and a cooled charge-coupled device camera. To study mitochondrial functions in cell proliferation and apoptosis, we successfully transfected BJ1 InfinityTM human fibroblast cells and bovine endothelial cells with plasmid DNA that encodes a fusion of enhanced yellow fluorescent protein (EYFP) with the mitochondrial targeting sequence for the cytochrome c oxidase. On our setup, we demonstrated that a femtosecond laser can precisely remove a subcellular structure, such as a single mitochondrion, without affecting the physiological function of other structures in living cells. We also looked at the extent of laser disruption by performing AFM on laser irradiated cell samples.

Proposed Work for FY05

We will continue to investigate how cells respond to femtosecond laser surgery. Questions we will address include: Does disruption of certain structures have more vital effects on cell functions than others? How does the cell respond over time to the disruption of nonessential organelles (e.g., a single mitochondrion)? Does laser surgery cause DNA damage? Single-cell laser surgery will also be used to disrupt individual mitochondria in the study of apoptosis. Our other focus will be testing the feasibility of delivering nanoparticle sensors through a live cell membrane by transiently creating a pore with the femtosecond laser surgery technique. The ultimate goal would be to use the nanoparticles to detect chemical changes in a cell after the laser disruption of mitochondria.

Mutations that Cause Human Disease: A Computational-Experimental Approach

Peter T. Beernink 03-LW-017

Abstract

In this project, we assess the effects of mutations that cause human diseases using a combination of computational and experimental approaches. This work is timely because the number of known human genetic variants proteins has grown exponentially in the past two years and the ability to integrate this data will depend on computational methods. Most disease-causing variants impact protein stability, therefore our objective is to calculate the structural properties of disease-causing proteins in cases where the structure of the normal protein is known. Predictions are validated with quantitative measurements of protein thermal stability. The resulting methods allow a rigorous analysis of over 100 proteins that cause single-locus genetic diseases and that have known, wild-type structures.

The project has developed a new computational approach for structure-based predictions of variant-protein stability, and obtained experimental data on stability and structure of certain proteins involved in cancer, which have elucidated the probable role of those variants in carcinogenesis. The experimental studies have validated the reliability of our

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predictions of the stability of variant proteins and their roles in human disease. By identifying specific effects of disease-associated genetic variation on protein structure and stability, this research will further scientific understanding of hundreds of disease-causing proteins.

Mission Relevance

This work supports the missions of LLNL in bioscience to improve human health and contributes to NIH missions by enhancing our understanding of natural human genetic variation, radiation biology, and cancer susceptibility. Genetic variation is hypothesized to lead to differences in individual susceptibility to cancer, which can be caused by environmental carcinogens, radionuclides, or spontaneous mutations.

FY04 Accomplishments and Results

In computational studies of protein stability, we developed a method using molecular dynamics (MD) simulations to calculate thermal stability. We showed a close correlation between experimental stability and the root-mean-square fluctuations in MD, which is well described by Lindemann theory and fits a binomial equation with an R^2 value of 0.97. We extended these studies to p53 tumor protein variants, the most commonly mutated protein in human cancer. We developed a high-throughput approach to examine the stability of many protein variants, which involves polymerase chain reaction and in vitro protein expression in 96-well format. The protein is quantified by fluorescence. In addition, we engineered variants of the Ape1 and XRCC1 DNA repair proteins, which will be used to examine the stability of cancer-associated variants.

Publications

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Beernink, P. T. et al. (2004). *Cancer-associated variants of Human Ape1 exhibit reduced enzyme activity and thermal stability*. Presented at the 9th Ann. Cancer Research Symp., U.C. Davis Cancer Center, Sacramento, CA and at the 1st Pacific Rim Conf. on Protein Science, Yokohama, Japan. UCRL-POST-200148.

Multiprobe Investigation of Proteomic Structure of Pathogens

Alexander J. Malkin 04-ERD-002

Astract

This project will investigate the proteomic structure of human pathogens through the combination of in situ high-resolution atomic force microscopy (AFM) with immunolabeling and mass spectrometry. A more comprehensive understanding of the structure and properties of pathogens would contribute significantly to an understanding of their life cycle and may lead to advances in diagnostic and immunological capabilities for biodefense. The

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research will characterize the protein structures of intact and dissected pathogens by AFM, correlate these structures with gene products using antibodies, and identify these protein components by mass spectrometry.

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

Mission Relevance

This work will support LLNL's national- and homeland-security missions by contributing to the development of techniques for identifying and characterizing organisms that might be used by bioterrorists. It also contributes to the Lab's mission in bioscience to improve human health by enabling more efficacious preventive and therapeutic measures for emerging diseases.

FY04 Accomplishments and Results

We used AFM to directly visualize in vitro, for the first time, species-specific high-resolution native structures of the spore coat of four *Bacillus* spore species. The direct visualization of the environmental response of individual spores revealed that the dormant spore is a dynamic physical structure, which changes in size and coat surface morphology when the environment changes from aqueous to aerial phase. The dimensions of individual spores were found to differ significantly depending on species, growth regimes, and environmental conditions. To map the proteomic structure of the spore coat, we produced and characterized several antibodies with immunochemical specificity for known protein components of the spore coat of several *Bacillus* species.

Proposed Work for FY05

We will utilize high-resolution in vitro AFM to develop molecular models of spore coat ultramicrostructures and models for visualizing the morphological changes of single endospores as a function of preparation procedures, environmental changes, targeted mutations, exposure to various decontamination agents, and corresponding changes in germination competency. To establish a direct correlation between the ultrastructure and the life cycle of bacterial spores, we will utilize monoclonal antibodies for the AFM imaging of cognate proteomic structures in dormant spores, activated spores, and germinating spores. This AFM analyses should help us define epitopes that are surface exposed, versus those that may be embedded in the surface of the pathogen.

Publications

Malkin, A. J., M. Plomp, and A. McPherson. (2004). *High-resolution visualization of pathogen signatures by in situ atomic force microscopy*. Materials Research Soc. Mtg., San Francisco, CA, April 12–16, 2004. UCRL-ABS-202134.

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bacterial spores visualized by atomic force microscopy. 2004 ASM Biodefense Research Meeting, Baltimore, MD, March 7–10, 2004. UCRL-ABS-202131.

Electronic Polymerase Chain Reaction

Shea N. Gardner 04-ERD-030

Abstract

The polymerase chain reaction (PCR) stands among the keystone technologies for analysis of sequence data. It is used at virtually every laboratory doing molecular, genetic, forensic, or medical research. Despite its ubiquity, we lack precise predictive capability to enable detailed optimization of the dynamics of PCR reactions. We are developing tools to perform virtual PCR (vPCR) by modeling the kinetic, thermodynamic, and biological processes of PCR. These tools will allow us to predict the effects of primers and reaction conditions on PCR products, and thus to optimize these variables. The algorithms we are building will enable, for the first time, the simulation of complete, multiple thermocycles. Applications of vPCR include the predictive, computational optimization of assays for the identification of genes and organisms present in complex microbial communities, DNA-polymerase-based gene synthesis, and forensic discrimination of closely related sequences.

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

Mission Relevance

Technology for vPCR will have applications in any field that uses PCR, including bioforensics, biodetection, and disease research, thus supporting LLNL missions in national security, homeland security, and bioscience and technology to improve human health.

FY04 Accomplishments and Results

After a midyear start, we (1) wrote the thermodynamics code to calculate reaction pathways and free energies; (2) linked this code with KINSOL, a nonlinear, simultaneous equation solver, to solve the thermodynamic equations; (3) developed the framework for efficient data transfer between the kinetic and thermodynamic modules; (4) developed algorithms for kinetic simulations; and (5) wrote prototype code to perform Gillespie's formulation of stochastic chemical kinetic simulations of PCR.

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Proposed Work for FY05

In FY05 we will (1) add polymerization (extension) to the kinetic and thermodynamic simulations; (2) modify the collective capability to handle the denaturation of primers during a thermocycle (i.e., the reverse reaction); (3) develop the capability to dynamically handle reaction vessel temperature ramps; (4) optimize parallelization at multiple levels; (5) perform experiments using real-time PCR to test the models; and (6) prepare a manuscript for submission to a peer-reviewed journal.

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

Julio A. Camarero 04-ERD-040

Abstract

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

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

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we accomplished in vivo cyclization of an SH3 domain as model polypeptide. The circular structure of the SH3 protein domain was fully characterized by elucidating its structure by nuclear magnetic resonance (pdb code entries: 1M3A, 1M3B and 1M3C). Studies initiated in FY04 are still in progress to obtain biological-based libraries based on short circular polypeptide scaffolds (i.e. cyclotides). Anthrax LF toxin was cloned into an expression vector. A post-doc was hired and a new scientific collaboration was established with the University of California, Santa Barbara for in vivo screening of cell-based libraries.

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Proposed Work for FY05

Work planned for FY05 includes biosynthesis of the first libraries based on cyclotides (small circular peptides extremely stable to biological and chemical conditions), development of a chameleon reporter for LF factor toxin, cloning, expression and purification of all the proteins involved, and model studies in vitro and then in vivo.

Publications

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

Gabriela G. Loots 04-ERD-052

Abstract

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

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

Mission Relevance

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

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FY04 Accomplishments and Results

In FY04 we (1) developed new computational tools for comparing whole genomes and identifying candidate genes; (2) optimized high-throughput in situ hybridization expression analysis on frog embryos using five known genes; (3) determined limitations by examining the expression patterns of ten novel genes from human chromosome 19; (4) perfected an in situ technique using DNA clones of five known genes; (5) aligned the human and frog genome, identifying 70 novel candidate genes and selecting 48 conserved genes for testing; and (6) began testing 10 novel genes from human chromosome 19.

Proposed Work for FY05

In FY05 we propose to (1) create a catalog of all novel genes conserved between humans and frogs and all other sequenced vertebrate genomes obtained from whole-genome alignments; (2) computationally prioritize genes based on known folds and motifs, which includes prioritizing, for functional characterization, putative homeobox genes, secreted molecules, transcription factors, and members of signaling pathways; (3) determine detailed expression patterns of 30 new genes; (4) experimentally analyze their function in frog oocytes by overexpressing messenger RNA or by knocking down endogenous transcripts with a combination of morpholino and small, interfering hybrids; and (5) begin creating a gene expression database.

Publications

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Field-Deployable DNA Analyzer

Elizabeth K. Wheeler 04-ERD-074

Abstract

Identifying and tracking individuals is a vital capability to the intelligence community and law enforcement. The miniaturization of polymerase-chain-reaction- (PCR-) based forensics

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is a recognized strength of LLNL, as exemplified by the Handheld Advanced Nuclei Acid Analyzer (HANAA), the Autonomous Pathogen Detection System (APDS), and the Bio-Briefcase. We propose to leverage this expertise in developing a DNA-based process for finding and identifying individuals by creating a small, portable DNA tester designed specifically to test DNA samples against known profiles of specific individuals. The concept has four components: (1) an automated sample-collection system, (2) a device for DNA analysis, (3) a component for the PCR- based analysis of DNA signatures, and (4) a system to compare the DNA to a database. Our research will investigate the fundamental limits of sample type, sample acquisition, and analysis necessary for a field-deployable human forensics identification device.

The goal of this project is to determine (1) the feasibility of automating sample introduction and preparation, (2) the limits of tissue types for which this concept would be feasible, (3) the minimum amplification times, and (3) the appropriate analysis technique for identification. Accomplishing these goals will enable the future development of an integrated portable DNA analyzer device.

Mission Relevance

This project leverages LLNL strengths and expertise in developing PCR-based technologies to advance the development of faster and smaller PCR-based forensics in support of the Laboratory’s mission in homeland security.

FY04 Accomplishments and Results

We investigated the fundamental limits of sample type, acquisition, and analysis necessary for a field-deployable human forensics identification device. We optimized a sample-preparation scheme for small sample sizes and one-touch operation. Assays minimized both the use of benchtop equipment and the times necessary for each step. This device is potentially useful in haplotype analyses for forensic applications in which multiple DNA sources are present. (The short tandem repeats technique used in law enforcement typically fails when a sample contains DNA from many subjects.) In addition, we designed, fabricated, and tested a rapid, antibody-based assay and device to determine exposure to bioterror agents.

Molecular Radiation Biodosimetry

Andrew J. Wyrobek 04-ERD-076

Abstract

This project will determine the feasibility of using molecular biomarkers to develop a new class of triage tools and medical assessment capabilities for use by first responders and medical clinics for consequence management of civilian radiation exposures. Our objectives are to develop valid biomarkers of an individual’s radiation exposure and to build and evaluate prototype devices using human models of radiation exposure. Biological dose will be based on early biochemical changes in cells from bodily fluids obtained within minutes to a few days

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after exposure. This project will evaluate specific DNA-damage-associated proteins and candidate gene transcript and protein biomarkers, and develop new human models for testing candidate biomarkers and prototype devices that utilize them.

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

Mission Relevance

This project is relevant to LLNL’s national security and homeland security missions, especially for the consequence management of persons exposed to ionizing radiation from radiological or nuclear events. The biomarkers to be developed in this project will support triage by first responders to identify those with severe exposure, and will enable followup medical assessment in a clinical setting to confirm exposure dose and tissue damage, which are critical for treatment to maximize long-term survival.

FY04 Accomplishments and Results

In FY04, we (1) applied gene-transcript microarrays to identify >50 radiation-sensitive genes; (2) performed pilot studies of radiation changes in DNA-damage proteins as radiation-specific biomarkers; (3) applied various protein discovery technologies (e.g., immuno-panning and mass spectrometry), identified >100 radiation-sensitive protein biomarkers, and prepared protein antibody arrays for parallel evaluation; (4) started protocol development for mouth, saliva, and blood cells for two new human models of radiation exposures (irradiated cells from normal donors, and cells from patients receiving radiotherapy); and (5) identified private-sector partners for developing a first-generation prototype triage device using the biomarkers validated under this project.

Proposed Work for FY05

We will (1) demonstrate the feasibility of gene-transcript biomarkers and DNA-damage-associated proteins for individualized radiation biodosimetry; (2) apply a high-throughput protein screening platform to down-select promising radiation biomarkers and validate their utility for mouth and blood samples; (3) develop human exposure models that use mouth and blood cells from normal donors (ex vivo model) and mouth cells from radiotherapy patients (in vivo model); and (4) identify additional biomarkers in mouth and blood cells of irradiated persons using novel LLNL technologies. This work will set the stage for building prototype triage and clinical devices for the optimal biomarkers and testing those devices in our human exposure models.

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

Lisa J. Stubbs 04-ERD-084

Abstract

This research seeks to establish new strategies and technologies for characterizing transcription factor (TF) proteins and the regulatory pathways in which they are involved in specific types of living cells. Understanding TFs and their regulatory "targets" is a key step in constructing regulatory network models. Beginning with characterizing a subclass of human TFs, we will develop a pipeline for characterizing TF proteins and pathways with the goal of building robust regulatory network models. All basic methods and expertise will be fully extensible to regulatory network modeling in any species, from microbes to mammals.

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

Mission Relevance

Accurate models of gene regulatory networks will be key to understanding biological mechanisms that govern form and function in all types of living cells. Understanding TFs and their regulatory "targets" is key to constructing regulatory network models, a goal important to both the National Institutes of Health (NIH) and DOE's biology programs, including Genomics:GTL. By building expertise in regulatory biology, biochemistry, and network modeling, this project supports LLNL missions in national security, environmental management, and bioscience to improve human health.

FY04 Accomplishments and Results

In FY04 we used bioinformatics approaches to identify more than 300 uncharacterized TF genes and selected 10 genes for initial studies. A bioinformatician we recruited developed a project database and designed and executed a strategy to identify novel TF genes. The team created protein expression constructs and used these constructs to establish robust protocols for gene manipulation experiments. We also recruited a postdoctoral fellow with expertise in protein biochemistry and established a protocol for chromatin immunoprecipitation (ChIP) experiments.

Proposed Work for FY05

During FY05 we will (1) select 50 zinc-finger (ZNF) genes for cloning into appropriate vectors; (2) complete a manuscript describing the human ZNF gene family and begin work

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to catalog these proteins in other species; (3) systematically apply protocols established in FY04 to analysis of 10 of the 50 selected proteins; (4) complete development of ChIP-based strategies for binding-site determination and complete ChIP analysis on the 10 proteins; (5) develop new methods for identifying DNA binding motifs in vitro—including a high-throughput version of the systemic evolution of ligands by exponential enrichment—and apply them to at least 5 proteins and a set of known-protein controls; and (6) begin work to derive pathway models using data derived for specific ZNF proteins.

Contaminant Uptake and Demography of the Loggerhead Shrike (Lanius Ludovicianus) at the Lawrence Livermore National Laboratory Site 300

Michael G. van Hattem 04-ERD-091

Abstract

The loggerhead shrike (*Lanius ludovicianus*) is a predatory passerine that is declining throughout its range. Because recent studies suggest that some contaminants can be detected in avian feathers, blood, and eggs, our study was designed to investigate the usefulness of biological materials (e.g., blood and feathers) collected from shrikes for analyzing contaminant load. We performed nonlethal collection of tissue samples in conjunction with territory mapping, bird banding, and demographic observations to determine if shrikes accumulate contaminants at Site 300, and if so, how these contaminants affect shrike reproductive success and survival. We tested the hypotheses that shrike contaminant loads are higher in contaminated areas, and that reproductive success of shrikes is negatively correlated with contaminant loads.

From this study, we expect to determine what effects contaminants have on the Site 300 shrike population. Past studies of contaminant loads in wildlife required sacrificing the study organism to obtain liver and fat tissues. The unique analytical capabilities of LLNL's Center for Accelerator Mass Spectrometry (CAMS) enabled us to detect lower levels of contaminants in these tissues compared to standard toxicology laboratories. Our study took the extra step to relate contaminant loads to reproduction and survival of this species, which may be a sentinel species for contaminant uptake.

Mission Relevance

This research will provide much-needed information regarding detectability of contaminants in special-status species, and contributes to the Laboratory's environmental management and assessment mission by determining whether the loggerhead shrike could serve as a sentinel species for low-level contaminant uptake. Because similar species are widespread throughout the world, the results of this research are applicable to other DOE facilities.

FY04 Accomplishments and Results

In FY2004, we (1) determined the distribution of shrike breeding pairs at Site 300 and compared the locations to contaminant distribution maps; (2) selected a suite of 11 metals to analyze on the basis of the contaminants found at bird territory locations; (3) used

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samples collected at Los Vaqueros Watershed as our control for contaminant uptake comparisons; (4) banded 36 nestlings and color-banded 36 fledged and adult birds; (5) determined fledging success; (6) assessed site fidelity with territory monitoring; (7) collected tissue samples; (8) measured metal content of samples at CAMS; and (9) analyzed contaminant load data in conjunction with demography information. Site 300 loggerhead shrike clutch size and hatching success appears to be consistent with published results for this species despite the fact that our preliminary evaluation of metal loads in blood samples shows that metal levels appear elevated when compared to the control site. Further examination of data is needed. Our study has shown that Loggerhead shrikes appear to be year-round residents at Site 300 and are an ideal organism for predator–prey contaminant uptake monitoring because of their predatory ecology and small territory size. A report detailing the methodology and results of this pilot study is in preparation.

Investigating the "Trojan Horse" of *Yersinia Pestis* Virulence

Joseph P. Fitch 04-ERD-101

Abstract

The plague-causing bacterium, Yersinia pestis, has caused three major pandemics. LLNL scientists speculate that *Y. pestis* is able to evade defeat using a "Trojan Horse" strategy to protect itself from the host immune response until a sufficient quorum of bacteria is available to overcome the host's defenses. The project proposed modeling "omic" and "real-time" studies to elucidate how the combined bacterial virulence and host defenses contribute to the pathogenesis of *Y. pestis*. "Omic" efforts focused on high-throughput proteomic and "functionomic" studies, and the "real-time" studies utilized a fluorescent reporter to continuously monitor virulence levels in living cells. The project was conducted in collaboration with a recognized Y. pestis expert at the University of Texas, Galveston.

By gaining a mechanistic understanding of virulence, the project results will be far-reaching and have considerable impact on the study of virulence in general and on overall biodefense preparedness. Our goal is to publish results in a high-profile journal like *Science or Nature*. We expect that this work will lead to follow-on projects conducted under programmatic sponsorship of agencies such as the NNSA, the Department of Homeland Security, or the National Institutes of Health (NIH). Results will have immediate application to pathogen detection, characterization of engineered threats, and to next-generation therapeutic/vaccine development for plague.

Mission Relevance

This work supports the Laboratory's mission in biodefense, with applications for detecting highly virulent, engineered and/or emerging pathogens. In addition, the work supports LLNL's mission in bioscience to improve human health by defining novel therapeutic and vaccine targets.

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FY04 Accomplishments and Results

In FY04, bioinformatic/modeling approaches were used to map proteomic, phenotypic and host response data (from previous host–pathogen interaction studies of *Y. pestis*) onto functional pathways to provide a systems biology view of virulence. The ultimate goal is to produce a predictive capability to elucidate virulence pathways and rapidly identify effective therapeutics. Molecular biology experiments to produce real-time reporter clones for dozens of virulence factors was also completed. Real-time characterization of virulence support the postulated "Trojan Horse" mechanism, explaining how *Y. pestis* is able to effectively elude the host immune response by presenting as "self" rather than "foreign." Two manuscripts are in preparation and a follow-on National Institutes of Health grant is targeted for FY05 submission.

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

Patrick S. Chain 04-ERD-103

Abstract

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

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

Mission Relevance

The novel search algorithm developed in this project will provide an important tool for genome, transcriptome, and systems biology. It will allow scientists to explore gene regulation to better understand regulatory networks in prokaryotic organism(s), and to apply this pathway information to those microbes relevant to the Laboratory's environmental (understanding pathways involved in microbial clean-up of metal contaminated sites) and national security (biodefense: understanding pathogen virulence pathways) missions.

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FY04 Accomplishments and Results

After a mid-year start, we made a significant number of accomplishments. Our first step was to obtain an accurate dataset for intergenic regions of the target bacterium, which involves a rigorous gene-calling and annotation format to accurately reflect the true coding potential and intergenic spaces in each genome. We have obtained the genomes of several strains of *Y. pestis* and *Y. pseudotuberculosis* and have curated the annotations to extract the intergenic regions. We have also performed a preliminary analysis of expression microarray data for Yersinia grown at 26 and 37°C, to begin characterizing the pathogen's gene (and pathway) response to temperature shift, and identify co-regulated genes.

Proposed Work for FY05

We will make use of available comparison and alignment tools to identify DNA regions that appear to be conserved amongst temperature regulated genes, and may be involved in their regulation. This is expected to yield a number of known and novel transcription factor binding sites and may specifically help in the identification of yet unknown virulence factors in *Yersinia*. We will also compare upregulate vs. downregulated predicted motifs and evaluate this general technique and complement it by looking at predictive capability without microarray data. We expect to implement numerous iterations of software development to improve this prediction capability. For informatically and informationally validated regulons, we will begin to design regulatory motif knockout experiments.

TOF-SIMS Measurement of Metabolites from Single Cells

Kristen S. Kulp 04-ERD-104

Abstract

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

This research will advance imaging mass spectrometry to an unexplored arena: the characterization of metabolites from an individual bacterium. This proposal will result in (1) quantitative demonstration of enhanced TOF-SIMS instrument sensitivity, (2) a reproducible sample-preparation method that renders bacterial metabolites accessible to analysis, and (3) a useful multivariate spectral pattern recognition technique that can compare metabolite production from multiple individual cells. These experiments will be the first-ever analyses of bacterial metabolites in single cells. This work will provide preliminary data to understand pathway fluxes and population interactions of environmentally stressed bacteria.

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Mission Relevance

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

FY04 Accomplishments and Results

After a late-year start, we (1) acquired a gold ion source to increase the analysis sensitivity; (2) designed and manufactured a sample preparation device to process bacterial samples; (3) completed preliminary experiments in cutting bacterial cells with a focused ion beam (a possible sample preparation method); (4) developed a spectral library of amino acid and phospholipid membrane standards to identify peaks in sample mass spectra; (5) implemented principle component analysis methods to analyze spectral data sets; and (6) successfully determined the masses responsible for the differences in closely related cell types.

Proposed Work for FY05

In FY05 we will (1) explore matrix-enhanced SIMS and alternative primary ion sources to enhance instrument sensitivity and mass range in order to study the sensitivity and fragmentation patterns of metabolite standards and cell homogenates spiked with metabolites; (2) develop physical or chemical methods of sample preparation to expose metabolites for identification, thereby improving the collection of molecular information from the cells; and (3) further refine available statistical analysis techniques to interpret spectra and monitor metabolite production, specifically by investigating sequential application of these techniques and measuring metabolite fluxes in single cells.

Publications

Knize, M. G. et al. (2004). "Analysis of single cell extracts by time-of-flight secondary ion mass spectrometry." UC Davis Cancer Center 10th Ann. Cancer Research Symp., Sacramento, CA, Oct. 20–21, 2004. UCRL-ABS-206566.

Calcium Dynamics in Human Bone

Darren J. Hillegonds 04-ERI-009

Abstract

Previous work has shown the value of ^{41}Ca accelerator mass spectrometry (AMS) in tracking bone health, primarily related to osteoporosis. This proposal broadens the ^{41}Ca assay to include impact on cancer treatment and survival. Our goal is to develop a minimally invasive, preclinical method to detect the transition from the primary cancer site to the skeleton. Such an early detection protocol could vastly improve patient quality of life and survival by allowing the use of anticancer therapies prior to significant skeletal damage. We will also develop the ability to closely monitor the progression of skeletal tumor growth,

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which could lead to significant reductions in the debilitating consequences associated with this disease.

Our deliverable will be a minimally invasive method to detect the transition from the primary cancer site to the skeleton, as well as the ability to closely monitor the progression of skeletal tumor growth. If successful, our technique will represent a significant improvement over current methods to assess calcium metabolic parameters, especially bone turnover rate.

Mission Relevance

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

FY04 Accomplishments and Results

We demonstrated that the ^{41}Ca assay provides information on Ca dynamics in healthy individuals, and we developed a predictive kinetic model for the interpretation of urinary and serum $^{41}Ca/Ca$. We also began a preclinical study using an established mouse model where human cancer cells grow in immunocompromised mouse skeletons. Our collaborative team has been expanded to include additional researchers at UC San Diego and the UC Davis Cancer Center. We gave invited talks on AMS ^{41}Ca and presented posters at scientific conferences. One of these posters was judged the best student poster at the conference and resulted in the publication of an invited paper.

Proposed Work for FY05

We plan to recruit and dose metastatic cancer patients and compare their results to the healthy volunteers already dosed. Using the results from a preclinical mouse project, we will predict the impact of skeletal metastases in a human volunteer. We expect to find significant differences between healthy and diseased individuals, and that these differences will be useful in determining the state of disease progression and the efficacy of subsequent treatment.

Publications

Hillegonds, D. J., F. Roughead, and J. S. Vogel. (2004). *Ultra low dose ^{41}Ca: Testing the detection limits of accelerator mass spectrometry-based analysis in nutrition research*. Presented at Exper. Bio. 2004. UCRL-POST-204237.

Hillegonds, D. J. and J.S. Vogel. (2004). *A new technique for measurement of small changes in bone turnover rates in individual human subjects*. Presented at the 4th Intl. Conf. Cancer-Induced Bone Diseases. UCRL-POST-202081.

Hillegonds, D. J. et al. (2004) "High throughput measurement of ^{41}Ca by accelerator mass spectrometry." *J. Assoc. Lab. Automation* **9**, 99. UCRL-POST-202162.

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

Amy L. Hiddessen 04-ERI-015

Abstract

This project aims to elucidate molecular mechanisms that regulate cellular behavior in response to chemical signals. The hierarchical process whereby hematopoietic stem cells (hsc) proliferate and/or differentiate into highly specialized blood cells exemplifies the complexity of signal-dependent cell behavior. While the functional heterogeneity in hsc is well recognized, the molecular mechanisms underlying hsc fate decisions are poorly understood. Similarly, little is known about cellular response to viral stimuli, particularly the host-cell cytoskeletal response to viral docking and entry. Subcellular labeling techniques will be used with fluorescence videomicroscopy to characterize real-time single cell response as cells are controllably exposed to stimuli in bioengineered environments.

The overall goal is to develop and apply bioengineered tools to elucidate molecular mechanisms of cellular behavior in response to chemical signals, with a focus on host-cell cytoskeletal response to pathogens and the molecular regulation of stem cell fate decisions in response to chemical cues. A new experimental capability for systematic analysis of single-cell processes will be established, while experimental data gathered could further the development of computational approaches designed to predict cellular/systems behavior. Results from this work will contribute to the development of drugs or therapies for preventing and/or treating disease.

Mission Relevance

By developing a new method for systematically examining the behavior of single cells in response to chemical/pathogenic signals, this work will contribute to biodefense applications in support of the Laboratory's national security and homeland security missions as well as its mission in solving basic scientific problems. This research will also strengthen the Laboratory's collaborative relationship with the University of California.

FY04 Accomplishments and Results

In FY04, tools needed to visualize cytoskeletal response to chemical stimuli were developed by fluorescently labeling actin filaments and microtubules in living fibroblast and epithelial cells via transfection and imaging using fluorescence microscopy. Experiments demonstrated that the cytoskeletons of these labeled cells can dynamically respond to chemicals introduced to them in artificial environments. To characterize the responses of both actin and microtubules within the same cell, cloning techniques were used to construct actin and tubulin fluorescent protein vectors with nonoverlapping emission spectra. Finally, micropatterned substrates were made that allow for directed deposition of proteins into spatially defined capture sites for single cells.

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Proposed Work for FY05

A major goal of FY05 research is to array single cells within an artificial bioengineered microenvironment (e.g., micropatterned substrates or microfabricated cell holders) to develop a method for tracking the behavior of numerous single cells, while exposing subsets of these cells to controlled levels of cytokines, siRNAs, or pathogenic peptides. Assays will be developed for delivering such chemicals to arrayed cells, and single cell responses will be analyzed using cell labeling and phase/fluorescence videomicroscopy. This approach will be used to identify or characterize, at the molecular level, the intrinsic functional heterogeneity of isolated hsc, genes and external factors which alter hsc proliferation or differentiation, and host cell morphogenesis in response to pathogenic stimuli.

Biomechanics of Spinal Fracture

John H. Kinney 04-LW-008

Abstract

Spinal fractures are the most common injuries associated with osteoporosis. We believe that most, if not all, of the tenfold decrease in the sustainable load of osteoporotic vertebrae can be explained by the age-related deterioration of trabecular architecture. We hypothesize that vertebral failure in older adults is initiated by loss of stability, or buckling. To test this hypothesis, we will use advanced imaging and computational methods. Vertebrae from the skeletal archives of the Smithsonian Institute will be imaged at the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory, and geometric, nonlinear, finite-element analyses will be used to simulate load-displacement behavior from these as-built models. A positive test of the hypothesis will influence how osteoporosis is diagnosed and treated.

If successful, the proposed efforts will demonstrate a need for geometric nonlinear analysis in simulating bone failure. The work will change the diagnostic focus from simply measuring bone mass to considering factors that influence buckling, such as the aspect ratio of the trabeculae. Not only will risk estimates improve, but it will be possible to select the best treatment strategies for an individual. Finally, a successful outcome will also provide the necessary infrastructure for as-built modeling from computed tomography images.

Mission Relevance

This simulation effort in this project has applications relevant to advanced simulation and computing in support of stockpile assessment, for which the ability to perform as-built, multiscale modeling is of the highest importance. In addition, the imaging tasks add capabilities that may prove useful for characterizing fusion-class laser targets. Most directly, the application to nonlinear materials modeling of the osteoporotic fracture is relevant to LLNL's mission in bioscience to improve human health.

FY04 Accomplishments and Results

The first high-resolution, three-dimensional (3-D) images of an entire human vertebra were obtained at the ALS. We developed an algorithm to extract and analyze the shapes of the

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trabeculae, and found that the aspect ratios of the trabecular elements increased with age. The 3-D images were meshed directly and simulated with a parallel version of the code NIKE3D on the Multiprogrammatic and Institutional Computing Capability Resource at LLNL. We found that (1) a transition from geometric linear to geometric nonlinear behavior occurs with age and (2) failure in the vertebra of a 63-yr-old male was initiated by loss of stability. This information was used as preliminary data to support our stability hypothesis. This work sets the stage for further investigations. Several papers were published in peer-reviewed journals.

Publications

Kinney, J. H. (2004). *Three-dimensional reconstruction of the human vertebra with synchrotron radiation*. UCRL-TR-206524.

Kinney, J. H. and J. S. Stolken. (2004). "Response to Keaveny." *Bone* **34**, 913. UCRL-JC-153684.

Kinney, J. H. and J. S. Stolken. (in press). "Spinal fracture in osteoporosis: Strength of stability? A new hypothesis." *Journal of Theoretical Biology*. UCRL-JRNL-206862.

Kinney, J. H. and J. S. Stolken. (2004). *The importance of geometric nonlinearity in finite-element studies of yield in trabecular bone*. UCRL-ABS-206195.

Kinney, J. H. and J. S. Stolken. (2004). "On the importance of geometric nonlinearity in finite-element simulations of trabecular bone failure." *Bone* **33**, 494. UCRL-JC-153684.

Kinney, J. H. et al. (in press). "An orientation function for trabecular bone." *Bone*. UCRL-JRNL-207238.

Nalla, R. K. et al. (in press). "Fracture in human cortical bone: Local fracture criteria and toughening mechanisms." *Journal of Biomechanics*. UCRL-JRNL-206523.

Nalla, R. K. et al. (2004). "Effect of aging on the toughness of human cortical bone." *Bone* **35**, 1240. UCRL-JRNL-207263.

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

Erik W. Draeger 04-LW-048

Abstract

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

The expected result of this work is to elucidate the catalytic mechanism of human P450 CYP1A2 oxidizing caffeine by using newly developed cutting-edge tools for advanced

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simulations. The completion of this project will also result in a powerful new computational tool that will allow research groups currently using ab initio methods to study significantly larger systems over longer time scales.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we developed a working dynamic QM/MM code, although several features must be implemented before it can be applied to the P450 system. In preparation for this, we performed pseudopotential and energy cutoff convergence tests for an iron porphine system and obtained structural properties that are in good agreement with previous quantum chemistry studies and experimental measurements. Using the water dimer as a test system, we tested other methods for modifying electron–ion QM/MM interactions to prevent the quantum electron density from unphysically localizing. Upon finding them not sufficiently general for a multipurpose tool, we began exploring more general alternatives for representing these interactions.

Proposed Work for FY05

FY05 will be devoted to carrying out the majority of the QM/MM simulations, as well as exploring further algorithmic improvements to make the QM/MM tool more flexible and efficient. We will continue coupling the ab initio molecular dynamics code GP to the large-scale classical MD code NAMD. Efficient electrostatic interactions between quantum and classical regions will be implemented using multipole expansions and covalent bonding across QM/MM boundaries using link atoms. We will perform QM/MM calculations of a porphyrin-ring model system and compare to experimental results as well as our FY04 fully ab initio results. Full dynamical calculations of the P450 enzyme system will then be performed, and the oxidation mechanism in the presence of a caffeine substrate will be studied in detail.

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

Ceslovas Venclovas 04-LW-049

Abstract

The objective of the project is to structurally characterize AAA+ proteins and complexes involved in DNA replication, recombination, and response to DNA damage. Because DNA carries all of the genetic information for life, understanding DNA metabolism is of utmost importance at the molecular level, where the AAA+ proteins play critical roles and function as protein machines. The project will apply advanced sequence-comparison methods, comparative

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modeling, and molecular dynamics simulations to characterize all 20 currently identified eukaryotic AAA+ protein families and their complexes involved in DNA management processes.

We expect this project will provide new insights into the structure, interactions, and functional mechanisms of the eukaryotic AAA+ protein families involved in DNA metabolism. The computationally obtained structural models will greatly facilitate future experimental biochemical studies.

Mission Relevance

The application of computational methods to understand some of the central processes involved in faithfully copying and maintaining genetic information supports LLNL's mission in bioscience and technology to improve human health and supports National Institute of Health and National Cancer Institute missions.

FY04 Accomplishments and Results

During FY04, we performed an initial sequence-based analysis of all known families of eukaryotic AAA+ proteins and established a system for continuous updates of the sequence and structure information that can be utilized for molecular modeling. Our initial modeling efforts focused on the RCF1, Rad17, and Ctf18 members of the AAA+ family. We conducted a phylogenetic analysis of RFC1, Ctf18, Rad17, PCNA, and the proteins of the 9-1-1 complex. We identified that amino acids Val422 and Leu423 will potentially interact with the human DNA clamp protein, PCNA and predicted that the extended C-terminus of Ctf18 could mediate an additional protein interaction between Ctf18 and the DCC1 or Ctf8 proteins.

Proposed Work for FY05

During FY05, we will test our computational predictions through several experimental methods, including examining protein–protein interactions in the clamp loader and clamp proteins through co-immunoprecipitation and surface plasmon resonance. We have initiated a collaboration with Dr. Jerard Hurwitz at the Sloan Kettering Institute, who will test our computationally designed point mutant of the Ctf18 clamp loader protein through in vivo assays of clamp loading of the PCNA clamp protein.

Publications

Venclovas, C., M. S. Kavanagh, and D. Sawicka. (2004). *The alternative clamp loader subunit Ctf18: A model for its interaction with PCNA, evolutionary conservation and relationship to RFC1 and Rad17*. Presented at the ASM Conference on DNA Repair & Mutagenesis: From Molecular Structure to Biological Consequences, Bermuda, Nov.14, 2004. UCRL-POST-207252.

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

Daniel Barsky 04-LW-069

Abstract

We propose a combined experimental and computational approach to investigate the

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interaction between DNA and a sliding-clamp protein. By encircling DNA, sliding-clamp proteins can freely slide past more than 10,000 DNA bases in less than a second. This movement defies expectations since the protein's high positive charge should create a strong electrostatic attraction with the negatively charged DNA. It is also mysterious that the clamp proteins can proceed past sizeable distortions in DNA. By single-molecule fluorescent energy transfer (FRET) experiments, analytical calculations, and molecular dynamics simulations, we will study the details of the motions of the bacterial sliding clamp along DNA, including the speed, direction, and rotation of the clamp.

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

Mission Relevance

This project is relevant to LLNL's mission in bioscience to improve human health and has possible applications in LLNL's homeland-security mission to prevent and respond to bioterrorism.

FY04 Accomplishments and Results

We expressed the pol III beta subunit (beta clamp) and labeled it with a FRET donor dye (Alexa 488); we also designed and procured a DNA oligomer labeled with a FRET-acceptor (Alexa 647) and annealed it to a long (7500-base), circular DNA plasmid. Simultaneous fluorescence of both the beta clamp and the labeled plasmid was observed. In the final months of FY04, we successfully loaded the beta clamp on the DNA plasmid as evidenced by the presence of a FRET signal. Computationally, our 2-ns molecular dynamics simulations display a widening of the inner ring of the beta clamp, suggesting greater conformational freedom observed in the crystal structure.

Proposed Work for FY05

Fluorescence correlation spectroscopy (with FRET) will be used to achieve two goals: monitoring the lifetime (time to fall off) and observing the translation speed of the beta clamp on plasmid DNA. The first goal will be attained by loading the clamp onto a FRET-enabled 45-base restricted region of the DNA and observing the FRET signal decay. The second will be achieved by annealing an additional unlabeled DNA oligomer adjacent to the labeled oligomer and observing the on/off FRET signal as the beta clamp translates between the FRET-labeled and unlabeled portions of the DNA, and by using a polymerase to complete the DNA plasmid so that the clamp can translate over the entire plasmid. A third goal will be to attach a large mass to the clamp protein and use anisotropy to measure the rotation of the clamp on the DNA.

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Protein–Protein Interaction Mapping of the Human DNA Damage Response Pathway

Joanna S. Albala 04-FS-014

Abstract

The goal of this project is to determine the feasibility of applying automated, high-throughput cloning and yeast two-hybrid analysis for protein interaction (interactome) mapping of the human DNA damage response pathway. Understanding the complex cellular signaling pathways underlying the processes of response to DNA damage will further our understanding of the molecular basis of cancer initiation. In addition, these techniques can generate scientific resources for comparative microbial proteomics and protein production and characterization for efforts such as the GtL:Genomics Program.

If successful, the project will map all the protein–protein interactions that take place at the cellular level in response to DNA damage. This mapping will identify the complex network of cellular pathways involved in damage response, which may lead to a greater understanding of the basic mechanisms underlying cancer. Future work could then optimize these methods for mapping the protein interactions in a microbial cell. As a microbial cell has far fewer proteins to measure, the entire proteome of a microbe could be mapped to identify the subcellular networks needed for single-cell function.

Mission Relevance

By providing the expertise for high-throughput cloning and high-throughput yeast two-hybrid automated analysis for comparative proteomics, this project will contribute to understanding the complexes and regulatory pathways of microbes in support of the Laboratory's mission in science to improve human health. The project will also contribute to developing microbe interactome maps in support of the Laboratory's mission in homeland security.

FY04 Accomplishments and Results

The first version of the open reading frame (ORF) of all protein-encoding human genes (the "ORFeome") was generated using a newly improved recombinational cloning approach. Leveraging the Mammalian Gene Collection resource, we successfully cloned 8,187 human ORFs, representing at least 7,354 human genes. These were assembled into the human ORFeome collection. After assessing the overall quality of this version we described the use of the human ORFeome for heterologous protein expression in two different expression systems at proteome scale. This represents a central resource for the cloning of large sets of human ORFs and will serve as the foundation for generating the human interactome network using an automated yeast two-hybrid analysis procedure to identify the interactions of various proteins.

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Direct Probing of Protein-Protein Interactions

Aleksandr Noy 04-FS-018

Abstract

This project explored the feasibility of using the single-molecule force spectroscopy (SMFS) of tethered protein systems to extract information about equilibrium protein–protein interaction potentials. We explored the applicability of two approaches for using SMFS to determine the equilibrium interaction potential characteristics in tethered ligand systems. We combined experimental studies of a cancer-marker protein–antibody system with Monte Carlo simulations of a tethered ligand unbinding. We also applied our data analysis techniques to existing atomic-force microscopy (AFM) data on DNA duplex unbinding.

Our goal was to demonstrate an ability to (1) use tethered systems to measure protein–protein interactions and (2) characterize equilibrium interaction potentials using these measurements. This project was intended to provide a roadmap for efforts in the direct characterization of protein–protein interactions, a capability that is important for many applications, from fundamental biophysical studies to high-throughput protein screening.

Mission Relevance

Protein interaction potentials contain information about specific binding and recognition and the ability to form complexes and superstructures. Direct measurement of these quantities is important for developing new binders and ligands, which is essential for efficient diagnosis and detection in support of LLNL national and homeland security mission areas of countering bioterrorism. In addition, protein characterization and information about interaction potentials is important for understanding the function of biological machines, in support of LLNL's mission of bioscience to improve human health, in particular for the research goals of the DOE's Genomics: GtL program.

FY04 Accomplishments and Results

We demonstrated the feasibility of using nonlinear Jarsynski averaging to recover equilibrium interaction potentials from AFM experiments. The reconstruction showed nearly quantitative agreement with the thermodynamic parameters. These results were published in *Applied Physics Letters*. We also used Monte Carlo simulations to analyze the applicability of this approach to tethered systems. Finally, we measured unbinding forces for the interactions of the cancer marker Muc1 and polyclonal antibody, using dynamic force spectroscopy to determine equilibrium unbinding potential width. From this, we developed a data-analysis routine to account for the nonlinear tether stiffness.

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Cell-Type-Specific Genome-Wide Expression Profiling after Laser-Capture Microdissection of Living Tissue

Francesco Marchetti 04-FS-021

Abstract

The purpose of this study is to develop and evaluate microgenomic tools for investigating genome-wide expression of very small numbers of cells isolated from whole tissues. Tissues contain numerous cell types that play varied roles in organ function and responses to endogenous and exogenous toxicants. Accurate gene-expression analysis requires the study of specific cell types in their tissue environment but without contamination from surrounding cells. This project focused on developing small sample gene-transcript analysis protocols to define genome-wide expression profiles of isolated cells and using laser-capture microdissection (LCM) to identify and isolate target cells from tissue sections, using histochemical labeling while retaining cellular messenger RNA integrity.

The technologies developed in this project are expected to contribute to advances in mechanism-based biodosimeters of exposure and the identification of molecular targets for reducing or preventing tissue damage after exposure to physical, chemical, or biological agents. This technology will have applications in characterizing host responses of specific cell-types in tissues, chemical toxicology profiles of cell-types in tissues and gene expression analyses of select microbial populations on biofilms derived from complex communities. The ability to isolate specific cell types from tissues will have wide application for expression analyses of gene-transcripts and protein as well as functional studies of other cellular biomolecules.

Mission Relevance

This study aims to develop accurate and sensitive microarray assays that reveal single-cell expression profiles, which are otherwise obscured in whole tissue samples or heterogeneous population of cells. New technologies for the analyses of single cell types within a complex cellular environment are expected to have broad applications to both mammalian and bacterial biosciences. Applications of this technology will advance LLNL missions in bioscience and technology to improve human. This work also has applications in LLNL's environmental management and national security missions.

FY04 Accomplishments and Results

In this feasibility project, we successfully isolated total RNA from small numbers of cells. Good quality RNA was consistently isolated from as few as 1000 cells, and, on one occasion from as few as 250 cells. Small-sample gene labeling protocols for DNA microarray and RT-PCR studies were developed. We also worked out a methodology for preparing sections from various mouse tissues (i.e., small intestine, kidney and testis) for LCM and performed experiments to test parameters such as medium for embedding tissues and staining procedures on the quality of RNA produced from microdissected cells. Further work is needed to refine tissue preparation protocols for LCM to improve the quality of isolated RNA.

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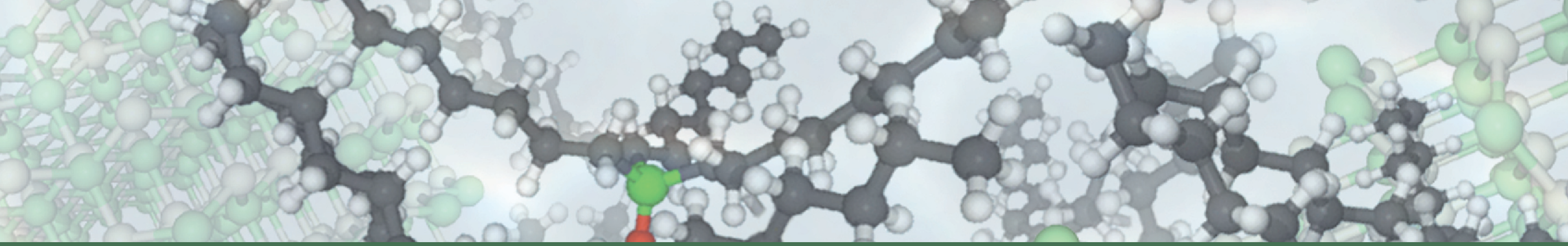
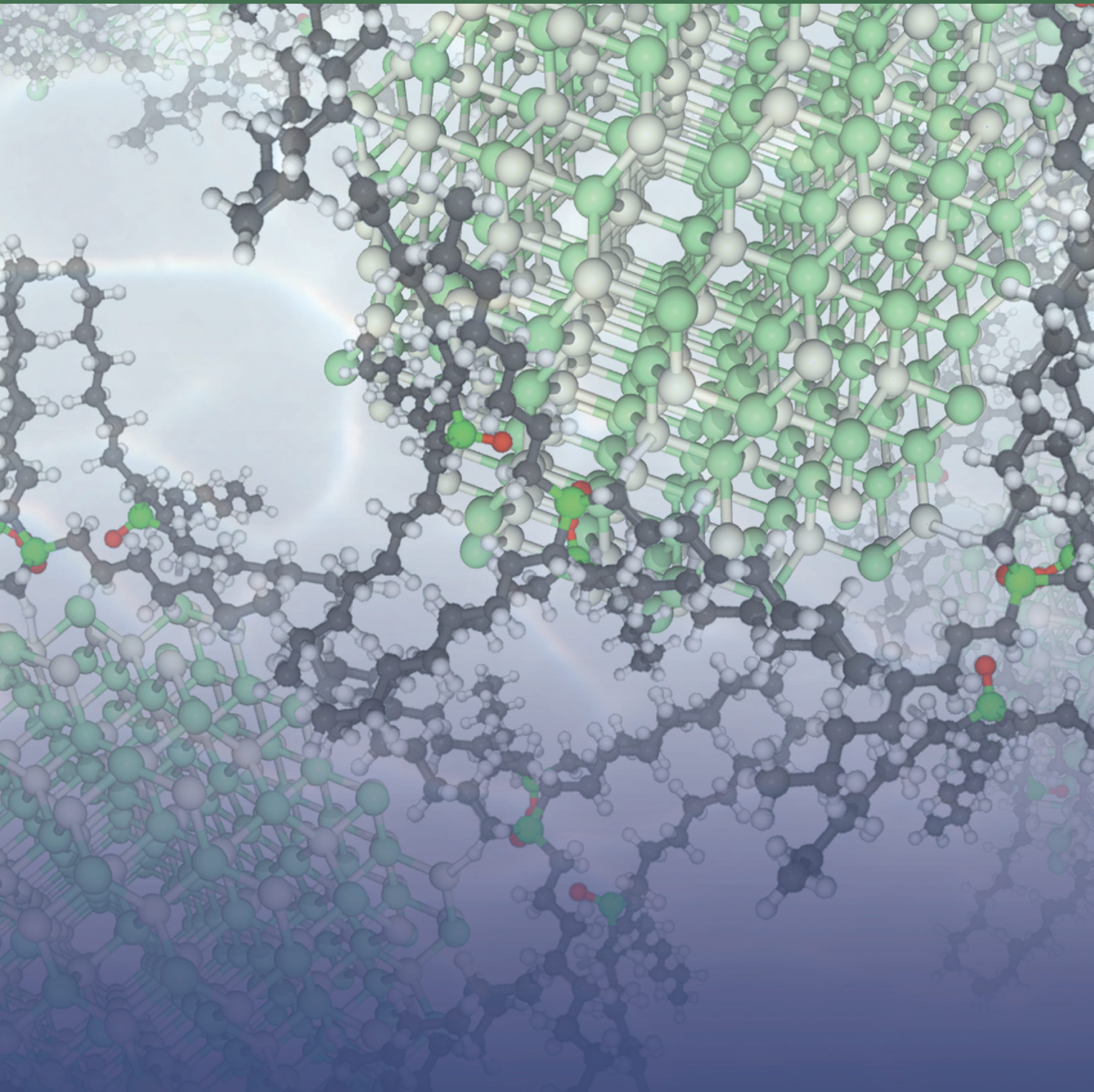


Chemistry



Modern Chemistry Techniques Applied to Metal Chelation with Medical and Environmental Applications

Mark Sutton

02-ERD-021

Abstract

This project aims to modernize and optimize the scientific approach to developing metal chelators using analytical and synthetic chemistry combined with thermodynamic and structural modeling. A screening process has been developed and used to choose Be chelators that have promise for environmental, animal, and clinical trials. In addition, similar protocols will be used to assess treatments for heavy-metal poisoning and to improved treatments for future use.

By the end of the project, we will have rigorously investigated the chemistry of chelation therapy for reducing or removing Be lung or body burden, including optimum chemistry, structure, environment, efficiency, and selectivity. A potential new environmental cleanup application using chelator-incorporated aerogel will also be developed. We also studied the chemistry of uranium in biological fluids. The results will benefit health of DOE workers, industrial workers, medical research, and environmental cleanup.

Mission Relevance

This work is a unique combination of medical toxicology and chemical science expertise and supports two LLNL/NNSA missions: bioscience to improve human health, with improved chelation therapy; and environmental management, with chelation techniques for environmental cleanup.

FY04 Accomplishments and Results

In FY2004, we again explored both medical and environmental applications. After a successful preliminary round of animal trials, we performed a full study of the effect of our chosen Be chelator molecule on Be-exposed mice. The results showed that we were able to reduce the Be body burden in mice while increasing the urinary Be excretion. A dose-response effect was observed and no visible side effects were noted. Additionally, we successfully removed Be from LLNL's Site 300 Contained Firing Facility test debris using our chosen chelator molecule. Furthermore, we studied the binding of Be to aerogel-activated carbon material, and we made some initial efforts to explore how our approach might be extended to other elements by examining the chemistry (solubility and speciation) of uranium in biological fluids.

Publications

Sutton, M. and S. R. Burastero. (in press). “Uranium(VI) solubility and speciation in simulated elemental biological fluids.” *Chem. Res. Tox*. UCRL-JRNL-20385.

Sutton, M. and S. R. Burastero. (2004). *Beryllium chemical speciation in elemental human biological fluids*. Presented at the Intl. Beryllium Research Symp., Denver, CO., January 2004. UCRL-PRES-201993.

Sutton, M. et al. (2004). *Pilot study of beryllium-specific chelators in mice*. Presented at the Intl. Beryllium Research Symp., Denver, CO., January 2004. UCRL-PRES-201995 and UCRL-ABS-201493.

Szechenyi, S. C. et al. (2004). *ICP-MS determination of environmental beryllium collected on air filters and aerogels*. Presented at the ICP-MS Winter Conference, Florida, January 2004. UCRL-POST-201469.

Local-Scale Atmospheric Reactive-Flow Simulations

Charles K. Westbrook 02-ERD-027

Abstract

This project developed an atmospheric airflow model that includes chemical reactions of gases and aerosols released into the air. The model describes chemical evolution over distances of a few kilometers of variable terrain, including obstacles such as buildings. This work modified a previously existing LLNL finite-element atmospheric-flow code, FEM3MP, already in use on massively parallel computers, by adding subroutines that simulate gas-phase chemical reactions and aerosol-surface reactions of species carried along by airflow. The emphasis is on chemicals and aerosols related to chemical and biological weapons. Effects of humidity, sunlight, temperature, and concentrations of other chemical species on reaction rates are included in the evolution submodels. Applications of this code capability have been carried out to test the combined code and illustrate its capabilities.

The project has produced a computational tool for use in a variety of nonproliferation, counterterrorist, and environmental applications. In addition, the code can provide analysis of past events and evaluate possible causes of those events, or it can be used as a predictive tool for mitigating anticipated atmospheric releases. The tool can also provide insight into the basic physics and chemistry of aerosols, which will help with efforts to mitigate the adverse consequences of atmospheric aerosol releases.

Mission Relevance

This work provides computational analysis tools for nonproliferation, counterterrorist, and environmental applications in support of LLNL missions in national security and environmental management.

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FY04 Accomplishments and Results

A final version of the code, completed in FY2004, was used to examine a hypothetical gaseous release of a chemical or airborne aerosol within the Salt Lake City area using atmospheric conditions that prevailed during the 2002 Winter Olympics to evaluate possible hazards. Influences of ambient humidity were emphasized in this analysis. The code performed its tasks very well. In addition to tracking the motion and dispersion of the airborne gas plume, in good agreement with the measurements that had been made during an actual test, the code demonstrated its capabilities of predicting the chemical evolution of the gases in the plume. Although the chemical evolution was not measured in the experiment and this capability was not therefore evaluated, the computed results were in good agreement with other laboratory-based experiments, and the combination of the two types of verifications indicates that the code is ready for field-work applications.

Transport and Biogeochemical Cycling of Iodine-129 from Nuclear Fuel Reprocessing Facilities

Jean E. Moran 02-ERD-058

Abstract

The unique biological and chemical properties of iodine—it is thermodynamically stable in multiple oxidation states and occurs in many chemical forms at the Earth's surface—make iodine isotopes excellent tracers of nuclear fuel reprocessing activities on a local or regional scale. This project has developed methods for measuring a long-lived radioactive isotope of iodine (^{129}I) released from reprocessing facilities and in downstream environmental reservoirs. These data will be used to assess the physical parameters that govern iodine transport and cycling.

We have developed capabilities to make fast measurements of ^{129}I and stable iodine in natural samples such as rainwater, river water, soil and groundwater. The results of these analyses were used to provide the scientific basis for predicting the transport and biological uptake of ^{129}I from releases from fuel-reprocessing facilities. It is hoped that these transport models will aide in unambiguous identification of its source.

Mission Relevance

The study of ^{129}I movement in the environment supports the Laboratory's counterproliferation mission by providing a tool for detecting potential proliferation-related activities, especially nuclear fuel reprocessing. The project also supports the Laboratory's environmental assessment and restoration mission by providing a capability to determine the likely fate of iodine at various DOE sites.

FY04 Accomplishments and Results

Benchtop soil-column experiments were used to help predict the environmental distribution of iodine. Environmental samples from affected areas, such as the Hanford Reservation and the Nevada Test Site, were analyzed for iodine speciation, properties affecting transport of different iodine species, and the ratio of ^{129}I to ^{127}I. A comparison

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between plasma-source mass spectrometry and accelerator mass spectrometry (AMS) techniques was carried out and showed that AMS allows much smaller samples and a greater concentration range for the tracer.

Publications

Hu, Q. et al. (in press). "Sorption and transport of iodine species in sediments from the Savannah River and Hanford sites." *J. of Contaminant Hydrology*. UCRL-JRNL-204421.

Hu, Q. and J. E. Moran. (in press). "Simultaneous analyses and applications of multiple fluorobenzoate and halide tracers in hydrologic studies." *Hydrological Processes*. UCRL-JRNL-202001.

Hu, Q., J. E. Moran, and P. Zhao. (2004). *Transport of iodine species in the terrestrial environment*. UCRL-CONF-200061.

Exploring the Linkage between Impurities and Optical Properties in Rapid Growth of Crystals

Ruth A. Hawley-Fedder 02-ERD-070

Abstract

LLNL has an international reputation in the solution growth of large potassium dihydrogen phosphate (KDP) crystals. Impurities are known to play a key role in both growth habit and final material properties of a crystal. Understanding impurity effects is critical to the development of KDP crystals that meet the size and quality requirements for advanced laser applications. The project will investigate impurity effects on KDP crystal growth using a multiscale experimental and simulation approach that includes atomic-force microscopy (AFM), small-scale (20-L) and large-scale (1000-L) growth runs, and kinetic Monte Carlo (KMC) and molecular modeling. In this way we plan to link nanoscale understanding of impurity–surface interactions to experimental observations of growth rate, growth morphology, and post-growth structure and chemistry.

Results from this project will further the fundamental science underlying crystal growth from solution, and enable crystals to be grown rapidly with the desired material and optical properties for a variety of applications.

Mission Relevance

Developing the scientific foundation and materials research for a rapid-growth system that can produce a variety of high-optical-quality crystals will benefit the broad array of efforts to develop crystalline materials for laser-system applications, optical components, and photonics applications. These areas of research are particularly important to the Laboratory's stockpile stewardship mission.

FY04 Accomplishments and Results

ignificant progress was made in understanding the role of Al in KDP crystal growth. We used AFM to develop a quantitative understanding of the consequences of Al incorporation on step morphology and velocity. We upgraded our KMC modeling to include Al effects on

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step motion, yielding results agree well with the AFM observations. Molecular modeling was also expanded to include Al incorporation. The Al^{3+} distribution coefficient was determined as a function of temperature using 20-L and 1000-L growth runs. We explored the relationship between Al concentration and inclusion formation, and investigated the effects of ethylenedinitrilotetracetic acid on the incorporation of Al. We successfully used Al doping to control aspect ratio and relative growth rates in 1000-L growth runs.

Publications

Salter, A., A. Wierzbicki, and T. A. Land. (in press). "Ab initio study of Al(III) adsorption on stepped {100} surfaces of KDP crystals." *Structural Chemistry*. UCRL-JC-203527.

Salter, A., A. Wierzbicki, and T. A. Land. (2004). "Ab initio studies on stepped {100} surfaces of KDP." *Int'l J. Quantum Chem*. **100**, 740. UCRL-JC-155816.

Thomas, T. N. et al. (2004). "Emergence of supersteps on KH_2PO_4 crystal surfaces." *Physical Review Letters* **92**, 216103. UCRL-JC-152310.

Thomas, T. N. et al. (2004). "AFM investigation of step kinetics and hillock morphology of the {100} face of KDP." *Journal of Crystal Growth* **260**, 566. UCRL-JC-152309.

Thomas, T. N. et al. (2004). In situ atomic force microscopy investigation of the {100} face of KH_2PO_4 in the presence of Fe(III), Al(III), and Cr(III)." *Langmuir* **20**, 7643. UCRL-ABS-202287.

Thomas, T. N. et al. (2004). "The growth morphology of the {100} surface of KDP (Archerite) on the molecular scale." *Journal of Colloid and Interface Science* **280**, 18. UCRL-JC-205497.

Environmental Fate of Organo-Phosphorus Compounds Related to Chemical Weapons

M. Lee Davisson 03-ERD-022

Abstract

This work investigates the behavior of the nerve agent VX and its manufacturing residues in environmental media. Because existing experimental data are limited, the persistence and degradation pathways of these compounds cannot be predicted at environmentally relevant conditions. However, the fate of VX is an important component in threat assessment, civilian response, and counterterrorism. This project strives to determine underlying chemical mechanisms controlling the persistence of VX and related chemicals in water and soil media. Controlled experiments will use the combined analytical capabilities of the Forensic Science Center and other laboratories within LLNL to determine rates of chemical reactions responsible for the destruction of VX.

We anticipate gaining data that characterize the environmental behavior of VX and its precursors and byproducts. These data will provide a quantitative basis for predicting VX behavior under a variety of environmental conditions, including the determination of (1) preferential sites of sorption and signature persistence, (2) environmental conditions that promote VX persistence at toxic levels, and (3) conditions that form persistent toxic VX byproducts. We will produce papers for publication in peer-reviewed journals, and we anticipate establishing a unique capability for research on dilute, live chemical-weapon agent transport and fate.

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Mission Relevance

This work addresses a knowledge gap in the LLNL mission areas of counterterrorism and homeland security by addressing the serious problem of the proliferation of weapons of mass destruction. This work will advance our understanding of the signatures of chemical weapons manufacturing and provide basic understanding of how nerve agents persist in the environment after release.

FY04 Accomplishments and Results

We (1) developed a new liquid chromatography–mass spectrometry in environmental media method that identifies and quantifies VX and its hydrolysis products in environmental media; (2) derived aqueous-mineral sorption coefficients, VX degradation pathways, and half-lives of VX under variable pH, ionic strength, initial concentration, and mineral/organic matter ratios; (3) quantified iron-oxide catalysis of VX under dehydrated conditions; and, most importantly, (4) demonstrated that the first-order hydrolysis rates of VX are strongly dependent on the type of solution buffer, also deriving degradation rate predictions that are improved over previously available models.

Publications

Love, A. H. et al. (2004). "Investigating the affinities and persistence of VX nerve agent." *Chemosphere* **57**, 1257. UCRL-JRNL-203337.

Love, A. H. et al. (2004). "Quantification of VX nerve agent and its degradation products using HPLC-ESI-MS." 52nd Conf. Am. Soc. of Mass Spectrometry, May 2004, Nashville, TN. UCRL-POST-204205.

Development of a "Virtual Crystallizer"

Teresa A. Land 03-ERD-051

Abstract

We have developed a computer model, the "virtual crystallizer," capable of predicting growth from a 1-cm-size seed to a large, 60-cm crystal utilizing actual crystallizer system conditions. The capability to predict how crystals grow is a valuable tool for any industry involved in commercialized crystal production. Our model utilizes equations governing the fundamental physics and chemistry of crystal growth along with mass transfer to crystal faces determined using a computational fluid dynamics (CFD) model into a large-scale crystal growth model that calculates growth rates for the individual faces and allows them to evolve in time to form a 3-D crystal. The model will be validated with actual potassium dihydrogen phosphate (KPD) crystals grown from 1 to 60 cm.

Expected results include an understanding of the sensitivity of growth to various parameters, a method for optimizing growth conditions, and the ability to grow reproducibly a crystal with desired dimensions and properties. The immediate impact of our investigation will be to increase the quality and yield of rapid-growth KDP crystals. These crystals are used for

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frequency conversion and fast optical switches in large-aperture inertial-confinement-fusion (ICF) laser systems worldwide.

Mission Relevance

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

FY04 Accomplishments and Results

The computational fluid dynamics (CFD) modeling and experimental effort at NASA Ames have focused on four test cases: 30cm and 60cm crystals each at two rotations. The CFD model has been modified to use a heat transfer equation, and results are now reasonably consistent with experiment and growth data. CFD simulation and experiment predict, by heat transfer analogy, that approximately 60% of the bulk supersaturation is present at the crystal surface due to mass transport limitations. A working 3-D version of the virtual crystallizer growth model that includes effects of both mass transfer resistance and simplified surface growth kinetics for prismatic and pyramidal faces of KDP crystals has been developed and used to predict several growth runs a priori.

Proposed Work for FY05

In FY05 we will focus on performing additional experiments to elucidate kinetic versus mass transfer contributions to crystal growth and incorporate these results into the virtual crystallizer growth model. The experimental effort has three major components designed to determine growth kinetics and mass transfer coefficients: NASA Ames half-scale heat/mass transfer experiments, large-scale crystal growth runs, flow cell and 20-L-tank experiments. A 1000-L growth station will be modified to grow a crystal at constant temperature in order to more easily sort out the mass transfer as a function of crystal size. The virtual crystallizer model will be expanded to include growth profiles for all eight crystal faces and include the capability to project final crystal shape from initial growth data.

Publications

Dylla-Spears, R. et al. (2004). *Mass transport modeling of 1000 L KDP rapid crystal growth system*. Presented at the AACGW, June 19, 2004. UCRL-POST-204559.

Laser-Initiated Nanoscale Molecularly Imprinted Polymers

Bradley R. Hart 03-LW-047

Abstract

We propose to synthesize nanoscale molecularly imprinted polymers (MIPs) that will benefit applications such as chemical and biological sensing and microscale separations

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and catalysis. This work couples recent advances in the design, synthesis, and evaluation of MIPs with LLNL's expertise in lasers, microscopy, and advanced materials synthesis, and will establish an entirely new and versatile route to practical MIP systems. This will be accomplished by using a focused laser source to pattern polymer features on a surface covered with a solution containing polymerizable monomers, a photoinitiator, and template molecules. We expect that this work will overcome the limitations associated with traditional MIP systems and establish a new and versatile route to practical MIP systems. .

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we refined our ability to prepare polymer features while controlling size, shape, and position on the surface. Additionally, we can now form polymers of differing compositions on the same surface while maintaining the spatial arrangement. We also demonstrated that the materials can be layered, one on top of the other, to control polymer composition (as well as size and shape) in all three dimensions. As an alternative detection scheme for nanoscale MIPs, we experimentally evaluated the use of surface-enhanced Raman spectroscopy (SERS), but complications were encountered relating to the compatibility of the SERS particles with the polymer matrix.

Proposed Work for FY05

We will examine the layering of a thin, inert polymer on top of the selective material and the use of polymer-bound fluorescent probes, as methods to prevent much of the nonspecific binding on the surface of the imprinted material. We will also investigate an approach recently reported in the literature that suggests solutions to problems with the SERS-based detection for bulk MIPs systems, which involve surface functionalization of the SERS particles before polymerization.

Dynamic Combinatorial Libraries for Target-Driven Ligand Development

Julie Perkins 04-ERD-007

Abstract

The objective of this project is to develop disulfide- and hydrazone-based dynamic combinatorial chemistry that enables the formation of ligands specific to a protein by using the target protein as a template and a small set of ligand building blocks. Ligands specific to a protein would have numerous uses in detecting biological agents in homeland-security

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applications. The use of antibodies is the current state of the art for detecting proteins, but more robust and reliable methods of detection are needed. The success of this project will enable the generation of ligands for a protein even before that protein is identified. Our approach combines thermodynamically controlled chemistry with advanced characterization by mass spectrometry.

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

Mission Relevance

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

FY04 Accomplishments and Results

The building blocks for a disulfide library for the C fragment of tetanus were synthesized and used in dynamic combinatorial library (DCL) experiments. Tetanus is an ideal starting target protein because it is well characterized, and ligands that bind to tetanus have previously been identified. Initial DCLs with the synthesized disulfide building blocks show that the starting disulfides exchange with one another to give all possible products in a one-pot reaction under the conditions in which tetanus toxin is stable and soluble. Initial experiments to determine the effect of adding the target protein (tetanus toxin C fragment) to the DCL are ongoing. Preliminary results show that the protein affects the position of the dislufide exchange equilibrium.

Proposed Work for FY05

A major goal for FY05 is the full development of a dialysis method combined with mass spectrometry. This will enable the constant monitoring of a DCL when a protein target is part of the reaction mixture. We will also optimize conditions to possibly amplify the amount of ligand formed in a single reaction, and also investigate the suitability of hydrazone formation for ligand development. If this is successful, a number of building blocks with suitable functionality will be synthesized, and DCLs from a larger number of building blocks will be created and investigated.

Designer Nanocellular Materials for Targets and Other Physics Applications

Joe H. Satcher

04-ERD-022

Abstract

Despite their importance for a myriad of applications, sol-gel materials are underutilized due to the difficulty of reliably controlling and predicting their final properties. Successfully demonstrating a fundamental understanding of and control over the processes that influence materials properties would revolutionize the synthesis of sol-gels and expand their potential applications. We propose to develop a predictive synthetic capability for preparing a new class of sol-gel materials with tailored physical properties. Using nuclear magnetic resonance (NMR) as an "in process" characterization technique, we will develop a fundamental understanding of the mechanisms associated with the sol-gel process and establish relationships between initial conditions and final structure.

This work will enable the template-assisted preparation of a new class of nanocellular metal foams with cell sizes and densities not attainable by conventional methods. This project will establish a new foaming technology for synthesizing novel nanocellular metal foams with many applications, including sensors, waste remediation, catalysis, separations, ceramics, fuel cells, and nanocomposites.

Mission Relevance

If successful, the proposed sol-gel design model will have applications in the design and production of high-energy-density (HED) physics targets. Because current techniques do not allow for the preparation of foams that meet all of the current and projected compositional and mechanical requirements of HED physics experiments, the proposed sol-gel design model will benefit the Laboratory's stockpile stewardship mission.

FY04 Accomplishments and Results

We (1) developed an Al_2O_3 foam that produces two materials differing in both morphology and mechanical properties; (2) used solid-state NMR to follow the kinetics of nucleation and growth in both systems; (3) characterized speciation changes occurring in the gel state of the reaction process; (4) produced, by dealloying, gold foams with a relative density of 40% to 30%; (5) characterized the gold foams by transmission electron microscopy, x-ray diffraction, and nanoindentation; (6) prepared copper–titanium alloy foams through melt infusion in carbonized resorcinol-formaldehyde (CRF) templates; (7) prepared Ni foams through electroless deposition onto CRF templates; (8) synthesized organic foams with different cell sizes; and (8) characterized the mechanical properties of these templates and the replicate metal foams.

Publications

Baumann, T. F. et al. (in press). "Synthesis and structural characterization of low-density aluminum oxide aerogel monoliths." *Chemistry of Materials*. UCRL-JRNL-204928.

Baumann, T. F. et al. (2004). *Mechanistic studies of the formation of aluminas from sol-gel methods by multinuclear NMR*. 17th European Experimental NMR Conference, Lille, France, Sept. 2004. UCRL-POST-206236.

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Biener, J. (2005). "Nanoporous Au: a high-yield-strength material." *Journal of Applied Physics* **97**, 024301. UCRL-JRNL-204115.

Hodge, A. M. et al. (2004). *Ductile/brittle behavior of nanoporous Au.* AVS Nanoscience and Bionanoscience Res. Mtg., Stanford, CA, Sept. 23, 2004. UCRL-POST-206680.

XChem

Ricky Chau 04-ERD-024

Abstract

This project addresses the fundamental nature of dynamic chemical processes at high temperatures, very high pressures, and short timescales during the passage of a strong shock wave. By exploiting the properties of a steady shock wave produced by a gas gun or laser, we can enhance our spatial and temporal resolution using optical spectroscopy to probe the development of fast chemical processes in a simple molecular fluid. Benzene, cyclohexane, and nitrobenzene will be the prototype systems for this study, which will focus on understanding several fundamental processes such as carbon formation, dissociation, and the formation of intermediate species. Experimental data at the 10-ps timescale from this project will be compared to computational results.

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

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we completed a hydrodynamic laser-target design that gives a sufficiently flat and steady shock wave at 20 GPa for a 5-ns, 10-J laser pulse. Engineering of the laser target was completed, and manufacturing of the targets was nearly completed. We also began preliminary optical-absorption experiments on a two-stage gas gun to test diagnostics and produce preliminary data. These experiments were performed on benzene at two different pressures using both a streak camera for subnanosecond time resolution and a photomultiplier tube for longer times. We found that increasing pressure and temperature dramatically increases the rate of molecular dissociation. At 17.7 GPa, for example, the sample became opaque within 10 ns.

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Proposed Work for FY05

In FY05, we will continue the preliminary absorption experiments on the gas guns on benzene and cyclohexane to compare the effects of carbon bonding on dissociation. We will then move the experiment to LLNL's COMET laser for initial laser experiments. We will also set up coherent anti-Stokes Raman spectroscopy (CARS) experiments. In the second half of FY05, we expect to conduct full laser experiments on LLNL's JANUS laser and carry out preliminary CARS experiments on gas guns.

Bioforensics: Attribution of Biological Weapons Agents by NanoSIMS

Peter K. Weber

04-ERD-039

Abstract

The objective of this project is to use nanoscale secondary ion mass spectroscopy (NanoSIMS) analyses to identify elemental and isotopic markers in spore material that can be used for attribution of biological weapons, including markers that allow the identification of the production method, the location of production, and the date of production. The NanoSIMS is ideally suited to the characterization of biological weapons agents because it combines high spatial resolution with high sensitivity. NanoSIMS capabilities allow us to work with very small amounts of evidentiary material, to analyze spores separate from weaponizing materials and environmental contaminants, and to characterize structures within individual spores.

This project has established a new method for characterizing biological weapons that attracted funding from national law enforcement agencies. The links that we are working to establish between chemical signatures and production will help law enforcement narrow their investigations, address critical attribution questions about the identity of perpetrators, and perhaps assess the likelihood of a follow-on attack.

Mission Relevance

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

FY04 Accomplishments and Results

In the first year, we established sample-preparation methodologies, initiated spore characterization by NanoSIMS, and began experiments to test our ideas for a new method to date spores. Our most significant result to date is the discovery of a mechanism by which elemental diffusion can occur in spores on timescales relevant to biological weapon production and release (hours to years). Using deuterated water (D_2O), we demonstrated that water is taken up by spores and either retained or exchanged with water in the spore. This result suggests a mechanism by which short-time-scale diffusion into and out of the spore takes place along hydration pathways.

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Proposed Work for FY05

In FY05, we are initiating experiments to constrain the controls on trace-element signatures in spores. We will examine both the bulk composition and the zonation of trace elements; study elemental diffusion into spores, starting with light elements such as Li and F that can potentially be used as the shortest period chronometers; and then progress to heavier elements that could serve as longer period chronometers. These experiments will also elucidate our trace-element signature studies. In addition, we plan to work with spores produced from isotopically distinct media to measure spore isotopic composition. Based on this work, we will begin to establish a flow chart of analyses for the attribution of spores.

Publications

Wainwright, M. et al. (2004). "Studies on bacteria-like particles sampled from the stratosphere." *Aeorobiologia* **20**, 237. UCRL-JRNL-205583.

Ionization Chemistry of High-Temperature Molecular Fluids

Laurence E. Fried 04-ERD-069

Abstract

This research studies the phase diagram of simple polar molecular fluids (H_2O, NH_3) and mixtures under conditions of extreme pressure (>10 GPa) and temperature (>1000 K). Under these extreme conditions, the neutral molecular form of matter transforms to a phase dominated by ions, the boundaries of which are unknown. Tightly coupled modeling and experiments will be performed to determine the phase diagram of simple fluids in this region, and proposed novel lattice states involving mobile H atoms will be investigated. For the first time, acid–base chemistry under extreme conditions will be investigated in a diamond anvil cell, and the chemistry of synthetic planetary interiors will be addressed by using molecular mixtures.

We expect this project will lead to the first understanding of ionization and acid–base chemistry under extreme conditions. To date, our experimental and computational results indicate the existence of nonmolecular phases of water over 50 GPa. This project may discover qualitatively new superionic phases of water and ammonia that have been predicted previously. By mixing water and ammonia under extreme pressure and temperature, we expect to understand how fundamental chemical notions such as acid–base chemistry are modified in extreme environments. This project will increase our understanding of planetary interiors and will guide the modeling of high-explosive detonations.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we obtained direct evidence of the existence of superionic water. First, theoretical calculations determined that water undergoes a phase transition from a molecular to an ionic fluid phase at 50 GPa. This result was then confirmed experimentally. In Raman experiments, water was laser heated to 1500 K, and the resultant spectra showed a gradual broadening of the OH stretch mode at pressures above 30 GPa, followed by a sudden and complete loss of the OH stretch at 59 GPa, which indicates a phase transition to a nonmolecular phase. Our calculations predicted a previously unknown superionic glassy phase at 80 GPa. The close agreement between experiments and theory up to 59 GPa provides strong evidence for the existence of glassy superionic water.

Proposed Work for FY05

Our plan for FY05 is to fully investigate superionic water. We propose a direct measurement of the Raman spectrum at pressures over 80 GPa and temperatures of ~1500 K to verify experimentally the transition into a phase with fixed oxygen atoms. Two possibilities exist for such a phase: (1) an amorphous phase with disordered oxygen atoms and (2) a crystalline phase. Raman spectra combined with x-ray synchrotron measurements would resolve this issue. We will also study ammonia to determine how common ionic and superionic phases are in the studied temperature and pressure range (2000 K, 20–60 GPa). To address this issue, we plan to conduct computations that calculate Raman intensities directly rather than vibrational densities of states.

Publications

Goldman, N. et al. (2004). *Covalency in the superionic phase of water.* UCRL-JRNL-205696.

Scott, H.P. et al. (2004). "Generation of methane in the Earth's mantle: In situ high pressure–temperature measurements of carbonate reduction." *Proc. Nat. Acad. Sci. (USA)* **101**, 14023. UCRL-JRNL-206515.

Is the Island of Stability Centered at $Z = 114$?

Joshua B. Patin 04-ERD-085

Abstract

We propose to investigate the western edge of the nuclear Island of Stability to determine whether or not the closed proton shell is located at Z = 114. We will accomplish this via three different paths: (1) irradiating ^{243}Am with ^{48}Ca ions to produce enough events to determine the structure of the alpha decay spectrum of the recently discovered element 115 isotope 287, (2) irradiating ^{249}Cf with ^{48}Ca ions to positively identify the decay properties of the as yet undiscovered element 118 isotope 294, and (3) participating in experiments to positively identify the atomic mass of an element 114 isotope through the use of Mass Analyzer of Super

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Heavy Atoms (MASHA), a new isotope separator. This project will be carried out in collaboration with the Joint Institute for Nuclear Research in Dubna, Russia.

If successful, we will not only discover yet another new element (element 118), but also better establish the decay properties of the nuclides of elements 115 and 113 and determine whether the Island of Stability is centered at 114 protons or at a higher atomic number. Such results would help extend and improve theoretical models used to calculate the decay properties and nuclear shapes of the heaviest elements. Stability against dynamic deformation of these nuclei provides fundamental information on the fission process. Fabrication techniques we will develop for the plutonium ceramic target that is the heart of the apparatus will contribute to target-preparation technology for the primary target of the Rare Isotope Accelerator fragment separator.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we shipped the ^{249}Cf target material to Russia, participated in preliminary chemistry experiments in which we isolated ^{268}Db, a decay descendant of element 115 isotope 288. We completed additional supporting radiochemical separations of ^{268}Db, measuring its half life and fission decay energy. We also planned future experiments with our Russian colleagues.

Proposed Work for FY05

In FY05, we will (1) complete planning of the ^{249}Cf + ^{48}Ca experiment; (2) carry out preliminary bombardments to check and calibrate equipment, and continue to analyze experimental data that become available; (3) conduct testing with the beam at MASHA (in Dubna) with a target fabricated out of ^{242}Pu or ^{239}Pu at LLNL; and (4) complete a ^{48}Ca + ^{243}Am run, including data analysis. The ^{249}Cf + ^{48}Ca experiment, with the potential discovery of element 118, will be performed late in FY05 or in FY06.

Ion Mobility Spectrometry

W. Henry Benner 04-FS-004

Abstract

This project seeks to develop a way to improve ion mobility spectrometers by devising time-of-flight drift tubes with increased resolution, defined as the distribution of the arrival time of ions at the drift-tube end. If this design proves feasible, improved resolution would allow more reliable differentiation of explosives from interfering substances, detection of

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individual peptides in high-throughput medical diagnostic tests, and more accurate determination of aerosol particle-size distributions.

To determine the factors that limit resolution, we will couple software codes that model the influence of fluid flow and electrostatic forces on ions. Next, we will work to overcome the limitations of previous designs, principally by optimizing the shape of electrostatic fields inside a drift tube.

We propose to create a design for an ion mobility spectrometer that improves drift-tube resolution. Resolution will be improved by designing a two-stage drift tube. A countercurrent flow of gas in the first stage will preclude interfering substances from entering the second stage, a static-gas region in which ion drift time is measured. We expect to use computation fluid dynamics to determine how to reduce swirling of the gas so that ion trajectories are not compromised. The electrostatic model will be used to design an electrostatic field that transports ions from an electrospray source and guides them to the center of the drift tube. The fluid model will be used to calculate flow streamlines in the drift tube so that gas velocity can be added to an electrostatic model of ion motion.

Mission Relevance

Improved ion mobility instrumentation will increase the reliability of explosives screening at airports and enable rapid medical surveillance, particularly the determination of exposure to biological agents, an important aspect of homeland security. The successful outcome of this research will therefore directly impact the Laboratory's missions in national security and bioscience to improve human health by providing more reliable instrumentation for such applications.

FY04 Accomplishments and Results

We met our FY04 goals to (1) evaluate how to combine data fluid-flow and electrostatic models and (2) use the results to guide the design of an improved spectrometer. We also identified design criticalities that need to be met for operating a drift tube at 1 Torr (i.e., atmospheric pressure). Two commercially available software packages for modeling ion motion in gas were evaluated. One was determined to be appropriate for use in designing a drift tube. Several new polymeric materials were investigated and found to be suitable substitutes for ceramic components in a drift tube. This project also allowed us to identify specific measurement applications that could benefit from drift tube technology.

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

Julio A. Camarero 04-FS-009

Abstract

The objective of this project is to develop a new entropically activated ligation method based on the naturally occurring protein trans-splicing process. This method will be used to generate spatially addressable arrays of multiple protein components by standard photolithographic techniques. Protein arrays are the best tool for rapid, in parallel analysis of

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whole proteomes, thus helping to identify all the protein–protein interactions in a living cell. The protein trans-splicing process will allow us to create a truly generic and highly efficient method for the covalent attachment of a proteins through its C-terminus to any solid support.

This feasibility study is expected to produce several important results: (1) a generic and highly efficient method for attaching any protein to solid supports through its C-terminus; (2) a highly selective process not requiring the purification of the proteins to be immobilized; and (3) chemical modification of the C-intein polypeptide with a photocleavable protecting group allowing use of standard photolithographic techniques for the creation of protein chips. The combination of in vitro expression systems, photolithographic techniques and protein attachment by protein trans-splicing developed in this feasibility study has the potential to revolutionize the proteomics field by making possible the rapid and efficient creation of protein chips with thousands of different protein components. Such protein arrays can be used as powerful biosensors.

Mission Relevance

We believe that the development of new methods for the rapid and efficient creation of protein chips will lead to basic science and technology breakthroughs in the biosciences, including genomics and proteomics, disease susceptibility, and health-care technologies. It will provide a valuable new technical tool for DOE's GtL Program and will be an enabling technology for biosensors that support Laboratory missions in national and homeland security.

FY04 Accomplishments and Results

In FY04, we accomplished chemical synthesis of the C-intein polypeptide; demonstrated cloning, expression, and purification of the N-intein polypeptide as a maltose binding protein (MBP); and successfully labeled MBP with a small model peptide by using protein trans-splicing in solution. Work continues, under follow-on funding, to reproduce the same results on a solid surface.

Publications

Camarero, J. A., Y. Kwon, and M. A. Coleman. (2004). "Chemoselective attachment of biologically active proteins to surfaces by expressed protein ligation and its application for protein chip fabrication." *J. Amer. Chem. Soc.* **126** (45), 14730-14731. UCRL-JRNL-205482.

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Rapid Generation of a Nanocrystal-Labeled Peptide Library for Specific Identification of the Bacterium Clostridium botulinum

Beng-Hoei Tok 04-FS-015

Abstract

Botulinum toxins are extremely potent neurotoxins produced by the *Clostridium* genus of anaerobic bacteria. In response to increased concerns about the threat of bioterrorism, an ultrasensitive method is needed to rapidly detect the bacterium *C. botulinum*. In this project, we propose to develop a rapid, in vivo biosensing technology that selects for peptide-affinty ligands that recognize protein signatures unique to *C. botulinum*. This biosensing technology will identify peptide sequences, optimized via screening randomized peptide libraries attached to nanocrystals, to both penetrate and exhibit specific binding of unique protein fragments to the bacterium. Then, the optimized nanocrystal-conjugated peptide sequence can easily be identified using fluorescence-activated cell sorting technology.

Optimized peptide sequences that specifically recognize the target bacterium, *C. botulinum*, will lead to a highly efficient and portable biosensing device that rapidly identifies the deadly pathogenic organism. Furthermore, due to the presence of the conjugated quantum dots, dynamics of the localization pathway into the bacteria cell's cytoplasm or nuclear compartment will also be investigated through fluorescence microscopy and immunohistochemistry. Generating a chemically diverse nanocrystal-labeled peptide library could also be extended to identify other significant pathogenic targets, e.g. *Bacillus anthracis*. The ability to provide advance warning to personnel in an area contaminated with lethal bacteria is the ultimate objective of this work.

Mission Relevance

By using "tailor-made" nanoparticle-derived peptide affinity binders to rapidly detect deadly pathogens, this project has applications in countering bioterrorism for the Laboratory's missions in national security and homeland security.

FY04 Accomplishments and Results

In FY04, we had two major accomplishments: (1) peptide library synthesis and (2) a fluorescent-based assay for screening peptide libraries. We completed the synthesis of four peptide libraries using the "one-bead/one-compound" approach, in which each library consists of approximately 1.8 to 2.5 million unique peptide sequences immobilized on Tentagel resins. Each of the libraries was subsequently interrogated with the binding domain of *Clotridium* tetanus. Applying the calorimetric assay approach, unique peptide ligands have been isolated and sequenced through a peptide sequencer. We successfully employed a fluorescent microscopy-base assay, modified from a previously reported approach by Schreiber and coworkers, to enable more efficient screening of our large collection of peptide-coupled library resin beads.

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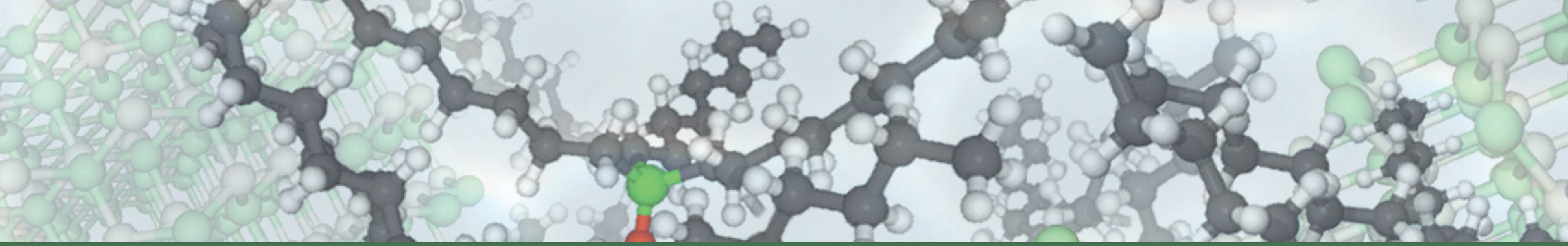
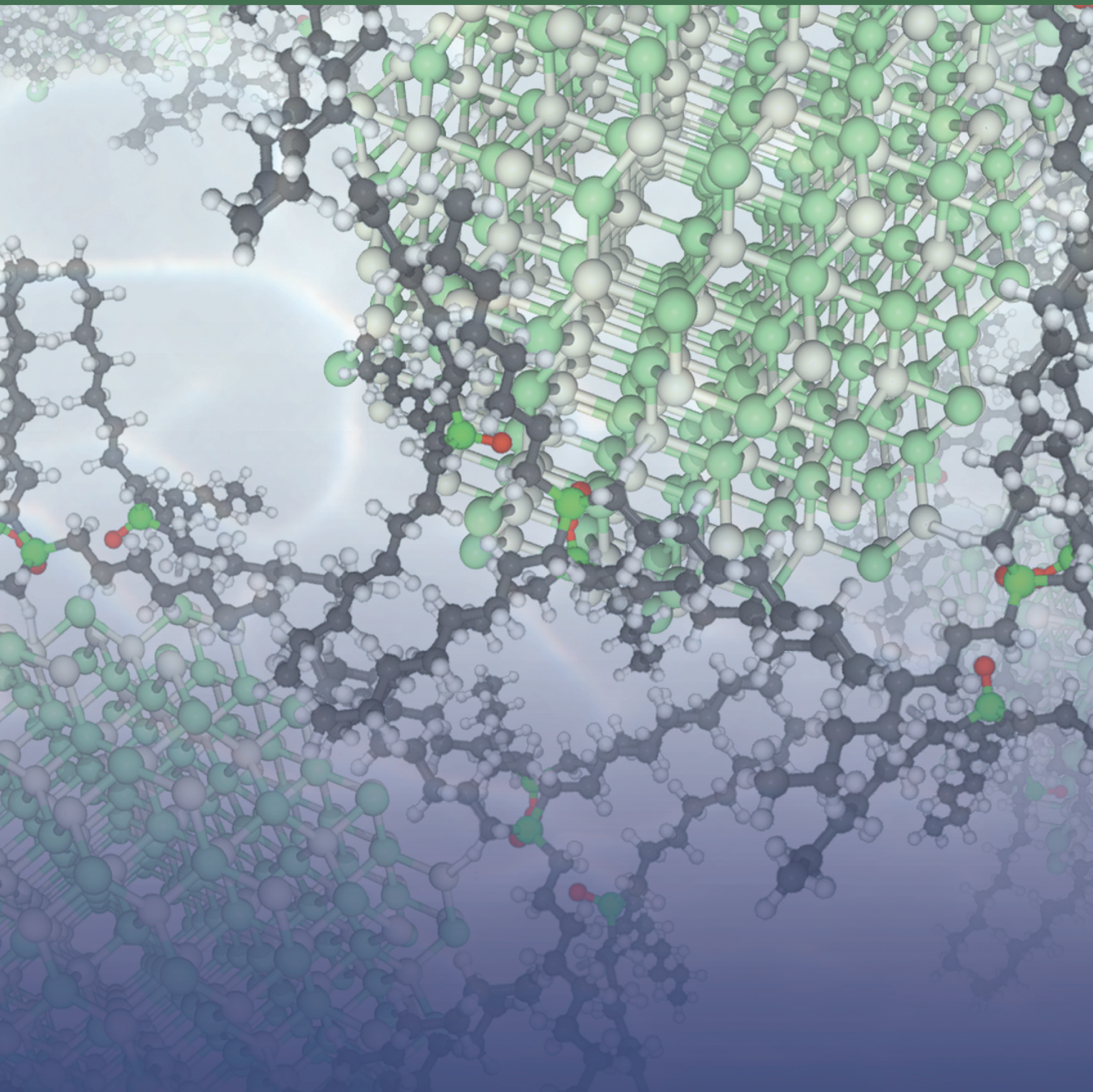


Earth and Space Sciences



Earth and Space Sciences

Direct Imaging of Warm Extrasolar Planets

Bruce A. Macintosh

02-ERI-005

Abstract

More than 100 giant planets orbiting other stars have been detected via spectroscopy. However, this technology can detect only planets located in the inner part of their systems. Such systems have overturned models of planet formation but have no room for habitable, Earth-like worlds. By contrast, direct imaging of the planets is sensitive to planets in wide orbits. However, imaging is challenging: mature planets are a billion times fainter than stars. To overcome this difficulty, this project used adaptive optics (AO) to look for infrared (IR) emission from young planets orbiting young stars. Using AO, we searched for young planetary companions to 200 stars and developed image-processing techniques to allow detection of sources in the scattered starlight.

Our large-scale survey of young stars produced some of the first direct detections of IR emission from planets orbiting other stars. We have the ability to detect planets with masses two to five times that of Jupiter in orbits of 10 to 20 astronomical units (AU)—planetary systems slightly bigger than our own. Even if such systems do not exist, their absence will strongly constrain planet formation, showing that giant planets form only in the central parts of solar systems and then migrate inwards. We also developed new techniques for high-contrast processing of AO images. These techniques will exploit correlations in noise patterns and will be applicable to a wide variety of problems.

Mission Relevance

Astronomical AO has driven the development of AO technology now being used to address a variety of problems in lasers, remote sensing, communications, and medicine. Tackling one of the most difficult AO imaging problems—direct detection of planets—focused research on high-contrast imaging and image processing, with applications that support the Laboratory's national security mission, including secure communications and remote sensing.

FY04 Accomplishments and Results

During FY04 we completed a Keck AO survey of more than a hundred young nearby stars. Analysis of these data is ongoing, but our results clearly indicate that young extrasolar planets in wide (>50 AU) orbits are rare, which helps exclude some classes of planet migration models. We also used the Keck telescope for a deep survey of a smaller sample of carefully selected target stars, using thermal IR observations and advanced image processing techniques. Around the best of these targets we could have seen a Jupiter-like planet in a solar-system-like (5 AU) orbit, the most sensitive direct imaging search yet carried out.

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Publications

Duchene, G., C. McCabe, A. Ghez, and B. Macintosh. B. (2004). "A multiwavelength scattered light analysis of the dust grain population in the GG Tauri circumbinary ring." *Astrophys. J.* **606**, 969. UCRL-JRNL-204388.

Adaptive Optics Views of the Hubble Deep Fields

Claire E. Max 03-ERD-002

Abstract

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

This effort will vastly extend our knowledge of how galaxies formed and evolved and will detect significant numbers of active galactic nuclei at high redshifts. The project is developing new methods and procedures for efficient use of the LLNL laser guide star AO systems, new methods for data analysis, and new ways to use our new data to model the early evolution of galaxies in the distant universe.

Mission Relevance

The AO technology and precision optics developed in this project have application to long-range surveillance for homeland security and to high-power lasers for stockpile stewardship and DoD applications. In addition, the project will contribute insights into galaxy formation and evolution, which support LLNL's mission in breakthrough science. Being at the forefront of a highly visible and exciting area of scientific inquiry, this research will also attract new talent to LLNL.

FY04 Accomplishments and Results

We continued our observational campaign using Keck AO systems to resolve key subcomponents of galaxies—bars, spiral arms, and regions of star formation. Our catalogue includes more than 150 galaxies, each with both AO near-infrared images and Hubble Space Telescope visible-wavelength images. We extended our work to the Lick and Keck Observatory laser guide star systems, with excellent results. Special software packages were used to measure galaxy profiles, identify merging systems, and study the bulge and disk properties of these galaxies. A new group of objects with separations from bright galaxies between 0.5 and 1 arcsec was discovered. These are probably similar to our own Magellanic clouds, but are likely to merge with the host galaxy within a billion years.

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Proposed Work for FY05

We will continue our imaging of distant galaxies using laser guide star AO at the Lick and Keck observatories. We will also use, for the first time, the new OSIRIS spatially resolved spectrograph at Keck. This new spectrograph will allow measurements of internal dynamics on sub-kiloparsec scales in "normal" galaxies in the early universe. A primary science driver for OSIRIS is the "dissection" of high-redshift galaxies to study stellar populations and to measure internal kinematics. Spatially resolved spectra will be used to search for central black hole activity and to measure the star formation rate and stellar ages in the disk and bulge. Spectra will also be used to measure rotation rates within the galaxies and, from this, to determine the mass distribution.

Publications

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Analyzing the Long-Range Transport of Asian Aerosols using an LLNL Atmospheric Model and CAMS/NOAA Measurements from Northern California

Philip J. Cameron-Smith 03-ERD-021

Abstract

Significant amounts of natural and anthropogenic aerosols episodically blow to California across the Pacific from Asia. These aerosols can cause adverse health effects, degrade visibility, and alter the climate. Aerosols potentially represent a large term in the radiative balance of the atmosphere that may be masking effects from greenhouse gases, but the effect of aerosols is regionally heterogeneous. In this project, we experimentally analyzed time- and size-resolved aerosol samples collected at sites in California, Oregon, and South Korea to determine aerosol mass and composition. Coupling this analysis with detailed modeling of the source and transportation of dust using IMPACT (LLNL's 3-D global chemistry transport model), we validated model processes and improved our understanding of the measurements. Virtually all properties of aerosols and clouds depend strongly on their size distribution.

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We expect this project will produce significant scientific results, including determining the extent to which observations in the U.S. can identify pollution sources in Asia; learning to bridge the spatial-scale gap between modeling results and surface observations; developing a scientific connection between observational capabilities at UC Davis, analysis capabilities at the Center for Accelerator Mass Spectrometry, and modeling capabilities at LLNL; developing the capability to input dust-related data to ocean models for iron fertilization and carbon sequestration; and improving and helping to validate LLNL's IMPACT model.

Mission Relevance

The validation of the IMPACT model, plus the high-resolution and microphysics capabilities implemented by this project support LLNL's mission in environmental management and contribute to LLNL's role in DOE programs related to climate change. In addition, dust relates to carbon management, because it is known to fertilize the growth of microorganisms in the oceans, which leads to sequestration of CO_2.

FY04 Accomplishments and Results

Our major accomplishments in FY04 were (1) simulating the impact of intercontinental transport of dust on the U.S. at high resolution (1 × 1 degree); (2) using the measurements to validate physical processes within the IMPACT model (aerosol lofting, transport, settling and deposition); (3) demonstrating that most airborne dust in the far north of California and Oregon during the spring comes from across the Pacific; and (4) upgrading the IMPACT model from fixed aerosol size and composition representations to a full microphysical treatment. The implementation of aerosol microphysics will reduce the forcing uncertainties associated with aerosol size distribution, chemical composition, and state of mixing, and allow us to place our estimates of aerosol radiative forcings on a more quantitative foundation. Agreement of the timing, width, and magnitude of dust events between the observations and the IMPACT model was remarkable, and validated many of the aerosol processes in the model (lofting, transport, settling, and deposition). The comparison also highlighted a subgrid-scale phenomenon that is not resolved by our model: down-slope flow on mountains at night

We expect these accomplishments will lead directly to journal publications.

Thermally Driven Processes and the Atmospheric Transport and Dispersion of Stable Macroparticles

Roxana M. Greenman 03-ERD-039

Abstract

Weapons of mass destruction (WMD) pose a significant threat to our military forces and civilian populations. This project examined the fate and dispersion of chemical agents released by a properly functioning warhead and by a successful low- and high-altitude missile intercepts, which are not well understood. We computationally and experimentally investigated the coupled aerodynamic and thermal response of agents to produce the source term for atmospheric fate and transport studies. This research effort aimed to provide better understanding of the fate of chemical agents from an atmospheric release,

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better quantification of the risk associated with different atmospheric release events, and the capability for the rapid analysis of both types of release events within the atmosphere.

This project will lead to enhanced computational capabilities in multiphase flow, which will address issues in homeland defense related to understanding of the fate of chemical agents released atmospherically and the risks and consequences of different atmospheric releases. Finally, we will develop an integrated, multidisciplinary tool that can predict the fate, dispersion, and ground effects of low- and high-altitude releases of chemical agents.

Mission Relevance

This research will further LLNL's mission in national security by addressing the serious problem of proliferation of WMD producing a new, integrated capability to determine the fate, dispersion, and ground effects of chemical and biological agents released by either a properly functioning warhead or successfully intercepted missile.

FY04 Accomplishments and Results

We investigated the thermal fate, atmospheric dispersion, and deposition of chemical agents. The atmospheric frictional heating effects were investigated by conducting coupled fluid dynamic and thermal calculations. We increased National Atmospheric Release Advisory Center (NARAC) capabilities by adding higher altitude drag effects, dynamic particle momentum, higher altitude turbulence and adding NASA's GEOS-4 high-altitude dataset. We developed a capability for addressing the wide range of meteorological conditions possible for a given location. The K-means clustering method was used to create a manageable set of homogeneous classes from heterogeneous high-dimensional multivariate data. These classes can be characterized stochastically for use in event planning and consequence assessments and in sensitivity/uncertainty analyses.

Publications

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Nourgaliev, R. R. et al. (2004). *Direct numerical simulation of disperse multiphase high-speed flows*. Presented at the 42nd AIAA Aerospace Sciences Meeting and Exhibit, AIAA 2004-1284, Reno, NV, January 5-8, 2004. UCRL-CONF-202429.

Predicting the Effects of Climate Change and Variability on Water Availability

Philip B. Duffy 03-ERD-042

Abstract

Global warming will affect California's hydrological cycle in ways that may necessitate construction of new elements of the water-related infrastructure. New strategies for operating the existing infrastructure will certainly be needed. Predictions of how climate change will affect California's hydrological cycle are needed to support decision-making by policymakers and water managers. Using a hierarchy of models (global climate, regional climate, and surface hydrology) we are projecting how climate change and year-to-year natural climate variability will affect surface temperatures, precipitation amounts, and the amount and timing of flow through rivers. By examining results from a range of models, we are estimating uncertainties in these projections.

This project is producing the best assessment to date of the effects of climate change and variability on the availability of water in California. Using state-of-the-art models at fine spatial resolution, we produced a set of excellent results. To help to quantify uncertainties in these results, we made multiple simulations based on the results of a wide range of climate models. In addition, we are using multimodel ensemble techniques to quantify uncertainties in surface hydrology models. Thus, besides improving understanding of how climate change will affect regional hydrology, our work has significantly advanced the state of the art in the modeling of regional climates and surface hydrology.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we achieved several major project milestones. We completed simulations of California's climate with the MM5 regional climate model at high (9-km) resolution, driven by a global model at high (75-km) resolution. These results demonstrated excellent fidelity. During this process we obtained the highest sustained megaflop (millions of operations per second) rate ever achieved with the MM5 model. We performed simulations of river flows from seven major California watersheds based on meteorology from 10 different global climate models (to estimate uncertainties). Finally, we began studying the predictability of regional precipitation associated with El Nino; we think we now understand that most El Nino winters in California are very wet, while some are dry, because of different patterns of sea-surface temperatures.

Proposed Work for FY05

We will continue to collaborate with regional, State, and Federal water managers to help them incorporate results of our simulations into their planning processes. They will do this

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by using our simulations of unimpeded river flows to drive their own models of the water storage and conveyance system (e.g., dams and reservoirs). This will provide us valuable feedback in our effort to design the simulation tools so they will have maximum impact.

Our research will focus on extreme years and provide quantitative answers to four questions: (1) Will there be more extreme precipitation and flood risk from El Nino when the climate gets warmer? (2) Will El Nino events become more frequent or intense? (3) Are multicentury droughts in California predictable or chaotic? (4) Is multiyear drought in California related to modes of natural climate variability?

Publications

Coquard, J., P. B. Duffy, and K. E. Taylor. (in press). “Present and future surface climate in the Western U.S. as simulated by 15 global climate models.” *Climate Dynamics*. UCRL-JRNL-205916.

Duffy, P. B. et al. (in press). “Simulations of present and future climates in the Western U.S. with four nested regional climate models.” *J. Climate*. UCRL-JRNL-204740.

Environmental Transport and Fate of Endocrine Disruptors from Nonpotable Reuse of Municipal Wastewater

George B. Hudson 03-ERD-065

Abstract

Endocrine-disrupting compounds (EDCs) affect normal hormonal activity and are contaminants of concern in drinking-water systems. This project studies the impacts of EDCs introduced into groundwater by wastewater recycling. We have applied new technologies to accurately determine the transport and fate of EDCs as they move from the surface into groundwater and potentially impact drinking water supplies. We used advanced isotope-tracer techniques, such as dating based on tritium/helium-3 ratios, to track groundwater movement, developed methods for measuring EDCs using state-of-the-art tandem mass spectrometry, and tested sensitive bioassay techniques to measure estrogenic activity from groundwater samples.

This work provides significant and necessary data for evaluating the use of recycled wastewater for irrigation. Water is a limited resource, and recycling of wastewater is an important option for enhancing water supplies. Issues related to contaminants governed by drinking water standards are well understood; however, impacts from EDCs are largely unknown. We will produce information and data needed to quantitatively account for the subsurface transport of EDCs.

Mission Relevance

This project supports the Laboratory’s environmental-management mission by providing significant data for evaluating the use of recycled wastewater for irrigation. This interdisciplinary investigation matches the fundamental strengths of LLNL with significant and enduring national needs in management of water resources.

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FY04 Accomplishments and Results

We obtained multiple samples from monitoring wells at the study site that showed no detectable nonylphenol (<0.01 ppb), whereas the irrigation water averages about 5 ppb nonylphenol. Based on isotope tracer measurements, dilution and dispersion can be ruled out as significant factors controlling the nonlyphenol concentration; we conclude that nonylphenol is efficiently removed during recharge and does not impact local groundwater. Based on the results from this project, the California State Water Resources Control Board decided to begin funding the project in FY2005. The primary focus will continue along the same lines, but with added emphasis on finding wastewater compounds that can be used to detect wastewater contributions to groundwater.

Nitrate Biogeochemistry and Reactive Transport in California Groundwater

Bradley K. Esser 03-ERD-067

Abstract

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

The motivation for this work is the need to develop science-based approaches to characterizing and managing nitrate contamination in groundwater. If successful, we will develop a robust approach to characterizing nitrate biogeochemistry in the subsurface. When coupled with our ability to characterize groundwater flow and to model reactive transport, this approach will allow an accurate assessment of the future distribution of nitrate in California aquifers and of the impact of different management practices on nitrate input to groundwater. Such assessment is vital to making cost-effective management and policy decisions regarding land use and groundwater remediation. This research will also allow quantitative assessment of the kinetics of microbially mediated denitrification in groundwater systems.

Mission Relevance

This project supports the Laboratory's mission in environmental management by developing models and technologies to assess the environmental consequences of toxic materials and to manage risk. This type of multidisciplinary investigation matches the fundamental strengths of LLNL with significant and enduring national needs.

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FY04 Accomplishments and Results

In FY04, we (1) developed a quantitative polymerase chain reaction (qPCR) method for functional nitrite reductase genes to assay denitrifier populations; (2) determined denitrification rate constants for an autotroph with Fe monosulfide; (3) used geochemical models with public water-quality data to show that high nitrate levels are associated with fertilization; (4) installed several multilevel wells at a Central Valley dairy, showing denitrification in stratified groundwater, and developed a conceptual model for groundwater flow; (5) analyzed groundwater in the Llagas Basin for age, recharge temperature, and excess nitrogen; (6) and developed a structural model and a geostatistical model for basin permeability in the Llagas Basin.

Proposed Work for FY05

In FY05, we will (1) validate the use of cell number as an indicator of activity by using pure cell culture to determine the correspondence between the specific denitrification rate and the messenger RNA transcript copy number of the nitride reductase gene as determined by reverse-transcription, real-time qPCR; (2) use real-time qPCR to determine denitrifier populations in soil cores taken from the dairy wells; (3) correlate cell density to sediment and water properties; (4) develop groundwater flow and biogeochemical reactive transport models to validate microbial kinetic rate expressions; and (5) develop a reactive transport models to simulate the transport and distribution of shallow groundwater nitrate contamination to quantify the impact of different management practices.

Publications

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Tectonic Morphochronology of the Southern San Andreas Fault System

Frederick J. Ryerson 03-ERI-001

Abstract

The San Andreas Fault (SAF) accommodates a major fraction of the motion between the North American and Pacific tectonic plates. How slip is distributed among the many segments of the fault is not well understood and has implications for seismic risk. South of the Transverse Range, the SAF divides into a number of strike-slip segments and thrust zones. Deformation is the focus of geodetic observations performed there, but only two millennial (long-term) slip-rate measurements exist for the SAF. We will begin developing a millennial slip-rate map of the western U.S., initially focusing on sites within the SAF system. Slip rates will be determined by measuring landscape offsets caused by active faults and then using cosmic-ray surface-exposure dating to determine their age.

We expect that this project will add to our understanding of the interactions of fault motion in continental settings, with important implications for the assessment of earthquake risk. We will use cosmogenic dating, which has not been widely used in the western U.S., to develop a method for determining millennial slip rates. Along with paleoseismic data and geodetic slip rates, these millennial slip rates will help to identify stress accumulation on the various segments of the fault, because disparities in long- and short-term slip rates influence earthquake frequency. If successful, this method will provide a new tool in evaluating crustal deformation that can be widely used in seismically active regions throughout the world.

Mission Relevance

This analysis of satellite imagery to identify tectonic features furthers radar interferometry and contributes to understanding the mechanics of the Earth's crust. These topics are also related to explosion monitoring and analysis, which supports nonproliferation. In addition, the structure and tectonics of the western U.S. are important in enhanced test readiness, geothermal energy, and the disposal of radioactive waste, in support of missions in national and energy security.

FY04 Accomplishments and Results

In FY04, we completed investigations in the rate of slip along the Mojave segment of the SAF and the uplift rates in the Sierra Madre-Cucamonga thrust region. The Sierra Madre rate is 50% less than geodetic rates for the Los Angeles Basin, implying that other structures are involved. We also obtained new aerial photographs of Biskra Palms on the Mission Creek segment of the SAF and collected subsurface samples to constrain the concentration of inherited nuclides; collected and dated samples from Cajon Pass, which will allow us to extend the previously measured slip rates further into the past; mapped and obtained preliminary ages at three sites between Biskra Palms and San Gorgonio Pass; and obtained new ages at Anza on the San Jacinto Fault.

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Proposed Work for FY05

The proposed work for FY05 will concentrate on four specific, related sections of the southern SAF system: (1) the Mission Creek segment of the fault between San Gorgonio Pass and Biskra Palms/Indio, (2) Cajon Pass, (3) subsurface samples at Biskra Palms, and (4) the San Jacinto Fault at Anza. We will investigate the spatial and temporal variation in the lateral slip rate along the strike-slip segments of the southern San Andreas and San Jacinto faults. We also will investigate the chemical extraction of spallogenic ^{14}C from quartz, to obtain better temporal resolution for samples younger than 20,000 yr. This method will be applied to a complete subsurface vertical section of an alluvial fan so that climate data can be retrieved.

Publications

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Dense Nonaqueous-Phase Liquid Dissolution in Porous Media: Multiscale Effects of Multicomponent Dissolution Kinetics on Cleanup Time

Walt W. Mcnab 04-ERD-001

Abstract

The goal of this project is to determine factors that control the rate at which dense nonaqueous-phase liquid (DNAPL) contaminants are released from source areas into groundwater. A thorough understanding of this process is needed to predict the overall dissolution rate, the consequences of contaminant flow and transport, and the time required for cleanup. The dissolution of DNAPL in the subsurface will be studied both experimentally and through the use of numerical models to gain insight about the multiphase dissolution processes of DNAPL at the microscopic scale, then appropriately scaling up this formulation for application at the field scale. The resulting model will be used to predict long-term behavior of contaminants in source areas and to reevaluate and inspire new remediation technologies.

The project will conduct pore- and mesoscale experiments and develop numerical capabilities to simulate the experiments and predict DNAPL behavior at the field scale. The experiments will be used to calibrate the pore-scale model that incorporates an improved understanding of the processes using Navier-Stokes equations and multiphase physics. The

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model will yield an interphase mass-transfer flux to be upscaled and validated on a series of mesoscale experiments. The upscaled mass-transfer flux will then implemented in a field-scale solute-transport model. The field-scale model will be tested using groundwater-monitoring data from the on-going LLNL cleanup efforts and will be used to assess new remediation technologies and reevaluate existing ones.

Mission Relevance

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

FY04 Accomplishments and Results

Progress during FY04 included (1) constructing a pore-scale experimental apparatus for directly imaging and quantifying dissolution of a nonaqueous phase in transparent analog porous media and completion of a series of experiments, (2) developing and testing a pore-scale model of multiphase dissolution and fluid flow by solving Navier-Stokes and transport equations using an adaptive-mesh finite-element code (a model for generating synthetic porous media was developed in parallel for 3-D simulations), (3) investigating multicomponent DNAPLs cosolvency effects on the contaminant concentrations using LLNL field-scale monitoring data, and (4) extending the pore-scale numerical model to assess the hydrodynamic instabilities of DNAPLs and supercritical fluids such as CO_2.

Proposed Work for FY05

Objectives for FY05 include (1) building a mesoscale experimental apparatus using components obtained from Sandia National Laboratory; (2) conducting micro- and mesoscale experiments for single- and multicomponent DNAPL dissolution; (3) improving the microscale model to include multiphase multicomponent dissolution of DNAPLs, (4) developing a time-dependent mass-transfer flux that represents the evolution of the entire source area body to be used in our existing field-scale model; and (5) developing a set of coupled, scaled-up flow and transport equations, from the microscale to the field scale that describe transient dissolution and aqueous-phase transport of DNAPL.

Magnetic Dynamos and Stars

Peter P. Eggleton 04-ERD-027

Abstract

We will upgrade Djehuty, a code that models stars in three dimensions (3-D), by incorporating magnetohydrodynamics (MHD). Djehuty already includes (1) an accurate equation of state, (2) radiative heat transport, (3) a full network of nuclear reactions, (4) self-consistent gravity in the spherical approximation, (5) Lagrangian hydrodynamics, and (6) an algorithm for adjusting severely distorted meshes. We will add (7) the magnetic-force term

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to the hydrodynamics and (8) the induction equation in the frozen-in approximation, enabling the code to model a wide range of inherently 3-D problems currently beyond computational possibility, including dynamo activity in stars and planets and ultimately the Sun's well-known but poorly understood magnetic cycle.

We expect to produce code that incorporates MHD and has improved abilities to model a broad class of astrophysical phenomena. This will be the first code to tackle and understand, for example, the generation of huge but transient (1- to 3-yr) star spots observed on some red giant stars; similar, shorter-time-scale phenomena in red dwarf stars; magnetic behavior in gaseous planets such as Jupiter and many recently discovered extrasolar planets; energy transport in contact binaries comprising two red-dwarf stars; and the solar cycle, which strongly impacts the Earth and Earth-orbiting satellites. These highly significant advances will enhance LLNL's role in the astrophysical world. This project has also attracted collaboration with the University of California.

Mission Relevance

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

FY04 Accomplishments and Results

Work in FY04 focused on ensuring that the magnetic field remains solenoidal. We tested and validated Djehuty's ability to compute turbulent convection and its interaction with rotation—two vital ingredients in stellar dynamo activity; reworked some algorithms in Djehuty to incorporate MHD; began including the Lorenz force term in the hydrodynamics; and started to include the induction equation in the infinite-conductivity approximation.

Proposed Work for FY05

In FY2005, we will incorporate two new features in algorithms that are already deeply embedded in the code: (1) adding a mesh-remapping procedure that avoids progressively more distortion near topological defects and (2) doubling, in each of the two dimensions, the number of cells near the surface. These accomplishments should make it possible to follow MHD in the important outer layers.

Publications

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Dynamic Data-Driven Event Reconstruction for Atmospheric Releases

Gayle A. Sugiyama 04-ERD-037

Abstract

For atmospheric releases, event reconstruction answers the following critical questions: How much material was released? When and where was it released? What are the potential consequences? Inaccurate estimation of the source term can lead to gross errors, time delays during a crisis, and even fatalities. We are developing a capability that seamlessly integrates observational data streams with predictive models to provide the best possible estimates of unknown source term parameters and optimal and timely situation analyses. Our approach utilizes Bayesian inference and stochastic sampling methods [Markov Chain Monte Carlo (MCMC) and Sequential Monte Carlo (SMC)] to reformulate the inverse problem into a solution based on efficient sampling of an ensemble of predictive simulations, guided by statistical comparisons with data.

We are developing a flexible and adaptable data-driven event-reconstruction capability for atmospheric releases that provides (1) quantitative probabilistic estimates of the principal source-term parameters (e.g., the time-varying release rate and location); (2) predictions of increasing fidelity as an event progresses and additional data become available; and (3) analysis tools for sensor network design and uncertainty studies. Our computational framework incorporates multiple stochastic algorithms, operates with a range and variety of atmospheric models, and runs on multiple computer platforms, from workstations to large-scale computing resources. If successful, the project will result in a multiresolution capability for both real-time operational response and high-fidelity multiscale applications.

Mission Relevance

This project directly contributes to the Laboratory's homeland security and national security missions by addressing a critical need for atmospheric release event reconstruction tools to step beyond current capabilities of the rapidly growing number of operational detection, warning, and incident characterization systems being developed and deployed by the Department of Homeland Security (e.g., BioWatch, BWIC system, national BioSurveillance Initiative) and Department of Energy (e.g., Nuclear Incident Response Team assets and deployments). The event-reconstruction and sensor-siting capabilities being developed by this research project will find application in studies of vulnerability and the protection of critical infrastructures and borders

FY04 Accomplishments and Results

In FY04, we (1) implemented a 2-D prototype Bayesian MCMC atmospheric event reconstruction capability and tested it against a variety of scenarios and synthetic data; (2) developed an MCMC capability using the National Atmospheric Release Advisory Center (NARAC) 3-D Lagrangian particle-dispersion code LODI and tested it against field experiment data; (3) developed an SMC methodology for time-dependent problems and tested it using synthetic data; (4) investigated optimization methods for determining proposal distributions and improving convergence; (5) evaluated the relative performance of

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MCMC and SMC methods using synthetic data; (6) designed an MCMC and SMC computational framework on massively parallel platforms; and (7) demonstrated the effectiveness of the SMC methodology for detecting moving sources using synthetic data.

Proposed Work for FY05

In FY05, we will (1) implement an SMC atmospheric event reconstruction capability based on NARAC operational models; (2) develop procedures and tools for input and model error quantification; (3) extend the event reconstruction capability to handle complex terrain and an extended range of potential release types; (4) develop a multiresolution capability for problems requiring multiple levels of models to accurately characterize the release event; (5) continue developing and testing efficient stochastic sampling and convergence algorithms; (6) explore methods for incorporating alternative input data types (e.g., remote sensing, qualitative, imagery); and (7) enhance performance on massively parallel platforms for efficient event reconstruction of complex atmospheric releases.

Publications

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Kosovic, B. et al. (2004). Dynamic data-driven event reconstruction for atmospheric releases. Presented at the 2nd Sandia Workshop on Large-scale PDE-Constrained Optimization, Santa Fe, NM, May 19–21, 2004. UCRL-POST-204204.

Kosovic, B. et al. (2004). Dynamic data-driven event reconstruction for atmospheric releases. Presented at the 8th Ann. George Mason Univ. Transport and Dispersion Modeling Conf, Fairfax, VA, July 2, 2004. UCRL-PRES-205169.

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Sugiyama, G. et al. (2004). Markov-Chain Monte-Carlo and Sequential Monte-Carlo approaches to dynamic data-driven event reconstruction for atmospheric releases. Presented at the 2nd Workshop on Monte Carlo Methods, Boston, MA, August 27–28, 2004. UCRL-POST-206484.

Coupling Micromechanics and Reactive Fluid Flow in Fracture Networks

Russell L. Detwiler 04-ERD-046

Abstract

In a broad range of energy-related problems, geological fractures provide dominant pathways for flow and transport, yet the effects of coupled mechanical deformation and geochemical alteration are poorly understood. Our research investigates the importance of coupled mechanical stresses and geochemical reactions on the evolving permeability of fractured subsurface media. We will develop and apply coupled computational models that explicitly incorporate the small-scale physical processes that control these mechanisms within individual fractures. The coupled small-scale processes will be scaled up to explicitly tie

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individual fractures into a discrete fracture network model, bridging a critical gap between our understanding of small-scale processes in individual fractures and field-scale observations.

The proposed fracture network model will explicitly couple small-scale processes in individual fractures within fracture networks. This will complement existing large-scale dual-continua models (e.g., the LLNL-developed reactive transport model, NUFT) by providing physically based, quantitative descriptions of sub-grid-scale response to coupled geomechanics and geochemistry. This modeling approach will enhance our ability to predict complex field-scale behavior based on laboratory measurements of fractured rock properties. Furthermore, we will apply our model to the problem of CO_2 sequestration to gain new insights into the role of the coupled, and potentially competing, processes of geochemical alteration and mechanical deformation of cap rocks during long-term CO_2 storage in saline aquifers.

Mission Relevance

Our coupled modeling approach provides a framework for integrating geochemistry, rock mechanics, and reactive transport. This will provide an enhanced understanding of complex coupled processes that are critical to a broad range of problems that support LLNL's mission in energy security and environmental management. These problems include geologic CO_2 sequestration, enhanced oil recovery, engineered geothermal systems, and radioactive waste isolation.

FY04 Accomplishments and Results

In FY04, we produced a model, SFRT, that effectively couples single-fracture models of reactive flow and transport and mechanical deformation. This is a critical first step in developing a discrete fracture network model that faithfully represents these coupled processes within individual fractures. We evaluated SFRT through detailed comparisons with recent experiments and enhanced the model to incorporate nonlinear reaction kinetics to simulate local rock-water interactions. To quantify permeability changes in individual fractures caused by mechanical deformation, we developed a single-fracture implementation of the code LDEC, which predicts normal and shear displacements, and incorporated Flex, a computationally efficient model of normal displacement.

Proposed Work for FY05

We plan to develop a discrete fracture network implementation of the coupled reactive transport and mechanical deformation models. This will involve incorporating the SFRT model into an existing discrete fracture network model that currently employs simple constitutive relationships to model the behavior of each fracture. In addition, we will develop and implement an approach for directly coupling LDEC with the extended reactive-transport model. Our ability to simulate these coupled processes in fracture networks will build upon our ongoing efforts to develop effective upscaling techniques for the mechanical deformation and geochemical alteration within individual fractures of the network.

Publications

Detwiler, R. L., H. Rajaram, and W. W. Cheung. (2003). *The role of Peclet number on the alteration of variable aperture fractures by dissolution: a comparison of physical experiments with*

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computational simulations. American Geophysical Union Fall Meeting, San Francisco, CA, Dec. 8–12, 2003. UCRL-JC-155067-ABS.

Advancing Climate and Carbon Simulation

Starley L. Thompson 04-ERD-051

Abstract

Until our recent efforts, climate models could not predict atmospheric CO_2 concentrations. This project proposes to upgrade LLNL's climate–carbon model, which predicts CO_2 concentrations based on rates of fossil fuel burning. We plan to perform a series of multicentury simulations to quantify climate and carbon feedbacks, move our terrestrial biosphere model into a new climate model, and improve key aspects of the terrestrial biosphere model.

Model improvements and simulation results from this project will help us better constrain the range of climate change that could occur during the 21st century. These activities will help define the new field of comprehensive interactive-climate and carbon-cycle simulation.

Mission Relevance

The work supports LLNL's environmental-management mission by advancing the science base for environmental regulation and accurately modeling global climate conditions.

FY04 Accomplishments and Results

We performed a set of six, 230-year runs with our modeling system that helped quantify and bound the role of ocean and terrestrial feedbacks on future changes in the carbon cycle. We found that uncertainties in the response of land vegetation to CO_2 increase cause the largest uncertainties in the positive feedback between climate and the carbon cycle. The feedback uncertainty caused by poorly constrained climate sensitivity is almost as large. We moved our IBIS2 terrestrial biosphere model into a new climate model (CCSM3) to keep our system current. Finally, we updated the IBIS2 model to allow for changes in agriculture or other non-natural land covers or land uses.

Publications

Govindasamy, B. et al. (in press). "Increase of carbon cycle feedback with climate sensitivity: results from a coupled climate and carbon cycle model." *Tellus*. UCRL-JRNL-203401.

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

Thomas P. Guilderson 04-ERD-060

Abstract

The fixation of inorganic carbon by phytoplankton in the ocean and its subsequent export out of the surface ocean (the "biological pump") is one of the fundamental controls on atmospheric CO_2. Production in the ocean is expected to change over a variety of time scales as a result of future climate change and nutrient loading. This project will analyze sediment trap and dissolved organic carbon samples at the molecular level to provide a unique and key perspective on the mechanisms controlling the remineralization of organic carbon in the ocean, including the role of microbial mediation and processes. The data will be used to optimize and test a food-web-based primary productivity and remineralization model, LLNL's Ocean-Biology Global Climate Change Model (OBGCM).

The data–model comparison and ocean-biology model enhancements that result from this study will increase the accuracy by which we can predict future changes in the ocean carbon cycle. In particular, this project will facilitate the study of future climate change scenarios and the efficacy of iron-fertilization strategies for carbon sequestration.

Mission Relevance

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

FY04 Accomplishments and Results

Accomplishments in FY04 include (1) splitting and general characterization of Cariaco Basin and Santa Barbara Basin sediment trap samples; (2) sterol extraction and characterization of Cariaco sediment trap samples; (3) splitting of open Atlantic Ocean sediment trap samples; (4) establishment of column chemistry protocol; (5) extraction of sufficient oceanic organic carbon to develop nuclear magnetic resonance (NMR) methods; (6) initiation of data mining; (7) porting the discharge-estimation code PISCES into OBGCM; (8) analysis of bulk carbon isotopes (^{13}C and ^{14}C); (9) completion of methods for NMR analysis; and (10) insertion of carbon isotope code into PISCES.

Proposed Work for FY05

Tasks for FY05 include (1) compound-class analyses of ^{13}C and ^{14}C; (2) additional bulk analyses of particulate organic carbon; (3) NMR quantitation; (4) sensitivity tests of PISCES using sediment-trap data; (5) Fe-fertilization and climate change scenario sensitivity tests in OBGCM; and (6) data–model comparisons using data from the Southern Ocean Iron Enrichment Experiment and the Iron-Ex experiments.

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

Peter M. Celliers 04-ERD-065

Abstract

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

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

Mission Relevance

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

FY04 Accomplishments and Results

We achieved three major sets of results in FY04. First, we produced, for the first time, dense He states that are metallic, and collected EOS data for He at the highest pressures to date. Second, we measured the onset of metallization for dense fluid H at several different densities and temperatures. Extrapolation of the measured onset of metallization for dense fluid H suggests a maximum in the H melt curve. This is the first experimental insight into why such a maximum would exist. Furthermore, the measured onset of metallization was consistent with recent quantum molecular dynamics simulations that predicted a plasma phase transition in H. Third, we produced metallic states in dense N.

Proposed Work for FY05

In FY05, we will produce planetary mixtures of H_2–He. Because they are immiscible in cryogenic ambient conditions, these mixtures have never been studied at high pressures and temperatures. Understanding these mixtures at high densities is one of the grand challenges

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in planetary science. Our studies of H_2–He will begin to address the issues of H_2–He miscibility, the transition to conducting states in the mixture, and the effect of the mixture on the EOS. In addition, we plan to reach higher states of precompression and complete target designs for advanced compression schemes.

Publications

Loubeyre, P. et al. (2004). "Coupling static and dynamic compressions: first measurements in dense hydrogen." *High Pressure Res.* **24**, 25. UCRL-JC-15138.

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

Kem H. Cook 04-ERD-070

Abstract

The next generation of imaging surveys in astronomy, such as the Large Synoptic Survey Telescope (LSST), will require multigigapixel cameras that can process enormous amounts of data read out every few seconds. This huge increase in data throughput (compared to megapixel cameras and minute- to hour-long integrations of today's instruments) calls for a new paradigm for extracting the knowledge content. We propose to develop the foundations for this new approach. LSST data will probe the distribution of dark matter and the evolution of dark energy via weak lensing and supernovae studies. In this project, we will study the systematics of extracting the dark matter distribution from weak lensing and the efficiency of finding and following supernovae. We will produce significant near-term scientific breakthroughs by developing new methods to probe both the elusive time and spatial variations in astrophysics data sets from the SuperMACHO (Massive Compact Halo Objects) survey, the Lowell Observatory Near-Earth Object Search (LONEOS), the Livermore Optical Transient Imaging System (LOTIS), and the Taiwanese American Occultation Survey (TAOS).

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

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we developed a fast photometry code; made significant advances in optimizing image subtraction; obtained the LONEOS data and began reductions to identify RR Lyrae

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stars; reported real-time detection of microlensing events in the SuperMACHO project; and began initial work on a weak-lensing simulator. In addition, we investigated the RR Lyrae distribution in the halo of the Milky Way; incorporated our new photometry code into the SuperMACHO pipeline; and finished bringing MACHO image data to LLNL to test pipelines and extract new science using difference image analysis. We also began a systematic study of the effect of the atmosphere on the apparent shapes of galaxies to better estimate weak lensing, designed the schema for a prototype LSST database, and developed typical queries.

Proposed Work for FY05

We will develop rapid, robust algorithms for the linkage of multiple observations of a moving object, which is a key challenge for LSST. Using the TAOS data set, we will develop a new class of techniques to look at wide-field, subsecond time-variable phenomena; use MACHO and SuperMACHO data to prototype high-throughput pipeline analysis systems for efficiently producing real-time alerts; and use the data and tools acquired to probe the variable universe to better understand the physics of variable stars, the mass distribution of the Galaxy, and the local distance scale. We will continue to probe the effect of the atmosphere on weak lensing measurements, and begin to examine the effects of the telescope on these measurements.

Publications

Alcock, C. et al. (2003). "The MACHO Project large Magellanic cloud variable-star inventory. XI: Frequency analysis of the fundamental-mode RR Lyrae stars." *Astrophysical Journal* **598**, 597. UCRL-JC-153183-PT-11.

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Schlaufman, K. et al. (2004). *Simulations of turbulence-induced ellipticity over large fields of view: the first step towards enabling LSST weak-lensing science*. UCRL-POST-205862.

High-Fidelity Fluid Mechanical Model for the Early Time Prediction of Atmospheric Contaminant Clouds From Nuclear Explosions

Allen S. Grossman 04-ERD-082

Abstract

This research proposes to evaluate a state-of-the art code to predict the motion of atmospheric contaminant clouds resulting from near-surface nuclear explosions. The code will cover the time interval between the very-early-time explosion phenomenology (<1 s) modeled by detailed weapons codes and the late-time (>1000s) atmospheric-dispersion models used in LLNL's National Atmospheric Release Advisory Center (NARAC) modeling system. The project team will work with Lawrence Berkeley National Laboratory (LBNL) personnel to import, adapt, and evaluate a low-Mach-number, adaptive-mesh-refinement (AMR) code developed at LBNL to provide the basis for an enhanced model that will be developed by this project.

Successful completion of this project will establish the feasibility and provide the basis for developing an advanced, predictive, early-time nuclear-contaminant cloud model. The proposed model will replace or supplement semi-empirical models currently used in the 1- to 1000-s time regime to describe radiological, nuclear, and explosive sources. This work will contribute to developing a state-of-the-art nuclear-effects prediction capability.

Mission Relevance

This project supports the DOE and LLNL national security missions by contributing to a capability to predict the atmospheric releases and subsequent plume movement that stem from nuclear explosions. This project also supports the Laboratory's counterproliferation mission by providing an enhanced capability to more accurately assess the atmospheric collateral effects of nuclear explosions.

FY04 Accomplishments and Results

In FY04 we (1) installed the LBNL Low-Mach-Number Code (LMC) and used it to contrast ~1-m simulations in a stratified atmosphere that made use of the anelastic approximation with simulations that did not use this approximation; (2) developed an idealized 1-kiloton source model to drive the LMC; (3) completed a model for an actual atmospheric nuclear test program event; (4) implemented an idealized tracer for the weapon debris; (5) tested an interface between the LMC and NARAC dispersion model LODI; and (6) ran a LODI simulation of tracer-particle distribution, calculated by the LMC code, for a 3-h period that used a well-defined set of actual meteorological data. After the LDRD project ended, validation of the code using nuclear test data began under subsequent programmatic sponsorship.

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Development of Generalized Mapping Tools to Improve Implementation of Data-Driven Computer Simulations

Michael E. Pasyanos

04-ERD-083

Abstract

The Stochastic Engine (SE) is a data-driven computer simulation for predicting the behavior of complex systems, providing decision makers with the ability to weigh various courses of action or determine what data would be required to reach a decision. Relieving the need for customized mapping for each input data type would significantly enhance the use of SE for a variety of potential applications. This project will develop a framework for incorporating different data sets into probabilistic inverse techniques like SE. We will characterize the transformation types and generalize the implementation of each into generic mapping classes. The goal is to reduce response time and make the approach more accessible. While the framework is general, we will demonstrate the benefits with an example of seismic structure of the Korean Peninsula.

From this research, we expect to gain answers to the following questions: (1) Does the approach of generalizing the mapping enhance the implementation of SE problems? (2) How does this generalization improve the integration of new data and speed its operation? (3) What are other possible generalizations we can make to improve efficiency? Where possible, we will develop a set of generic tools for various data types. Finally, for a relevant earth science problem (a high-resolution regional seismic model), we will use the tools to develop the stochastic model. We will provide the resulting inversion models and publish the results, introducing the methodology to regional-scale earth science problems.

Mission Relevance

By improving the efficiency of implementing the SE, this project will benefit several Laboratory missions. Data-driven simulations of complex earth and atmospheric science systems will support missions in environmental technologies and in energy and water security, while the nuclear-explosion monitoring application will support national security missions in nonproliferation and arms control.

FY04 Accomplishments and Results

We performed a census of model transformations and classified theoretical model transformation types, thereby developing a framework for set of transformations. Using these tools, we advanced the development of a high-resolution regional seismic model for the Yellow Sea and Korean Peninsula region and submitted a paper on the results to a high-profile peer-reviewed journal. Results from the model will aid nuclear explosion monitoring in the region by helping to lower the seismic monitoring threshold. The tools have also been used to develop a plan for using available data to build a seismic model for the Las Vegas Valley and southern Nevada for the Nevada Test Site Readiness Program. We also explored other strategies for improving the SE approach.

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Publications

Pasyanos, M.E., G.A. Franz and A.L. Ramirez. (in press). "Reconciling data using Markov Chain Monte Carlo: An application to the Yellow Sea - Korean Peninsula region." *Geophys. Res. Lett*. UCRL-JRNL-206357.

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High-Resolution Aerosol Modeling: Decadal Changes in Radiative Forcing

Douglas A. Rotman 04-ERD-089

Abstract

Atmospheric aerosols are important in mediating the radiation balance of the Earth's atmosphere. Fine spatial resolutions are required to model the effects of regional pollution on the global atmosphere and the effects of long-range transport of aerosols on regional climate. In addition, the short lifetime of aerosols (approximately 1 wk) results in large spatial and temporal variations of sulfate concentrations that also require fine resolutions. This project has carried out high-resolution calculations of direct sulfate forcing. Simulations integrated LLNL's global chemistry-aerosol model (IMPACT) with the LLNL high-resolution global climate model simulation output (horizontal resolution as high as 50 km) to examine the temporal evolution of sulfate forcing since 1950.

The objective is to assess the radiative effects of anthropogenic sulfate aerosols on a region-by-region basis. By looking at historical emissions, this project examined the changing regional character of aerosol forcing over the past five decades. This work studied the historic direct sulfate forcing at much finer spatial resolutions than ever attempted before.

Mission Relevance

Identifying, understanding and remediating environmental consequences of energy use form an important part of the Laboratory's energy security and environmental management missions. Continuous refinement in modeling aerosols in atmospheric releases also plays an increasingly important role in the Laboratory's ability to carry out its missions in nonproliferation and homeland security, especially in event management.

FY04 Accomplishments and Results

We analyzed regional aerosol distributions over the last five decades and examined their direct radiative forcings. Meteorological information was provided by the National Center for Atmospheric Research CAM3 model. IMPACT simulations used aerosol emissions representative of the five decades from the 1950s to the 1990s to predict aerosol distributions and radiative forcings. Results show the effects of the growing magnitudes and changing geographical pattern of sulfur emissions. In 1950 the United States emitted 41% of the world's sulfur aerosols; this dropped to 15% by 1990. However, Asian emissions jumped six fold during the same time, from 7% in 1950 to 44% in 1990. These emissions drive a change in forcing, with those in Asia being equal to or larger than forcings over Europe and North America.

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Publications

Bergmann, D. J., C. C. Chuang, and D. A. Rotman. (2004). *LLNL scientists use NERSC to advance global aerosol simulations*. UCRL-JRNL-207209.

Mission to Very Early Earth

Ian D. Hutcheon 04-ERI-004

Abstract

This project will conduct a combined analytical and experimental program to study mineral and melt inclusions in ancient zircons (age >4.4 billion yr) to determine when conditions suitable for life first emerged on Earth. After gathering the geochemical data as a function of zircon age, we will relate these data to the respective parent materials. The proposed work involves dating and isotopic and chemical analysis on the zircons and the mineral and melt inclusions in them, as well as a search for graphitic "chemofossils." The experimental portion focuses on the partitioning of trace elements, notably uranium and actinides, between zircon and melt. The investigation utilizes a broad array of new analytical facilities that also support a variety of nuclear forensic applications.

The most important scientific output of this project is the chemical characterization of Jack Hills zircons as a function of age. Based on these new data we will (1) gain understanding about how the fossil record of past life is preserved today, (2) determine the time when conditions suitable for supporting life first emerged on Earth, and (3) get a more accurate picture of the evolution of the atmosphere and hydrosphere over the first 160 million years of Earth's history.

Mission Relevance

This work supports both the stockpile-stewardship and basic-science missions of the Laboratory by providing an important testing ground for the microanalytical instrumentation required by many DOE programs and is closely related to ongoing investigations of fissile materials for stockpile stewardship and optical components for future fusion-class lasers.

FY04 Accomplishments and Results

During FY04 we (1) developed analytical protocols for the analysis of elemental compositions and oxygen-isotope abundances in micrometer size inclusions with NanoSIMS technology (secondary ion mass spectrometry for nanometer-scale samples), (2) applied these procedures to inclusions in Jack Hills zircons with ages >4 billion years, (3) mapped inclusion populations in new Jack Hills zircons, and (4) began experiments to measure trace element partitioning between zircon and silicate melt. The NanoSIMS images revealed a diverse population of inclusions, including apatite, xenotime, quartz, muscovite, feldspar, and a possible melt inclusion. One apatite inclusion analyzed in detail contained a number of heretofore unrecognized features, most notably chemical zonation on a 500-nm scale and a 1- to 2-mm-wide zone enriched in O, Mg, and Fe surrounding the inclusion.

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Proposed Work for FY05

In FY05 we will (1) continue the chemical and structural characterization of mineral and melt inclusions in ancient zircons from Jack Hills using electron microprobe, micro-x-ray absorption fine structure (XANES) and NanoSIMS, (2) determine the oxygen-isotope composition of zircons and their inclusions with mapping of internal zonation by in situ NanoSIMS analysis, (3) correlate variations in oxygen-isotope composition with inclusion chemistry and age, and (4) use new partitioning and diffusion data to constrain the thermal history of the Jack Hills zircons and the long-absent host rock. This would be a major step forward in our understanding of conditions on the Earth's surface during the Hadean epoch.

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

Gregory J. Nimz 04-ERI-013

Abstract

This project creates a capability to analyze ^{129}I by accelerator mass spectrometry (AMS) and builds a foundation for its scientific application by developing chemical processing technology needed for environmental and nonproliferation use. The chemical behavior of ^{129}I allows the isotope to escape into the environment during nuclear processing, making it an indicator of clandestine nuclear activities. Capabilities in AMS require optimization of interrelated instrumental parameters; required processing technologies focus on sampling and chemical extraction. As a proof of principle of this new capability, anthropogenic ^{129}I in soils and groundwater will be analyzed.

This research develops (1) the ability to analyze small quantities of materials with low $^{129}I/I$ ratios, (2) a new generation of iodine-sampling technology for low-level detection, (3) technology for iodine extraction from a wide range of natural materials (milk solids, soils, plants), and (4) an understanding of labile ^{129}I migration in the environment. This facilitates basic research in environmental science, nutrient cycling, hydrology, and oceanography; it impacts biomedical research on iodine deficiency disorder, fetal brain development, and thyroid functioning; and it permits monitoring and detection of clandestine nuclear activities. Immediately publishable results will include methods for redox-sensitive sampling and the movement of labile iodine in soils and groundwater.

Mission Relevance

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

FY04 Accomplishments and Results

In FY2004, we achieved excellent results: measurement linearity of standards for $^{129}I/^{127}I$ ratios in the AMS-relevant range of 10^{-14} to 10^{-9}, instrumental backgrounds for $^{129}I/^{127}I$ less than 2 ×

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10^{-14}, excellent 127I beam currents greater than 155 A, accelerator transmission better than 11%, and electrostatic operation at 1110 kV, permitting energy/charge states required for the analyses. We investigated alternate detector configurations and window materials and optimized target materials and cathode compositions. Background levels and ionization efficiency were optimized to permit low-level (205 g) iodine analysis, a unique capability. We are now capable of high-quality $^{129}I/^{127}I$ analysis of standard solutions and in FY05 will turn our attention to natural matrices and the potential analytical problems they present. FY04 concentrated on AMS instrumental development; subsequent years concentrate on processing technology, proof of principle, and collaborative research on the fate and transport of iodine in the natural environment.

Proposed Work for FY05

In FY2005, we plan to develop sample processing and ^{129}I extraction technology for natural matrices through a series of activities. The first examines soil migration of anthropogenic ^{129}I. Transport of dissolved 129I will be contrasted with ^{129}I complexed with both oxyhydroxide and organic acid colloids. This first-of-its-kind study will elucidate several aspects of anion transport. Three hydrology projects will be initiated in FY2005, in collaboration with UC faculty and students. These projects utilize natural and anthropogenic ^{129}I as a hydrologic tracer. We will investigate: (1) how changes in climate affect drainage from the Sierra Nevada; (2) mechanisms for solute migration away from a radionuclide contaminated stream; and (3) interaction between young meteoric groundwater and older geothermal water.

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

Paula J. Reimer 04-ERI-014

Abstract

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

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

Mission Relevance

The proposed research will enhance understanding of soil carbon cycling in support of LLNL's

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mission in environmental management, and it will provide valuable input for various DOE programs investigating the effects of increased levels of atmospheric carbon dioxide on the Earth's climate. The project will also strengthen LLNL collaborations with two UC campuses and Lawrence Berkeley National Laboratory (LBNL).

FY04 Accomplishments and Results

We completed several tasks related to sample collection, archiving, and processing for assessment of DOC and estimating mean carbon turnover time. We sampled soil pits and installed lysimeters at one terrace; set up a soil water sampling program and hired and trained a student assistant in sample collection and processing; sampled soil and vegetation three other terraces; archived soil, water, and vegetation samples; and helped establish a controlled access soil biogeochemistry lab at LBNL for soil analysis and sample preparation. We expect to complete the analysis of archived material by October 2004.

Proposed Work for FY05

We plan to hire a postdoc dedicated to this project in early FY05. We will continue with root, soil, and DOC sampling and analysis to measure natural variability in samples, and incorporate preliminary results and additional pathways into the soil model as required. In additional, we will collaborate with the Sierra Nevada Research Institute at UC Merced, where a Ph.D. student will help with soil separation and expand the carbon characterization using technologies such as Fe- and Al-based x-ray absorption spectroscopy and Fourier-transform infrared analysis. These data will be used by our collaborators in the terrace studies and may set the stage for future work.

Publications

Swanston, C. W. et al. (in press). "Initial characterization of processes of soil carbon stabilization using forest stand-level radiocarbon enrichment." *Geoderma*. UCRL-JRNL-201893.

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

Jeffery J. Roberts 04-LW-077

Abstract

This project will conduct an integrated study of the electrical conductivity, texture, and permeability relationships of silicate-sulfide partial-melt samples to provide much needed constraints on models of planetary core formation. Tasks include synthesis and characterization, electrical conductivity measurements, and x-ray computed microtomography (CMT) 3-D imaging. The goal is a unified model of the bulk conductivity and permeability of a partial melt that incorporates the melt geometry. Material synthesis will occur in the piston cylinder and the multianvil devices at pressures in excess of 25 GPa. Recovered samples will be characterized using x-ray CMT.

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

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to near-micrometer spatial resolution. Successful development of a cell for in situ measurement of electrical conductivity in LLNL's multianvil (D-DIA) device will be a significant advance, setting the stage for measurements made on high-pressure silicate phases, such as perovskite in contact with melt. These results will constrain models of core segregation and compositions of the melt phase.

Mission Relevance

The development of microscale imaging techniques and the distribution of melt or minority phases on a microscale is relevant to obtaining a better understanding of weapons performance issues, in support of several NNSA national security missions, including stockpile stewardship. In addition, development of microscale transport models will benefit the Laboratory's mission in environmental management.

FY04 Accomplishments and Results

Silicate-sulfide partial melts with a range of compositions and melt fraction were synthesized and electrical conductivity measurements were performed on these samples. Key findings are the change in behavior between 3 and 6 wt% non-wetting sulfide melt, and the high conductivity observed for samples containing more than 6 wt% melt after reaching 1250˚C. We made significant progress in the imaging performed at the Advanced Light Source (ALS) beamline (14 samples), and computed tomography (CT) scans of standard porous materials have been used to test an implicit lattice Boltzmann flow simulation code for calculating permeability. Work in this area resulted in first-of-a-kind images of flow velocity in porous media and led to a new collaboration with University of Oregon researchers. A significant accomplishment was the development of an experimental cell to measure conductivity in situ in the D-DIA apparatus.

Proposed Work for FY05

In FY05, we plan to perform a systematic set of simultaneous high-pressure and temperature electrical conductivity experiments on olivine and Fe-S-Ni partial melts. These experiments will include varying wetability by controlling melt chemistry, varying melt fraction, and performing measurements over sufficient temperature range to affect crystal–melt interaction. We will also perform x-ray micro-CT imaging on recovered run products at ALS and lattice-Boltzmann code simulations on digital volumes to predict permeability of each run product, compare the permeability predicted from lattice-Boltzmann methods to those from measured electrical conductivity, and place results in context of models of core formation. Expected results include knowledge of olivine Fe-S systems in terms of melt interconnectivity and melt extractability.

Publications

Du Frane, W. D. et al. (2004). *Anisotropy of point defect mechanisms and electrical conduction in single crystal olivine*. Presented at the Fall American Geophysical Union Meeting, San Francisco, CA, Dec. 13–17, 2004. UCRL-ABS-206406.

Roberts, J. J. et al. (2004). *Electrical properties of olivine-FeS partial-melts*. Presented at the 2004 Western Pacific Geophysics Meeting, Honolulu, HI, Aug. 2004. UCRL-ABS-203509.

The Innermost Inner Core: Fact or Artifact?

Hrvoje Tkalcic

04-FS-019

Abstract

There is currently considerable debate in the Earth sciences community over the composition, thermal history, and dynamics of Earth's inner core. Due to the uneven distribution of large earthquakes and recording stations around the globe, the details of structural models are based on precious few seismological observations of travel times of primary waves traversing the outer core (PKP waves). Using state-of-the-art signal-processing techniques to measure and compile a unique set of global travel time data of surface-reflected seismic waves that propagate through the center of the Earth (PKPPKP waves), we propose to investigate the characteristics of the innermost inner core. We will carry out a systematic investigation to determine the configuration of inner core anisotropy, previous studies of which are biased by a limited spatial sampling of the inner core by PKP waves.

We expect to demonstrate whether or not the existence of innermost inner core features can be supported by seismological data, and we hope to provide major constraints on the amount of inner core anisotropy.

Mission Relevance

This project enhances and extends a critical Laboratory core competency in seismology for national security, specifically for ground-based nuclear explosions monitoring. This high-profile science project also supports LLNL's mission in basic science by leading to a better understanding of deep Earth structure, physical properties and models used to describe the evolution of the Earth's core.

FY04 Accomplishments and Results

In FY04, we acquired a large number of broadband seismic waveforms from global broadband and several regional and temporary networks for large earthquakes and explosions that satisfy our criteria regarding the epicentral distance, event depth, and magnitude. Our work focused on (1) preparing and analyzing waveforms that satisfy our source–receiver criteria, from 1990 to the present day, and (2) observing clear PKPPKP arrivals and their precursors in both time and frequency domains. We presented our results at the Fall 2004 Meeting of the American Geological Union and a paper is in preparation for submittal to a peer-reviewed journal.

Proposed Work for FY05

In FY05, we plan to finalize the observational part of our project and analyze and interpret absolute and differential travel times for both polar and equatorial paths corresponding to PKPPKP waves and their precursors. We also intend to analyze the observed PKPPKP waves in frequency domain. By analyzing travel times and frequency content of the observed PKPPKP waves, we plan to (1) determine the extent and configuration of anisotropy in the inner core consistent with these new PKPPKP observations and (2) evaluate if the previously hypothesized innermost inner core structure is supported by these observations.

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Publications

Tkalcic, H. (2004). *Probing the deep Earth with seismology: the Earth's structure in the core–mantle boundary region and the core*. UCRL-ABS-201978.

Feasibility of Space-Based Seismometry Utilizing a Satellite-Borne Real Aperture Radar

William Foxall 04-FS-023

Abstract

This high-risk project proposes to explore the feasibility of a real-aperture radar satellite mission to image propagating seismic waves, which are extremely subtle and may be impossible to detect. The resolution capability of present analyses to characterize and discriminate seismic sources is severely limited by the sparse sampling of two-dimensional (2-D) complex wave fields afforded by regional and global seismograph networks. We will partner with NASA's Jet Propulsion Laboratory in a workshop that will define scientific and engineering aspects of the mission, including the design of a scaled proof-of-concept mission using current-technology radar flown on a stratospheric balloon and targeted at planned seismic events.

The outcome of this project will be a report on the feasibility of the proposed satellite mission, including documentation of the scientific and engineering data constraints and on the feasibility assessment process. If the overall concept is judged feasible, the next step will be to develop the balloon mission as the second phase of the joint LLNL/NASA project. The proposed radar mission would revolutionize seismic analyses by imaging complete seismic wave fields over large regions.

Mission Relevance

The ability to provide satellite-launched seismic source discrimination to detect signatures of nuclear testing and assessment of foreign program capabilities will contribute to NNSA and LLNL's mission to provide analysis and support for nuclear nonproliferation and arms control. The remote sensing R&D developed in this project also has potential application to LLNL support for Department of Defense missions.

FY04 Accomplishments and Results

Wave fields of buried explosions and earthquakes in the range Mb3–6 were simulated using the finite difference code E3D and were used to define minimum requirements for amplitude detection threshold, measurement accuracy (including atmospheric turbulence SNR limitations), time sampling, spatial resolution, and ground footprint as a basis for initial mission design. Preliminary assessment suggests that a middle Earth-orbit synthetic-aperture radar mission might meet the spatial resolution requirements more readily than a geostationary real-aperture mission. The mission-design workshop planned for Sept. 28–29 was postponed by JPL. We will complete the mission concept report after the workshop.

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Publications

Rodgers, A. (2004). *Ground motion simulations to investigate the feasibility of space-based seismometry*. UCRL-TR-207266.

Tralli, D.M. et al. (2004). *Laser- and radar-based mission concepts for suborbital and spaceborne monitoring of seismic surface waves*. To be presented at the 2005 IEEE Aerospace Conference. UCRL-CONF-206807.

Constraints on Strong Earthquake Ground Motion from Nuclear Explosion Data

Lawrence J. Hutchings 04-FS-029

Abstract

The long performance-assessment periods mandated for high-level nuclear waste repositories may result in extremely high estimates of earthquake-related ground motion that many seismologists believe to be physically unrealizable. However, no generally accepted method exists to bound ground motions from potential earthquakes. The extreme nature of nuclear explosions suggests that information about ground motion obtained from underground testing could provide valuable constraints on effects of extreme earthquakes. We propose to study recorded ground motion and rock-property data collected from past underground nuclear explosions to determine the feasibility of using these data to place bounds on potential ground motion from large earthquakes. If the waves are found to be inelastic, we will determine their decay rate. If found to be nonlinear, we will attempt to describe the cause of this nonlinearity.

If the project is successful, we expect to produce a significant data set that can be used to guide calculations that predict the limits of the ground-motion response of repository structures during large earthquakes.

Mission Relevance

This project investigates the feasibility of using a unique data set to assess the performance of a high-level nuclear waste repository in support of LLNL's energy-security and environmental-management missions.

FY04 Accomplishments and Results

We categorized the large amount of ground motion data and other information archived from LLNL underground tests and assembled a detailed database of this information. We compiled a similar database containing the location and availability of LANL and Sandia ground motion data, and an archive of analog recordings at DOE Nevada Operations Office. Our initial analyses of selected LLNL data recorded in tuff and alluvial rocks show that they are useful in constraining in-situ material properties governing failure and wave attenuation at high strains, including transition from highly non-linear to elastic wave propagation. We presented these findings at a DOE-sponsored workshop on extreme ground motions from earthquakes and at a DOE Yucca Mountain Project (YMP) workshop on ground motion saturation. The success of this feasibility study has led to follow-on funding from YMP.

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Geophysics Experiments on High-Powered Lasers

John J. Zucca

04-FS-031

Abstract

An area of research important for monitoring underground nuclear explosions is cavity decoupling, which has the potential to determine whether the signature from a seismic event indicates that the event was a nuclear explosion in an underground cavity. Using actual nuclear explosions for such research is precluded, and high-explosives tests produce inadequate data because pressure from the high-explosive gases confuses the results. We propose to investigate the feasibility of using high-power, laser-driven experiments to create scaled explosion systems to study cavity decoupling. We have the opportunity to demonstrate this concept of scaled geophysics experiments at the Z-Laser at Sandia National Laboratory (SNL) in Albuquerque, NM. This study will demonstrate that such scaled experiments are possible, paving the way for larger-scale experiments to explore the critical issues in cavity decoupling using fusion-class lasers.

Mission Relevance

This study supports LLNL's national-security mission by answering questions on cavity decoupling, a high-priority topic in global nuclear explosion monitoring.

FY04 Accomplishments and Results

The proposed laser experiment will have two targets, one fully coupled and one fully decoupled. Initial calculations indicate that the pressure pulse near the outside of the rock-like material (Macor) in the target would show a large difference in peak amplitude between the fully coupled and fully decoupled targets. In FY04 we completed the preshot calculations, the detailed design of the experiments, and the manufacture of the test assemblies.

Proposed Work for FY05

The effort for FY05 will be to conduct the laser shots at the Z-laser at SNL. Following the shots, the targets will be inspected at LLNL, data reduction and interpretation will be performed, and the final report completed. The ratio of the peak pressure recorded at the surface of the test units is based on detailed computational work completed in FY04. We expect that the measured increase in radius of the two units will also have about the same ratio, thus demonstrating the feasibility of using high-power lasers to conduct scaled earth-science experiments.

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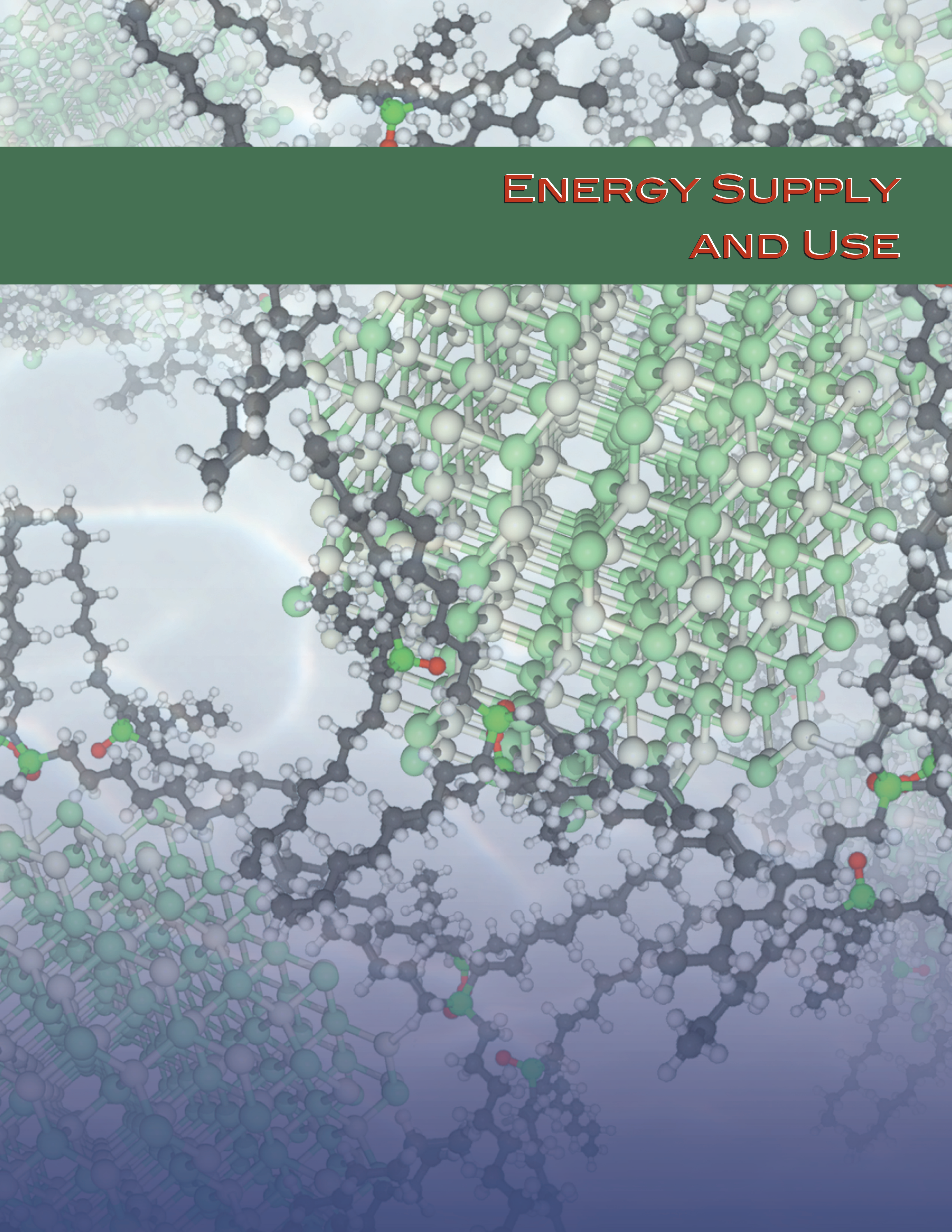
Energy Supply and Use



Energy Supply and Use

The Kinetic Stabilizer: A Route to a Simpler Magnetic Fusion System

Richard F. Post 02-LW-043

Abstract

This research is a study of a new stabilization concept, the kinetic stabilizer (KS), as applied to magnetic-mirror fusion systems. The KS stems from theory by Ryutov, who showed that magnetohydrodynamically- (MHD-) unstable, axisymmetric mirror systems are stabilized by the presence of low-density plasma in the field outside the mirror. The KS allows the concept to be used to make tandem-mirror (TM) fusion systems that are much simpler than present systems. The KS employs ion beams injected into the magnetic field outside the mirror. Compression and reflection of these ions forms the stabilizer plasma. The basic feasibility of the KS having been shown, we are now using analyses and computer codes to verify and extend these results in preparation for proof-of-principle experiments.

Success in these studies could result in a major change in the prospects for fusion power. Magnetic fusion research has been dominated by a search for ways to handle plasma turbulence, which is endemic to closed approaches such as the tokamak. Turbulence determines the scale of such systems and impedes theoretical prediction of performance, requiring an empirical approach involving increasingly large and expensive apparatus. The KSTM offers the possibility of fusion power systems that are simpler but are also amenable to prediction, in that performance would not be dominated by turbulence. The studies should provide a firmer theoretical basis for designing fusion-relevant experiments, leading to a fusion experimental reactor that could be much faster to construct than a tokamak such as ITER.

Mission Relevance

The KS concept aims at a pressing need expressed by the DOE Magnetic Fusion Program for simpler and faster-to-develop approaches to fusion energy in support of LLNL’s energy-security mission. The KS would allow MHD-stable operation of tandem mirror fusion power systems whose magnetic fields are axisymmetric (i.e., ones that are generated solely by circular magnet coils). Such systems should be able to exploit fully the potential of magnetic confinement, something not possible with other systems. In addition to the fusion power application of the KS, the stabilization of axisymmetric mirror systems will open up other new possibilities, including design of compact pulsed-compression neutron sources of interest to other NNSA missions.

FY04 Accomplishments and Results

The FLORA MHD stability code was used to demonstrate KS stabilization of fusion-relevant TM “plug” plasmas with beta values of 40%. A new KSTM transport code, SYMTRAN, was written; it predicted deuterium–tritium fusion ignition and burn in KSTMs with magnetic fields and plasma radii much smaller than those for tokamak systems such as ITER. A Fokker-Planck code, developed by the Japanese for their TM studies, was received from them and operated on LLNL computer systems. Plasma conditions required for the avoidance of

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certain non-MHD residual instability modes were established and new operating modes of the KSTM were explored, aimed at better avoiding these residual instability modes. Overall, these studies more firmly established the fusion-relevance of the KSTM concept.

Publications

Post, R. F. et al. (in press). "Axisymmetric tandem mirrors: Stabilization and confinement studies." *Trans. Fusion Sci. & Tech* **47**, 49. UCRL-CONF-205283.

Post, R. F. et al. (2004). *Axisymmetric tandem mirrors: Kinetic stabilizer studies*. Presented at the Innovative Confinement Concepts Workshop 2004, Madison, WI, May 25–28, 2004. UCRL-ABS-203437.

Molecular Engineering of Electrodialysis Membranes

William L. Bourcier 03-ERD-060

Abstract

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

Our goal is to develop molecular separations methods based on a molecular-level understanding of membrane transport processes. Our methodology will be applicable to a variety of separation needs in water treatment. We also expect numerous industrial applications, including processing the water produced during extraction of fossil fuels and treating the large volumes of water used in power plants. Our target application for this project is nitrate-specific, energy-efficient membranes used to remove nitrate from contaminated water supplies. We plan to synthesize membranes whose design is based on our models of ion transport through metallized or functionalized charged membrane pores. We will test the membranes in laboratory and field settings. We then hope to license the technology to the water treatment industry.

Mission Relevance

In addition to supporting the Laboratory's role in environmental management, this effort will contribute to LLNL's homeland security mission by developing a means to protect water, a resource that is vital to national security. To meet the goals of a safe, economic supply of water, it is important that clean water be generated in a way that minimizes energy use. Technologies developed under this research project would provide clean water in a cost-effective manner for the U.S. Future applications of this technology may target removal of nuclear, biological, and chemical species from water supplies contaminated during warfare or from terrorist attacks.

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FY04 Accomplishments and Results

In FY04, we (1) developed a new numerical method for calculating electrostatic fields near charged nanostructures, and a new method for calculating dielectrophoretic forces on molecules, which resulted in two publications and a record of invention; (2) synthesized nanopores of controlled size in polycarbonate sheets using ion track etching, gold-coated these materials, and applied an external charge to the surface; (3) developed test procedures for measuring membrane resistivity and selectivity for ion transport; (4) measured the transport rates of salts through membranes that demonstrated continuous through-going pores and ion permselectivity; and (5) set up and tested a benchtop electrodialysis unit into which we will place our up-scaled nanoporous membranes for field tests.

Proposed Work for FY05

FY05 work will include (1) continuing the fabrication and testing of nanoporous polycarbonate membranes, with emphasis on optimizing species selectivity and maximum permselectivity, development of durable metallized coatings, membrane functionalization, and testing in mixed electrolytes; (2) developing new modeling capabilities to determine electrostatic and dielectrophoretic forces on molecules near the membrane pore surfaces that incorporate quantum descriptions of the target species; (3) carrying out simulations of ion–membrane interactions that will guide the design, synthesis, and testing of new membrane geometries; and (4) scaling up membrane synthesis for proof-of-concept testing in a benchtop electrodialysis unit.

Publications

Schaldach, C. M. et al. (2004). "Dielectrophoretic forces on the nanoscale." *Langmuir* **20**, 10744. UCRL-TR-203675.

Schaldach, C. M. et al. (2004). "Electrostatic potentials in the vicinity of engineered nanostructures." *J. Colloid and Interphase Chemistry* **275**, 601. UCRL-TR-203010.

Schaldach, C. M. et al. (2004). *Manipulation of molecules by active nanostructures*. Presented at the American Membrane Technology Association Annual Meeting, San Antonio, TX, Aug. 2004. UCRL-POST-203469.

Persistent Monitoring Platforms

Charles L. Bennett 03-ERD-076

Abstract

In this project, we plan to build and test a model of the power plant for a stratospheric aircraft powered by thermal energy from the sun. Such an aircraft could maintain station over a designated ground location almost indefinitely, because it would not need fuel. We are developing a thermally coupled system with an efficiency nearly an order of magnitude better than the state of the art (Helios) by creating the technology for a sun-tracking solar-heat collector, thermal storage reservoir, and a high-efficiency heat engine. We will also develop physics models for thermal transport, materials interactions, loss mechanisms, and engine performance in the stratosphere's environment.

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We will develop and validate the physics models to prove the principles involved in a solar thermal-powered aircraft in preparation for constructing a prototype scale-model aircraft to demonstrate station-keeping capability at sea level. Successful demonstration of this technology would set the stage for construction of a stratospheric-altitude prototype capable of circumnavigating the globe.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04, we constructed a lithium hydride (LiH) thermal battery core and developed a layered containment structure that can safely contain high-temperature, reactive LiH in equilibrium with a significant pressure of hydrogen gas. This technology could enable a thermal battery based on LiH, which would have a specific energy capacity more than an order of magnitude higher than the most advanced rechargeable electric batteries. We developed a comprehensive suite of physics models for each of the components in the thermal-powered aircraft. This modeling study culminated in the submission of a patent application broadly covering the multiple aspects of a high-efficiency solar-powered aircraft.

Proposed Work for FY05

In FY05, we have two primary goals. First, we will develop a heat engine whose thermal efficiency is at least 60% of the Carnot limit. To this end, we plan to closely couple computational fluid-dynamics models with experiments on a prototype Stirling engine. Second, we will conduct laboratory experiments on the LiH thermal battery invention developed in FY04. We will use nondestructive evaluation techniques to monitor, in real time, the cooling characteristics of the thermal battery as it discharges, and compare experimental observations with theoretical predictions.

Metal-Containing Organic and Carbon Aerogels for Hydrogen Storage

Joe H. Satcher 04-ERD-053

Abstract

The goal of this project is to prepare metal-loaded carbon aerogels (MCAs) that meet or exceed the target (6 wt% H_2) set by DOE for the storage and transportation of hydrogen. Though carbon nanostructures are well suited for H_2 storage due to their low molecular weight, tunable microporosity, and high surface areas, fundamental understanding of the interaction between H_2 and the carbon adsorbant is needed in order to optimize the H_2 storage properties of these materials. We used nuclear magnetic resonance (NMR) techniques

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to elucidate H_2 uptake and release in MCAs. Results were used to identify structural features critical to the adsorption and desorption of H_2 over a wide range of temperatures and pressures and to synthesize MCAs with optimized properties for H_2 storage.

Our work with these novel MCAs will provide significant insight into the nature of hydrogen-carbon-metal interactions and help optimize the next generation of carbon-based H_2 storage materials.

Mission Relevance

Hydrogen storage is an important component of the national initiative to develop alternate energy sources to reduce America's dependence on oil. Competing with oil as an energy source, however, requires the ability to transport and utilize H_2 stored at or above the 6 wt% H_2 target set by DOE. Because current technology falls well short of this target, this research on the design of new H_2 -storage materials is an important component of LLNL's energy-security mission.

FY04 Accomplishments and Results

In FY2004, we synthesized a variety of MCAs, using iron, cobalt, and nickel, and characterized the structural properties of these materials using gas adsorption, electron microscopy and x-ray diffraction techniques. This characterization showed that the materials possess high surface areas (~700 m^2/g) and contain metal nanoparticles intimately mixed with graphitic structures. We have begun interactions with researchers at the National Renewable Energy Laboratory (NREL), who are currently studying the H_2 storage properties of these materials. These interactions have led to our incorporation in the DOE Center for Hydrogen Storage at NREL with funding beginning in FY2005 for conducting NMR experiments that examine hydrogen interactions with our MCAs.

Hydrogen Absorption in Fluids: An Unexplored Solution for Onboard Hydrogen Storage

Gene D. Berry 04-ERD-075

Abstract

This project's goal is to determine the basic feasibility of H_2 absorption storage for automobiles. Multiple hydrogen storage approaches have been examined since the 1970s, but each faces fundamental limits of weight (metal hydrides), volume (compressed gas storage), or poor thermal endurance liquid hydrogen. We have begun studying the feasibility of a new automotive H_2 storage approach: H_2 absorption in liquid nitrogen. The concept promises to store hydrogen more compactly than compressed-gas storage, in a vessel that is lighter than metal hydrides, and with a substantially greater thermal endurance than liquid hydrogen tanks. The fundamental physical phenomena underlying these advantages are the attractive van der Waals forces between H_2 and N_2 molecules, which are stronger than the H_2 to H_2 interactions or the N_2 to N_2 interactions.

Recent advances in lightweight materials, hybrid-electric and/or fuel-cell drive trains have now made it conceivable to eliminate automotive air pollution, greenhouse gases, and

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petroleum use with efficient (~60 to 100 mpg equivalent) H_2 automobiles. Storing enough fuel (4–7 kg H_2) on these automobiles for a 400-mi driving range is perhaps the greatest challenge. The ultimate choice of onboard storage impacts not only vehicle design and performance, but also has strategic consequences for the scale, shape, and energy requirements of the H_2 refueling infrastructure.

Mission Relevance

This project supports LLNL's carbon management mission by developing a new option for hydrogen as a carbonless transportation fuel. Hydrogen absorption in fluids will enable safer and more practical hydrogen storage on board vehicles. Adoption of such vehicles could ultimately eliminate greenhouse gas emissions from automobiles and the Nation's reliance on imported petroleum, thus improving U.S. long-term energy security.

FY04 Accomplishments and Results

We measured the molar volumes of H_2/N_2 mixtures at hydrogen mole fractions of 0.50, 0.60, and 0.70 at room temperature (273 K) and pressures up to 30,000 psi. Upon cooling to the boiling points of argon (87 K) and nitrogen (77 K) we observed that the pressure dropped by a factor of 8 to 10, indicating deficiencies in the way the attractive force are represented by current calculation techniques. Our results also verified rapid absorption of H_2 into liquid nitrogen at these temperatures. Measurements of overall mixture density at 77 K and 87 K for 0.70 mole fraction H_2 mixtures indicate the potential to store 30 to 45 kg H_2/m^3 at pressures of 1750–5500 psi. This meets the 2010 volumetric H_2 storage goal set by DOE. Experiments using liquid nitrogen cooled to ~65 K indicate pressures can be further reduced to 1250–3200 psi.

Interaction of Viruses with Membranes and Soil Materials

Charlene M. Schaldach 04-ERD-079

Abstract

This project is aimed at determining the binding energies of a specific viral particle to a charged surface in a fluid environment. We determine the viral-surface interaction by calculating (1) the direct electrostatic interaction between charges on the material surface and intrinsic viral charges; (2) the hydrophobic force; (3) surface charge polarization; and (4) the viral particle-surface van der Waals and electron-overlap repulsion forces. We have developed a model of the forces between a virus (the nonpathogenic cowpea virus) and a charged surface in a fluid of varying pH and ionic strength. We calculate the force as a function of distance from a Gouy-Chapman plane, varying the ionic strength of the fluid. Polarization of the plane by the virus is included. Van der Waals and electron-overlap repulsion interactions appropriate to gold surfaces will be employed and comparisons with atomic force microscopy experiments at LLNL will be made. Calculations of the deactivation of the virus by the electric field from a

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charged surface will also be performed to assess the feasibility of designing active membranes to deactivate viral particles.

The scope of this project is broad. Many of the techniques developed here will apply to other viruses and to proteins known to cause membrane biofouling and ground water contamination, which are issues of vital importance to water quality.

Mission Relevance

Understanding and quantifying the fundamental forces governing the transport of viruses in the subsurface may make it possible to design better methods for virus collection and detection in water supplies, thus supporting the Laboratory's role in advancing water technology for national energy security and environmental management missions of the DOE. The techniques developed in this project will also find applications in support of the Laboratory's national security and homeland security missions.

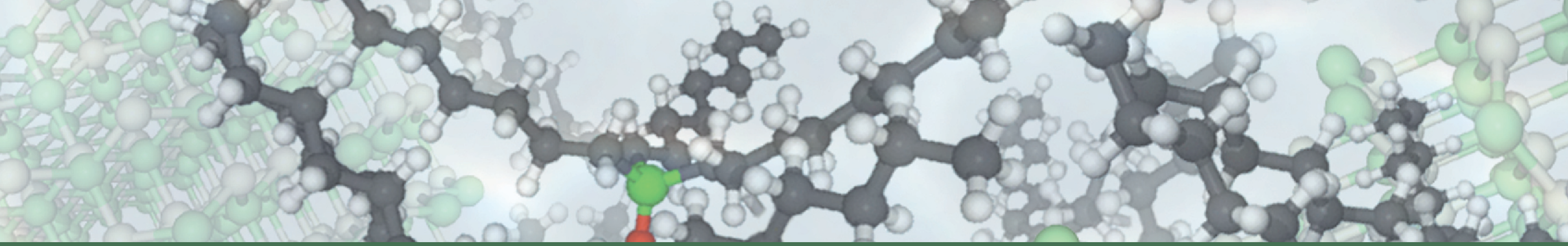
FY04 Accomplishments and Results

We developed a model of the forces between a virus and a charged surface in a fluid of varying pH and ionic strength. The virus is modeled as a particle comprised of ionizable amino acid residues surrounding a spherical RNA core of negative charge. We treat the surface as a Gouy-Chapman plane with an atomistic region of opposite charge, which is allowed to polarize in the field of the virus. Interactions are modified by the ionic strength of the fluid. Our model's results for virus surface potentials agree with experimental zeta-potential measurements. We extended the model to address deactivation of viruses by surfaces. These results indicate that we may be able to design tailored surfaces, optimized to deactivate viruses. We submitted a manuscript describing our results to a peer-reviewed journal.

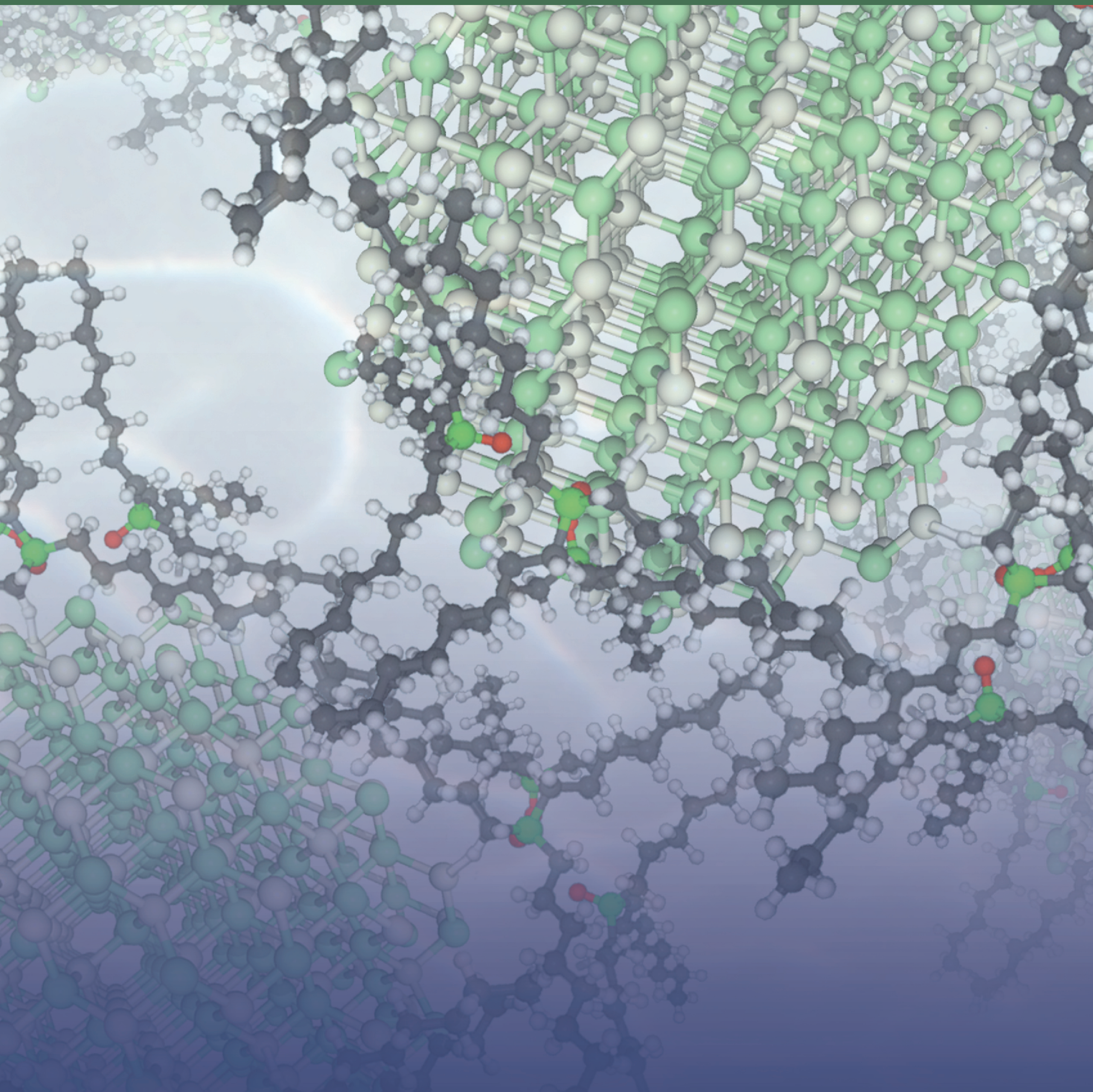
Publications

Schaldach, C.M. et al. (2004). *The influence of ionic strength on the interaction of viruses with charged surfaces under environmental conditions*. UCRL-JRNL-207332.

Viani, B.E. et al. (2004). *Electrostatic interaction of viruses with heterogeneously charged surfaces under environmental conditions,* Presented at the American Geophysical Union Meeting; San Francisco, CA, Dec. 13–17, 2004. UCRL-ABS-206427.



Engineering and Manufacturing Processes



Engineering and Manufacturing Processes

Extremely-High-Bandwidth Diamond Tool Axis for Weapons Physics Target Fabrication

Richard C. Montesanti

02-ERD-008

Abstract

This project advances the state of the art in high-bandwidth, precise motion-control mechatronic devices, and will enable the fabrication of three-dimensional (3-D) surface features needed for high-energy-density physics experiments on large lasers. Currently, LLNL is limited in its ability to fabricate the surfaces needed to provide quantitative corroboration of the physics codes being developed for stockpile stewardship. Ultraprecision, single-point machining with the fast tool servo (FTS) to be developed in this project will enable the fabrication of the desired features. The project will deliver a 10-kHz bandwidth, high-accuracy, high-stiffness servo axis designed specifically for these target-surface requirements.

The goal of this project was to advance the state of the art in high-speed precision machine tools—specifically, to develop a fast tool servo capable of producing a closed-loop motion of the tool tip at frequencies of up to 10 kHz while producing optically smooth surfaces on diamond-turnable materials. A 10-kHz bandwidth is an order of magnitude greater than commercially available devices and will enable fast production rates of the desired target surfaces and meet the cutting speed requirements of certain plastic target components. Besides the immediate application with high-energy-density physics targets, high-bandwidth motion control has application in the fabrication of precision optics that are non-axisymmetric or have short spatial wavelength variations.

Mission Relevance

A state-of-the-art capability in high-frequency, ultraprecision fabrication at LLNL supports the Laboratory's stockpile stewardship mission by enabling the fabrication of 3-D surface features needed on target components for high-energy-density physics experiments.

FY04 Accomplishments and Results

We successfully developed a 10-kHz rotary FTS capable of 10-µm peak-to-peak motion at low frequencies and 2.5-µm peak-to-peak motion at 10 kHz (500 g). The FTS acts as a high-speed axis for a diamond-turning machine, moving the tool towards and away from a workpiece in synchronization with the spindle rotation angle. This enables the fabrication of non-axisymmetric or textured surfaces. We also developed software tools based on first-principles physics and closed-form engineering equations and used them to design and optimize the highly integrated mechanical-magnetic-electrical systems for the actuator and its payload. We also filed the second of two patent applications resulting from this project. This project set the stage for future work that could entail assembling the hardware, characterizing FTS performance, and performing cutting tests with a diamond-turning machine.

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Publications

Montesanti, R. C. and D. L. Trumper. (2004). "Design and implementation of the control system for a 2 kHz rotary fast tool servo." *Proc. Am. Soc. for Precision Engin. 2004 Spring Topical Meet.* UCRL-PROC-203295.

Nanoscale Fabrication of Mesoscale Objects

Raymond P. Mariella 02-ERD-014

Abstract

Laser ablation produces periodic roughness patterns on material surfaces. Several applications of laser ablation (e.g., fabricating laser targets for physics experiments) would be enhanced if surface roughness could be reduced. In this project, we plan to create a technique for fabricating and characterizing mesoscale (millimeter-size) objects with micrometer-scale contours and nanometer-scale precision using femtosecond laser pulses and ion beam etching. This project will integrate LLNL expertise in precision engineering, microtechnology, and nondestructive characterization along with special capabilities in short-pulse laser–matter interactions and numerical simulation of material properties at multiple scales.

The immediate impact of our investigation will be a much better understanding of the chemical and physical processes that influence materials that are exposed to femtosecond laser pulses. More broadly, we expect to develop the capability for fabricating and characterizing mesoscale objects using femtosecond laser pulses and ion-beam etching for applications such as fabricating laser targets, developing miniature fuels cell, remote sensors, and medical technologies.

Mission Relevance

These experiments will support the Laboratory's national security mission by providing data for corroborating the models in improved physics codes for stockpile stewardship and by developing new capabilities at LLNL for the fabrication and characterization of mesoscale objects with nanoscale precision.

FY04 Accomplishments and Results

In FY04, we combined, for the first time, the one-dimensional (1-D) hydrocode HYADES with the 3-D molecular-dynamics simulator MDCASK in modeling studies. We investigated the laser ablation of carbon, including laser-enhanced chemical reaction on the carbon surface for both vitreous carbon and carbon aerogels. We studied the process both in a vacuum and in air. We used vitreous carbon because it is readily available and because it was the easiest carbon to model. Through our collaboration with Lawrence Berkeley National Laboratory, we also showed that oxygen-ion beams could micromachine smooth features in vitreous carbon. Finally, we were able to micromachine simple features in carbon aerogels using laser ablation.

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Publications

Gilmer, G. H. (2003). "Thin film deposition and manipulation of surfaces using laser beams: Atomistic modeling." Presented at the DOE-NSET Workshop on Artificially Structured Nanomaterials, Gatlingburg, TN, Oct. 13–15, 2003. UCRL-PRES-200845.

Gilmer, G.H. (2004). "Short-pulse laser ablation of metals: Large-scale molecular dynamics simulations." Presented at the 2nd Intl. Workshop on Strength and Fracture, Berkeley, CA, Jan. 7–9, 2004. UCRL-ABS-201337.

Gilmer, G. H., et al. (2004). *Laser ablation of metals: Large-scale atomistic simulations and experiments*. UCRL-JRNL-200163.

Mariella, R. (2004). *MEMS-based sensor systems*. Presented at the NNSA Future Technologies Conference, Washington, D.C., May 17–19. UCRL-CONF-204129.

Shirk, M. D. et al. (2003). "Mesoscale laser processing using excimer and short-pulse Ti: sapphire lasers."*Proc. Intl. Conf. on Applications of Lasers and Electro-Optics*, M702. UCRL-JC-154591.

Concealed-Threat Detection at Multiple Frames per Second

John T. Chang 02-ERD-061

Abstract

Personnel protecting special nuclear material and others responsible for homeland defense cannot easily detect hidden threats such as concealed weapons or persons behind walls. This fact defines a need for fast and accurate technologies to help search for concealed threats in real time. This project investigated the efficacy of real-time [10 to 30 frames per second (fps)] array imaging to rapidly detect hidden threats, such as weapons and people, through clothing, smoke, fog, or walls. The project first demonstrated that detection is measurably enhanced by real-time techniques, using simulations and statistical analyses. Next, we developed methodologies of data processing, using novel array beam-forming hardware and software that assist real-time human perception of a threat, and we designed, assembled, and tested a prototype array (radar or acoustic) to demonstrate detection capability. This research produced important information on the feasibility of a system for the rapid, accurate, real-time detection of hidden threats.

Mission Relevance

This project supports the Laboratory's national security and homeland security missions by demonstrating that an accurate, real-time array device can be developed to detect hidden threats.

FY04 Accomplishments and Results

In FY04, we developed a computational simulator system to evaluate and predict the performance of the imaging system. Prototype radar camera hardware was built and used to study and characterize the phenomena of ultrawideband (UWB) image generation. We demonstrated a laboratory-based imaging system that can represent images at approximately

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15 fps, and we demonstrated and quantified the effects of UWB beam forming and focusing ability. To enable beam steering, we researched array geometry and radiating element distribution. We successfully integrated the benchtop array with field-programmable gate arrays for both data acquisition and image representation. Finally, we documented quantitative improvements in perception studies with human subjects. Two records of inventions were submitted, and we plan to submit two manuscripts for publication in peer-reviewed journals.

Publications

Chang, J. et al. (2004). *Ultrawideband radar methods and techniques of through barrier imaging*. Presented at the IEEE APS/URSI, Monterey, CA, June 2004. UCRL-ABS-201914.

Eliassi-Rad, T. and E. Chow. (2004). *Using ontological information to accelerate search in large semantic graphs.* UCRL-CONF-202002.

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

Jerald A. Britten 03-ERD-059

Abstract

The objective of this project is to develop a state-of-the-art capability for producing meter-scale, multilayer dielectric (MLD) diffraction gratings for pulse compression in high-energy, petawatt-class (HEPW) laser systems. We plan to pattern dielectric coatings on monolithic substrates with grating lines approximately 0.2 µm wide, ~0.7 µm high, and ~0.5 µm apart, uniformly over areas of 1 m by 800 mm. This effort requires the development of tools and processes for large, heavy substrates and involves laser-interference lithography, reactive ion-beam etching, wet-chemical processing, and critical-dimension metrology.

The need for meter-scale, high-damage-threshold, pulse-compressor optics has been identified by all NNSA labs as the key HEPW technology hurdle. HEPW pulses would enable the generation of critical, high-energy x-ray backlighters; access to new states of matter via isochoric heating; and studies of physics relevant to a full-scale demonstration of fast ignition. Successful completion of this project will represent a breakthrough in producing large-scale diffractive optics for HEPW lasers in the U.S. and abroad.

Mission Relevance

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

FY04 Accomplishments and Results

We successfully demonstrated a full-aperture (800 × 400 mm) 1780-lines/mm multilayer dielectric grating having a diffraction efficiency of >90% and a wave-diffracted wavefront

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error of less than 1/4 wave in conditions of use (1053-nm wavelength, 73.5° incidence angle) over the entire aperture. We also developed processing technology to improve laser-damage thresholds and successfully demonstrated record laser-damage thresholds on witness gratings at use conditions—up to 4.5 J/cm^2 (normal incidence fluence) at normal incidence for 1053 nm, incident at 76.5° at a pulse length of 10 ps.

Proposed Work for FY05

We will (1) develop densified, multilayer dielectric coatings that will be more environmentally insensitive and more durable to withstand the rigors of grating manufacture; (2) develop a 351-nm lithography technology to replace our existing 413-nm system, which will allow us to make even bigger gratings with the same collimating optics; (3) continue to improve the designs and laser-damage resistance of MLD gratings; (4) develop tools to better collimate our exposure system and to measure gratings at the angle and wavelength of use, with the aim of improving wavefront flatness of the diffracted beam; and (5) complete the tools and processes required for the ultraclean priming and photoresist coating of large-grating substrates.

Publications

Britten, J. A. et al. (in press). “First demonstration of a meter-scale multilayer dielectric reflection grating for high-energy petawatt-class lasers.” *Optics Letters*. UCRL-JRNL-205887.

Britten, J. A. et al. (in press). “Multilayer dielectric gratings for petawatt-class laser systems.” *Proc. SPIE* **5273**. UCRL-CONF-154956.

Acoustic Characterization of Mesoscale Objects

Diane J. Chinn 04-ERD-013

Abstract

Mesoscience is an emerging area of science that focuses on the study of millimeter-size objects with micrometer-size features. The goal of this project is to research acoustic characterization of mesoscale objects using noncontacting laser ultrasonic techniques (LUT). Although LUT frequencies of 100 MHz to 10 GHz are required to acoustically characterize features ranging in size from 0.5 to 5 µm, LUT in this frequency range is not currently available.

The main research goals of this project are threefold: (1) Establish gigahertz acoustic attenuation relationships for laboratory materials at distances to 1 mm to confirm acoustic testing as a viable method for mesoscale characterization, (2) extend our acoustic models to mesoscale dimensions, and (3) research laser-generated gigahertz ultrasound modeling and hardware for mesoscale characterization. Achieving these goals will broaden the field of acoustic testing by filling the existing gap in acoustic characterization capabilities. Work in this area has never before been performed. Our results will be reported in nondestructive-testing and material-evaluation journals.

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Mission Relevance

Advances in this promising technique will provide an acoustic characterization tool for many mesoscale applications related to LLNL missions. For example, this work will benefit high-energy-density physics efforts that support LLNL's stockpile stewardship mission. Other applications could support LLNL's energy-security mission by enabling developments in fuel cells or providing understanding of geochemical processes. Study of tissue and cell abnormality would support LLNL's mission in bioscience to improve human health.

FY04 Accomplishments and Results

The primary objective for FY04 was to determine the viability of mesoscale ultrasound by measuring propagation distances for materials of interest. We assembled a set of 27 samples including metals, polymers, and aerogels of various thicknesses chosen to represent potential mesoscale applications. Material-attenuation coefficients, the parameters by which propagation distance can be modeled, were measured using gigahertz immersion ultrasonic testing equipment at Oak Ridge National Laboratory's High-Temperature Materials Laboratory and LUT equipment at Boston University. The LUT system gave very promising results: Propagation distances greater than 0.2 mm were achieved in most metals and polymers. These results establish viability of LUT as a characterization tool for mesoscale objects.

Proposed Work for FY05

Our objectives for FY05 are to model gigahertz wave propagation resulting from a thermoelastic source and build a prototype gigahertz-laser acoustic microscope based on the preliminary design made in FY04.

Publications

Huber, R. D., D. J. Chinn, and D. H. Chambers. (2004). "Optical mapping of the acoustic output of a focused transducer." *Proc. 147th Acoustical Society of America Meeting*. UCRL-ABS-201964.

Development of a High-Stiffness Hybrid Passive/Active Magnetic Bearing for Precision Engineering Applications

Lisle B. Hagler 04-FS-001

Abstract

The objective of this research was to demonstrate the feasibility of a dynamically stable, high stiffness, hybrid passive/active magnetic bearing with low error motion for ultraprecise machining of parts. This project, which leveraged previous work on passive magnetic bearings performed at LLNL in connection with flywheel energy storage, achieved error-motion tolerances of 50 nm or better by constructing a practical, dynamically stable, stiff, hybrid passive/active magnetic-bearing system. The system consists of a passive magnetic bearing coupled with separate, actively controlled electromagnetic steering for high precision.

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This research produced a preliminary design for an entirely new class of bearing—a totally passive magnetic bearing with high stiffness and high rotor-dynamic stability, coupled with an electromagnetic steering device. This type of bearing will allow unprecedented precision in machining operations without frictional wear, making it maintenance free. This new technology will advance LLNL's state of-the-art precision-engineering capabilities to routinely manufacture parts at previously unattainable tolerances.

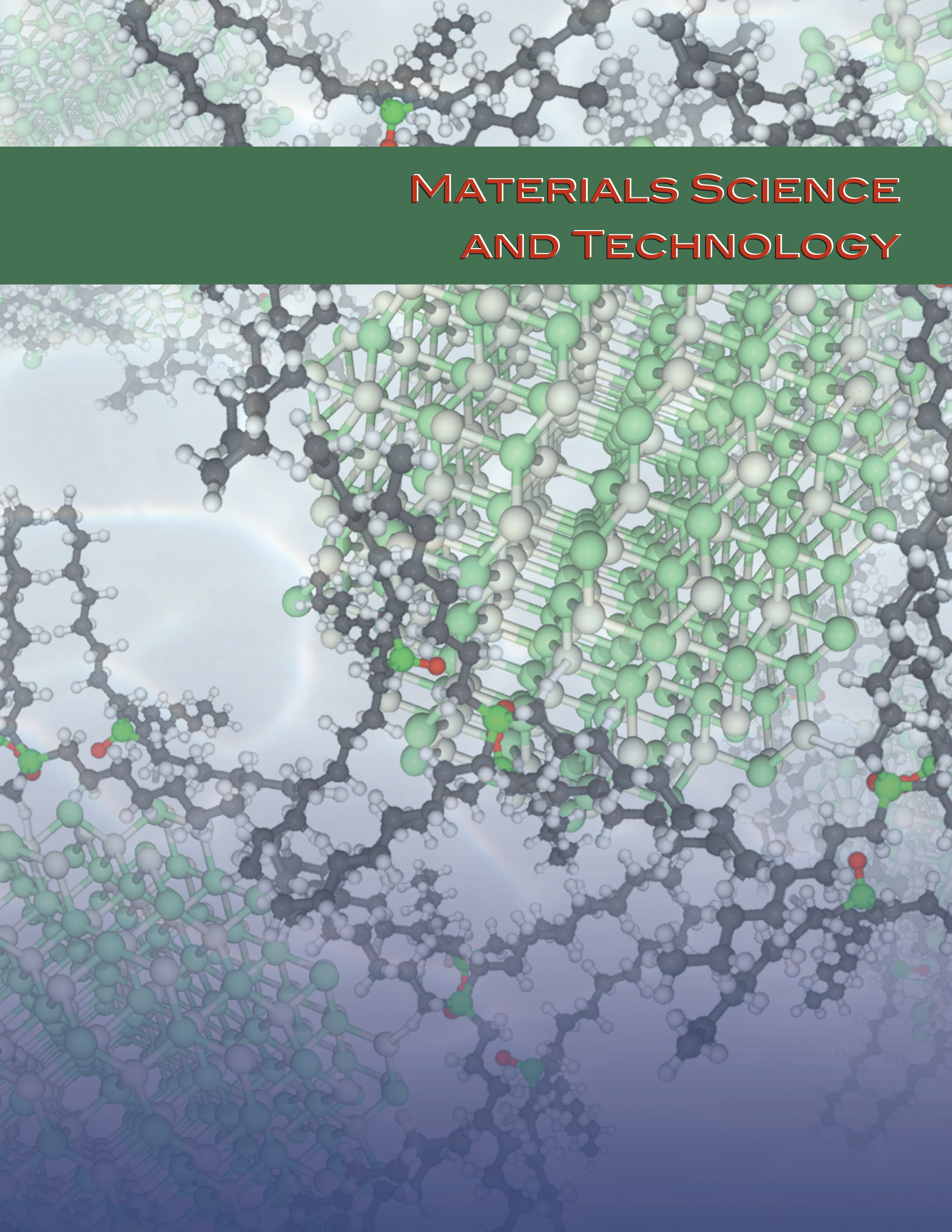
Mission Relevance

Applications of improved precision machining provide better mirrors for laser-optic systems, improved precision of fusion-class laser optical components, and high-energy density physics targets in support of Laboratory missions in national security. In addition, applications for improved telescope mirrors and optical components support LLNL's basic science mission.

FY04 Accomplishments and Results

This study has shown, theoretically, that rotor error motions due to mass imbalance and internal/external excitation sources can be rendered negligible. Additionally, we developed a methodology to initially align the rotor to meet our tolerance constraints. At the same time, we developed an active control scheme to take care of the residual rotor error motion. Finally, the project produced a preliminary design of a prototype machine employing a hybrid magnetic bearing.

Materials Science and Technology



Materials Science and Technology

Spectroscopy of Shock-Compressed Deuterium

Neil C. Holmes 01-ERD-098

Abstract

This project will perform experiments to test and challenge the predictions of first-principles simulations of the molecular vibrational spectrum and optical properties of shocked deuterium (D2). Spontaneous Raman spectroscopy, using a crossed-beam method, will be used to investigate the frequency and line shape of the D2 vibration at high pressures; absorption spectroscopy will be used to observe the opacity at pressures below 13 GPa. Once these spectroscopic techniques are demonstrated with single shocks at the gas gun, they will be transferred to laser experiments for use at higher pressures.

We expect to develop a new capability to perform Raman and absorption spectroscopy under shock compression in fluids at high temperature and pressure to elucidate the molecular properties of such compressed fluids. This project will result in the first Raman measurements on a shocked cryogenic sample. An experimentally validated theory of compressed hydrogen and D2 is important for modeling the performance of explosives and of capsules for future fusion-class lasers, and is also required for an accurate description of the interior structure and evolution of giant planets and brown dwarf stars.

Mission Relevance

The techniques developed in this project are fundamental for understanding explosives and the destruction of chemical weapons and for simulations of weapons and inertial confinement fusion experiments for stockpile stewardship. They also support LLNL's basic science mission by providing a new ability to conduct Raman and absorption spectroscopy under shock compression in fluid systems at high temperature and pressure.

FY04 Accomplishments and Results

In FY04, the spontaneous Raman diagnostic was completed, and static Raman signals were obtained from benzene and water. We completed target designs for benzene, liquid nitrogen, and liquid hydrogen and successfully fabricated benzene targets. A broadband white-light source for complementary optical absorption experiments was also completed.

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A Hydrophobic Silica Aerogel/Granular Activated Carbon Composite for Removing Arsenic and Hexavalent Chromium from Groundwater

Sabre J. Coleman 01-ERD-106

Abstract

Contaminants in water limit the quantity of potable water. As the population increases, it is important to increase the availability of drinking water. The objective of this project is to prepare and use composites made of functionalized hydrophobic silica aerogel with granular activated carbon (GAC) to adsorb As from groundwater. Our approach is similar to past efforts with composites for U and hexavalent Cr. Bench-scale testing, using flow-through columns, will be used to test the adsorbency of the composites. Preliminary testing of a composite looks promising for As removal from water. The optimization process will include determining (1) the appropriate balance of the functional groups, aerogel, and GAC and (2) the adsorptive capacity of the composites for As in simulated groundwater.

We expect to prepare at least one composite that can effectively adsorb As from water. Aerogel–GAC composites could be used at LLNL, other DOE sites, and Department of Defense sites as part of environmental remediation efforts or for treatment of drinking water. For example, a composite could be used to treat groundwater that is purged form a borehole as part of a well installation process.

Mission Relevance

By developing aerogel–GAC composites for removing As, U, and hexavalent Cr from groundwater, this work supports LLNL's mission in environment management. For instance, LLNL has a regulatory driven requirement to clean up contaminated groundwater at the Livermore Site and Site 300. The composites we develop can be incorporated into pump-and-treat activities at both sites.

FY04 Accomplishments and Results

In FY04 we produced two As-specific composite formulations, one with an Fe functional group and the other with Fe and Mn functional groups. We tested the composites by conducting comparison flow-through column testing of simulated groundwater with known low concentrations of As, in the range of 50 to 200 (μg/L). Using this method, we were able to adsorb more than 95% of the As contaminants, although we encountered some difficulties with stabilizing composite materials specifically formulated for As. After reformulation, we were able to successfully remove As. These results confirm that this composite is a feasible technology for the removal of low concentrations of As from water.

We collaborated with researchers at Arizona State University and Clemson University to (1) validate proof-of-concept testing for iron-enriched GAC and aerogel–GAC composites as viable adsorbents for removing As from groundwater and (2) conduct technical and economic feasibility assessments of these innovative processes.

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The Properties of Actinide Nanostructures

Alex V. Hamza 02-ERD-025

Abstract

The project goal was to create an experimental tool to provide a predictive description of the structural and electronic properties of actinide materials such as Pu. Band-mapping experiments on actinide materials are needed to benchmark current band-structure theories, and studying clean actinide nanostructures is key to meeting this goal. Earlier work on this project produced actinide nanostructures by laser ablation, but due to their high reactivity, initial oxidation always remained. For FY04 we sought to produce unoxidized actinide nanoparticle films, create an environment in which their lifetime is maximized, and analyze these films with photoemission and scanning probe techniques to characterize the active oxidation processes.

The properties of the 5f elements represent one of the last frontiers of condensed-matter physics. Through proper manipulation of the ablation environment, we hope to eliminate the initial oxidation encountered in our earlier studies. If successful, the project will produce clean actinide films suitable for studying the characteristics of uncontaminated material. As laser ablation inherently produces nanoscaled material in a range of sizes, the study of 5f electron-correlation effects in a quantum confinement regime becomes possible if the sample lifetimes can be sufficiently extended. As one of the most complex aspects of the actinide elements is the correlated behavior of the 5f electrons, such studies could define the contributions of the various electron-correlation effects.

Mission Relevance

This project contributes to LLNL's long-term goal of predicting Pu behavior over tens of years, in support of the stockpile stewardship mission. A confirmed electronic structure for Pu will make this possible by enabling predictions of vacancy mobility and dislocation motion.

FY04 Accomplishments and Results

The first laser deposition of a clean actinide thin film (160 nm of depleted U) was achieved. Oxidation levels of the freshly deposited films are equivalent to the best U films grown to date and are measurably oxygen-free immediately following ablation. A study of the in-vacuum oxidation of these films was carried out and the results modeled. Films were allowed to oxidize fully and were found to have good adhesion where oxidation occurred at reduced pressure. Exposure to air and subsequent oxidation of the films at atmospheric pressure produced stresses in the films which separated them from the substrate.

Publications

Glade, S. C. et al. (2004). "Characterization of uranium particles produced via pulsed laser deposition." *Matls. Res. Soc. Symp. Proc.* **802**, DD5.5.1. UCRL-PROC-200965.

Trelenberg, T. W. et al. (2004). "Instrument for the investigation of the nanostructure of

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Pu." *Rev. Sci. Instr.* **75**, 713. UCRL-JC-152913.

Trelenberg, T. W. et al. (2004). *Techniques for the study of actinide nanostructures*. UCRL-ABS-204282

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Rapid Resolidification in Metals using Dynamic Compression

Frederick H. Streitz 02-ERD-033

Abstract

We investigated the effect of kinetics on the rapid resolidification of metals under compression by designing and performing novel time-resolved dynamic compression experiments using the LLNL light gas gun (LGG). The project focused on the rapid solidification in molten Bi and water. We (1) designed isentropic compression experiments with highly optimized layered impactors; (2) developed novel, time-resolved diagnostic tools for use with the LGG facility; and (3) performed atomic-scale simulations of the resolidification process. Creating an isentropic compression capability for the LGG represents a major new experimental capability.

The primary deliverable of this project is the ability to experimentally control pressure and temperature loading rates at the LGG (and elsewhere) by using highly optimized layered impactors constructed from graded-density material. This capability has substantially broadened the range of phase space that can be accessed with dynamic compression experiments and will eventually lead to a better understanding of the dynamic response of materials, including the kinetics of phase transitions in nonequilibrium situations.

Mission Relevance

Kinetic limitations play a large role in the dynamics of solidification during freezing, but the role of kinetics in solidification at high temperatures and pressures is not understood. Detailed knowledge of such transitions under highly nonequilibrium conditions is critical for LLNL's stockpile stewardship mission. The experiments conducted in this project will improve our interpretation of equation-of-state (EOS) experiments and our ability to predict phase behavior and material strength at high pressures and temperatures relevant to the Stockpile Stewardship Program.

FY04 Accomplishments and Results

We continued our study of rapid resolidification in molten Bi and water using graded-density flyers to produce near-isentropic compression with the LGG. We have quasi-isentropically compressed molten Bi to pressures that would solidify the sample; however, the time scale of resolidification and imprecise nature of our diagnostic precluded a distinct signature of solidification. Computer simulations also indicate the possibility of very long time scales for metal solidification. Our experiments in water have unambiguously demonstrated solidification during quasi-isentropic compression. We have tentatively identified the time scale for this solidification and have mapped out a part of the dynamic melt curve for water.

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Publications

Nguyen, J. H. et al. (2004). *High-pressure tailored compression: Controlled thermodynamic paths.* UCRL-JRNL-201279.

Nguyen, J. H. et al. (2004). "Specifically prescribed dynamic thermodynamic paths and resolidification experiments." *AIP Conf. Proc.* **706**, 1225 (Shock Compression of Condensed Matter, July 20-25, 2003, Portland OR). UCRL-JRNL-201169.

Streitz, F. H. et al. (2004). *Rapid resolidification in metals: Simulation and experiment*. Presented at the Workshop on Multiscale Modeling of Strength and Fracture, Berkeley, CA January 7-9, 2004. UCRL-ABS-201390.

Magnetic Transition Metals and Oxides at High Pressures

Valentin Iota-Herbei 02-ERD-046

Abstract

We are investigating the electronic and magnetic properties of 3d transition metals and oxides at high pressures, with a focus on determining spin transitions and the magnetic phase diagrams of Fe, Co, and Ni. In addition, we are using the same experimental techniques to investigate Mott electronic transitions in simple 3d oxides (MnO, FeO, CoO, and NiO). To obtain electronic structure information, we are adapting, for use in a diamond anvil cell, two new x-ray spectroscopy techniques sensitive to the configuration of the valence band: x-ray emission spectroscopy (XES) and x-ray magnetic circular dichroism (XMCD). The experiments are being carried out at the Advanced Photon Source (APS) at Argonne National Laboratory.

This study delivers detailed understanding of the pressure-induced changes in the electronic structure of transition metals and oxides, including pressure changes in the valence band spin polarization (spin state). By monitoring changes in the spin state at high pressure, we will construct the magnetic phase diagrams for Fe, Co, and Ni, and study the systematics of magnetism suppression in these 3d transition metals. Our measurements on Fe and Fe oxides provide physical data for Earth-core models, directly addressing the incompletely understood issue of the origin and stability of the geo-dynamo.

Mission Relevance

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

FY04 Accomplishments and Results

In FY04 we developed XMCD measurements at high pressures, using partially drilled diamond anvils. Using this method, we performed highly accurate XMCD and extended x-ray absorption fine structure measurements in Fe (to 24 GPa), Co (100 GPa), and Ni (80 GPa). Our results show that magnetism suppression in Fe starts at the structural phase

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transition, but that a small magnetic moment persists to 24 GPa, well into the high-pressure hexagonal close packed (hcp) phase. By contrast, in Co, the reduction in magnetism occurs in the low-pressure (i.e., hcp) phase preceding the structural transformation to face-centered-cubic structure, at around 100 GPa. An extrapolation of our results to higher pressures predicts the suppression of magnetism in Ni above 100 GPa and suggests the existence of a nonmagnetic phase of Ni.

Proposed Work for FY05

For FY05 we propose to complete the XMCD measurements on the 3d magnetic metals under pressure. Our plan is to increase the maximum pressure to >100 GPa to confirm the existence and stability domain of the nonmagnetic phase of Ni and to establish in detail the nature of the electronic changes observed in the high-pressure phase of Co. Second, we will study the Mott transition in FeO by XES measurements up to 150 GPa. The existence and pressure boundary of the metallization of FeO are highly controversial and are of crucial importance to understanding the origin of Earth's magnetic field. Third, we will develop low-temperature XMCD measurements at APS for studying magnetism in f transition metals.

Exchange Coupling in Magnetic Nanoparticles Composites to Enhance Magnetostrictive Properties

Harry B. Radousky 02-ERI-006

Abstract

This project is investigating the basic materials properties of magnetostrictive materials which have a variety of applications as actuators, sensors and damping materials. Our goal is to create coated nanoparticles and embed these particles in elastomer composites using the techniques of spark erosion and chemical synthesis. This project will provide insight into the spark-erosion process, investigate the physical and magnetic properties of the particles as a function of size, and study the properties of the composites. In the final year of the project, we added the goal of coupling magnetostrictive and thermostrictive materials to piezoelectric materials to create compound thermoelectrics for energy-harvesting applications.

The results of this project will enhance our understanding of the magnetostrictive response of magnetoelastomers containing small particles. Our results will validate the concept of using hollow particles in these materials and will give us an understanding of the spark-erosion process used to create these types of particles. We will also validate the concept of using compound thermoelectric materials for energy-harvesting applications, as well as an improved understanding of the magnetostrictive and thermostrictive materials that are used.

Mission Relevance

This work supports LLNL's mission in breakthrough science, particularly as related to the design, simulation, synthesis, processing, and properties of existing and novel materials. Potential applications to other mission areas include improved actuators for Department of Defense (DoD) applications, potential microelectromechanical devices for health-care

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technology, and compound thermoelectrics for a variety of DOE and DoD energy-harvesting applications.

FY04 Accomplishments and Results

In FY04, we completed our work with Ni and Fe-Ga composites. The results for the hollow Ni and Fe-Ga particles was published in *Applied Physics Letters* (September 2004) and the *Journal of Applied Physics* (August 2004). We optimized the spark-erosion process for controlled production of a wide range of materials in particulate form and published the results in the *Journal of Applied Physics* (February 2004). We also completed a study of the $Gd_5Si_2Ge_2$ system and demonstrated its viability as a volume magnetostrictive composite. This result was published in *Applied Physics Letters* (June 2004). A spinoff of our work on $Gd_5Si_2Ge_2$ resulted in the creation of a new type of material we call a compound thermoelectric, and a provisional patent was filed.

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

Jeffrey D. Colvin 03-ERD-018

Abstract

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

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

Mission Relevance

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

FY04 Accomplishments and Results

In FY04 we (1) fabricated a number of four-layer targets with different initial grain sizes using a vapor-deposition technique; (2) designed, fabricated, and tested a jig to hold the target and facilitate the preheating and post-shot active cooling of the target; (3) performed several test shots with the Trident laser at Los Alamos National Laboratory, which led to some modifications in the detailed target design and the heating and cooling system; (4) completed additional shot series with Trident and an LLNL laser; and (5) began post-shot analysis of the recovered samples and the in situ diagnostic data. This includes the first-ever data from a nanosecond-scale resistivity diagnostic that was designed and fabricated as an integral part of the target.

Proposed Work for FY05

We will extend our knowledge about how a very rapidly cooled elemental metal liquid structures itself on resolidification to investigate how a metallic alloy would behave differently. (The behavior of alloys could be different and more complex than that of pure metals, because the alloy presents different nucleation sites for resolidification.) We will therefore conduct a similar set of experiments with pure Bi and a Bi-Sn alloy, or some other suitable Bi alloy, and publish the results of these experiments.

Publications

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A Two-Particle Formulation of Electronic Structure

Antonios Gonis 03-ERD-064

Abstract

The problem of correlated electrons in elemental solids and alloys figures prominently in the Laboratory's efforts to understand the behavior of Pu metal and its alloys and to predict such properties as phase and dimensional stability under self-irradiation and as a function of time. We have developed and are model-testing a formal approach that combines the two diverse methodologies customarily used to study the electronic structure of solids: band theory and many-body theory. This is done through the study of two-particle units rather than single particles, as is done presently. The codes we develop will thus simulate the Coulomb interaction and its effects on materials properties with greater accuracy than is possible with traditional methodology.

This work will provide theoretical and practical electronic structural information about important materials and systems for which conventional methods are currently inadequate, particularly complex systems such as Pu and its alloys. In the long term, we will develop realistic models for the study of heavy elements and their alloys and apply them to materials central to the Laboratory's mission. Further applications to the electronic properties of materials, such as transport, will become more directly accessible than at present, taking account of electron–electron interactions more accurately than is currently possible.

Mission Relevance

Because understanding the electronic structure and properties of materials, especially actinides, is crucial to nuclear science and relevant for stockpile stewardship, this project supports the Laboratory's national-security mission.

FY04 Accomplishments and Results

In FY04, we developed the formal aspects of the methodology that combine band theory and many-body theory into a single coherent framework. Based on this methodology, we carried out tight-binding (TB) calculations of correlated electrons in model systems, both ordered and disordered, accounting both for the Coulomb interaction and disorder effects. We compared exact results for the density of states with those obtained within the two-

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particle approximation and found the agreement to be very good and much improved over that obtained in the single-particle picture.

Proposed Work for FY05

In FY05, we will calculate the x-ray photoemission spectrum of Ni, based on the most recent formulation of the theory, and compare our results with previous calculations. The self-energy and spectra will be computed for disordered model systems (of the type described above) so that we can assess their analytic properties, accuracy, and transferability. We will develop the computational framework for carrying out self-consistent calculations and will compare and contrast the new methodology with recently developed formalism for the study of correlation effects based on model calculations and realistic Hamiltonians.

Optics Performance at 1 omega, 2 omega, and 3 omega

John Honig 03-ERD-071

Abstract

The interaction of intense laser light with dielectric materials is a fundamental applied science problem that is becoming increasingly important with the rapid development of ever more powerful lasers. To better understand the behavior of optical components in large fusion-class laser systems, we are systematically studying the interaction of high-fluence, high-power laser light with high-quality optical components, with particular interest on polishing/finishing and stress-induced defects and surface contamination. We will focus on obtaining comparable measurements at three different wavelengths, 1ω (1053 nm), 2ω (527nm), and 3ω (351 nm). Modeling at both microscopic and macroscopic scales will support interpretation of the experimental results.

Materials research that enhances our understanding of the complex mechanisms describing the response of optical materials to intense laser irradiation will benefit worldwide efforts to field ever more powerful, fusion-class laser systems and allow them to operate efficiently and reliably at or above their design specifications. Our modeling efforts will in turn allow the operators of fusion-class laser systems to predict optics lifetime effects of upcoming experimental campaigns and enable campaign planning to be optimized to minimize incremental costs that results from having to replace optical components. Additionally, this effort will help advance and ensure LLNL's continued leadership in the fields of optical materials science and laser technology.

Mission Relevance

This work will add substantially to the knowledge base of optical materials under the intense laser illumination (i.e. fusion-class laser systems) and allow us to validate our theoretical and stochastic models. This work supports NNSA and Laboratory missions in stockpile stewardship by providing data that can be used to validate computer simulations related to the safety and reliability of the nuclear stockpile.

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FY04 Accomplishments and Results

In FY04, our systematic studies of defect initiation and growth at 2ω and 3ω in the presence of unconverted light provided essential data for validating our models. A model of the evolving defect state was developed using equations to describe defect initiation densities, growth parameters, and initial initiation site dimensions and to take into account, material type, wavelength, fluence, and pulse length as well as the recipes for finishing and post processing. Model validation is in progress. Experiments showed that conditioning of potassium dihydrogen phosphate (KDP) was most effective using short pulses (800 ps) and the associated pulse-scaling relationship was determined. Forensic studies of the morphology of individual bulk KDP defect sites has led to enhanced understanding of defect initiation.

Proposed Work for FY05

In FY05, we expect to determine impact of unconverted light on the growth of defects in KDP. We will measure defect density/cm^2 at fluences above 14 J/cm^2 in fused silica to understand the impact the 3-wave mixing (hot focusing by frequency conversion crystals) that was discovered two years ago and validated this past year in Optical Science Laboratory (OSL) experiments. We plan to determine how the fluence for a given damage level scales with pulse length in KDP crystals to allow the prediction of crystal bulk damage in a high-energy, high-power laser system using our campaign model formalism; and improve predictions of damage in on-line experiments by combining within the framework of the model data gained from clean, well-controlled, offline experiments with on-line damage observations where variables cannot be as carefully controlled.

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Novel Methods for Bonding Disparate Materials

Michael W. McElfresh 03-ERD-074

Abstract

This project will use a systematic strategy to develop new processes for bonding disparate materials for fabricating experimental samples (e.g., laser targets). The project will focus on methods that yield extremely thin, uniformly thick, strong bonds that have minimal impact on the properties of the materials to be bonded. We will focus on long-chain, polymer-tethered molecules covalently attached as monolayers to the surfaces to be bonded. Force spectroscopy will be used to study bond strength and the ability for tethers to span gaps that would be associated with surface roughness in real materials. Patterned, tethered molecular systems will be used to study chemically directed assembly.

The expected result is a more scientific understanding of possibilities for bonding between disparate materials. This study will allow us to identify bonding systems and strategies for using these systems with materials common to laser-target fabrication. These strategies may involve using appropriate solvents to modify the tethered molecule systems so they will bridge the gaps that exist when bonding together pieces of material. Chemically directed assembly will offer a new strategy for aligning small parts during fabrication. This research will result in publications in refereed journals and provide new concepts for mission-relevant work.

Mission Relevance

If successful, this project will support the Stockpile Stewardship and the Inertial Confinement Fusion Programs by providing multilayer targets for important classes of experiments that study materials under precisely controlled shock conditions driven by a gas gun or laser. In these experiments, the bonding of the multilayer targets must be sufficiently strong so as not to delaminate prematurely, and the bonding layer must be extremely thin to meet experimental design specifications.

FY04 Accomplishments and Results

The hydroxide bonding method was used to bond aluminum to quartz and copper to aluminum. This method was investigated for target fabrication, and other materials were

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tested. Chemical surface derivatization was extended to new classes of active groups tethered on long-chain molecules. Various sulfur–gold bonds were studied to better understand their potential for attaching tethered molecules to surfaces. Force spectroscopy results indicated that equilibrium configurations for tethered molecules can be altered by changes in the solvent. Multiple bonds (i.e., multivalency) on the same targeting molecule were characterized by force spectroscopy for the Concanavalin A–mannose system.

Proposed Work for FY05

Using force spectroscopy, we will characterize the strength and frequency of tethered molecular systems as a function of tether length and active bonding group. The ability of tethered molecules to extend and span a gap will be studied by time-delay force spectroscopy, a novel technique under development. Using solvents to control the molecular configuration of the long-chain molecules, we will determine the relationship between solvent and equilibrium extension of the polymer. Time-delay force spectroscopy will allow us to determine the dynamic extension properties of the polymer. Patterns of tethered molecules will be used to direct assembly. The ability to align and bond parts will be studied as a function of the density of functional groups and solvent conditions.

Plutonium and Quantum Criticality

Michael J. Fluss 03-ERD-077

Abstract

This project will map the phase diagram of Pu near $T = 0$ K. Plutonium's Mott-like f-electron properties may be the signature of a second-order phase transformation or quantum critical point (QCP), which could explain the anomalous phase stability of delta-Pu. We will vary Pu properties with alloying [Pu(Am) of various Am concentrations], pressure, and magnetic field to search for non-Fermi liquid characteristics at low temperatures using measurements of resistivity, magnetic susceptibility, heat capacity, and Hall coefficient. In collaboration with Los Alamos National Laboratory (LANL), we will conduct positive muon spin relaxation (+μSR) experiments at the TRIUMF Laboratory in Canada to search for local moments. We also are collaborating with Oak Ridge National Laboratory on the Pu(Am) alloy experiments.

Our goal is to identify a hypothesized QCP in Pu, which can be identified by a singularity at $T = 0$ K. By mapping the low-temperature phase diagram of Pu, we will determine the region of non-Fermi liquid behavior. We also will develop evidence for the existence or absence of local moments in the Pu system through experiments with point defects and muon relaxation. This work will provide new insight into the organizing principles of Pu and a more fundamental understanding of the origin of its six solid-state phases and its many anomalous physical properties, including its large volume changes and its extraordinary phase sensitivity to impurities.

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Mission Relevance

Understanding the organizing principles of Pu is one of the great solid-state physics problem, and better understanding its physical properties is particularly important to stockpile stewardship. Today, an intense scientific debate is examining the importance of correctly introducing many-body and other correlation effects into the first-principles description of Pu and Pu alloys. Our mapping of the low-temperature phase diagram for Pu will provide the high-fidelity measurements of its physical properties that are needed to validate emerging theories.

FY04 Accomplishments and Results

In FY04, we built a low-temperature, solid-state laboratory consisting of two instruments: a magnetic-properties measurement system (MPMS) and a physical-properties measurement system (PPMS). The PPMS was used to conduct diamond-anvil pressure–resistivity and heat-capacity experiments on cold nonradioactive specimens. Initial results indicate that a large magnetic anomaly is associated with the accumulation of radiation damage in alpha-Pu, and a heat-capacity phase diagram mapped a magnetically tunable quantum critical system, $Sr_3Ru_2O_7$. Plutonium experiments were begun in the MPMS. In July 2004, LLNL and LANL completed initial experiments at the TRIUMF Laboratory, conducting a +μSR search for local moments in alpha- and delta-Pu.

Proposed Work for FY05

We will complete the characterization of defects in delta-stabilized Pu(Ga) and alpha-Pu combining susceptibility, resistometry, and heat capacity. The Pu(Am) alloys will be synthesized to confirm the compositional magnetic anomaly reported at Pu(25% Am). We will then use our tools to map the low-temperature phase diagram for Pu(Am) alloys. The +μSR experiments on alpha- and delta-Pu will continue and, if feasible, we will study the local moments at interstices and at point defects. We also expect to complete the heat-capacity measurements of temperature versus field on $Sr_3Ru_2O_7$.

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Development of Sample Handling and Analytical Expertise for the Stardust Comet Sample-Return Mission

John P. Bradley 03-ERI-007

Abstract

On January 2, 2004, the Stardust spacecraft successfully passed through the tail of Comet Wild-2, resulting in the capture of micro- and nanometer sized cometary particles in the dedicated silica aerogel collectors. As comets are “time capsules” from the earliest stages of solar system formation, the study of the Stardust particles will give a unique insight into early solar system processes. The focus and scope of this project are twofold: (1) to develop extraction methods to isolate the captured particles from aerogel and (2) to develop procedures to analyze material in situ and isolated aerogel-free particles. We are utilizing the extensive analytical capabilities of LLNL and are collaborating with researchers at Stanford and UC Berkeley.

The development of extraction and analysis skills of particles embedded in silica aerogel will enable the analysis of cometary particles captured by the Stardust spacecraft. When samples are returned to Earth in February 2006, we will be able to answer some fundamental scientific questions: What is comet dust? Is it interstellar dust, (inner) solar system dust, or both. These answers, in turn, will provide insight about circulation and large-scale transport within the solar nebula and the formation of comets. What is the source of organic matter in the solar system? Do comets have any relevance to astrobiology? Do the captured cometary particles resemble material in primitive meteorites or interplanetary dust particles collected on Earth and in the stratosphere?

Mission Relevance

Since the collected samples are natural nanomaterials, the techniques and analytical procedures developed under this LDRD are directly applicable to nanotechnology critical for key Laboratory missions in national security, including homeland security and stockpile stewardship, and in basic science. The proposed work will prepare for full scientific exploitation of the Stardust samples in support of NASA’s sample-return missions.

FY04 Accomplishments and Results

The project has been driven on two fronts over the past year, the first of which was technology development for the routine handling and extraction of particles from aerogel. This included the first section of a micrometeoroid captured in aerogel using the focused ion-beam (FIB) microscope and detailed analysis of captured micrometeoroids using the nuclear microprobe. The second thrust was new science from the analysis of interplanetary dust particles, including isotopic measurements using the NanoSIMS ion probe at LLNL. The first far-infrared measurements were carried out on the synchrotron beamline at the Advanced Light Source. Floss et al., in *Science*, reported on the first observation of correlated carbon and nitrogen isotopic anomalies in heteroatomic organic matter from an interplanetary dust particle.

Proposed Work for FY05

We propose to continue the multianalytical approach of the project with specific focus areas on (1) the extraction and analysis of particles embedded in aerogel from both laboratory impacts and those captured in space using diamond microtools and (2) characterization of micrometeoroids from low-Earth orbit recovered from aerogel and interplanetary dust particles collected in the stratosphere using the nuclear microprobe, NanoSIMS, dual beam FIB-field-emission scanning electron microscopy, the new 200 keV monochromated transmission electron microscopy, synchrotron x-ray microprobe techniques (x-ray fluourescence, x-ray absorption near-edge structure, and extended x-ray absorption fine structure), and synchrotron infrared spectroscopy. To understand the mineralogy and chemistry of the extraterrestrial materials studied, we will synthesize analogues in the laboratory under similar simulated astronomical conditions.

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Infrared Diagnostics for Dynamic Events

Steve J. Deteresa 04-ERD-005

Abstract

This research investigates the thermodynamics of dynamic deformation and failure of materials using high-speed and spatially resolved infrared (IR) measurements of temperature. During deformation, mechanical work is converted to different forms of energy depending on the deformation processes, but how this work is converted to energy is not well understood. It can be dissipated as heat in purely plastic deformation, stored as strain energy in dislocations in metals and in oriented polymeric molecular structures, and expended during the generation of new surfaces during damage and fracture.

We expect this project to produce a more thorough understanding of the thermomechanical behavior and failure of materials under dynamic loading. The experimental results will provide an excellent dataset for testing existing models and may lead to new discoveries in material response. The dynamic IR temperature measurement capability developed here will have applicability to other problems involving dynamic temperature gradients, such as the interaction between lasers and materials. This project addresses fundamental problems in thermomechanics and will lead to journal publications and conference presentations.

Mission Relevance

Improved models of materials behavior under extreme dynamic conditions that are developed in this project will support the Laboratory's stockpile stewardship mission. Future applications of the IR temperature-measurement technology include safety aspects of explosives, verification of existing material models, and contributions to the development of new material models, all relevant to the Laboratory's national security mission. This technology may also contribute to armor/antiarmor applications of materials in support of Department of Defense missions and to penetrator-case and explosive-containment-vessel materials for NNSA applications.

FY04 Accomplishments and Results

We completed a series of experiments to resolve a controversy regarding the percentage of plastic work that is converted to heat for metals. One of our results shows that there is no significant difference between IR temperature calibrations performed in air or vacuum and that the emissivity does not change due to surface oxidation for the materials we are studying. We used the LS DYNA code to perform thermomechanical simulations of the uniform compression of metals at different strain rates. These models showed that adiabatic conditions were met at intermediate strain rates. Preliminary tests with Ta at intermediate strain rates yielded different values of temperature when measured using IR and a thermocouple. Ongoing work is aimed at characterizing the emissivity of deformed specimens to resolve this discrepancy.

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Proposed Work for FY05

If the characterization work begun in FY04 shows that a significant fraction of work is stored as a change in internal energy, we will study how the remaining mechanical work is stored in a plastically deformed material. This will initiate a new effort to understand the physical mechanisms of dynamic plastic deformation. We also plan to characterize the temperature gradients due to strain localization from shear bands in dynamic deformation experiments with metals, using the high-speed detector we acquired in FY04. A key part of this effort will be measuring the spatial resolution of the detector system. Work with mock high explosives will determine the energy conversion and dissipation mechanisms under uniform dynamic deformation.

Time-Resolved Dynamic Studies using Short-Pulse X-Ray Radiation

Art J. Nelson 04-ERD-010

Abstract

This project proposes to continue development and demonstration of real-time, in situ interrogation of the dynamic response of materials under extreme conditions, e.g., response to laser-induced shocks, together with simulations that include the time-dependent electronic structure during equilibration between electrons and ions, and the resulting motion of the atomic nuclei. Short-pulse (subpicosecond) x-ray radiation will track changes in the physical properties in tandem with measurements of the atomic and electronic structure of materials undergoing fast laser excitation and shock-related phenomena. Time-resolved x-ray diffraction, x-ray absorption and photoemission spectroscopy, and x-ray scattering will be used to study phase changes in materials with subpicosecond time resolution.

The results of experimentation—laser pump–x-ray probe and x-ray pump–laser probe interrogation of ultrathin metal foils—will be compared with the first-principles electronic structure methods and molecular-dynamics simulations of materials under extreme conditions. The simulations include the time-dependent electronic structure during equilibration between electrons and ions, and the resulting motion of the atomic nuclei. Thus we will introduce the notion of an electronic temperature–pressure phase diagram that differs from conventional phase diagrams, in which ion temperature is used. The combination of modeling and experiment developed in this project will be used to answer important scientific questions about the dynamic response of materials under extreme conditions.

Mission Relevance

Ultrafast materials response under extreme conditions, together with state-of-the-art models and computational facilities, are directly related to and support LLNL's missions of national security and stockpile stewardship.

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FY04 Accomplishments and Results

We successfully demonstrated the optical pump–x-ray laser probe capability to observe the evolution of changes in the electronic structure of isochoric laser-heated ultrathin Cu with picosecond time resolution. We produced (1) the first measurements of the disassembly dynamics of femtosecond-laser-heated ultrathin Cu foil; (2) the first time-resolved photoemission spectra of laser-heated ultrathin Cu foil showing depopulation of the d-band as the electron temperature increases and a smearing of the Fermi-Dirac electron energy distribution; and (3) the first-of-their-kind atomistic and molecular-dynamics simulations of void nucleation in Cu thin films. Our results were presented at the 2004 Workshop on Ultrafast X-ray Science.

Proposed Work for FY05

In FY05, we will further develop the pump-probe technique with experiments and modeling extended to other metals of interest and to higher-shock loading. Specifically, we will (1) demonstrate the pump-probe technique on actinides (e.g., ^{238}U); (2) further utilize the Sub-Picosecond Photon Source at the Stanford Linear Accelerator Center for time-resolved studies and two-photon ionization of the Cu K-edge, with subsequent observation of the Auger cascade of the excited state; and (3) develop a two-dimensional level-set model for describing large-scale morphology.

Publications

Dunn, J. et al. (2004). "Picosecond x-ray laser photoelectron spectroscopy of room-temperature and heated materials." *Proc. 9th Intl. Conf. X-ray Lasers*, Beijing, China, May 23–28, 2004. UCRL-CONF-204309.

Dunn, J. et al. (2004). *Recent developments in tabletop x-ray laser sources and applications.* Presented at the 28th European Conf. Laser Interaction with Matter, Rome, Italy, Sept. 6–10, 2004. UCRL-ABS-204479.

Nelson, A. J. (2004). *Time-resolved x-ray photoelectron spectroscopy with an x-ray laser source.* Presented at the 2004 Workshop on Ultrafast X-ray Science, San Diego, CA, April 28–May 1, 2004. UCRL-ABS-203032.

Nelson, A. J. et al. (in press). "X-ray laser induced photoelectron spectroscopy for single-state measurements." *Applied Physics Letters*. UCRL-JRNL-205272.

High-Strain-Rate Deformation of Nanocrystalline Metals

Eduardo M. Bringa 04-ERD-021

Abstract

Although current understanding of the deformation mechanisms of nanocrystalline (nc) metals (grain sizes <500 nm) at high strain rates (>10^6/s) is limited, improved understanding could lead to the design of improved materials with higher or lower resistance to plastic deformation. We are studying the high-strain-rate deformation of nc-Ni using both laser-induced shocks and isentropic compression, along with molecular dynamics and continuum

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simulations. Samples are characterized using electron microscopy, x-ray diffraction, laser profilometry and nano-hardness testing before and after loading. We aim at validating current constitutive models using atomistic molecular dynamics and continuum simulations performed at the boundary of their computational possibilities to match experimental scales.

If successful, our experiments and atomistic/continuum simulations will validate constitutive models for plastic behavior and will determine the deformation mechanism of nanocrystals of different grain sizes. These results will be unique because proposed strain rates have never been attained in nanocrystals and because experiments and atomistic simulations will cover the same length and time scales. Our atomistic simulations to date show that materials under these loading conditions display an ultrahigh flow-stress never reached with other deformation paths, opening the possibility for new applications of these materials.

Mission Relevance

This work will (1) validate constitutive models (e.g., grain-size correction and mobile dislocation densities) important to the Stockpile Stewardship Program (SSP); (2) enhance our understanding of nc metals important to SSP; (3) map deformation processes at high strain rates; and (4) develop massively parallel simulation capabilities to study these processes. The findings that result from this project will also find application in basic science research in several areas ranging from warm dense matter to multiscale modeling that will benefit LLNL's national security mission.

FY04 Accomplishments and Results

We developed recovery capabilities at the Janus and Omega lasers and performed shots using shockless and shock drives. A total of 12 samples were loaded and recovered. Transmission electron microscopy results indicate large defect densities in both grain growth and grain refinement, which is in agreement with nanohardness and x-ray diffraction measurements and with our atomistic simulations. We developed computational tools necessary for large-scale simulation of nanocrystalline samples and performed continuum calculations with 2D-CALE to model laser loading. Molecular dynamics simulations show substantial differences in the deformation mechanisms as pressure increases, with an apparent disappearance of grain boundary sliding at small grain sizes, and a consequent ultrahigh flow stress.

Proposed Work for FY05

Samples of nc-Ni with grain sizes between 10 and 100 nm will be deformed at strain rates of 10^6/s to 10^8/s and pressures of 15 to 60 GPa. We will use electron microscopy as the main observation tool, with nanoindentation providing mechanical testing. X-ray diffraction will provide grain size distributions, and positron annihilation spectroscopy will measure free volume, indicating densification and annealing of grain boundaries. Atomistic simulations will provide the grain size dependence of the shock-induced deformation type, plastic and twinning thresholds, etc., at the length and time scales of interest; results will be used to improve continuum models. A novel comparison between atomistic and continuum simulations and experiments for grains of 50 nm will be carried out.

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Publications

Bringa, E. (2004). *High strain rate deformation of metals.* Presented at the Congress on Numerical Methods and their Applications, November 2004. UCRL-ABS-207319.

Fission Fragment Sputtering

Bartley B. Ebbinghaus 04-ERD-026

Abstract

Fission fragments born in the first 7 μm of the surface of Pu metal can eject a thousand or more atoms per fission event. This project will combine experimental data with molecular dynamics simulations to develop a continuum model for this internally initiated sputtering process. The study will investigate the amount of material removed per fission event and its dependence on the depth in the material of the nucleus that fissioned, the kinetic energy distribution of the sputtered atoms, and the effects of temperature and the surface condition of the material on the sputtering process. For the experiments, Pu materials will be irradiated at the McClellan Nuclear Radiation Center (MNRC).

The primary goal of this project is to provide a continuum model that can accurately predict the amount, size, and energy of Pu or U atoms that are sputtered per fission event. The model will also include best estimates of the effects of surface condition, Ga content, and temperature, but accurate quantification of these effects will likely require follow-on work.

Mission Relevance

The continuum model of Pu fission events has potential application to the Laboratory's stockpile stewardship mission. By helping define the physical parameters that cause Pu to migrate during handling, this project will contribute to safer handling practices for unencapsulated Pu. The models developed for fission fragment sputtering may also explain the dispersal of interstellar dusts, in support of LLNL's mission in basic science.

FY04 Accomplishments and Results

We designed irradiation experiments at MNRC to elaborate on limited existing data showing that the velocity for the nominally 10-nm-diam ejecta from Cf_2O_3 exceeds 20 m/s, with about 1% of the ejecta exceeding 2,000 m/s. We also modeled the electron cascade using the code TRIPOS-E, with a fraction of this momentum used as input for the nuclei momentum in the code MD-CASK. These simulations showed the formation of chunks or spray depending upon the momentum distribution of the electron cascade. We modeled this momentum distribution rigorously and began the validation and application in simulations necessary to ready the data for publication.

Proposed Work for FY05

About a dozen irradiation experiments will be conducted at MNRC. For the first set of experiments, natural U foils will be irradiated. Some experiments will be conducted under vacuum, others conducted under a reduced pressure of inert gas. The irradiated samples will

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be returned to LLNL and examined by transmission electron microscopy for particle size and number density. Modeling of the fission fragment ejection process will continue using a modified version of TRIPOS-E and MD-CASK. The rigorous momentum distribution model of the electrons will be validated and tested with simulations. The next phase of the modeling will look at effects of phase (i.e., solid vs. liquid), atomic mass, and electron density of the material that interacts with the fission fragments.

Nanosecond Ultrasonics to Study Phase Transitions in Solid and Liquid Systems at High Pressure and Temperature

Patricia A. Berge 04-ERD-033

Abstract

This project will combine ultrasonic and multianvil technology to make precise measurements of elastic moduli, attenuation, and dispersion at high pressure (≤ 40 GPa) and temperature (≤ 2200 K) on large-volume (≤ 10 mm^3) samples. By incorporating recent innovations in high-frequency contact ultrasonics and wave-propagation analysis methods developed for geophysics, the project will study materials with large grains and fabrication-dependent textures, as well as time-dependent behavior in melts and phase transitions. For this research, we will adapt the gigahertz interferometer developed at the University of Colorado and use it in our multianvil (D-DIA) apparatus for polycrystalline and liquid samples.

We expect to develop a capability in static high-pressure methods for studying time-dependent material properties under extreme conditions, enabling, for the first time, the direct determination of the viscoelastic properties of liquids and crystalline mush and the kinetic phase boundaries of materials. We will be able to (1) measure precisely, at high pressure and temperature, the properties of polycrystalline materials with grain sizes larger than a few hundred micrometers and (2) make high-temperature and -pressure measurements of strength-related properties (e.g., frequency-dependent shear modulus) and test equation-of-state (EOS) assumptions about the thermal independence of the Grüneisen gamma parameter.

Mission Relevance

This work supports the Laboratory’s stockpile stewardship mission by providing improved adiabatic EOS information, such as precise velocity measurements for the Grüneisen gamma parameter and shear data for strength estimates and attenuation data for understanding loss mechanisms, to validate computer simulations of explosion phenomena. The high-pressure and -temperature ultrasonic measurements obtained in this project also support the Laboratory’s mission in breakthrough science by addressing questions about the composition and thermodynamic state of the Earth and other planetary bodies.

FY04 Accomplishments and Results

In FY04, we (1) built, tested, and calibrated a prototype ultrasonic system operating from 4 to 400 MHz (at ambient pressure) for liquid samples; (2) used the new system to obtain preliminary data for water (for calibration) and began obtaining data for a Ga eutectic point

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(Ga is liquid at 20˚C); (3) acquired equipment needed for an improved system, including data acquisition and high-frequency, high-power radio-frequency sources for transducer excitation; and (4) obtained beam time at the National Synchrotron Light Source and made preliminary measurements using prototype ultrasonics from our Stony Brook collaborators, combined with the LLNL D-DIA apparatus. We measured elastic constants for Ta to 5 GPa and 1000˚C.

Proposed Work for FY05

The initial work in FY05 will be to interpret data collected in FY04, improve experiment design, and set up and test the apparatus upgrades purchased at the end of FY04. We will continue measurements on Ta and Mo at high pressure and Ga alloys at low pressure, and we will begin measurements on Bi to look for temperature dependence of elastic moduli near phase transitions. Our primary focus will be temperature effects, such as the temperature-dependence of shear velocity in Ta. We will submit our results for publication in peer-reviewed journals

Multiscale Characterization of Body-Centered-Cubic Crystals Deformed to Large Extents of Strain

Jeffrey N. Florando 04-ERD-036

Abstract

The objectives of this proposal are to provide large-strain data for body-centered-cubic (bcc) crystals to develop crystal-plasticity models and validate dislocation dynamics simulations. The accuracy of such simulations is based on the ability of the underlying theory to capture the necessary physics; currently, the large-strain deformation behavior of bcc single crystals is not well understood. Because materials of interest to the Laboratory are deformed to relatively large strains, the need exists to develop new experimental techniques to measure the deformation behavior of bcc single crystals to large extents of strain. The new experimental design will include using a non-contact method to measure strains, in conjunction with stress analysis and multiscale characterization.

Computer simulations require experimental data to develop and validate multiscale crystal plasticity models, but little work has been done to develop these models for large extents of strain. Specifically, crystal plasticity models are an intricate part of a multiscale modeling effort in which information is passed between simulations conducted at different length scales, with the eventual goal of predicting the deformation response of complex materials under various loading conditions. Insight gained from our large-strain experiments has the potential to impact crystal plasticity theory and advance predictive modeling capabilities.

Mission Relevance

Because material-strength models that are accurate under extreme conditions of high pressure, high strain rate, and large extents of strain are highly relevant to the Stockpile

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Stewardship Program, they strongly support LLNL's national security mission. Such models may also play a role in simulating future large-scale laser experiments.

FY04 Accomplishments and Results

We acquired a three-dimensional image-correlation system and used it in deformation experiments to measure strain non-uniformities in a Mo sample. Experiments were conducted to verify the accuracy of the image-correlation technique. Detailed finite-element modeling was performed on a deformed sample to examine the non-uniformities in the stress state. The analysis showed that at 2% strain, the uniaxial stress in the sample varied by ±20%. Based on this analysis, a sample was remachined to achieve larger strains. We also conducted an experiment on a symmetrically oriented sample at 500K to 10% strain for later comparison with dislocation dynamics simulations.

Proposed Work for FY05

In FY05 we will (1) incorporate a second pair of cameras in our existing experimental setup; (2) continue to remachine and test single-crystal Mo samples to achieve larger strains; (3) continue characterizing deformed samples in collaboration with UC Berkeley; (4) begin experiments on single-crystal Ta; and (5) compare the results of our experiments and characterization data with simulations to further our understanding of the deformation mechanisms in these materials and to begin validating our dislocation dynamics codes.

Ultrafast, in-situ Probing of Shocked Solids at the Mesoscale and Beyond: A New Paradigm for Materials Dynamics

Keith S. Bradley 04-ERD-071

Abstract

This project will conduct the first unambiguous in situ measurements of the microstructural evolution of metals under high loading rates using ultrafast x-ray and electron probes to produce diffraction, scattering, and imaging measurements. By directly probing lattice response in real time down to nanometer and nanosecond resolution, we will provide critical insight about the fundamental mechanisms governing macroscopic behavior. We will develop these novel dynamic measurements using electron microscopes, laser-produced x-ray sources, and accelerator x-ray facilities. We will shock samples under study with high power lasers and electric mini-flyers.

We expect that measurements from this research will produce new understanding of solid-to-solid phase transformations and damage evolution. Specific physical observables will include kinetics of crystalline transformations, elastic-to-plastic transitions, dislocation and defect concentrations, and void growth, all in real time. Measurements will directly connect lattice response with continuum behavior, important to materials science and practical applications of metals under shock loading.

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Mission Relevance

Success in stockpile stewardship, energy research and other mission areas demands a fundamental understanding of materials behavior under dynamic loading. Techniques demonstrated in this project will be applicable to current and future Laboratory experiments in support of stockpile stewardship. Capabilities demonstrated will drive development of computational tools for predicting performance, safety and reliability of nuclear weapons and find application in complex issues important to high-power laser research.

FY04 Accomplishments and Results

Our goal for FY04 was to demonstrate that our approach could produce useable data on the spatial and temporal scales of interest. We have demonstrated x-ray diffraction in shocked Ti, measuring preliminary lattice response as a function of shock pressure. Dynamic electron microscopy has measured a solid-state phase transition in Ti and imaged a solid–liquid interface under transient melting conditions. Laser-driven x-ray imaging experiments have produced high resolution, time-resolved images and have enabled the set up of a theoretical framework to optimize dynamic experiments. We have developed a portable, small-scale apparatus for shock loading foils at synchrotron sources and made static scattering measurements of submicrometer void distributions in vanadium.

Proposed Work for FY05

In FY05, we plan to conduct several ground-breaking measurements to establish the value of in situ probing under dynamic loads. We will utilize laser-based, x-ray backlighter techniques for dynamic diffraction and imaging, specifically for studying the alpha-to-omega phase transformation in Ti using diffraction and the shock-induced void growth in Al using x-ray imaging. We will perform measurements with a novel tool, a dynamic transmission electron microscope, currently nearing completion at LLNL, to investigate phase transformations in Ti with high temporal and spatial resolution. Finally, we plan to utilize an electronic miniflyer to produce shocks in metals to conduct experiments at accelerator light sources such as the Advanced Photon Source at Argonne National Laboratory.

NanoBIS Determination of the Unoccupied Electronic Structure of Plutonium

James G. Tobin 04-ERD-105

Abstract

This project is using nanofocused bremsstrahlung isochromat spectroscopy (nBIS) to obtain direct measurements of the conduction band (unoccupied electronic structure) of Pu and other actinides. By using a scanning electron microscope (SEM) as the electron beam source in nBIS, we will be able to probe microscale single-crystalline grains in polycrystalline samples. Furthermore, by working at sufficiently high energies, we will sample preferentially for bulk properties, reducing the impact of surface effects.

We expect to determine the unoccupied conduction-band electronic structure of Pu and other actinides (and possibly rare earths) in a phase-specific fashion with emphasis on bulk

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contributions. Our results will resolve the 4f and 5f electronic structure controversies by allowing direct comparison to the predictions made with dynamical mean field theory.

Mission Relevance

In this project, we will validate Pu equation-of-state theories and provide experimental benchmarking of modeling for stockpile stewardship, in support of the national security mission.

FY04 Accomplishments and Results

We used our two months of funding in FY04 to order and set up our experimental equipment, including the soft x-ray spectrometer, the SEM, and a variety of vacuum equipment.

Proposed Work for FY05

We will perform in-house nBIS experiments using the SEM and the soft x-ray spectrometer as the focused electron source and the photon detector, respectively. We will begin our studies using less-radioactive actinides such as depleted U (^{238}U) and rare earths such as Ce, which is the rare-earth analogue of Pu in that similar unknowns exist about its multiphase electronic structure. We will also prepare for experimentation on Pu to resolve the 4f and 5f electronic structure and to clarify Pu structure–property relationships.

Mapping Phonons at High Pressure: Phase Transformation, Phase Stability, and Elastic Anisotropy

Daniel L. Farber 04-ERD-106

Abstract

This proposal represents LLNL's participation in an international collaboration to develop new techniques for mapping phonon dispersion curves (PDCs) at high pressure in the diamond anvil cell using high-resolution inelastic x-ray scattering (HRIXS). The project will focus on probing PDCs in a number of physically novel systems: (1) Ce (an element often used as a surrogate for Pu) at or near the solid–solid singular point; (2) V at ultrahigh pressures; and (3) the high-pressure form [hexagonal closely packed (hcp)] of Fe.

If successful, this project will open a new field of research directed at probing the dynamics of systems at extreme conditions with HRIXS. Understanding the role of phonons in phase transitions is critical to our ability to describe the underlying physics that controls phase stability and a range of transport properties in lanthanide and actinide systems. This work will likely result in publications in high-profile journals.

Mission Relevance

The study of physical properties of Ce promises to provide the key to understanding other lanthanide and actinide systems of importance to the Laboratory's stockpile stewardship mission. We will also investigate the pressure evolution of the elasticity tensor in V, another material relevant to stockpile stewardship.

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FY04 Accomplishments and Results

In FY04, we procured single-crystal samples (Ce and V) and set up diamond anvils and diagnostic equipment for our experiments. We also fabricated the first generation of 40-μm-diameter single crystals, designed a high-pressure loading system, and worked on developing the computer codes for the data reduction.

Proposed Work for FY05

We will probe two very important and poorly understood types of phase transitions: the transition from f-electron localization to itinerancy in face-centered-cubic Ce and the Lifshitz or electronic-topologic transitions in hcp metals (Os and Cd). We recently documented a strong anomaly in the lattice parameters of Os upon compression to above 25 GPa, which may be the fingerprint of a possible Lifshitz transition. Our studies of Os and Ce will allow us to probe the dynamical effects of electronic phase transitions while developing new high-temperature, high-pressure HRIXS capabilities.

Nanomaterials for Radiation Detection

Tzu F. Wang 04-ERD-107

Abstract

This project will fabricate 3-D nanomaterials for radiation detection as improvements over current semiconductor- and scintillation-based technologies. By providing a significant leap beyond the current 2-D nanotechnology, the project has the potential to significantly improve radiation-detection technologies, which are currently based on crystal growth. We will address two important areas: gamma ray and neutron detection. For gamma-ray detection, our first goal will be to use porous nanomaterials to achieve scintillation output several times better than that of NaI(Tl). For neutron detection, we will construct a 3-D structure using a doped nanowire "forest" supported by a boron matrix. In principle, this technology will determine both neutron energy and neutron direction, which is not possible with current technology.

We expect improvements over semiconductor-based and bulk-crystal-based technologies in flexibility, cost, durability, surface area, and uniformity of charge collection. Scintillation detection will be improved in output and wavelength, thereby improving the energy resolution of gamma-ray detection. If successful, this project will also enable future development in wide-area radiation monitoring for both gamma-rays and neutrons.

Mission Relevance

This work will have a significant impact on nuclear science for the Laboratory's national security mission, especially in radiation detection technologies for both gamma rays and neutrons. In addition, the improved capability in neutron spectroscopy can help with (n,n') (n,2n) and cross-section measurements for stockpile stewardship.

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FY04 Accomplishments and Results

We performed proof of concept experiments for the efficient conversion of gamma rays into visible photons by using a 60-μm-thick resin film containing 10 mg of CdSe/ZnS core shell quantum dots per milliliter that was cured between two glass microscope slides. These dots present a green luminescence at 540 nm. The scintillation output recorded with an ^{241}Am source (10 μC), and with a ^{137}Cs source show significant differences from background. The data demonstrate that the conversion of incident gamma rays into green photons has been realized by the film of semiconductor quantum dots. Higher efficiencies are expected for thicker films.

Proposed Work for FY05

For gamma-ray detection, we will fabricate luminescent, porous silicon samples and test their scintillation for various radioactive sources to understand the physics behind the process. For neutron detection, we will fabricate basic, single-layer semiconductor nanowire samples and study the material properties at the nanoscale. For both types of detection, we plan to file a record of invention and a patent and publish a peer-reviewed paper on the results.

New Shape-Memory Polymers for Actuators

Thomas S. Wilson 04-LW-054

Abstract

We propose to develop new shape-memory polymer (SMP) compositions that bridge the gap between current SMPs and shape-memory alloys. The project will design and synthesize acrylic and urethane polymers that have a 10× increase in recoverable stress due to increased crosslink density and the incorporation of covalently coupled carbon nanotubes. The nanotubes will be oriented to further enhance their reinforcing effect by shear and applied electric fields. Resulting polymer actuators will have the attributes of high recoverable stress and energy, low energy consumption for actuation, and high biocompatibility for engineering and biomedical applications. Finally, we will demonstrate the new SMPs in defense (micro-gripper), biomedical (stent), and space (expandable foam) applications.

We expect that the novel class of actuating materials created in this project, SMPs with significantly increased recovery forces, will expand the possibilities for the use of SMPs in many engineering and biomedical applications. This work will attract and retain young scientists and lead the way to new materials development. The intellectual property developed will increase LLNL's portfolio in SMP technology.

Mission Relevance

The new nanotechnology-based SMPs created in this project have applications in microelectromechanical components for weapons-detection and -inspection systems in support of the Laboratory's homeland security and stockpile stewardship missions. In

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addition SMPs developed here are applicable to biomedical microdevices for stroke, dialysis therapy, and implants in support of LLNL's mission in bioscience to improve human health.

FY04 Accomplishments and Results

In FY04, a series of highly crosslinked acrylics and polyurethanes were synthesized and shown to have shape-memory behavior. The new acrylic SMPs have advantages over previously reported SMPs due to their glass-like optical clarity, higher modulus (>10× over commercial SMPs), and expected improved biostability. In addition, moisture-curable acrylic SMPs were produced, which would allow for melt processibility. The new urethane SMPs display glass-like optical clarity, improved modulus (3× over commercial SMPs for neat polyurethane or 10× with a silica filler), and extremely sharp glass transitions. Both have adjustable transition temperatures. The new SMPs were fabricated into both micro-gripper and thrombus-removal coil applications, which were demonstrated on the benchtop.

Proposed Work for FY05

In FY05, we propose to further investigate SMPs modified with carbon nanotubes. We will examine the effect of nanotube orientation as achieved through processing (shear) or imposed fields (electric) during cure. We expect significantly enhanced mechanical properties in samples with nanotube orientation over macroscopically unoriented materials. The advantages of this orientation will be demonstrated in two applications: (1) neurovascular stents and (2) expandable foams for aerospace applications. In both applications, a comparison will be made to commercial SMP-based devices.

Publications

Wilson, T. S, J. P. Bearinger, and D. J. Maitland. (2004). *New shape-memory polymers for actuators*. Presented at the Polymer Networks 2004, Bethesda MD, Aug. 15–19, 2004. UCRL-PRES-206090.

Application of Light-Emitting Polymers to Detect Pathogen DNA in Blood

Rodney L. Balhorn 04-LW-065

Abstract

The primary goal of this project is to develop improved, highly sensitive, and specific DNA and DNA damage-detection methods for national security and medical applications that use light-emitting polymers (LEPs). To accomplish this goal, we will conduct a series of experiments using LEPs and fluorescence resonance energy transfer (FRET) to determine the sensitivity limit for detecting specific pathogen signature DNA sequences and investigate the utility of using LEPs for monitoring DNA adduct concentrations in human serum. In a final set of experiments, we will also determine if LEPs can be used to increase the sensitivity of protein activity assays.

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Numerous applications in national security and medical surveillance can benefit from developments that enable identification of specific DNA sequences in real time and with high sensitivity. Applications include forensic and genetic mutation analysis, detection and monitoring of food-borne pathogens, and biodefense. A 100-fold increase in the detection sensitivity of PCR and protein activity assays would have a direct impact on the development of next-generation biosecurity monitoring systems. Increased sensitivity for the detection of DNA adducts in blood would provide a new approach for monitoring the response of cancer patients to chemotherapy, enabling the development of individualized treatment regimes for cancer.

Mission Relevance

Successful application of LEPs for detecting pathogen DNA and adduct DNA can result in patentable, highly sensitive chemical sensors for applications that support the Laboratory's missions in national and homeland security and in bioscience to improve human health. Potential applications include cancer detection, customized treatment for cancer radiation and chemotherapy, more sensitive protein-activity assays, and non-PCR-based methods for pathogen detection.

FY04 Accomplishments and Results

We developed a protocol for obtaining DNA from the blood of lung cancer patients undergoing adriamycin chemotherapy and obtained Institutional Review Board approval for it. We designed two different forms of the LEP, both are positively charged (via the quaternary amine) and water-soluble. Synthesis and purification of the first form was in progress at year's end. Calf thymus DNA was used to prepare both the adriamycin (1 adduct/5bp DNA) and benzo[a]pyrene (1 adduct/28bp DNA) adducted DNA standards. Experiments were performed using commercially available LEPs. Three- to four-fold enhancement in adriamycin adduct emission was obtained, even under conditions with limited DNA binding.

Proposed Work for FY05

Using the water soluble LEPs and synthetic DNA adducts we synthesized in FY04, we will complete a series of experiments using the LEPs and fluorescence resonance energy transfer to determine the sensitivity limit for detecting specific pathogen signature DNA sequences and investigate the utility of using LEPs for monitoring DNA adduct concentrations in human serum. We will also determine if LEPs can be used to increase the sensitivity of protein activity assays using an LEP-tagged trypsin substrate. In a final set of experiments, we will use DNA samples obtained from the blood of lung cancer patients undergoing adriamycin chemotherapy and patients with nasopharyngeal carcinomas to determine if LEPs enable the detection of adriamycin DNA adducts and Epstein Barr virus DNA, respectively.

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Ceramic Lasers

Thomas F. Soules 04-FS-006

Abstract

The objective of this project is to make laser amplifiers and components by sintering ceramic powders. Currently, the use of large single-crystal slabs for high-power lasers such as the solid-state heat-capacity laser (SSHCL) is hampered by the difficulty in obtaining boules. Ceramic parts would alleviate this problem and have advantages over single crystals, including the possibility of larger apertures, different shapes, and tailored structures. This project will (1) determine whether we can make transparent yttrium-aluminum-garnet (YAG) onsite, (2) provide an understanding of issues in making transparent ceramics, (3) identify commercial sources of ceramic YAG:Nd parts, (4) determine their viability as laser amplifiers, and (5) turn our ideas for using ceramics in high-powered lasers into intellectual property.

First, we will produce small pieces of transparent sintered YAG and determine the most important parameters that effect the optical quality of the final ceramic and also model the process. Second, we will design ceramic amplifiers and determine their performance in the SSHCL. Third, we will produce intellectual property on a number of novel uses of ceramics, including our multifunctional monolithic designs, in high-powered lasers. This work could bring about a major change in the way laser amplifiers are made and in the design of high-power lasers.

Mission Relevance

Transparent ceramic amplifier slabs would be an alternative to hard-to-obtain single-crystal slabs for small, high-power lasers that could be used in applications such as clearing land mines, defending against mortars, and other applications in support of LLNL's national security mission. Ceramic amplifier slabs could provide enhanced, more robust and compact designs for high-peak-power fusion-class lasers for stockpile stewardship and inertial-confinement fusion, and high-average-power lasers for inertial-confinement fusion energy and theatre missile defense.

FY04 Accomplishments and Results

In FY04 we (1) made translucent YAG ceramic pieces using nanoparticle YAG powders subjected to vacuum sintering at 1750°C and field-enhanced SPS sintering, obtaining 100% of theoretical density, with translucent optical quality; (2) made, for the first time at LLNL, nanoparticle YAG using sol-gel processing; and (3) obtained a transparent 10 × 10 × 0.6-cm ceramic YAG amplifier slab and measured absorption, emission, scattering, and gain. The data suggested that this material can be used in some high-power lasers, and helped determine the specifications for 10 × 10 × 2-cm piece for in-laser testing.

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A Bright Source of High-Energy X Rays

Jeffrey D. Colvin 04-FS-007

Abstract

When intense laser light impinges a solid metal, a high-temperature plasma is formed that emits x rays. Such sources are used as radiography back-lighters for laser targets, but the drawback is their extremely low x-ray conversion efficiency (XRCE). At sufficiently low metal-target density, however, a laser beam should be able to supersonically and volumetrically heat the material, providing much higher XRCE than otherwise possible. The project objective is to fabricate a pure metal foam via a novel technique based upon ion-tracking lithography.

In this technique, a polycarbonate foil is irradiated at random angles with heavy-ion beams. The damaged foil then undergoes a chemical etching process that preferentially etches the foil along the ion tracks, leaving cylindrical holes. Although any material that can be electroplated is suitable, including some semiconductors and insulators, this project focused on Ni. Our goal is metal foam with density <1% of the solid.

This study demonstrated the feasibility of fabricating thin sheets of very low-density metal foams for adaptation as bright sources of high-energy x rays for fusion and material science experiments. The technique employed to produce such low-density foams involved the formation of a network of nanoscale metallic struts. Because the struts produced by this fabrication technique are solid intersecting cylinders, the foam is expected to be very strong, with virtually all the metal contributing to the strength of the material. Assembling the sheets by a novel crumpling technique (or other means) into a larger, 3-D volume may achieve even lower average density, suitable for an x-ray back-lighter target.

Mission Relevance

Future large, fusion-class lasers are of importance to LLNL missions in stockpile stewardship and energy security. Several different experiment campaigns planned for such lasers require radiographic imaging of dense materials. Demonstrating the feasibility of fabricating these foams would help provide a viable new diagnostic option for such experiments.

FY04 Accomplishments and Results

We determined that 14.8-MeV Ar+ ions provide the penetration and electronic energy loss required for ion-tracking the 3-μm-thick commercially obtained polycarbonate foils. The foils were tracked at normal, off normal, two angles, and four angles. All samples were oxygen-plasma etched and some have been chemically etched. The four-angle specimens (azimuthal angles of 0, 90, 180 and 270 degrees, each at 45 degrees off normal), irradiated at a dose of 1.3×10^9 Ar/cm^2, have the greatest promise for creating metal foam structures, with density 0.6% solid for tracks etched to 10 nm. Our collaborator, Prof. Searson, and students at

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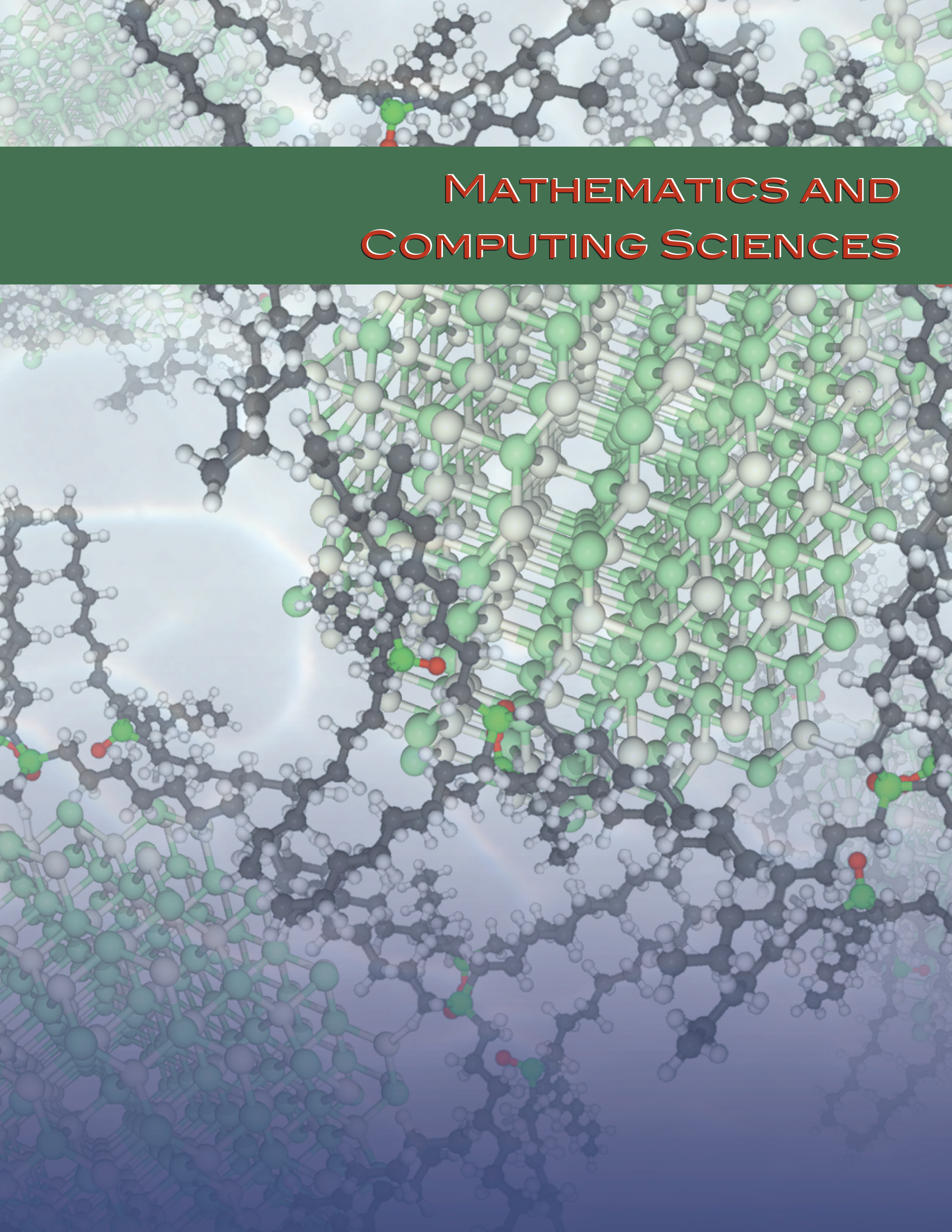
Johns Hopkins University have done the metal electrodeposition into the pores and the chemical etching of the polycarbonate. Scanning electron microscope images are expected at the end of October.

Publications

Felter, T. E. and J. D. Colvin. (in press). "Fabrication of very-low-density pure metal foams as bright high-energy x-ray sources." *Bull. Amer. Phys. Soc.* UCRL-ABS-206199.

Mathematics and Computing Sciences



Mathematics and Computing Sciences

ViSUS: Visualization Streams for Ultimate Scalability

Valerio Pascucci 02-ERI-003

Abstract

This project developed a suite of progressive visualization algorithms and a streaming infrastructure to enable interactive exploration of large scientific data sets. The methodology optimizes the data flow in a pipeline of processing modules. Each module reads and writes a multiresolution representation of a geometric model, providing the flexibility to trade speed for accuracy, as needed. The data flow is streamlined with progressive algorithms that map local geometric updates of the input into immediate updates of the output. A prototype streaming infrastructure demonstrates the flexibility and scalability of this approach for visualizing large data sets on a single desktop computer, a cluster of personal computers, and heterogeneous computing resources.

The ViSUS project benefits the Laboratory at least at two levels. At the deployment level, the improved efficiency in the use of hardware resources reduces the cost of visualization-hardware infrastructures. At the scientific level, the developed technology reduces the overall time required for the design, simulation, and visualization cycle. The ability to remotely monitor large and expensive simulations saves computing resources by providing the opportunity for early termination and restart of erroneous test simulations, for instance. Runtime steering is now possible for simulation codes with mechanisms for dynamic modification of running conditions.

Mission Relevance

Use of our innovative, high-performance visualization techniques allows interactive display of very large data sets on simple desktop workstations and the monitoring (or steering) of large parallel simulations. This will have valuable applications to several LLNL missions that use large-scale modeling and simulations, including stockpile stewardship, nonproliferation, energy security, and environmental management.

FY04 Accomplishments and Results

In FY04 we brought ViSUS to a sufficient level of maturity and robustness for its use by targeted users. The main steps completed this year include developing new techniques that accelerate isosurface extraction with occlusion culling, graphics hardware, and view-dependent refinements. A test viewer has been developed for datasets from the HYDRA code simulation. We provided a stable library, with full implementation of our streaming technology, that can be used by simulation codes for saving rectilinear grids in ViSUS IDX format and released a new version of the Progressive Viewer with full slicing, isocontouring, and volume-rendering capabilities. We began working in collaboration with the MIRANDA code-development team to start using the IDX format as output of choice for Blue Gene/L runs.

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Publications

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Enabling Large-Scale Data Access

Terence J. Critchlow 02-ERI-007

Abstract

The goal of this project was to develop a metadata infrastructure capable of providing scientists with access to large numbers of data sources through a single, intuitive interface. The infrastructure employs a user-specified description of a service class to crawl the Web. When an interface in the service class is identified, a wrapper that supports an XML-based query interface is automatically created. We developed an initial prototype of this infrastructure and performed initial testing of the system. Using BLAST, a sequence-similarity search tool, we plan to demonstrate the feasibility of our approach by automatically identifying sequence-similarity search interfaces across a variety of Web sites on genomics.

This project will contribute to several technology areas, including data integration, bioinformatics, metadata, web-aware agents, and wrapper generators by developing an infrastructure for accessing extensive data sources through a single interface. The interface combines local data, for example, simulation data, with related information publicly available over the Internet, such as scientific publications, databases, materials descriptions, and production techniques.

Mission Relevance

The infrastructure developed in this project will support almost every LLNL mission by improving and simplifying scientists' access to large-scale data sources, which will enable them to better synthesize information from multiple sources. For example, the expertise gained in this project will support LLNL's mission in biodefense by helping scientists to gain insight into gene-sequencing.

FY04 Accomplishments and Results

In addition to demonstrating end-to-end automatic wrapper generation for BLAST interfaces, we extended the service class description to handle more complex data types such as citations, which cannot be easily represented by regular expressions. We performed several short Web crawls that successfully demonstrated the application of our infrastructure

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on both well-structured BLAST interfaces and poorly structured publication interfaces by identifying previously unknown sites in both domains.

Biological and Synthetic Nanostructures Controlled at the Atomistic Level

Giulia A. Galli Gygi 03-SI-001

Abstract

This research combines theoretical and experimental investigations of Group IV semiconductor nanostructures with the goal of designing structures having desired labeling and sensing properties. Our focus is on C, Si, Ge, and SiC dots, as well as Ge and Si rods and wires and their properties in solution. In addition we are studying structural and electronic properties of CdSe dots and rods to make contacts between advanced simulations and well-established experimental results. Finally, we have started a computational study of water and solvation properties of nanostructures, with the ultimate goal of addressing transport properties of dots, wires, and tubes in wet, confined environments.

The project will result in new expertise, improved computer codes, colloidal-synthesis techniques, and surface-sensitive spectroscopic characterization techniques for semiconductor nanostructures. In addition, we hope to identify novel compound nanostructures for building semiconductor-based chemical labels and biotags. Our research results have already had an impact on the nanoscience community, and we expect this impact to grow if we continue to be successful in publishing our results in high-profile journals.

Mission Relevance

This proposal will help predict, develop, and design semiconductor materials with tailored properties for advanced biodetection needs in support of LLNL's national-security mission. This will provide a scientific foundation for the creation of next-generation technologies based on manipulating and controlling matter at the nanoscale, in support of the Laboratory's mission in breakthroughs in fundamental science and technology.

FY04 Accomplishments and Results

In our experimental efforts, we succeeded in producing size-selected Ge dots in significant quantities to obtain reproducible x-ray spectra and in producing Ge rods, using a new procedure based on VLS technologies. Significant progress was made in doping Ge dots, and a project was started on doped Ge nanoparticles. We successfully carried out the first precise determination of the force between small molecules and single-walled carbon nanotubes (SCWNT). On the computational side, major progress was made in the study of solvation properties of nanoparticles in water, in understanding solvent influence on nanoparticle growth, and in determining the properties of catalytic nanoparticles needed for an efficient growth of nanotubes.

Proposed Work for FY05

We propose to work on dots, wires, and rods and to compute optical and electronic properties of interacting nanoparticles, including thermal conductivities and nonlinear

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optical properties. We will also continue our simulations of solvation processes and of water and other fluids in confined media to study transport in channels. Experimentally, we will attempt to produce water-soluble nanoparticles and control rod and wire growth to enable their use in macroscopic assemblies. Finally, we will continue our joint experimental and theoretical work on nanoparticle functionalization and on the study of molecule–SWCNT interactions.

Publications

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ICE: The Image Content Engine

James M. Brase 03-SI-003

Abstract

Advancements in imaging-sensor technologies (particularly for remotely sensed images) is resulting in volumes of data that overwhelm human analysts. The goal of this project is to develop a framework, the Image Content Engine (ICE), that will allow human analysts to interpret massive volumes of imagery in a timely fashion. The ICE approach focuses the attention of humans on relatively few small areas with specified attributes. This is achieved by capturing content extracted from images as nodes in semantic graphs whose links define relationships between pieces of image content and information obtained from non-image sources.

The ICE framework will allow content extracted from imagery, the most voluminous data source, to be seamlessly integrated with information from other sources. This will help break down the compartmentalized nature of analyst-based interpretation in the defense and

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intelligence communities. ICE will greatly increase the productivity of image analysts by very efficiently focusing attention on potential objects of interest. The ICE approach also applies to other areas of experimental science (e.g., physics, biology, and environmental science) in that mining massive archives of complex measurement data for new patterns and relationships is an emerging model for discovery in modern science.

Mission Relevance

The work supports LLNL's mission in national security by developing the technology to allow content extracted from images to be seamlessly integrated with information from other sources, thus enabling analysts to submit sophisticated queries relevant to defense, intelligence, counterterrorism, deployment strategies, determining the functions of enigma facilities, and detecting the production of weapons of mass destruction.

FY04 Accomplishments and Results

An infrastructure for a configurable feature-extraction pipeline was implemented for regions and model-based objects, allowing these feature-extraction tools to operate on multiple images of arbitrary size. Phase-sensitive detection for arbitrary 2-D object signatures was demonstrated. Algorithms for projecting 3-D target models into images have been developed and integrated with the phase-sensitive detector. This capability has been used to demonstrate a broad-area search capability for important national-security program targets. We put a baseline hierarchical image-feature model in place and demonstrated tile-based queries. We also implemented a baseline hierarchical data representation scheme for the semantic graph and metric-based matching schemes.

Proposed Work for FY05

We plan to complete the development and integration of tile-based feature extraction and query tools, extending them to multispectral imagery. Partial model matching will be implemented and evaluated for object detection. The hierarchical representation of graph data will be extended to include relevant link types and full search capabilities that employ metrics and morphological matching. Detection and query applications will be extended accordingly, with confidence estimation and relevance feedback incorporated, and performance analysis will be performed using real data where feasible. A parallel version of the pipeline will be implemented in a cluster environment for analysis of actual data streams from the Large-Area Synoptic Survey Telescope.

Publications

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Correction of Distributed Optical Aberrations

Scot S. Olivier 03-ERD-006

Abstract

The objective of this project is to demonstrate the use of multiple distributed deformable mirrors (DMs) to improve the performance of optical systems with distributed aberrations. This concept is expected to provide dramatic improvement in the optical performance of systems in applications where the aberrations are distributed along the optical path. Our approach uses multiple DMs distributed to match the aberration distribution. The project will simulate the performance of these multiple DM systems, develop a testbed to evaluate distributed correctors matched to the distributed aberrations, conduct experiments, and compare experimental results to the predictions of detailed modeling.

This project will also develop the underlying theoretical and experimental capabilities for correcting distributed optical aberrations, which will significantly increase the corrected field of view for tactical or surveillance imaging scenarios over long horizontal or slant paths. Applications include wave-front sensor and correction algorithms for the Large Synoptic Survey Telescope (LSST), which will enable the mapping of dark energy and dark matter in the Universe and surveys of near-Earth objects. This project will also numerically investigate field correction for short-pulse lasers using multiple DMs, which would enable new regimes of science to be investigated with existing and future short-pulse laser systems.

Mission Relevance

The development of advanced techniques for correcting distributed aberrations will support new efforts in remote sensing and imaging applications for the Laboratory's national-security and homeland-security missions, and will also enable forefront astronomical science on the next generation of giant telescopes in collaboration with the University of California.

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FY04 Accomplishments and Results

In FY04, we (1) successfully completed the field test of a single DM adaptive-optics testbed through distributed turbulence; (2) assembled and obtained initial results on a multi-conjugate adaptive-optics testbed using three DMs; (3) published six papers (three refereed and three conference papers) on our results; (4) developed a new multigrid algorithm for the reconstruction of gradient wave-front sensors and a tomography algorithm for the reconstruction of distributed turbulence from multiple field angles; (5) developed a new technique for the field correction of short-pulse lasers using the transport-of-intensity equation and two DMs; and (6) began investigating the most appropriate wave-front sensor technology for the LSST.

Proposed Work for FY05

We will (1) continue development of the transport-of-intensity approach to field correction for short-pulse laser systems to increase the energy and intensity delivered to target; (2) perform further experiments on the multi-conjugate adaptive-optics testbed to examine the correction of scintillation and field-of-view increases with multiple DMs; (3) continue evaluating possible technologies for the wave-front sensor for the LSST; (4) arrive at the baseline technology and design for the LSST wave-front sensor and control algorithms for removing thermally and gravitationally induced aberrations; and (5) implement the design in a scaled laboratory testbed and evaluate the performance of the system under realistic conditions.

Publications

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Adaptive Mesh Refinement Algorithms for Parallel Unstructured Finite Element Codes

Ian D. Parsons 03-ERD-027

Abstract

This project will produce algorithms and software for adaptive-mesh-refinement (AMR) methods used to solve practical solid mechanics and electromagnetics problems on multiprocessor, parallel computers using unstructured finite element meshes. The goal is to provide computational solutions that are accurate to a prescribed tolerance; adaptivity is the correct path toward this goal. The new tools will enable analysts to conduct more reliable simulations at reduced cost, both in terms of analyst and computer time. Research issues include effective error estimators for nonlinear structural mechanics and electromagnetics, local meshing at irregular geometric boundaries, and constructing efficient software for parallel computing environments.

The proposed technical approach will produce AMR tools with applications in state-of-the-art engineering codes, like Diablo for solid mechanics and Eiger and EMSolve for electromagnetics. All of these codes are based on unstructured mesh, finite element methods and will require similar algorithms for tasks such as mesh refinement and load balancing. Deliverables include the components of AMR techniques applied to unstructured finite element discretizations, such as treatment of irregular geometries, constraint enforcement, intermesh transfer of state variables, and partitioning of adaptively refined domains.

Mission Relevance

Application of our robust, scalable AMR capabilities with simplified mesh generation and lower computational demands will support LLNL's national-security mission by reducing the total costs of analyses that are performed for stockpile stewardship and other programs. Model verification will be significantly easier and more transparent by combining error analysis and refinement.

FY04 Accomplishments and Results

For solid mechanics, a serial AMR capability was added to Diablo, including the data structures and algorithms that refine a mesh and error estimators based on patch recovery and residual computations. Test problems confirm the correctness of the implementations. The AMR implementation performs both isotropic and anisotropic refinement and derefinement of a mesh. Both residual and patch-based error estimators are being developed for application to large-deformation solid mechanics problems with contact. Coupled thermomechanics simulations demonstrate the capabilities of the current AMR implementation. For electromagnetics, a global L2 projection based error estimator was developed and tested, and a local patch recovery based error estimator was under development at the end of the year.

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Proposed Work for FY05

The serial unstructured functionality developed for solid mechanics will be used to investigate the effectiveness of algorithms for estimating discretization errors and for interpolating state variables to refined meshes. Improvements will be explored. The AMR functionality will be parallelized for efficient performance on massively parallel processor systems using mesh partitioning capabilities for allocating load-balanced subassemblies of a mesh to the available processors. The underlying data structures will be modified to account for the resulting additional interprocessor communications. A hierarchical mesh refinement capability will be combined with the error estimators to form an automatic AMR capability for electromagnetics. Optimization of the algorithm for parallel efficiency will also be conducted.

Entity-Based Modeling of Population-Based Systems

Andrew J. Cleary 03-ERD-030

Abstract

This project designed and completed preliminary development on a toolkit for scalable entity-based modeling (EBM) of complex population-based systems, with a specific emphasis on epidemiology for use in bioterrorism response planning. In the academic community, discrete simulation technology based on individual entities has shown promise for these kinds of complex systems, but the technology has not been scaled to the problem sizes or computational resources of LLNL, and overall the technology is immature. Our developmental emphasis was on showing that this technology can be extended to parallel computers and to larger problems, as well as overall maturation of the technology from an academic to a more applied setting.

This project represents one of the few potentially viable methods for understanding highly nonlinear, complex, adaptive systems, also called chaotic systems. In many ways, the Lab's computational groups have only tackled the easiest of real-world phenomena: those that behave regularly and predictably. Recent events show us, however, that there are many important national security-related phenomena that are not so well behaved. If we are successful, we will provide the Laboratory a powerful tool in its arsenal for understanding, predicting, and controlling such phenomena. We expect this kind of model to be used as an "in silico" laboratory in which analysts conduct "what if" experiments to assess, for instance, potential hostile tactics, U.S. responses to and strategies for those tactics, and the efficacies of each.

Mission Relevance

Our project is directly relevant to LLNL's national-security mission, because EBM is a potential core computational capability with application to a range of multidisciplinary problems, such as simulation of disease transmission in complex, population-based systems. We are working with the major national security efforts at LLNL to tailor our technology for applications such as the spread of infectious disease and bioterrorism response models, starting with homeland security.

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FY04 Accomplishments and Results

Although ending three months into FY04, this project achieved two major milestones. First, we completed a design of two essential, innovative software layers. One layer is a highly scalable, conservative discrete-event simulation layer (DES) that enables and encourages hierarchy in the simulation design and exploits that hierarchy to achieve scalable parallel performance; the other is a layer for constructing entity-based models on parallel computers, and sits on top of the DES layer. Second, we completed a performance study of our epidemiological model, analyzing scalability as a function of the communication characteristics of the model.

Detection and Tracking in Video

Chandrika Kamath 03-ERD-031

Abstract

Video cameras are used for monitoring and surveillance in several applications. We are developing robust, accurate, and near-real-time techniques for detecting and tracking moving objects in video from a stationary camera. This allows us to model the interactions among the objects, thereby enabling us to identify normal patterns and detect unusual events. Our algorithms and software include techniques to separate the moving foreground from the background, extract features representing the foreground objects, track these objects from frame to frame, and post-process the tracks for display. We focus on video taken under less than ideal conditions, with objects of different sizes moving at different speeds, occlusions, changing illumination, low resolution, and low frame rate.

This project will produce robust and accurate technology for video detection and tracking under less-than-ideal conditions with occlusions, fog or changing illumination, or at a low resolution or frame rate. This project will enhance existing algorithms to address these situations, allowing us to better understand their limitations, which, in turn, will determine the conditions under which successful surveillance is possible. The algorithms and software are being applied to surveillance video as well as spatiotemporal data from computer simulations.

Mission Relevance

The capability to detect and track in video supports the national security mission of LLNL by enabling new monitoring and surveillance applications for counterterrorism and counterproliferation.

FY04 Accomplishments and Results

During FY04, we (1) created a software infrastructure to handle streaming video data; (2) implemented several background-subtraction algorithms and evaluated them on videos taken under different conditions; (3) proposed a new background-subtraction method that outperforms other methods, especially on low-resolution, low-frame-rate video; and (4) extracted features and used them in simple tracking algorithms. We also filed a provisional patent on the new method and summarized our work in two papers. We are currently

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adapting the tracking algorithms to work under adverse conditions. We collaborated with the University of Colorado, Boulder, on tracking people; the University of California, San Diego, on tracking under occlusions; and a summer student on object representations for tracking.

Proposed Work for FY05

In FY2005, we will (1) research more advanced tracking methods based on salient points and particle filters; (2) apply the tracking methods to understand and address the situations where they fail, especially in low-resolution, low-frame-rate video; (3) develop software to post-process the tracks; and (4) provide a user interface to handle specific conditions such as tracking a particular vehicle or highlighting tracks that pass through a region of interest.

Publications

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Moelich, M. (2004). *Autonomous motion segmentation of multiple objects in low resolution video using variational level sets*. UCRL-TR-201054.

Scalable Discretization-Enhanced Solvers

Robert D. Falgout 03-ERD-033

Abstract

We propose to develop scalable, optimal linear system solvers for discretizations of an important class of partial-differential equations (PDEs) that are handled poorly by existing linear system solvers. These PDEs are used in accurate modeling of many phenomena studied at the Laboratory, particularly electromagnetics. The performance of codes for these applications can be adversely affected by poor performance of the linear system solver. Development of scalable, optimal linear system solvers for this class of PDEs will enable faster, more realistic simulations. To develop novel multigrid algorithms, we will exploit discrete representations of the large, oscillatory near nullspaces that characterize these PDEs.

Our goal is to extend this linear solver capability to a whole new class of PDEs, which in turn will impact a large set of the Lab's physics codes. As a specific example, a scalable algorithm for implicit Maxwell's equations will enable the EMSolve application code to solve problems that are orders-of-magnitude larger (involving greater accuracy in the computed solution) than can currently be solved today, significantly enhancing the Laboratory's capability in electromagnetics design and analysis.

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Mission Relevance

These PDEs arise in several LLNL applications. Helmholtz equations arise in light-propagation models for laser-plasma applications and in single-frequency source models for accelerator design. Maxwell's equations arise in broadband-frequency sources for accelerator design and optical scattering. Extending this linear-solver capability to linear systems that result from discretization of these equations will impact physics codes applicable to LLNL missions, especially in the area of stockpile stewardship.

FY04 Accomplishments and Results

We developed and implemented a geometric multigrid algorithm that uses operator-based interpolation to solve variable-coefficient time-domain Maxwell equations. Numerical testing shows that this scales well for two and three-dimensional problems with coefficient jumps of 4 to 5 orders of magnitude. We also developed and tested physics-motivated boundary agglomeration interpolation schemes for variable-coefficient problems. The approach we used extends easily to unstructured grids, which is one of our main goals. Both geometric and unstructured algebraic solvers for Maxwell's equations are being tested on the HYDRA code.

Proposed Work for FY05

In FY05, we will produce a robust parallel multigrid code for solving the definite Maxwell equation on structured grids. Based on our promising existing numerical results discussed above, our current plan is to use the algorithm based on separate treatment of the curl-free and divergence-free components, in the context of the HYDRA code. We will also develop an AMG algorithm for solving the definite Maxwell equation on unstructured grids in the EMSolve code. Finally, we will investigate algorithms for solving the indefinite Maxwell equations, using techniques to design robust interpolation schemes for the variable coefficient problem.

Publications

Jones, J. F. and B. Lee. (2004). *A multigrid method for variable coefficient Maxwell's equations.* UCRL-JC-204137.

Parallel Graph Algorithms for Complex Networks

Edmond T. Chow 03-ERD-061

Abstract

In the intelligence community, huge amounts of data are being collected. A major challenge has been to sift through these data to identify potential threats by understanding the interactions among individual pieces or subsets of data. This project is developing parallel and scalable algorithms for searching very large, complex networks, with a focus on semantic graphs used in intelligence and other applications. This work will enable graph search on distributed parallel computers. We anticipate significant decreases in

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communication time by using edge partitioning and reductions in search complexity by using heuristic search. The project has two aspects: (1) develop efficient parallel implementations, which requires innovative data structures and algorithms, and (2) develop algorithms that exploit the structure of semantic networks.

Semantic graphs generalize networks to include heterogeneous nodes and links. The power of these representations lies not only in the structure of the links, but in the data that reside on them. We expect to develop new algorithms and software techniques that will enable complex queries to be carried out on large-scale semantic graph data on distributed parallel networks.

Mission Relevance

Semantic graphs are used to encode relationship data gathered by intelligence agencies for counterterrorism purposes. By exploiting the large aggregate memory of parallel computers, this research will enable the scaling to unprecedented sizes of complex network applications relevant to LLNL's national and homeland security missions. In addition, such parallel algorithms will also have application in complex networks in other disciplines, particularly the analysis of metabolic networks in support of LLNL's mission in bioscience.

FY04 Accomplishments and Results

We began in FY04 by implementing models of complex networks, including a new, realistic model for semantic graphs. Data were collected to verify the models, which involved converting data from various knowledge bases. Our work in graph partitioning led us to develop edge-partitioning approaches that are proving to be essential in reducing the communication time of parallel semantic graph searches. Simulations show a potential orders-of-magnitude improvement over traditional partitioning techniques. Another key accomplishment in FY04 was the development of heuristic search algorithms that reduce the complexity of point-to-point shortest path searches.

Proposed Work for FY05

In FY05, we plan to finish implementing a parallel graph library that uses edge partitioning, carry out theoretical and empirical studies to evaluate edge partitioning techniques for complex networks with different topological properties, develop more sophisticated search algorithms that utilize correlations of vertex and edge types in a semantic graph, and finally, study the performance of advanced data structures and implementation techniques to improve the interprocessor communication performance of our software.

Publications

Chow, E. (2004). *A graph search heuristic for shortest distance paths*. UCRL-JRNL-202894.

Eliassi-Rad, T. and E. Chow. (2004). *Using ontological information to accelerate search in large semantic graphs*. UCRL-CONF-202002.

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A Computational Design Tool for Microdevices and Components Used in Pathogen Detection Systems

David P. Trebotich 03-ERI-003

Abstract

We propose to develop new computational models to simulate complex biological flows in integrated microsensors and to validate these models experimentally. The proposed work is important because next-generation pathogen-detection systems being developed at LLNL leverage microfluidic technology. Our computational tool will (1) provide critical understanding of the fluid dynamics involved in microdevices, (2) shorten the design and fabrication process to reduce costs, (3) optimize prototypes, and (4) provide a predictive capability for new, more advanced designs. Computational work will be performed in collaboration with UC Davis, experimental work in collaboration with UC Berkeley.

We expect to produce computational tools for simulating complex biological flows in microdevices. With this capability, the design and fabrication cycle of microelectromechanical system devices, such as the "biobriefcase," can be shortened by months per cycle by eliminating the need for trial-and-error design. This tool will help to optimize current prototypes and will lead to new, more advanced designs.

Mission Relevance

The new computational models to simulate complex biological flows developed in this project will further LLNL's mission in countering the proliferation and use of weapons of mass destruction.

FY04 Accomplishments and Results

We (1) developed a complex geometry capability for flow described by incompressible Navier-Stokes; (2) extended our incompressible Navier-Stokes model in complex geometry to a higher order method for viscoelasticity; (3) coupled the particle model to the continuum model in complex geometry; (4) continued to develop the particle model to include more advanced constitutive modeling and used it to demonstrate shearing effects on a chain of particles, as well as particle-boundary interactions in a pillar array channel; (5) extended the complex geometry capability to the geometries of real device components and compared the results to experimental data; and (6) applied our code to the polymerase chain reaction (PCR) module of the biobriefcase to help evaluate DNA-capturing designs.

Proposed Work for FY05

We will extend our algorithms to adaptive mesh refinement and large-scale simulation frameworks to simulate 3-D flows, including the packed bed reactor of the biobriefcase PCR module. We will also investigate methods of sticking and breaking particle chains in our model; simulate flows in other microfluidic device components, including components of other modules in the bio-briefcase; and supplement our computational work with experimental data. Results should show how DNA molecules change conformation as they travel through a real microdevice, thereby helping to evaluate component designs.

Publications

Trebotich, D. et al. (2004). "A numerical algorithm for complex biological flow in irregular microdevice geometries." *Tech. Proc. 2004 Nanotechnology Conf. and Tradeshow* **2**, 470–473. UCRL-CONF-201479.

Long-Time-Scale Atomistic Simulations

Wei Cai 03-LW-027

Abstract

The limit of present-day atomistic simulations to nanosecond time scales precludes the study of many phenomena that occur on time scales of milliseconds or longer. We propose to develop a novel theoretical framework and its robust numerical implementation for effective atomic-scale simulations of rare yet fundamental processes that control the macroscopic behavior of complex systems. Our proposal promises to remove this fundamental constraint and open ways to heretofore impossible studies of scientific problems. We plan to combine the importance-sampling formalism and high-dimensional optimization techniques to conquer this challenge. Our effort will be to extend our new method, which has been successfully applied to small systems, to complex materials-science problems.

This research will establish a solid theoretical and practical foundation for long-time-scale simulations of complex processes. This will be a major advance in our ability to model and understand fundamental processes in condensed matter, which are of critical importance to several large programs at LLNL. Combining our methodology with the unique computational facilities at the Laboratory will significantly enhance our predictive atomistic modeling capabilities. In particular, our method should be of great value to applications involving long-time-scale biological processes.

Mission Relevance

This project will provide deeper insight into the atomistic processes controlling dislocation mobility and solid–solid phase transitions, which are of critical importance to several large research programs at LLNL, such as the Advanced Simulation and Computing Program's dynamics-of-metals effort, in support of stockpile stewardship.

FY04 Accomplishments and Results

During FY04, we implemented an efficient parallel Importance Sampling Monte-Carlo code, and applied it to the dislocation kink motion in silicon. The code employs an elaborate iterative minimization technique for optimizing judiciously chosen parameters for the importance function, such that the statistical error for the calculated transition rate is

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minimized. We also developed a new algorithm that optimally samples a reduced phase space containing the reaction coordinates. We also implemented a working code for this algorithm and demonstrated its efficiency and versatility when applied to the silicon dislocation motion problem.

Publications

de Koning, M. et al. (2004). *Adaptive importance sampling Monte Carlo simulation of rare transition events*. UCRL-JRNL-206340.

Three-Dimensional Vectorial Time-Domain Computational Photonics

Jeffrey S. Kallman 04-ERD-004

Abstract

Three-dimensional (3-D) time-domain (TD) design tools are fundamental to developing new technologies to create all-optical nonlinear logic systems for data generation, transmission, manipulation, and detection. This project proposes to develop state-of-the-art simulation tools for design and analysis of next-generation 3-D photonic integrated circuit devices. The simulation codes developed in this project will include models for optical gain and nonlinearities and microscopic, nonuniform, inhomogeneous structures. We expect to develop a new suite of 3-D tools general enough to be adapted to specialized problems in several areas, and flexible enough to embrace design of future mixed-signal systems and stand-alone systems in separate areas of the electromagnetic spectrum.

This project is expected to result in a set of sophisticated 3-D TD tools that designers can use to model new, complex photonic devices and to advance scientific understanding. By developing this sophisticated modeling capability LLNL will maintain a leadership role in the worldwide scientific and computational community. We expect to publish, patent, and license (where possible) the results of this work.

Mission Relevance

New capabilities developed in this project will benefit LLNL's national-security missions that rely heavily on advanced photonics. Applications include encryption circuits for secure communication and remote sensing for counter- and nonproliferation missions; high-bandwidth optics for advanced laser science and high-density optical interconnects for high-performance computing, both in support of stockpile stewardship; and microsensors for weapons miniaturization.

FY04 Accomplishments and Results

Work in FY04 focused on different approaches for two types of problems: those solvable using approximate methods and those requiring full-wave methods. We approached these two categories by enhancing two LLNL-developed codes: an approximate scalar 2-D nonlinear model (Quench2D) and a full-wave 3-D linear model (EMSolve). Quench2D was modified to

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incorporate a 3-D scalar fast-Fourier-transform-based beam-propagation kernel. In parallel, we added the ability to solve auxiliary partial differential equations that describe nonlinear effects for EMSolve. We also began numerical experiments to test our two approaches by comparison to literature examples and to experiment. Our codes were used in the RadSensor project and the Deterministic Nanoscale Fabrication of Mesoscale Objects project.

Proposed Work for FY05

Work in FY05 will extend the two approaches begun in FY04, specifically (1) extend the full-wave EMSolve code to model spontaneous emission, gain, and distributed sources and to simulate an in-bandgap Fabry-Perot interferometer; (2) modify the Quench3D code to model polarization effects, applying finite elements to both narrow-bandwidth Maxwell's equations and carrier density rate equations and to simulate waveguide-based x-ray detectors and gain-quenched laser inverters; and (3) begin incorporating quantum-well nonlinearities into both approaches, investigating appropriate semiconductor models and numerical methods for speeding computation of electromagnetic fields in a quantum well, such as implicit methods and mesh subcycling.

Publications

Bond, T. C. and J. S. Kallman. (2003). *Time-domain tools for the investigation of gain-quenched laser logic.* Presented at the 2003 Intl. Semiconductor Device Research Symposium, Washington, DC, Dec. 10–12, 2003. UCRL-JC-154843.

Locally Adaptive Mesh Refinement for Linear-Scaling Electronic Structure Calculations

Jean-Luc Fattebert 04-ERD-012

Abstract

In this project, we propose to investigate a new scalable algorithm for efficient electronic structure calculations, the most computationally expensive part of first-principles molecular dynamics simulations. In current application codes, the algorithm scales with the cube of the number of electrons and thus limits the size of tractable simulations. We hope to develop a new algorithm that scales linearly with the number of electrons—while preserving the accuracy of standard methods—by investigating a new way of representing the electronic structure using localized orbitals. Fine grids will be used to resolve the “center” of localized wave functions, and coarser grids will represent the tails of the functions. The new approach will use the SAMRAI parallel adaptive mesh refinement framework.

The final product of this research will be a fast and efficient algorithm for electronic structure calculations that may be used as the core of a first-principles molecular dynamics code. This algorithm could potentially reduce by more than one order of magnitude the cost of simulations of physical systems of the order of 1000 atoms, thus allowing larger simulations and longer molecular dynamics and opening the possibility of investigating new physical phenomena.

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Mission Relevance

Achieving an efficient, accurate, and scalable algorithm for computing the electronic structure of a system with many atoms would have a large impact in many research fields that have applications to stockpile stewardship, such as molecular dynamics simulation of materials or fluids at high temperature and pressure, and determining the equation of state of various materials.

FY04 Accomplishments and Results

In FY04, we were able to carry out two main developments simultaneously. First, we implemented a prototype parallel, standard finite-element electronic structure code based on SAMRAI, which includes some numerical kernels and algorithms we will use in our new algorithm. Second, new functionalities were implemented within SAMRAI to support our new algorithm. At the end of FY04, we began merging these two efforts to start testing the proposed algorithm. A parallel electronic structure code with local mesh refinement is already a unique tool—similar efforts have usually been limited to regular grids or serial codes.

Proposed Work for FY05

In FY05, we will develop and test an iterative algorithm to solve efficiently the Kohn-Sham equation in the context of multiple local adaptive-mesh refinements. In particular, we will develop an efficient multigrid preconditioner to speed up convergence; implement functionalities for full density-functional theory calculations; tune SAMRAI to achieve close-to-ideal scaling; and, by the end of the year, carry out scaling, accuracy and efficiency tests on realistic physical systems.

Publications

Fattebert, J.-L, and F. Gygi. (2004). "Linear scaling first-principles molecular dynamics with controlled accuracy." *Comput. Phys. Comm.* **162**, 24. UCRL-JRNL-202957.

Fattebert, J.-L. and F. Gygi. (2004). *Real-space methods: Towards O(N) first-principles molecular dynamics*. Presented at the APS Annual March Meeting, Montreal, Canada, March 22–26, 2004. UCRL-ABS-201307.

Exfiltration Interdiction Algorithm Development

David A. Knapp 04-ERD-047

Abstract

We will conduct basic research on generalized methods for detecting and interdicting data exfiltrated from classified to unclassified networks. Current approaches to the problem are limited to very application-specific techniques. Our goal is to invent and develop general techniques that are new and innovative.

Success in this project will lay the foundation for a large number of applications in information security. Protecting classified and sensitive information from exfiltration is a goal

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of all government institutions. This work will establish a cornerstone with application in many areas of information security.

Mission Relevance

This work will create new concepts that can be applied to the data exfiltration problem, in support of LLNL's mission in national security.

FY04 Accomplishments and Results

We investigated three possible approaches to interdiction and characterized and evaluated each approach by theoretical and empirical studies. We developed new tests and a deeper understanding of some key technologies useful in solving one of the general problems. A prototype detection component was implemented for inclusion into a standard framework, and we began the process of evaluating and characterizing its performance, which will continue in FY05.

Proposed Work for FY05

We expect to complete implementation and evaluation of a specific detection technique. The results will be presented in a national forum and integrated into an evaluation framework.

Protein Classification Based on Analysis of Local Sequence-Structure Correspondence

Adam T. Zeml 04-ERD-068

Abstract

We are developing a system for comparing protein structures to generate information about sequence correspondence between related protein structures. The ability to verify sequence-based alignments through comparison to the correct structural alignments will significantly improve the quality of protein structure modeling, protein classification, and protein function recognition. There are no fully automated libraries of protein folds that provide credible information about sequence correspondence (alignments) between protein structures. We will deliver a structure alignment program (STRAL) that will generate detailed information about the regions of global and local similarities between protein structures and will establish an automated database for the sequence-structure classification of proteins.

The software and database resulting from this project will have a significant impact on the classification of proteins by structure and function, and on protein structure prediction methods. This system will enhance our ability to identify regions on protein surfaces as candidates for ligand binding sites. Because accurate structural analysis is requisite to computational protein-based detection schemes, this work will improve the success rate and reduce the cost for choosing high-quality, unique protein surfaces for antibody or high-affinity ligand recognition, and will improve our ability to identify possibly cross-reactive proteins related to proteins targeted for detection.

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Mission Relevance

The proposed protein structure comparison system will have immediate applicability for research related to the Laboratory's homeland-security mission. Our proposed library of protein sequence–structure motifs will provide a significant advance in protein modeling capability that will enable us to predict more high-quality signatures of protein targets from pathogens of interest to homeland security.

FY04 Accomplishments and Results

In FY04, we developed a prototype version of STRAL that allows us to partition protein structures into arbitrary segments, make structural comparisons at multiple levels of granularity, score structural similarity, and classify proteins according to structural relatedness. Tests of modules developed for STRAL show that by using our multilevel method for detecting similarities between protein structures and our similarity scoring function, we can cluster proteins automatically into classes and folds that correspond to those determined manually in the Structural Classification of Proteins (SCOP) database. We also worked on the definition of scoring function and detection of residue–residue correspondence between compared molecules.

Proposed Work for FY05

In FY05 we will develop the protein sequence–structure database by (1) classifying proteins (and domains) into clusters according to their similar structural motifs, (2) analyzing and comparing our database with SCOP to refine our classification criteria and methods, (3) conducting pair-wise comparison of proteins in the database using high-performance computing resources, and (4) classifying and clustering proteins based on their structural similarity and sequence correspondence.

Publications

Chauhan, S., A. Zemla, and V. Motin. (2004). *Introduction of internal histidine affinity-tag into plasminogen activator of* Yersinia pestis. Presented at the ASM Conference, New Orleans, LA, May 23–27, 2004. UCRL-ABS-202437.

Goens, S. D. et al. (2004). "Bovine enterovirus type 2: complete genomic sequence and molecular modeling of a reference strain and a wild type isolated from endemically infected U.S. cattle." *J. General Virology* **85**, 3195. UCRL-JRNL-202639.

Kanterdjieff, K. A. et al. (2004). "Mycobacterium tuberculosis RmlC epimerase (Rv3465): A promising drug-target structure in the rhamnose pathway." *Acta Crystallographica* **D60**, 895. UCRL-JRNL-202415

Zemla, A. et al. (2004). *LGA protein structure analysis and AS2TS protein structure modeling systems.* http://as2ts.llnl.gov/. (retrieved Dec. 28, 2004). UCRL-WEB-206292.

Zhou, C. E. et al. (2004). *High-throughput selection of protein-based signature targets for detection of bio-threat agents.* ASM Conference, Portland, OR, Oct. 6–9, 2004. UCRL-ABS-206063.

Electro-Thermal-Mechanical Simulation Capability

Daniel A. White 04-ERD-086

Abstract

This project will research and develop numerical algorithms for three-dimensional (3-D) electro-thermal-mechanical (ETM) simulations and incorporate them into the ALE3D and Diablo computational mechanics codes. A coupled 3-D ETM simulation solves, in a self-consistent manner, the equations of electromagnetics, heat transfer, and nonlinear mechanics. Research will include advection of electromagnetic quantities in an ALE setting, algorithms for electrical contact and slide surfaces, and electromagnetic boundary conditions. Extensive algorithm analysis and code verification will be performed to ensure the model equations are solved correctly. To validate the simulation capability, we will compare simulation results to available measured data from magnetic flux compression experiments.

We expect to develop a unique, robust ETM simulation capability that will enable the high-fidelity simulation of explosively driven magnetic flux compressors, electromagnetic launchers, inductive heating and mixing of metals, microelectromechanical systems, and biophysics applications. Results of our research will be submitted to peer-reviewed journals.

Mission Relevance

The ETM simulation tools developed in this project will find application in weapons systems for the stockpile stewardship mission, as well as a variety of pulse power systems used for basic DOE physics research. In addition, ETM simulation tools would support the design of electromagnetic launchers that have been proposed for DOE equation-of-state research and Department of Defense missile defense and artillery systems.

FY04 Accomplishments and Results

In FY04, after a midyear start, we researched several alternative formulations for the electromagnetics equations, focusing on stationary geometry (i.e., the eddy current problem) as a necessary first step towards dynamic moving-mesh problems. We evaluated the various formulations and began writing a manuscript that will be submitted for publication in a peer-reviewed journal.

Proposed Work for FY05

In FY05 we will incorporate the equations of electromagnetics into the ALE3D and Diablo codes; research coupling approaches; and investigate the stability, conservation, and convergence properties of our approaches. We will work to solve simple, fully coupled ETM problems by year's end. We will also tackle the key research issues of this project: advection of electromagnetic quantities, electrical contact and slide surfaces, and electromagnetic boundary conditions.

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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 approach, based on a "natural neighbor" discretization technique, to model extreme deformation and for failure analyses involving processes like earth penetration and dam failure. This method is related to finite-element approaches except that arbitrary polyhedral elements are defined by non-Sibsonian shape functions. A free Lagrange approach can then be exploited such that mesh connectivity is redefined on the fly to eliminate mesh tangling. No re-mapping of quantities will be required, since they will be stored at node points. The method will be effectively meshless and will be incorporated into the DYNA3D code.

If successful, the new approach will provide an improved method for modeling extreme events, e.g., earth penetration and dam failure. For example, since the method is fully Lagrangian, better descriptions of anisotropic material damage for concrete can be applied; even water spilling over a damaged dam could be modeled with this approach. Being meshless, this approach could be used for applications where nondestructive evaluations are required, from weapons analysis to biomechanics. The method is more stable than smooth-article hydrodynamics methods, faster than element-free Galerkin meshless methods, and circumvents the advection required by arbitrary Langrangian/Eulerian (ALE) techniques, making it applicable to a much larger class of problems.

Mission Relevance

The proposed approach treats both solid and fluid mechanics and is applicable to several important LLNL national security and homeland security missions. Analyses of earth penetration and terrorist vulnerability evaluation of infrastructure elements are target applications for this new approach. As-built x-ray tomography of laser targets and in vivo magnetic resonance imaging imaging for biomechanics create "point clouds" and are good examples where some form of meshless method is needed for expediting stress analyses.

FY04 Accomplishments and Results

In FY04, definition of data structures and code module development for the DYNA3D implementation began, including (1) setting up general, ragged-type data structures to handle arbitrary connectivity of polyhedral elements in DYNA3D; (2) implementing Vornoi techniques for identifying nearest neighbors; (3) developing a method for determining which neighbors to include in a natural-neighbor set near convex boundaries; and (4) implementing non-Sibsonian interpolation modules in DYNA3D. The result of work in FY04 was a serial implementation not yet able to handle the connectivity updates needed to eliminate mesh tangling in the very large deformation problem.

Proposed Work for FY05

In FY05 we will finish implementing small deformations in NIKE3D and DYNA3D by extending work from FY04 to handle large deformations and damage, including (1) further evaluating different approaches for determining surface definition from a cloud of points; (2) evaluating the free Lagrange approach for large deformations; and (3) exploring different damage models.

Internet Ballistics: Supplanting Source IP as the Sole Arbiter of Internet Attribution

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 and counterintelligence efforts require adversary identification, but rely primarily upon easily spoofed or illicit source IP addresses. The proposed work seeks to supplant IP address with other evidence characteristic of the adversary. We will measure the influence on packet arrival patterns due to varied attack software, platform performance, and intervening network conditions by employing controlled experiments with reference platforms. Mathematical characterization will employ wavelets.

The project will be able to separate adversary-specific traffic signatures from network-specific traffic signatures and will provide a testable foundation for a system of attacker attribution that can be employed in augmenting and disambiguating adversary profiles. Importantly, this capability enhances adversary identification through passive means that cannot be detected by the adversaries in question. A prototype throughput system for continuous generation and update of scan signatures will be produced. Directions for further research in improving the underlying metrics will be indicated.

Mission Relevance

This work will support Laboratory and DOE/NNSA's national security missions in cybersecurity and will benefit counterintelligence efforts through increasing analysts' ability to relate what might otherwise appear as unrelated network activities. In the cybersecurity realm, added confidence in source attribution can lead to improved discovery of related intrusions that might have been overlooked due to deliberately obscured or multiplied source IP addresses. Correct and consistent identification of adversaries, despite obscured or ephemeral source IP addressing, is critical to the effectiveness of cybersecurity and counterintelligence missions.

FY04 Accomplishments and Results

Our network scan partitioning and wavelet scalogram characterization techniques have proven effective to support a reliable methodology for pattern-based network adversary identification. In over 60% of those cases where hostile scans were repeated after a interlude

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of days, the traffic-pattern matching techniques alone succeeded in correct matching to unique source IP address. Several data transformations and different wavelet measures were evaluated in combination to optimize the traffic characterization scheme. This work has provided a foundation to conduct controlled studies in our pattern matching to disambiguate the effects of adversary behaviors from those attributable to network topology and location. Improved scan capture tools have been approved for deployment at LLNL.

Proposed Work for FY05

To expand the effectiveness of our identification method, we will refine and apply our proximity metrics to more recent data, captured with tools providing higher timestamp resolution. We will also conduct controlled studies to generate and capture specific adversarial traffic, using attack tools commonly employed by the hacker community. These studies will control separately the effects of attack algorithm, source machine characteristics, and varied network location, seeking means to isolate their individual effects upon packet arrival patterns through adjustment of the vector space components. A paper detailing the methodology and results will be submitted for publication in a peer-reviewed journal.

Publications

Bartoletti, A. and B. Parno. (2004). *Internet ballistics: Retrieving forensic data from network scans*. Presented at the 13th Usenix Security Symp. UCRL-POST-205856

Petascale Simulation Initiative

David R. Jefferson 04-ERD-102

Abstract

This project will dramatically increase LLNL's capability in scalable multiphysics and multiscale simulations for stockpile stewardship and other applications. The focus will be development of algorithms and software for efficient coupling of multiphysics calculations in a Multiple Program, Multiple Data (MPMD) environment on massively parallel computers, like BlueGene/L, and on adaptive algorithms for multiscale modeling. The architecture developed will facilitate coupling existing, independently written codes, thus leading to new multiphysics and analysis capabilities. Advances to the programming environment will provide more flexible, and potentially more efficient, use of massively parallel computers coming to LLNL. Adaptive sampling will enable more efficient use of multiscale models.

A scalable, simulation-component ("symponent") architecture (SA) will be developed and implemented for multiphysics codes, and the core capabilities will be demonstrated with a full-scale simulation on a high-performance computing platform. Adaptive sampling algorithms will be created and tested, permitting significantly more efficient use of subscale physical models in full-scale simulations. The code coupling and adaptive sampling software will be portable, useable with other LLNL codes, and applicable to a broad class of material

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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 mission areas that require the multiphysics, multiscale synthesis of existing models. The SA will facilitate coupling existing codes, shortening development time for complex applications. MPMD capabilities exercised in the demonstration problem will help achieve greater machine efficiency. Adaptive sampling applied to multiscale material modeling will be useful for many applications requiring subscale physics. In addition, this work is unique and high risk; our progress will be highly visible and lead to publications and proceedings.

FY04 Accomplishments and Results

After a midyear start, we (1) completed a survey of the software architecture of 52 unclassified, parallel, LLNL simulation codes to guide the design of the SA (UCRL-TR-206653); (2) completed initial specification of the SA programming interface; (3) implemented initial serial versions of three adaptive sampling algorithms (one described in UCRL-CONF-206365) driving a crystal plasticity model; (4) began studying issues related to performance and accuracy; and (5) drafted a plan for porting Linux to BlueGene/L.

Proposed Work for FY05

In FY05 we will (1) write the first version of the SA interface specifications and implement it on the development platform; (2) convert the finite-element code ALE-AMR and a lightweight polycrystalline plasticity code to symponent form; (3) create the first parallel version of the multiscale coupling model; (4) write the first generation of parallel sampling and interpolation algorithms; and (5) run test problems to exercise the core SA and sampling algorithms; (6) assess BlueGene/L environment in light of the requirements of the SA and begin Linux port; and (7) begin porting the initial symponent architecture to BlueGene/L or the simulator to identify compilation or linking problems.

Publications

Rudd, R. E. and D. R. Jefferson. (2004). *Toward on-the-fly multiscale modeling of damage localization*. Presented at the 2nd Intl. Conf. on Multiscale Materials Modeling, Los Angeles, CA, Oct. 11–15, 2004. UCRL-CONF-206365.

Rudd, R. E. (2004). *Multiscale modeling of elasticity and dissipation in NEMS resonators*. Presented at the 2nd Intl. Conf. on Multiscale Materials Modeling, Los Angeles, CA, Oct. 11–15, 2004. UCRL-ABS-203665.

Tannahill, J. R. and R. C. Becker. (2004). *Survey of selected LLNL unlcassified parallel simulation codes.* UCRL-TR-207192.

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Simulation of Biochemical Pathway Adaptability using Evolutionary Algorithms

William J. Bosl

04-FS-008

Abstract

Systems biology seeks a quantitative and predictive description of cells, but theoretical and experimental methods necessary for such descriptions still need to be developed. This project will investigate a novel systems approach to understanding the adaptability of biochemical pathways. Yeast will be used as a model organism because of the availability of data, and evolutionary algorithms and ordinary differential equations (ODEs) will be used to simulate pathway adaptability.

For the explicit purposes of this project, and projects that are intended to result from it, a quantitative representation must include both the static structure of the network, showing interactions between various genes and proteins, and the dynamics of the network. The model need not be numerically precise, but only accurate enough to answer the questions being asked and to allow evolution of the model pathway. Furthermore, the model has to be constructed from available data. The project will include the development and testing of hypotheses to explain the observed adaptability of yeast biochemical pathways when the myosin-II gene is deleted, and development of novel, data-driven evolutionary computation as a way to connect exploratory computational simulation with hypothesis-driven experimentation.

The scientific goal for this project is to demonstrate a new quantitative approach to understanding biochemical pathway adaptability on a simple model system. Through our collaboration with researchers at Harvard University, this project will establish new capabilities in computational genetics, which will help us better serve mission-critical biological research. This work is high risk, and is expected to eventually result in one or more high-visibility publications. Furthermore, it will serve as an effective recruiting vehicle to attract top young systems biologists to the Laboratory.

Mission Relevance

Computational methods for building predictive models of cellular processes will be essential to realizing DOE missions in biodefense and in biological research to improve human health, including the Genomics:GtL. For instance, such work focuses on engineering the genomes of key organisms for environmental cleanup, climate-change mitigation, and energy production, all of which will require quantitative tools for understanding the evolvability of organisms. Biodefense work will benefit by using these tools to study host–pathogen interactions.

FY04 Accomplishments and Results

Our original goal was to base our biosystems model on kinetic equations and ODEs. After working closely with collaborators at Harvard, we found that this might not be the best approach for studying pathways where detailed kinetic information has not been measured and therefore would not achieve significant biological goals. Instead, we developed a new approach to systems biology using technology from systems engineering. Petri nets, fuzzy logic, and genetic algorithms were successfully demonstrated and will enable future work in this direction. The change of approach did not affect the original goals of the project, which were to develop a strategy for studying the evolvability and adaptability of biochemical pathways using the mitotic exit network in yeast as the model system.

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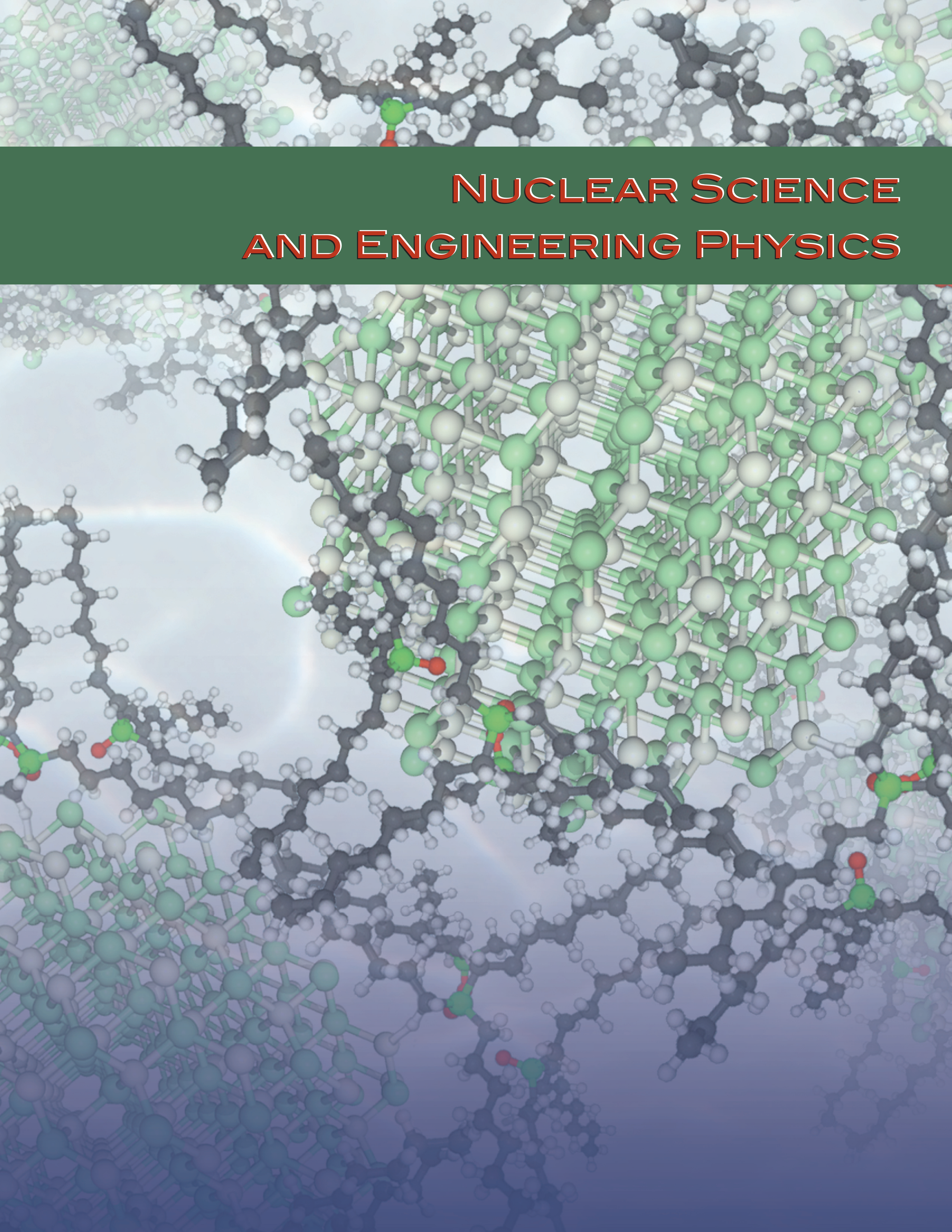
Nuclear Science and Engineering Physics



Nuclear Science and Engineering

Mononitride Fuel Development for STAR and Space Applications

Jor-Shan Choi

03-ERD-019

Abstract

The project will manufacture modified nitride-based uranium (UN) fuels for the small, secure, transportable autonomous reactor (SSTAR) concept. The UN fuel is also a good candidate for space nuclear reactor and research reactors. In the project, the thermal and mechanical properties of the modified UN fuel will be examined according to a set of criteria (e.g., compactness, proliferation-resistance, safety, reprocessing and waste disposal) for suitability of use in SSTAR. The fuel will be analyzed computationally to examine its performance under irradiation. We will also collaborate with UC Berkeley to develop a waste-disposal model for spent nitride-based fuel.

This project will develop a modified uranium- (U-) based fuel for use in compact, long-life reactor cores. Hafnium nitride (HfN) may be added to the fuel design as a burnable neutron poison. Zirconium nitride (ZrN) may be added to displace uranium nitride (^{238}UN) and limit the buildup of plutonium in the reactor or to increase the stability and life of the fuel. Uranium-235 enrichment will be tailored to optimize the fuel life. Uranium nitride is more suitable for compact reactors than UO_2 because of its high actinide density and high thermal conductivity. Modification of the composition with HfN, ZrN, or other materials may significantly increase the stability and lengthen the life of the reactor core. A success in this project will contribute to alternative fuels concepts for nuclear energy.

Mission Relevance

This project supports LLNL's national-security mission by advancing work in proliferation-resistant reactor designs. Such reactors would be fabricated and sealed by a trusted nation and shipped to other nations for up to ~30 yr of use, after which the sealed reactor would be returned to the trusted nation for disposition. This project will contribute to the emerging Advanced Fuel Cycle and Generation IV Initiatives and could also support NASA's space initiative, Prometheus, which calls for the development of a compact liquid-metal-cooled nuclear reactor for deployment in space. In addition, a modified UN fuel with high U density and good thermal conductivity, could be a good candidate as replacement fuel for research and test reactors worldwide.

FY04 Accomplishments and Results

In FY04, the computational code SPACEPIN was used to evaluate the irradiation data of the UN fuel manufactured for the SP-100 space reactor compositions (containing ZrN, HfN, or other materials) to optimize the long-life reactor core. Results of the SPACEPIN calculations (fission gas release, etc.) indicated good agreement with SP-100 fuel pellet irradiation data, thus establishing the computational capability for nitride-based fuel. A nitride-fuel

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manufacturing facility was also set up first to manufacture mono-uranium nitride fuel. The modified uranium nitride fuel compositions were studied based on a set of selection criteria, including compactness, proliferation-resistance, safety, reprocessing, and waste disposal.

Proposed Work for FY05

In FY05, we will modify the SPACEPIN code to include thermal and mechanical properties of ZrN and HfN, so that the irradiated fuel performance of the modified UN fuel can be analyzed. We intend to compare the SPACEPIN computational analysis results with other codes such as LIFE4Rev1 and SIEX3. We will fabricate the modified U-based nitride fuel, and will collaborate with Argonne National Laboratory and Westinghouse in nitride-fuel corrosion testing at a Westinghouse facility. Also, we will collaborate with UC Berkeley to develop a waste-disposal model for nitride-based fuel and will pursue a potential collaboration with DOE Naval Reactor laboratories and industrial entities to investigate and develop space reactor-fuel technologies.

Broadband Radiation and Scattering

Niel K. Madsen 04-ERD-017

Abstract

The goal of this project is to enhance LLNL's computational electromagnetics capability in the area of broadband radiation and scattering. Broadband analyses include electromagnetic interference and electromagnetic compatibility noise analysis, broadband radar, and accelerator wakefield calculations. LLNL analysis codes are limited by the accuracy of radiation boundary conditions (RBCs), which truncate space. The project will develop improved RBCs by (1) extending the perfectly matched layer (PML) approach to non-Cartesian meshes and (2) research and develop discrete-time-domain, boundary-integral techniques, which are compatible with high-accuracy, finite-element methods and capable of arbitrary accuracy. We will compare the two approaches for accuracy and efficiency for a variety of radiation and scattering problems.

The ultimate deliverable is an enhanced computational electromagnetics (CEM) capability that can provide accurate and efficient computational solutions to broadband radiation and scattering problems. The algorithms for improved RBCs will be incorporated into LLNL's existing EMSolve code. The result will be a 10× to 1000× improvement in the accuracy of simulations. Improved algorithms and our existing high-performance computer hardware will place LLNL's CEM activity among the top capabilities in the world. This research and the resulting capability will be documented in appropriate peer reviewed publications.

Mission Relevance

This project supports the national security mission by reducing the time and money spent in building and testing existing programs, and will enable computer simulations for new devices and systems, performance analysis of systems critical to nonproliferation efforts, and the design of micropower impulse radar and other microwave systems.

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FY04 Accomplishments and Results

In FY04, we extended the PML concept to higher-order unstructured grids and analyzed the performance of this approach. We performed novel, higher-order photonics simulations that would have been impossible without these extensions to the PML concept. Results were presented at a 2004 IEEE conference, and a manuscript has been submitted to the Journal of Computational Physics. For boundary integral techniques, we developed a prototype code that is used to evaluate various boundary integral formulations. Significant effort was spent on investigating numerical instabilities; we determined that temporal basis functions (vs. temporal differencing) are required for stability.

Proposed Work for FY05

The stability and accuracy of the boundary integral approach depends critically on the integration or quadrature rules. We will continue to research singularity extraction, polar integration, and Duffy transformation based quadratures. We plan to collaborate with Prof. Jandhyala, an expert on time-domain integral equations at the University of Washington, on time-integration methods and the hybrid finite-element boundary element formulation. We expect to have a fully functioning, although not fully optimized, time-domain boundary integral code working by the end of FY05. This will enable the full hybridization with finite elements in FY06.

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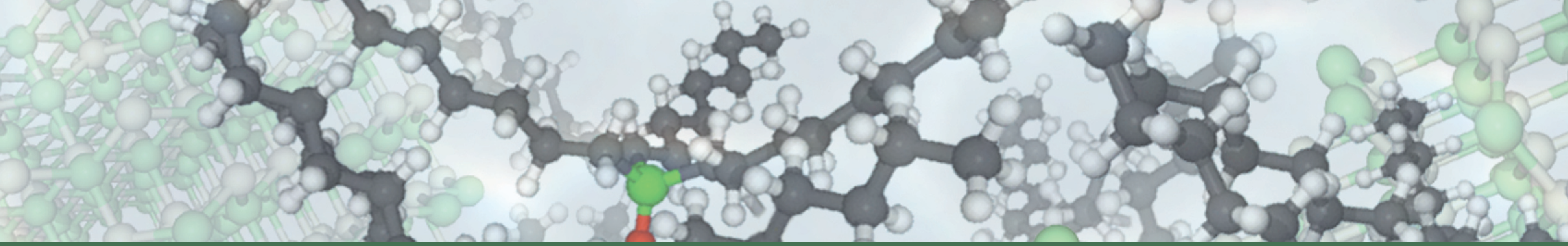
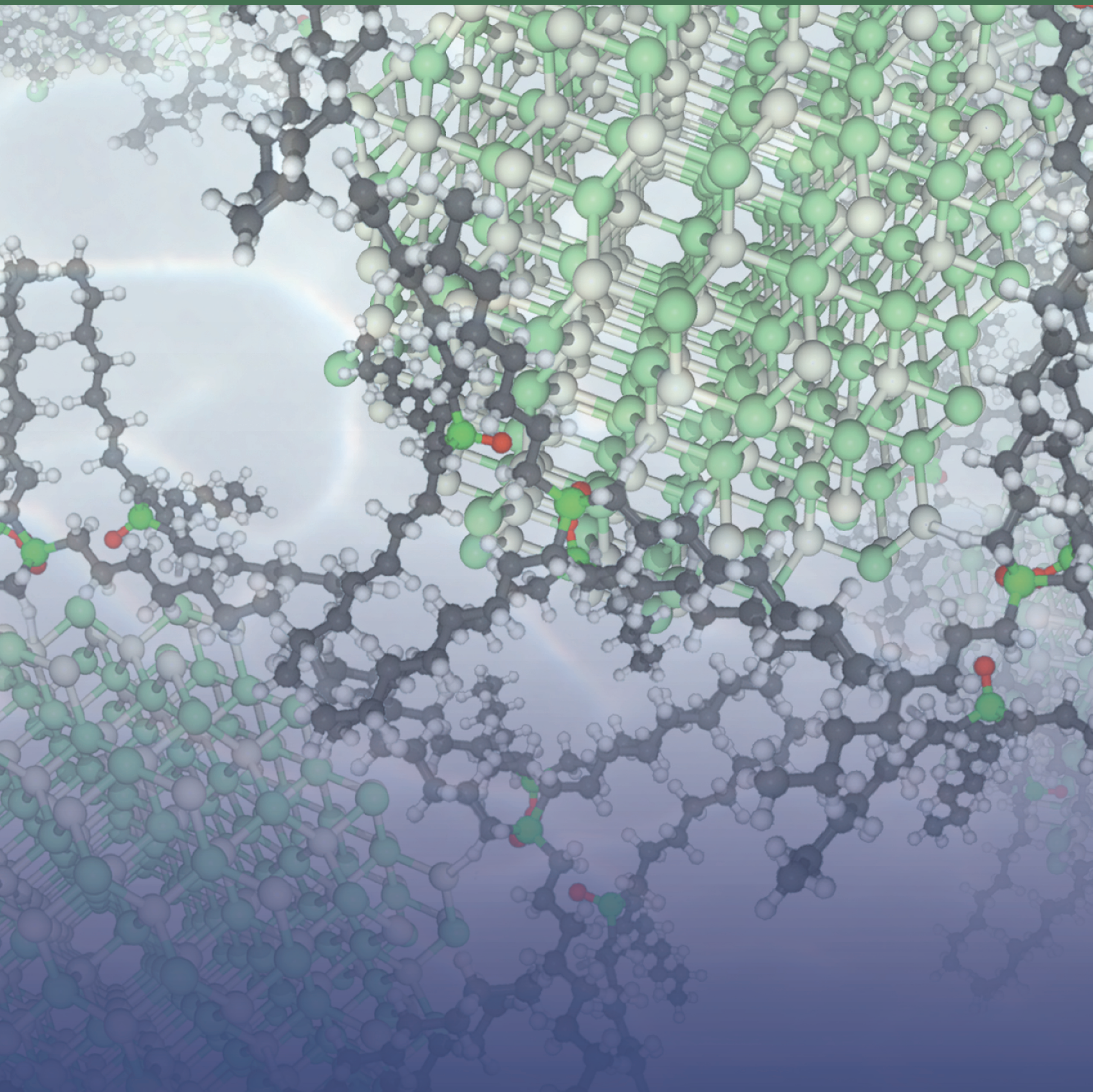


Physics



Physics

Structure and Spectroscopy of Black-Hole Accretion Disks

Duane A. Liedahl 02-ERD-004

Abstract

We are designing a computer model of accretion-disk atmospheres, COMPASS, with the goal of elucidating the high-radiation-density environments associated with mass flows in the curved spacetime near black holes. By combining our expertise in spectral modeling, LLNL's computational resources, and models of irradiated accretion disks developed by the project team, we will evolve the capability to generate realistic, theoretical x-ray spectra of accretion disks to explore the behavior of matter in the strong-field limit of gravitation.

COMPASS will give us dramatically improved modeling capabilities relevant to x-ray spectral formation in black hole and neutron-star accretion disks, a key scientific driver in the field of high-energy astrophysics. To this end, we will also advance the field of atomic modeling of plasmas in non-local thermodynamic equilibrium through the develoment of atomic models of near-neutral multielectron ions. Success in this major focus area of high-energy astrophysics will also help recruit talented astronomers and astrophysicists to the Laboratory.

Mission Relevance

This project maintains and extends LLNL's core competency in atomic modeling and computational radiation transport, which is vital to the Laboratory's stockpile stewardship mission. Furthermore, it will yield important new data sets useful for benchmarking our atomic physics calculations and for exercising our spectral synthesis and radiative transfer models. The project will also be an excellent recruiting magnet to attract young scientists with skills already closely allied with weapons physics.

FY04 Accomplishments and Results

To complete the radiative-transfer capability in COMPASS, we added resonant line scattering by incorporating an algorithm into our Monte Carlo code and conducted testing. This provided a crucial ingredient to COMPASS—line opacity. The final step to be included was a probabilistic line-splitting capability. We developed a geodesic solver for both non-rotating and rotating black holes, the latter of which was a key activity in FY04. The algorithm was tested and shown to run independently, i.e., in vacuum. A future task would be to incorporate this algorithm into the Monte Carlo code. However, we successfully ported the code to the FROST supercomputer, thus affording a major speed increase.

Publications

Mauche, C. W. et al. (2004). "Reprocessing of soft x-ray emission lines in black hole accretion disks." *Astrophys. J.* **606**, 168. UCRL-JP-200059.

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Reaching Isochoric States of Matter by Ultrashort-Pulse Proton Heating

Pravesh K. Patel 02-ERD-006

Abstract

This project is developing a method to obtain experimental equation-of-state (EOS) data of high-pressure, off-Hugoniot material states in the warm dense matter (WDM) regime (solid density plasmas, 1 to 10 eV) for the first time. To accomplish this, we will use an intense, ultrashort pulse of protons produced by LLNL's Janus ultrashort-pulse laser (JanUSP) to volumetrically and nearly instantaneously heat a material to high pressure (1 to 10 Mbar) and temperature (1 to 10 eV). In addition, the project will use a second proton beam to measure the rarefaction wave velocity in the plasma by two-dimensional spatial and time-resolved radiographs. This measurement will provide the first EOS data of any material on the isochore.

We expect to obtain, for the first time, experimental EOS data of high-pressure, off-Hugoniot material states, a previously inaccessible regime. The major result of this project will be a full understanding and optimization of the proton-production mechanism in laser–foil interactions.

Mission Relevance

Our project supports LLNL's national-security mission by investigating WDM, a topic of interest to the Stockpile Stewardship Program. In addition, our project's successful collection of EOS data in a highly complex and previously inaccessible regime will impact many core elements of LLNL's scientific program, including inertial confinement fusion target design, high-power laser experiments, planetary core physics, and high-pressure theory and molecular dynamics modeling.

FY04 Accomplishments and Results

In FY04 we continued experimental and modeling work, including analysis of data from FY03 experiments. Improvements were made to the modeling of proton heating, including the addition of temperature gradients in the heated sample. This enabled more realistic comparisons with the data and showed that time-dependent spectral emission from a sample could indirectly provide information on the interior conditions. The proton radiography results indicated that scattering was a problem at low proton energies and limited the spatial resolution obtainable with the JanUSP proton beam. In the future, scaling to the higher laser energies due to be available should enable the EOS technique to be demonstrated.

Publications

Mackinnon, A. J. (2004). *Proton probing of plasmas and shocked materials*. UCRL-ABS-155849

Snavely, R. A. (2004). *Proton beam focusing and heating in petawatt laser–solid interactions.* UCRL-ABS-152428

Town, R. P. J. (2004). *Calculations of proton radiography of magnetic fields in hohlraums.* UCRL-JC-152014-ABS.

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Proton Radiography of Laser–Plasma Interactions with Picosecond Time Resolution

Andrew J. Mackinnon 02-ERD-012

Abstract

This project has developed the capability to perform laser-driven proton radiography with picosecond resolution to diagnose electromagnetic fields and density perturbations in laser plasma conditions. Proton deflection experiments have directly measured highly transient electric fields of 7 keV/cm inside a plasma that is opaque to conventional probe beams. Simulations performed with a hybrid PIC/LSP code and the Lasnex hydrodynamic code agree well with the experimental data. These experiments have been conducted on the Janus ultrashort-pulse laser at LLNL, the Vulcan multibeam facility in the U.K., and the LULI laser at Ecole Polytechnique in France. Possible future experiments include investigating magnetic field generation in laser-produced plasmas.

Probing plasmas with picosecond proton probe beam has given us entirely new information on the generation of extremely high electric-magnetic fields inside a wide range of plasmas, from those occurring inside imploding shells to ignition conditions inside laser-driven fusion target capsules. These results are now being used to validate field generation models in important plasma simulation codes such as Lasnex and LSP.

Mission Relevance

The new diagnostic capability provided by proton probe beams with picosecond time resolution has enabled electromagnetic fields inside plasmas to be studied for the first time. These data will be used to validate plasma simulation models in areas that have not been possible until now. This has the potential to contribute crucial verification and validation information to the Inertial Confinement Fusion (ICF) Program and the Stockpile Stewardship Program in support of LLNL's national- and energy-security missions.

FY04 Accomplishments and Results

In FY04 we used results from experiments we conducted at the LULI and Vulcan lasers to develop a predictive capability for the proton probing of diagnosing fields and density inhomogeneities using LSP. This was used to predict deflectometry results expected in experiments to measure magnetic fields in a laser-produced plasma, thus meeting one milestone. The second milestone, which we also met, was to determine the utility of proton probing as a diagnostic for ICF experiments. The proton probing of density structures in an imploded cryogenic capsule and measurements of magnetic fields in a laser-driven hohlraum using the results of this project are currently under consideration.

Publications

Mackinnon, A. J. et al. (2004). "Proton radiography as an electromagnetic field and density perturbation diagnostic." *Rev. Sci. Instr.* **75**, 4183. UCRL-CONF-203614.

Mackinnon, A. J. et al. (2004). *Proton probing of plasmas and shocked materials.* Presented at the Conf. on High-Temperature Plasma Diagnostics. Apr. 19–22, 2004. San Diego, CA. UCRL-ABS-155850.

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Mackinnon, A. J. et al. (2004). *Generation and application of laser driven proton beams*. UCRL-PRES-203304.

Dense Plasma Characterization by X-Ray Thomson Scattering

Otto L. Landen 02-ERD-013

Abstract

Our goal is to demonstrate x-ray Thomson scattering as a robust and unique technique for the microscopic characterization of dense plasmas. Such information could be used to interpret laboratory measurements of material properties such as thermal and electrical conductivity, equation of state, and opacity. For example, the plasma ionization-balance predicted by various theoretical models in the warm dense plasma regime varies considerably and has never been tested against experiments. A series of x-ray Thomson scattering experiments will be completed at the Omega and upgraded Janus laser facilities, and the results compared to ionization-balance models and integrated radiation-hydrodynamic code predictions.

We expect the scattering experiments to distinguish between various existing ionization-balance models of low-*Z*, solid-density plasmas. The overall goal is to demonstrate x-ray Thomson scattering as relevant to high-energy-density physics (HEDP) and physical data research, for eventual application to the fusion-class lasers and other HEDP facilities. We hope to demonstrate the technique's applicability to kilojoule (not just >10 kJ) laser facilities. If we are fully successful, we expect the technique to eventually be used at planned facilities such as the Linac Coherent Light Source and the Z Machine.

Mission Relevance

This work provides new insight into dense-plasma statistical mechanics, a key competency at LLNL. For inertial-confinement fusion (ICF), which supports LLNL's energy-security mission, x-ray scattering could be used to measure the fuel adiabat in both planar- and converging-capsule geometries. For HEDP, which also supports stockpile stewardship, we plan to demonstrate that x-ray scattering can provide a noninvasive, time-dependent measurement of the internal temperature in radiation-dominated flow in foams and gases.

FY04 Accomplishments and Results

In FY04, we finished the demonstration and code comparisons of time-resolved scattering results from hohlraum-driven carbon foams at Omega. The diagnosis was extended to lower-density, hotter plasmas using a 2× higher energy (9 keV) zinc resonance line scattering source. We published a comparison of various ionization balance models with earlier results from volumetrically heated carbon foams. The publications have generated wide interest in the ICF plasma physics community, leading to a new collaboration with scientists at the Laboratory for Laser Energetics (Rutherford, England) measuring the adiabat of directly driven plastic capsules. In parallel, we have fielded a focusing version of our

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spectrometer, allowing us to record the first scattering data on a subkilojoule-class facility and a short-pulse facility.

Publications

Gregori, G. et al. (2004). "Electronic structure measurements of dense plasmas." *Phys. Plasmas* **11**, 2754. UCRL-JC-152388.

Pak, A. et al. (in press). "X-ray line measurements with high-efficiency Bragg crystals." *Rev. Sci. Instr.* UCRL-JC-154964.

Gaseous Laser Targets and Optical Diagnostics for Studying Compressible Turbulent Hydrodynamics

Michael J. Edwards 02-ERD-023

Abstract

Our objective is to use gas targets and optical diagnostics in laser-driven experiments to provide high-quality data on compressible (high-Mach-number) turbulent mixing, which cannot be obtained using traditional x-ray diagnostics. The Janus laser is being used to drive a blast wave to interact with a gas jet, producing a turbulent flow, that will be diagnosed by powerful optical methods. We are collaborating with the University of Texas at Austin.

If successful we expect to (1) obtain high-quality images resolving 2 to 3 orders of magnitude in spatial scale in compressible shock-driven turbulent flows; (2) obtain crude velocity information; (3) implement a laser-sheet diagnostic, which is the first step towards advanced diagnostic techniques; and (4) examine other potential advanced diagnostics. Unlike the majority of shock tube experiments, these experiments will provide high-Mach-number data for developing and validating turbulence models. Such data cannot be obtained using the traditional laser-driven technique of x rays and solid targets.

Mission Relevance

Turbulence modeling is an active and important part of stockpile stewardship because turbulence cannot be simulated directly but must be calculated using models, which are necessarily based on experiment. Virtually all existing high-quality data are from low-Mach-number experiments. This work supports LLNL's stockpile stewardship mission by providing improved data from high-Mach-number experiments for developing and validating turbulence models.

FY04 Accomplishments and Results

Our two main objectives for FY04 were developing (1) the gas target and (2) the laser-sheet diagnostic. Gas target development was successfully completed. After prototyping, a "minichamber" was designed and built, which sits inside the main vacuum chamber and allows targets to operate at atmospheric pressure. Full-up testing was successfully done on the

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Janus laser with very limited shots, and modifications to the design subsequently incorporated. The laser-sheet diagnostic was designed. The Janus upgrade in FY04 led to decreased shot opportunity during commissioning and an increase in user demand. This project received only a handful of shots for the entire funding period, thus preventing further progress.

Publications

Hansen, J. F. et al. (in press). "Laboratory observation of secondary shock formation ahead of a strongly radiative blast wave." *Phys. Rev. Lett.* UCRL-JRNL-205848.

Remote-Sensing Signatures for Ballistic Target Interceptions

Glen T. Nakafuji 02-ERD-035

Abstract

The objective of this project is to identify and characterize optical signatures for remotely assessing the destruction of intercepted missiles with nuclear, chemical, or biological payloads. We investigated candidate signatures using analysis, modeling, and experiments. Our focus was on nuclear signatures generated after a successful missile intercept, since identifying the type of incoming warhead is vital for determining operational, policy, and response options. We validated nuclear signatures using explosively driven experiments to examine the behavior and spectral emission of low- and high-Z surrogate materials in conditions analogous to those during a successful missile intercept.

Successful results obtained in this project will significantly reduce uncertainty in signature calculations for nuclear targets and enhance LLNL's capabilities in characterizing the signatures from devices with weak emission.

Mission Relevance

This work supports LLNL's national-security mission by characterizing signatures used for missile intercept, by extending LLNL's capability for identifying proliferant signatures, and by validating LLNL codes used in stockpile stewardship to model the temperature of metals in release and expansion.

FY04 Accomplishments and Results

In FY04 we measured the velocity, configuration and temperature of high-velocity Au jets driven by high explosive detonations, established an experimental technique for obtaining hydrocode benchmarking data, and observed the presence of jetting in low-Z shells using flash x-ray diagnostics.

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A Revolution in Biological Imaging

Henry N. Chapman 02-ERD-047

Abstract

This project will develop the science to enable ultrahigh-resolution imaging of biological materials with x-ray free-electron lasers (XFELs), which are under development. We plan to solve several critical problems in XFEL imaging, including modeling the atomic motion of the sample and image reconstruction. Our calculations will be used to determine limits to single-molecule imaging as a function of fluence and pulse length. We will assess the importance of XFELs and existing light sources to life-science programs, and experimentally test x-ray imaging techniques on a three-dimensional (3-D), micrometer-sized test object.

Early results from our models and experiments indicate that atomic-resolution imaging on XFELs should be feasible. The pioneering work performed in this project, such as the hydrodynamic model of the Coulomb explosion and the first lensless 3-D x-ray imaging, is establishing LLNL as a premier center for ultrahigh-resolution imaging. Our work will provide the basis to enable imaging at XFELs, and will determine a plan of research to develop the required technologies. Atomic-resolution imaging of macromolecules—the ultimate result of this work—will have an enormous impact on structural biology and medicine.

Mission Relevance

Improved tomography algorithms will benefit stockpile stewardship. As a specific example, diffraction imaging techniques can be applied to the study of warm dense matter, an important 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 biodefense and bioscience to improve human health. Our research also enhances the capabilities of the Linac Coherent Light Source, a high priority project of the DOE Office of Science, in support of LLNL's mission in breakthrough science and technology.

FY04 Accomplishments and Results

We extended our hydrodynamic model of the x-ray-induced damage of biological molecules to include trapping of the photoelectrons and three-body recombination effects. (The model and the results of an initial parameter study were published in *Physical Review E*. This study showed the required pulse lengths to be shorter than initially thought. We designed validation experiments to be conducted at the Sub-Picosecond Photon Source (SPPS) at the Stanford Linear Accelerator Center and at the Vacuum Ultraviolet–Free-Electron Laser (VUV-FEL) in Hamburg, Germany. We extended our x-ray image-reconstruction algorithms and performed the first experimental full 3-D lensless diffraction imaging from a rotation series of coherent x-ray diffraction patterns recorded at the Advanced Light Source. Our 10-nm-resolution image is the highest resolution full-3-D image ever made with x rays.

Proposed Work for FY05

We will (1) apply our hydrodynamic model to a full-parameter study of the diffraction imaging of macromolecules and theoretically investigate the effectiveness of damage-

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mitigation strategies, e.g., molecular orientational alignment; (2) continue the design of validation experiments at SPPS and the VUV-FEL, including the development of a particle injection system; (3) implement our image-reconstruction algorithms on a high-performance cluster to reconstruct our 3-D images at full resolution and image size; and (4) use the unique capabilities developed in this project to image a sample of aerogel at 10-nm resolution in all three dimensions, which will be as challenging as imaging single molecules.

Publications

Carrozzini, B. et al. (2004). "Phasing diffuse scattering: Application of the SIR2002 algorithm to the non-crystallographic phase problem." *Acta Crystallographica A* **60**, 331. UCRL-JRNL-203071.

Chapman, H. N., J. Hajdu, and K. Hodgson. (2004). *Report on the instrument development workshop for biological imaging experiments at LCLS*. Instrument Development Workshop for Biological Imaging, Menlo Park, CA, Mar. 15–16, 2004. UCRL-PROC-206061.

Chapman, H. N. (2004). *Introduction to the Instrument Development Workshop.* Presented at the Instrument Development Workshop for Biological Imaging, Menlo Park, CA, Mar. 15–16, 2004. UCRL-PRES-203061.

Chapman, H. N., et al. (2004). *Prospects for single-particle imaging at XFELs.* Presented at the IEEE LEOS Biophotonics Summer Topicals Meeting, San Diego, CA, June 28–30, 2004. UCRL-PROC-203932.

Chapman, H. N. (2004). *Diffraction imaging with x rays and electrons*. Presented at the First National Lab and University Alliance Workshop on Ultrafast EM, Pleasanton, CA, Apr. 15–16, 2004. UCRL-PRES-204134.

Hau-Reige, S. et al. (2004). *Pulse requirements for diffraction imaging of single biomolecules with x-ray FELs*. Presented at the Ultrafast X-Ray Science Workshop, La Jolla, CA, Apr. 28–May 1 2004. UCRL-POST-203935.

He, H. et al. (2004). "Use of extended and prepared reference objects in experimental Fourier transform x-ray holography." *Appl. Phys. Lett.* **85**, 2454. UCRL-JRNL-204107.

London, R. (2004). *The physics of x-ray–material interactions*. Presented at the University of Uppsala, Uppsala, Sweden, Sept. 27, 2004. UCRL-PRES-206131.

London, R. et al. (2004). *The physics of x-ray–material interactions.* Presented at the Ultrafast X-Ray Science Workshop, La Jolla, CA, Apr. 28–May 1, 2004. UCRL-POST-203788.

Marchesini, S. (2003). *Coherent X-ray diffractive imaging: Applications and limitations.* Presented at the C.A.S.I.S. Signal and Imaging Sciences Workshop, Livermore, CA, Nov. 20–21, 2003. UCRL-PRES-201069.

Single-Particle Nanotracking for Genomes-to-Life Applications

Stephen M. Lane 02-ERD-054

Abstract

This project is using real-time, single-particle imaging techniques to investigate the behavior of polymerase enzymes labeled with plasmon resonant particles (PRPs) that can

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be imaged with a dark-field video microscope. In experiments, these techniques will be used to (1) study the motion of molecular motors (in this case, DNA polymerase) to optimize rolling-circle amplification (RCA), a new LLNL-pioneered method of amplifying DNA, and (2) track the motion of individual biomolecules within a cellular environment. The experiments will generate information on enzyme trajectories, molecular forces, diffusion and binding rates, and molecular localization.

This project will result in a new capability for real-time imaging of labeled, single biomolecules and viral particles in vitro and within cells. Experiments will generate fundamental information at the single-particle level, e.g., how single enzymes replicate DNA, how particles (including toxins and viral agents) pass through cell membranes, and how particles are able to move within the cellular environment.

Mission Relevance

The project will support LLNL's national-security mission by enhancing competence in RCA, which has potential applications in bioterrorism agent detection. In addition to ongoing and expected national-security-related projects, the experiments are relevant to the new DOE Genomics:GtL initiative and will position LLNL to support existing and future DOE needs, e.g., pathogen detection, atmospheric warming, environmental cleanup, and energy production.

FY04 Accomplishments and Results

In FY04, we achieved a major milestone: development of a novel platform for the observation of single polymerase enzymes. This platform consists of a modified darkfield video microscope with a custom stage producing an adjustable magnetic field that stretches 50,000-base DNA strands that had been functionalized with magnetic beads. This allowed us to directly observe the behavior of PRP-labeled polymerase enzymes. Another major accomplishment was the development of a computer tracking code, which we used to produce trajectories and diffusion constants from video images for individual PRPs outside and inside living cells. This work has paved the way for future studies on the behavior of individual enzymes and PRPs.

A Tunable, Monochromatic, 1-Angstrom, Compton Scattering X-Ray Microfocus for Multiwavelength Anomalous Diffraction Experiments

Frederic V. Hartemann 02-ERI-004

Abstract

This project proposes to achieve the first demonstration of the microfocus multiwavelength anomalous diffraction (μMAD) concept, in which tunable, monochromatic x rays produced by a Compton-scattering microfocus are imaged on a small protein crystal to yield the full three-dimensional (3-D) structure of the macromolecule, with atomic resolution. The experiments will be performed in two steps by first obtaining diffraction data from simpler crystals, to limit data collection to a manageable time, followed by a study of K-edge imaging to demonstrate that the

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source can be tuned sensitively enough. The x rays are produced at LLNL's Picosecond Laser-Electron InterAction for the Dynamical Evaluation of Structures (PLEIADES) facility.

Proving that Compton light sources produce tunable x rays that are suitable to perform diffraction experiments and are tunable through the K-edge of a given material will pave the way toward the development of tabletop x-ray sources. Ultimately, by revealing the full 3-D structure of the macromolecular protein with atomic resolution, such compact, tunable x-ray sources will revolutionize x-ray protein crystallography and high-throughput structural genomics.

Mission Relevance

This work extends the Laboratory's expertise in laser/electron-beam, high-brightness, hard-radiation sources, a developing core competency that supports the Stockpile Stewardship Program's studies of materials at extreme conditions. In addition, the project contributes to the Laboratory's homeland security mission by furthering the systematic search for viral and pathogenic protein structures.

FY04 Accomplishments and Results

In FY04, two key experiments were performed. First, the x-ray absorption K-edge of Ta was scanned to show the required tunability of the Compton x-ray source and its relevance to μMAD phasing. Second, diffraction was measured on a small, well-diffracting crystal to help validate the μMAD concept. The results of the first experiment are in excellent agreement with theoretical predictions, and we have published five papers describing this experiment within the broader context of commissioning the PLEIADES x-ray source. The second experiment, performed with highly ordered pyrolitic graphite, with high mosaicity (0.9 degrees), shows that even for imperfect crystals, such as protein crystals, diffraction can be obtained with a tunable Compton x-ray source.

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Beta-Decay Experiments and the Unitarity of the Cabibbo-Kobayashi-Maskawa Matrix

Paul E. Garrett 02-LW-026

Abstract

A study of super-allowed beta decay, governed by the weak nuclear force, provides a unique way to test the Standard Model predictions of electroweak interactions. This project will test the unitarity of the quark-mixing (Cabibbo-Kobayashi-Maskawa) matrix required by the Standard Model. Beta-decay tests of unitarity require extreme precision in measuring beta-decay half lives and branching ratios, and calculated corrections must also be applied to the data. The project will test and constrain these calculated correction factors by performing accurate half-life and branching-ratio measurements at Canada's Tri-University Meson Facility (TRIUMF) radioactive beam, ISAC, for nuclei where different sets of calculated corrections give divergent results.

The ultimate goal of this project is a definitive conclusion regarding the unitarity of the quark-mixing matrix and adequacy of the Standard Model of particle physics. This significant and high-profile research in particle physics will attract talented young physicists to the Laboratory.

Mission Relevance

Tests of the Standard Model have been identified as a high priority in the DOE and National Science Foundation Long-Range Plan for Nuclear Science. These studies complement and support the experimental program at DOE Office of Science laboratories such as the Stanford Linear Accelerator Center and Fermilab, where investigations into other quark sections are being performed.

FY04 Accomplishments and Results

In FY04, we completed the setup and tuning of our advanced data-acquisition system for the refurbished 8-pi gamma-ray spectrometer at TRIUMF. Initial tests of the system had demonstrated that we could quite readily reach the desired statistical uncertainty of <0.05% in half-life determination, but we had to overcome several sources of systematic uncertainty. After that, these sources were successfully brought under control. A final test of the system using a ^{26}Na beam was completed. Preparations for the key measurement on ^{34}Ar were made. Other experiments on the superallowed beta-decay emitters ^{18}Ne and ^{62}Ga were conducted, and data analysis was begun.

Short Pulse: Enabling Relativistic Applications for Advanced Inertial-Confinement Fusion

Christopher P. Barty 02-SI-004

Abstract

We are researching and developing next-generation laser technology for high-energy, ultrashort-pulse scientific applications. These include fast ignition, hard x-ray radiography, and studies of high-energy-density matter. These science applications are of increasing importance and the subject of a national initiative of NNSA. Our work has four main facets: (1) developing an advanced laser front end for chirped-pulse generation; (2) developing the key enabling technologies for compression and focusing of high-energy petawatt (HEPW) pulses; (3) evaluating system concepts for HEPW lasers; and (4) coordinating research and development for conceptualizing short-pulse science applications. We expect to develop chirped-pulse generation and other technologies for advanced inertial-confinement fusion.

Mission Relevance

This research will investigate the use of high-energy lasers for stockpile stewardship and fusion energy experiments that require a combination of focused, ultrashort pulses in the relativistic regime with high energy in long pulses. This research is directly relevant to LLNL's national security and energy security missions.

FY04 Accomplishments and Results

We developed (1) a novel technology solution for the first deployment of two 2-kJ, 10-ps pulses from a single laser aperture at future large laser systems; (2) diffraction gratings with record damage thresholds; (3) the first fiber-based, chirped-pulse amplification seed pulse laser system for Nd:glass lasers, which was coupled to an existing preamplifier to produce an integrated test bed; and (4) concepts for a near-field damage test system for prototype gratings. We also explored concepts for phase-array gratings and novel metrology for such arrangements. Work on HEPW science was directed towards establishing the feasibility of conceptual applications, particularly high-energy x-ray radiography.

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Chemical Dynamics of High-Pressure Interfaces

Eric R. Schwegler 03-ERD-001

Abstract

Interfacial systems are central to many areas of science, ranging from the study of physical properties such as melting and phase transitions, to chemical properties such as catalysis and corrosion resistance. Despite the importance and significance of interfacial processes, very little has been known about them at a microscopic level until recently. In this project, we have worked to develop and apply ab initio simulation methods to study the complex interfaces that are encountered in high-pressure physics at the microscopic level. We have built on our existing expertise in first-principles molecular dynamics by incorporating two-phase simulation techniques. These simulation methods have been applied to a set of low-Z materials that are relevant to potential high-power laser experiments.

The primary benefit of this project is in the development and application of a new capability at the Laboratory for investigating interfacial systems under extreme conditions. The success of this project depends on utilizing the unique computational capabilities of the Laboratory. In particular, the application of first-principles molecular dynamics to interfacial systems is not feasible without access to a parallel computing facility and state-of-the-art simulations codes. The techniques on which we have focused are general and can be applied to a wide range of systems. Our research has already led to several high-profile publications and has stimulated new experiments. We anticipate that additional publications on lithium hydride, hydrogen, water, and carbon will be generated in FY05.

Mission Relevance

This research supports LLNL missions in national and energy security by providing a detailed understanding of materials that undergo changes under pressure, melting, and phase transitions. This understanding will particularly benefit the stockpile stewardship program.

FY04 Accomplishments and Results

During FY04, we focused our efforts in (1) continued application of the two-phase approach to compute phase boundaries in lithium hydride, hydrogen, water, and carbon; (2) the use of methods that go beyond density functional theory to improve accuracies (corrections based on diffusion Monte Carlo energies); and (3) evaluation of the magnitude of non-adiabatic effects on our computed melt curve for systems such as hydrogen and water. During the past year, we succeeded in publishing our work in the *Journal of Chemical Physics*, and *Nature*. In particular, our article in *Nature* was highlighted on the cover of the Oct. 7th issue.

Proposed Work for FY05

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During FY05, we expect to make significant progress in the following areas: (1) determine the B2–liquid and the B1–B2 phase boundaries in lithium hydride; (2) develop an approach for using quantum Monte Carlo energies to improve the accuracy of computed melting temperatures; (3) examine the mechanism behind the liquid–liquid phase transition in hydrogen and locate the proposed liquid–liquid–solid triple point; (4) perform path-integral molecular dynamics simulations to examine the influence of quantum effects on the hydrogen melt curve; (5) continue performing two-phase simulations of the melting of ice-VII at 50 GPa and determine if a superionic phase is involved in the melting process; and (6) examine the melting of carbon at 10 Mbar.

Publications

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Photon Collider Physics

Jeffrey B. Gronberg

03-ERD-003

Abstract

This project will develop the physics case and run plan for photon collider experiments at high and low energies. A photon collider experiment is unprecedented, and a detailed simulation of the signal and background is critical to quantifying its physics potential. This project will create Monte Carlo tools and use them to evaluate a number of interesting physics cases. A signal possibly representing new physics and well suited to study at a photon collider will be analyzed at the proton collider at Fermilab.

If successful, this project will create a suite of Monte Carlo tools that will allow the evaluation of other physics analyses possible at a photon collider proposed at the Stanford Linear Collider (SLC). This project will also produce publishable estimates of the reach of new physics and spin-zero heavy-quark physics at a photon collider, a publishable search for new physics at the Collider Detector at Fermilab (CDF), and the basis for justification of a new experiment at the Stanford Linear Accelerator Center (SLAC) in which LLNL would have technical and scientific leadership.

Mission Relevance

Photon colliders open an entirely new field of particle physics and are enabled by LLNL's unique competencies in high-average-power, short-pulse lasers and laser–electron beam interactions. This project drives high-average-power laser technology and the science of laser-electron beam interactions, both of which are relevant to innovative concepts for future brilliant light sources to be used in dynamic materials studies in the Stockpile Stewardship and Inertial Confinement Fusion Programs, in support of national security missions.

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FY04 Accomplishments and Results

The ability of an SLC-based photon collider to create gamma–gamma and e-gamma luminosity was quantified. It was shown that such a facility would be competitive in the energy range from 3 to 20 GeV center of mass energy. A study of the reconstruction of eta mesons into a proton-antiproton final state was completed. A paper describing these results is in preparation.

Publications

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Quantum Electrodynamics and Electron Collisions in the Superstrong Fields of K-Shell Actinide Ions

Peter Beiersdorfer 03-ERD-004

Abstract

Because of the ultrastrong nuclear field found only in the heaviest ions, the actinides offer rich new physics that is missing in all-electron collision codes used at LLNL and elsewhere. This project studies the physics in the ultrahigh fields of the heaviest elements available to record the world's first high-resolution K-shell x-ray spectra of hydrogenic and helium-like U, Cm, and Cf. We will determine electron-impact excitation cross sections, dielectronic recombination, energy levels, and the Lamb shift. The project combines several LLNL capabilities: The world's only source of stationary high-*Z* ions (SuperEBIT); access to the relevant actinide isotopes; high-resolution, hard x-ray detectors developed in collaboration with NASA; and unique theory capabilities.

The project will provide the most definitive test of high-field quantum electrodynamics ever undertaken, identify the role of new forces active in high-*Z* collisions and quantify their significance for spectral modeling of high-temperature plasmas. The project will also develop a new class of hard x-ray detector—a high-resolution microcalorimeter array operating near absolute zero—and a new ultrahigh-resolution extreme ultraviolet (EUV) spectrometer, adding one-of-a-kind experimental capabilities to the Laboratory.

Mission Relevance

This world-class science effort to investigate new physics in the ultrastrong fields of highly charged actinide ions will create new science competencies and test the range of validity of collisional theory important for LLNL's mission in stockpile stewardship. It builds capabilities in astrophysics and develops novel x-ray detection equipment, also with application to stockpile stewardship. This world-class science effort also extends the Laboratory's core competency in atomic physics and will attract talented scientists to the Laboratory.

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FY04 Accomplishments and Results

A new 16-pixel calorimeter was completed at the beginning of FY04 and successfully implemented in a low-energy run on an electron beam ion trap. This run produced a wealth of atomic data that we began preparing for publication. We then improved the calorimeter's resolution for high-energy x rays by a factor of 4 from FY03 by developing a new mode of operation. We used the instrument to address the long-standing puzzle of the existence of the ^{229}Th metastable ground state by measuring radiation as high as 60 keV and provided the first reliable measurement of its energy. Our measurements of U yielded the most sensitive test of high-field quantum electrodynamics ever. We also developed a rare-isotope-injection system.

Proposed Work for FY05

In FY05 we will (1) complete the analysis and publication of our results with ^{229}Th and U; (2) extend our investigations to transuranic elements using our rare-isotope-injection system, focusing on Cm and Cf; and (3) use our high-resolution calorimeter to measure K-shell dielectronic recombination rates and to determine the size of the Breit interaction in high-*Z* collision processes.

Publications

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Exploring Properties of Quantum Chromodynamics with Proton–Nucleus and Deuteron–Nucleus Collisions

Michael D. Heffner 03-ERD-005

Abstract

The standard model of strong interactions predicts the presence at high temperatures of a phase transition in which nucleons dissolve into a plasma of quarks and gluons. Relativistic heavy-ion collisions provide the only laboratory-controlled method available for producing

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and studying this phase of matter and is the primary goal of the Relativistic Heavy Ion Collider (RHIC). Experimental proof of the production of this phase relies on a set of observations that are currently inconclusive. The key to their resolution lies in studying baseline proton–nucleus collisions. We will upgrade the Pioneering High Energy Ion Experiment (PHENIX) experiment at RHIC to characterize the fragmentation of the nucleus in proton–nucleus collisions to help resolve whether the phase has indeed been observed.

In this project, a new detector will be installed in PHENIX to characterize proton–nucleus collisions at RHIC. This will aid in the systematic study of bulk nuclear matter at high energy densities and help resolve a long-standing question: Can we create a new phase of matter, the quark–gluon plasma, in the laboratory and study its properties? We expect to gain a much improved understanding of physics in this regime.

Mission Relevance

This work further the Lab's national-security mission by advancing cutting-edge techniques in nuclear radiation detection and state-of-the-art computer simulation of complex events. This project will also help recruit top scientists in these fields through forefront research in nuclear and particle physics.

FY04 Accomplishments and Results

The detector was built and installed at the PHENIX experiment and the detector systems were commissioned in FY03, just before the deuteron-gold (d-Au) run for which this detector was built. After commissioning, we directly entered the data taking phase and successfully operated the detector collecting data for the full d-Au run. The response and calibration of the detector were studied, and, through simulation and special calibration runs, we achieved a detailed model of the detector's response, which is needed for the next step of extracting the centrality information from the collisions and using this information to understand the dense matter generated in the collisions.

Proposed Work for FY05

We are approaching a final result for the d-Au run for which this detector was designed. The plan is to continue support of the detector for future runs and finish the analysis, which consists of a number of details: understanding run-by-run variations and formulating calibrations run by run, understanding the systematics of the centrality model and the best method of fitting the data to the model, and furthering our understanding of saturation and cross-talk effects within the detector and from a nearby calorimeter. We have released some of the results to the collaboration, and we are working towards a final result that will be used to understand the nature of high-energy nuclear collisions at RHIC.

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Electron Production and Collective Field Generation in Intense Particle Beams

Arthur W. Molvik 03-ERD-008

Abstract

The future of heavy-ion fusion could depend on predicting and controlling electron cloud effects (ECEs), which limit the performance of present electron and ion storage rings. To better understand the phenomena and develop mitigating mechanisms, this project is conducting theoretical and experimental studies of electron-cloud generation, trapping, transport, and effects on ion beams. Experiments are utilizing accelerators at the Heavy-Ion Fusion Virtual National Laboratory and at LLNL. Theoretical studies are being coordinated with a Lawrence Berkeley National Laboratory LDRD project that combines a code that models electron clouds with the particle-in-cell code WARP for modeling self-consistent beams in accelerators. The project will validate the models and tools through comparison with experimental data.

Results from modeling and experiments will lead to the development of definitive, validated tools for studying ECEs in advanced accelerators; preliminary versions of these tools are already being applied to other accelerators. Results from work with a 4-MeV accelerator at LLNL will increase our fundamental understanding of processes involved in ion-induced gas desorption, a major issue at the largest accelerators.

Mission Relevance

By addressing electron effects in ion beams, the models and diagnostic techniques developed in this project will be applicable to high-intensity accelerator technology in support of LLNL's stockpile stewardship and fusion energy missions.

FY04 Accomplishments and Results

In FY04, we developed diagnostics that measure sources of electrons that cause ECEs (gas ionization, beam tubes, and end walls). We demonstrated three mitigation techniques: (1) reducing electron and gas emission and ion scattering by using rough walls, (2) suppressing electron emission from end walls, and (3) clearing electrons between quadrupole magnets. Simulations with prescribed electrons showed that ion beams are robust enough to withstand various types of possible electron cloud perturbations, such as random variations of fixed electrons, and that resonance with a breathing mode is the most damaging to ion beams. Future simulations will be self-consistent to enable us to determine the likelihood of a particular electron distribution, as well as shape-related details that may be important in determining ion-beam phase-space distortions. However, self-consistency required resolving the electron cyclotron period, resulting in time-consuming computations. We also developed an electron mover that allows time steps ~25× longer than the electron cyclotron period—a major advancement with applications in magnetic fusion energy, astrophysics, and near-space physics. We developed most of the modules needed for the self-consistent modeling of electrons in positive beams and applied them to three-dimensional code runs.

Proposed Work for FY05

In FY05, we plan to complete the integration of our self-consistent modeling code and apply it to beams with long pulses to understand self-consistent effects, especially offset beam interactions and beam losses. We will also test diagnostics for measuring electron accumulation, then use our electron-source diagnostics to study electron accumulation from each source. In so doing, we will build on our measurements (which indicate that gas desorption scales with a power of the electronic component of ion energy loss) to study desorption from surfaces characterized in situ, then compare the results with molecular dynamics modeling.

Publications

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A Coupled Turbulence-Transport Model for Edge Plasmas

Thomas D. Rognlien 03-ERD-009

Abstract

This project's goal is to develop the first predictive simulation model of the edge-plasma characteristics in tokamak fusion devices. Experiments have shown that edge-plasma properties are key to high fusion-energy output and determination of wall plasma fluxes and that simulations are needed to reliably design large devices. This project uses parallel computer codes to solve magnetized-fluid equations for both the fast-timescale turbulence in three dimensions (3-D) and the much slower toroidally averaged transport (profile evolution) in 2-D. Because the turbulence is driven by the plasma profiles, and profile transport is determined by

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turbulent plasma fluxes, multiple timescale coupling techniques are developed to obtain self-consistent solutions, including plasma and recycled and sputtered neutrals.

The project will provide the first predictive model of the edge-plasma region of a magnetic-fusion-energy tokamak. The properties of this region are key to controlling the overall energy confinement, and thus fusion-power output, as well as wall heat loads. The model will be used to help design and guide operation of future devices such as the International Thermonuclear Experimental Reactor. The project is also an important precursor for a more comprehensive computer model of a fusion device being planned by DOE under the Fusion Simulation Project and the planned Multiscale Initiative; both the physics models and the coupling techniques will be directly applicable to these development projects. The coupling techniques developed here should be applicable to other complex fluid systems of national interest such as climate and combustion.

Mission Relevance

The project advances fusion-energy research in support of LLNL's energy-security mission and develops numerical algorithms for large, coupled physics simulations on LLNL's high-performance computers in support of stockpile stewardship; the 3-D turbulence code is especially computer intensive. The project also supports the Laboratory's mission in breakthroughs in fundamental science.

FY04 Accomplishments and Results

We completed two aspects begun in FY03: (1) coupling density fluxes for evolving density and velocity profiles and (2) 2-D domain decomposition for turbulence code. We also completed short-time full-turbulence evolution within the code BOUT and transport-evolution comparison between BOUT and UEDGE for model turbulence. We accomplished the first coupled simulations with four plasma variables, showing strong convective transport as measured experimentally in the DIII-D and C-Mod tokamaks. The platform-independent, PYTHON-based coupling infrastructure was completed, as was the model for neoclassical collisional transport.

Proposed Work for FY05

We will complete self-consistent edge-plasma turbulence and transport coupling by including the stabilizing radial electric field. The simulation should conclusively demonstrate improved core confinement through edge transport barrier formation. We will implement and test advanced implicit integration schemes for noisy (turbulent) systems and include reduced kinetic corrections to fluid BOUT for parallel Landau damping and finite gyroradius effects that can be calibrated from the full kinetic code. In addition, we will continue the benchmarking model with experimental data from the DIII-D, C-Mod, and National Spherical Torus Experiment tokamaks to establish validity for using the coupled codes for prediction as well as deeper understanding of edge plasmas.

Publications

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Strain Rate Scaling of Deformation Mechanisms

James M. McNaney 03-ERD-015

Abstract

Our objective is to determine material-deformation mechanisms at ultrahigh pressures and strain rates. Further, we intend to assess the scalability of these results across a wide range of very high strain rates to determine whether what we learn about deformation mechanisms at very high strain rates is applicable to other regimes. To accomplish our objectives, we will use controlled, shockless loading along with soft-capture recovery on three experimental facilities: the Omega laser at the University of Rochester, the LLNL gas gun, and a high-explosives facility in Russia. Post-shot characterization will allow us to determine deformation mechanisms. Multiscale modeling simulations will guide experimental planning and mechanistic interpretation.

We expect to demonstrate the methodology for determining deformation mechanisms over a wide range of strain rates and ultrahigh pressures. This methodology uses multiple facilities to produce the relevant test conditions along with recovery and post-mortem characterization to infer the operative deformation mechanisms. The results from this project will provide basic scientific information in unexplored regions of pressure–strain-rate space, as well as providing a demonstrable scientific underpinning for the high-pressure strength measurements critical to the Stockpile Stewardship Program. Additional benefits include initiating an effort to investigate solid-state material behavior under extreme conditions and recruiting young scientists.

Mission Relevance

Deformation mechanisms provide the scientific underpinning of strength measurements at ultrahigh pressures. This project supports LLNL's national-security mission by developing a methodology for both assessing these deformation mechanisms and determining the scalability of high-strain-rate results, which will be critical input for long-range, science-based stockpile stewardship.

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FY04 Accomplishments and Results

In FY04 we (1) used post-recovery characterization to identify the dominant high-pressure shockless deformation mechanism in copper over the pressure range of 10 to 40 GPa, finding major differences between shock and shockless material response; (2) conducted the shockless loading of copper along with soft recovery on the LLNL gas gun; (3) began preparing the recovered samples for analysis; (3) prepared experimental designs and samples for Ta-recovery experiments; and (4) began our high-explosives-driven experiments at a facility in Russia.

Proposed Work for FY05

In FY05, we will (1) construct a deformation map for the high-pressure response of copper; (2) continue soft-recovery experiments with copper on the LLNL gas gun (3) conduct a pressure scan on Ta using the Omega laser; and (4) analyze samples recovered from the gas gun and the high-explosives facility experiments to assemble the first strain rate scaling study.

Publications

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Phonon Dispersion Curves Determination in delta-Phase Plutonium–Gallium Alloys

Joe Wong

03-ERD-017

Abstract

The goal of this project is to measure the phonon-dispersion curves (PDCs) of face-centered-cubic (fcc) Pu–Ga alloys, as well as their temperature and pressure dependencies. Pure Pu exhibits six solid-state phase transitions, with large volume changes, along the path to melting: alpha, beta, gamma, delta, delta’, eta, and liquid. The delta phase has desirable mechanical properties. Alloying with Ga expands the delta-phase field from high temperature to below room temperature.

To measure the PDCs of Pu–Ga alloys, we will use x-ray scattering methods employing high-brightness synchrotron sources to eliminate both the neutron absorption problem of Pu and single-crystal requirements for conventional PDC measurements with neutron scattering.

We expect to obtain high-q phonon dispersion data for Pu–Ga alloys and their dependence on Ga concentration to elucidate the T(111) softening and a possible Kohn anomaly. The results will be used to delineate the crystal dynamics associated with the delta-to-alpha’ transformation, which is important for understanding phase stability in Pu–Ga alloys.

Mission Relevance

This project will provide much-needed basic lattice dynamical data for phase-stability and property simulations of Pu materials relevant to science-based stockpile stewardship.

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FY04 Accomplishments and Results

In FY04 we (1) demonstrated that thermal diffuse scattering (TDS) can be used to determine PDCs for Pu materials; (2) were awarded 40 shifts at the European Synchrotron Radiation Facility (ESRF) for low-temperature inelastic x-ray scattering (IXS) experiments and 30 shifts at the Advanced Photon Source for our Ga-dependence TDS study; (3) measured the (110) TDS pattern for delta Pu–Ga using a 20-μm beam; (4) fabricated two high-purity delta Pu–Ga alloys (0.4 and 1.2 wt% Ga) with complete chemistry; and (5) designed, fabricated, and successfully tested online at ESRF a cryochamber with a microjet system for IXS measurements at temperatures as low as 200K, with ±0.1K stability over 30 h.

Proposed Work for FY05

In FY04 we will (1) fabricate large-grain Ga–Pu alloys with detailed microstructural characterization; (2) perform high-resolution IXS measurements at low temperature in the 0.6-wt%-Ga alloy to determine whether softening of the TA(111) branch is associated with the delta-to-alpha' transformation; (3) perform TDS measurements for Ga content and at low temperature to elucidate the nature of the TA(111) softening observed at room temperature; (4) explore the use of a diamond anvil cell to perform TDS under pressure to elucidate the nature of the Kohn anomaly observed in the T_1(110) branch and to determine the Gruneisen parameters; and (5) publish our results in refereed journals. Our overall goal is to obtain key lattice dynamical data pertaining to delta-phase stability.

Publications

Wong, J. et al. (2004). "Imaging phonons in a fcc Pu-Ga alloy by thermal diffuse x-ray scattering." *Appl. Phys. L.* **84**, 3747. UCRL-JC-151455.

Wong, J. et al. (2003). "Phonon dispersions of fcc Pu-Ga by inelastic x-ray scattering." *Science* **301**, 1078. UCRL-JC-153065.

Wong, J. et al. (2004). "Probing phonons in plutonium." *MRS Proc.* **802**, 112. UCRL-PRES-153799.

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Propagation Models for Predicting Communication System Performance in Tunnels, Caves, and Urban Canyons

Hsueh-yuan Pao 03-ERD-023

Abstract

This project employed statistical descriptors and random boundary conditions to address the problem of characterizing the propagation of electromagnetic fields in the adverse environments that are encountered in enclosures such as caves and tunnels, where rough and complex surfaces can compromise signal quality. The project investigated the propagation physics, developed the mathematical models, derived the statistical

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descriptors, tailored the results to radio communication in tunnels and caves, and selected the models and methods best suited to the enclosures of interest.

The techniques and tools developed in this research will lead to engineering of wireless systems for communications, sensor networks, remote sensing, and radio frequency (rf) ranging in complex environments, with possible applications in urban-canyon environments. The novel techniques that were developed in this project can be directly extended to other electromagnetic problems in complex environments that have national security and commercial applications.

Mission Relevance

Today, the limited ability to maintain communications in complex enclosed environments prohibits the rapid deployment of wireless systems in caves, tunnels, and urban canyons. The results of this project will enable wireless systems for communications, sensor networks, remote sensing, and rf ranging in such complex environments. These results will have application in counterterrorism and other efforts that support LLNL's missions in national security, in the Yucca Mountain Repository Project for LLNL's energy security mission, as well as in military applications for Department of Defense missions.

FY04 Accomplishments and Results

In addition to our ongoing assessment of previous research, analytical work during FY2004 focused on investigating the excitation and propagation of electromagnetic fields excited by sources situated within straight caves and tunnels that have rough walls. We derived the statistical characteristics of the fields and rf signals in straight, perfect-electrical-conductor, rough-wall tunnels and extracted the wireless communication channel model in the tunnel from the physics-based models. The results of this work are presented in five peer-reviewed papers that are in progress, one of which is scheduled to appear in 2005.

Publications

Dudley, D. (in press). "Propagation in lossy circular tunnels." *IEEE Trans. on Ant. & Propag.* UCRL-JP-200387.

Colliding Nanometer Beams

Jeffrey B. Gronberg 03-ERD-044

Abstract

The goal of this project is to develop and demonstrate an alignment and metrology frame that will allow beam position monitors (BPMs) to reach nanometer resolution in the measurement of the transverse position of an electron beam for a linear collider. Future linear colliders will achieve high event rates by focusing electron beams down to spot sizes of a few nanometers. Stabilization systems to hold the final magnetic optics steady at that level are under development, but definitive tests cannot be conducted until after

construction of the linear collider. Stabilization system failure would be a showstopper. However, a stabilization system test using a single beam is possible with nanometer-resolution BPMs. A metrology and alignment frame for a set of BPMs will be designed, built, and operated to demonstrate nanometer resolution.

The project will determine whether existing cavity BPMs can achieve nanometer resolution. This would be a more than order-of-magnitude improvement in current beam diagnostics and would provide sufficient signal sensitivity to monitor bunch tilt, which is expected to have a significant effect on the luminosity of future accelerators. A working nanometer-resolution BPM would allow the stabilization scheme required for colliding beams at a linear collider to be tested, greatly improving our confidence in the chosen linear collider design.

Mission Relevance

The DOE advisory panel on high-energy physics has identified a teraelectronvolt-scale electron–positron linear collider as the next big project in high-energy physics, and LLNL is a charter member of the U.S. R&D consortium for the design of such a machine. This project will allow a potential showstopper to be tested before detailed design and construction. The project will also advance our capability in nanometer metrology, which will support DOE and LLNL's mission in fundamental science and help recruit young talent.

FY04 Accomplishments and Results

The BPM alignment frame was successfully installed at the Accelerator Test Facility (ATF) in Japan, and beam data were collected. The frame allowed a high-precision calibration of the BPM electronics. We then performed in situ metrology of the BPMs at LLNL, which indicated that the electrical centers of the BPM fell within the 100-µm range of adjustment of the frame. Initial data taken with the BPMs at high gain uncovered a coupling between the *x* and *y* measurements that will require improved analysis code to uncouple. A design for the metrology frame based on a composite of carbon fiber and carbon foam was created. Initial bench testing of the nanogrid sensors was also begun.

Proposed Work for FY05

In FY05, the BPM metrology frame will be installed at the ATF, and final data will be collected. We will test new algorithms for data analysis of the BPM waveforms to resolve *x*–*y* coupling effects. The resolution of the system will be measured and published. Thermal drift of the BPM positions will allow the metrology frame measurements to be correlated with BPM measurements, thereby validating the absolute position measurement accuracy of the system.

Publications

Fitsos, P. (2004). *Design, simulation, and testing of a precision alignment frame for the next linear collider.* Presented at the 4th Intl. Conf. European Soc. for Precision Eng. and Nanotech., Glasgow, Scotland, May 30–June 3, 2004. Glasgow, Scotland. UCRL-CONF-204801.

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Laser–Matter Interactions with a 527-Nanometer Drive

Warren W. Hsing 03-ERD-070

Abstract

The primary goal of this research is to develop an understanding of laser–matter interactions with 527-nm (2ω) light for studies of interest to numerous Laboratory programs, including ignition, material strength, radiation transport, and hydrodynamics. The potential of significantly greater energy delivered onto targets at 2ω allows us to access a wider range of parameter space for ignition (higher yield or more robust ignition), probe increasingly higher pressures in matter, and access a wider parameter space for hohlraum-driven experiments. As part of this project, we are also developing the enabling technology and prototype instrumentation to diagnose a high-fluence laser beam for energy, power, near field intensity profile, and full aperture backscatter at 2ω.

As a result of this work, we will resolve important scientific questions relating to laser–plasma interactions, laser–target coupling, preheat and hohlraum filling, and gain a better understanding of the advantages and limitations of 2ω illumination relative to 3ω for high-energy-density (HED) and inertial-confinement fusion (ICF) applications. We will also pave the way for critical stockpile-stewardship-related experiments to be performed in matter in regimes of pressure, temperature, and strain rate much higher than previously accessible.

Mission Relevance

Five key areas within the Stockpile Stewardship Program would significantly benefit from the ability to conduct experiments with a 2ω drive: ignition, material dynamics, experiments using special nuclear materials, nuclear weapons effects testing, and hydrodynamics. This project will also enable cutting-edge research in other LLNL mission areas such as ICF and HED physics.

FY04 Accomplishments and Results

In FY04 we (1) completed a comprehensive set of optical diagnostics on the Omega laser to measure 2ω beam energetics; (2) measured and modeled high-temperature gas-bag plasmas at Omega using Thomson scattering and the three-dimensional radiation hydrodynamics modeling code HYDRA; (3) measured backscattering by stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS) in these gas-bag plasmas with phase plate smoothing and smoothing by spectral dispersion (SSD), which improved laser–target coupling by twofold and effectively suppressed beam filamentation and spray; (4) measured hot electron production and its correlation with SRS at 2ω, finding that hot electron preheat can be controlled by the choice of target materials; and (5) measured a higher x-ray temperature for hohlraums using low-density high-Z foam hohlraums, suggesting that the efficiency of converting laser light to x rays is affected by the density of the wall material.

Proposed Work for FY05

In FY05 we plan to (1) compare and model 2ω propagation and backscattering in empty (HED) and gas-filled (ICF) hohlraums from experiments on Omega; (2) obtain SRS data from

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large-scale-length plasmas using polarization smoothing with a 2ω beam (i.e., Omega); (3) compare experiment and theory for 2ω vs. 3ω coupling in well-characterized ignition-relevant cryogenic hohlraums on Omega; (4) measure forward SRS on Omega in a high-density target and assess the effects of hot electron production due to forward SRS for HED and ICF targets under ignition-relevant conditions; and (5) complete prototype diagnostics and hardware modifications for 2ω operations at future large-scale laser facilities.

Characterization and Optimization of High-Energy K-alpha X-Ray Sources

Jeffrey A. Koch 03-ERD-072

Abstract

This project will explore the generation, characterization, and optimization of high-energy (20- to 100-keV) K-alpha x-ray backlight sources for application to high-energy petawatt (HEPW) radiography experiments at future large lasers for fusion research. Our goal is to develop the capability to predict and optimize source parameters in HEPW experiments. To this end, we will characterize key K-alpha source parameters as a function of laser and target parameters in experiments at LLNL's Janus-pumped, ultrashort-pulse laser facility and (if possible) at higher-energy, petawatt-scale laser facilities.

By careful characterization of source parameters, we expect to illuminate the physics responsible for these parameters and, by learning more about the physics, to optimize backlight targets for specific applications. In particular, we will measure x-ray source sizes and yields from high-atomic-number targets at up to 100 keV, determine whether and how laser prepulses affect source characteristics, and use shifted K-alpha line signatures to determine local heating that might affect electron-beam distributions and therefore source size. We will also investigate physically restricted sources, which could enhance source brightness.

Mission Relevance

The use of x-ray sources as backlights for radiography experiments is an established technique for studying hydrodynamics, equations of state, and other properties of materials under extreme conditions of high temperature and density. Our project will investigate how to most effectively perform experiments at future large lasers for fusion research by using high-energy x-ray backlights to study large-volume, high-atomic-number materials. Success in this effort is directly relevant to the stockpile-assessment activities in support of LLNL's national-security mission.

FY04 Accomplishments and Results

In FY04, we measured Ag K-alpha and bremsstrahlung source sizes, K-alpha yields from solid foil targets, and Sm K-alpha and bremsstrahlung source sizes as functions of laser intensity, pulse duration, laser spot size, target thickness, and viewing angle. We also measured the spectrally resolved K-alpha emission using single-hit charge-coupled device (CCD) spectroscopy, showing

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multiple peaks from different ionization stages. Tests were conducted to compare an imaging plate, a scintillator CCD, and a Cd–Te array for detection of 20- to 40-keV x rays.

Proposed Work for FY05

In FY05, we will take additional measurements of Au K-alpha and bremsstrahlung source sizes and K-alpha yields from solid foil targets as functions of laser intensity, pulse duration, and laser spot size. We also will measure the spectrally resolved Cu K-alpha emission using high-resolution crystal spectroscopy and determine how laser prepulses and angle of incidence affect source characteristics, including yield and source size. In addition, we will investigate the effects that physically restricted sources such as small foils have on yield and source size.

Publications

Koch, J. A. (2004). *NIF HEPW 2D Radiography Option Survey.* UCRL-TR-203996.

Kuba, J. et al. (2004). *High-energy K-alpha x-ray source development at 22 keV: effects of irradiation parameters.* Presented at the 46th Ann. Mtg. Division of Plasma Physics, Savannah, GA, Nov. 15–19, 2004. UCRL-ABS-205340.

Park, H-S. et al. (2004). *Characteristics of high energy k(alpha) and bremsstrahlung sources.* Presented at the 15th Topical Confer. on High-Temperature Plasma Diagnostics, San Diego, CA, Apr. 19–22, 2004. UCRL-ABS-155826.

Park, H-S. et al. (2003). *20-100 keV K(alpha) x-ray source generation by short-pulse, high-intensity lasers.* Presented at the 3rd Int. Conf. Inertial Fusion Sciences and Applications, Monterey, CA, Sept. 7–12, 2003. UCRL-JC-155162.

Wickersham, J. E. et al. (2004). *Imaging and photon counting detectors for 20-100 keV x-ray backlighters in HEDES petawatt experiments.* Presented at the 15th Topical Conf. on High-Temperature Plasma Diagnostics, San Diego, CA, Apr. 19–22, 2004. UCRL-ABS-155825.

A Compact Accelerator for Proton Therapy

George J. Caporaso 03-ERD-073

Abstract

High-gradient insulator technology developed at LLNL and coupled with the dielectric wall accelerator (DWA) concept has produced the potential for a compact, high-voltage, short-pulse accelerator. This accelerator is being developed as a flash x-ray radiography source for stockpile stewardship. The successful exploitation of this technology to build small proton accelerators could revolutionize cancer radiation therapy and make this effective treatment widely available. Proton therapy is superior to x-ray therapy yet is practiced only in a few locations in the U.S. because of the large size and high cost of these facilities. In addition, this work may lead to compact accelerators for other applications such as high-energy physics, Compton scattering, and free-electron laser (FEL) drivers.

We have two major objectives. The first is to examine the feasibility of a DWA architecture that meets the requirements for proton therapy. This architecture will be verified by

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simulation and by construction and testing of scalable prototypes. Second, we will determine the performance limits of the switches and components needed for the accelerator. At the conclusion of this effort, we will know whether the DWA can be applied to proton therapy.

Mission Relevance

The project focuses on using DWA in a regime that has the potential to produce the highest accelerating gradient of any non-plasma-based acceleration scheme. These accelerators could be used for medical treatment, Compton scattering sources, FEL drivers, and high-energy physics, in support of LLNL missions in national security, biotechnology to improve human health, and breakthroughs in fundamental science. Furthermore, it will enhance capabilities in induction linac technology for stockpile stewardship.

FY04 Accomplishments and Results

In FY04, we demonstrated first- and second-generation SiC photoconductive switches (including switches made from both “A” and “C” plane wafers), and implemented changes to a three-dimensional (3-D) electromagnetic simulation code to enable simulation of nonlinear dielectric materials. This feature is necessary to model the performance of voltage-transformation lines. We also characterized samples of candidate nonlinear dielectric materials to determine which ones are suitable for use in the prototype voltage-transformation lines.

Proposed Work for FY05

In FY05, we will test our prototype voltage-transformation system and determine the performance limits of the SiC photoconductive switching technology and of the various accelerator components to assess whether the proposed accelerator is feasible for proton therapy.

A Next-Generation Microlensing Survey of the Large Magellanic Cloud

Kem H. Cook 03-ERI-002

Abstract

This project conducted a microlensing survey, called SuperMACHO (Massive Compact Halo Objects), that showed a significant improvement over the MACHO project in the detection rate of microlensing events. The main goal of the SuperMACHO project is detecting microlensing events toward the Large Magellanic Cloud (LMC). The research addresses one of the outstanding problems in the physical sciences: the nature and distribution of dark matter. In MACHO, the detected rate of gravitational lensing events indicated that MACHOs comprise, at most, 20% of the dark matter halo. Our goal is to determine the nature of this lensing population, which may be more massive than all other known components of the galaxy. We will use Chile’s Cerro Tololo International Observatory’s 4-m telescope to conduct this survey.

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We expect to locate a significant fraction of the lenses in microlensing events toward the LMC. This will determine the baryonic fraction of the Milky Way's halo dark matter. If the lenses are in the halo, we will have detected the first significant component of dark matter. If not, we will make the best determination yet that dark matter is composed of exotic subatomic particles. We will achieve significant experience in handling large quantities of large-format images and analyzing them in real time. This experience supports the development of software and techniques that apply to both national-security imaging needs and the development of the National Academy's highly recommended project for astronomy, the Large-Aperture Synoptic Survey Telescope.

Mission Relevance

The project supports LLNL's national-security mission by advancing imaging capabilities, specifically, persistent surveillance through wide-field imaging that requires real-time analysis of large data streams. The project also supports DOE's and NNSA's goal of doing great science by characterizing the nature of dark matter and uses LLNL competencies in image analysis and database management. The research is a collaboration involving universities and national facilities.

FY04 Accomplishments and Results

In FY04, we participated in the collection and analysis of data using both image-difference techniques and object-based photometry; produced a list of microlensing candidates (on a "microlensing alerts" webpage) and distributed it to the astronomical community; conducted spectroscopic observations for the best microlensing candidates; worked to determine detection efficiency in real time and to optimize the survey procedure to maximize detections; made significant improvements to the image-reduction pipelines by optimizing the image-subtraction algorithm and the image-co-addition algorithm; and worked on optimizing the observing strategy.

Publications

Nikolaev, S. et al. (2004). *Object-based photometry pipeline for SuperMACHO Project.* Presented at AAS Meeting 203. UCRL-POST-207223.

Silicon Nanocrystal Laser

June Yu

03-FS-010

Abstract

The purpose of this project was to demonstrate the world's first silicon-nanocrystal-based laser. Such a silicon laser, made using conventional silicon-manufacturing technologies, would be the crucial component that would enable a completely silicon-based photonic system. It could be both extremely small (a few tens of micrometers) and compatible with complementary metal oxide semiconductor technology, enabling arrays of such lasers to be

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integrated with standard integrated-circuit (IC) chip technology. The ability to fabricate arrays of lasers on an IC chip would make possible a host of promising optical technologies and devices, such as optical interconnects, optical computing, and new types of optically based biological and chemical sensors.

Demonstration of an all-silicon laser would have a tremendous impact on optoelectronics by enabling many new, promising optical technologies and devices. Success in the feasibility study would strongly motivate further work to eventually demonstrate an integrated silicon laser based on an electrical pumping scheme.

Mission Relevance

As the world faces increasing threats from chemical and biological weapons, advanced portable sensors offering increased sensitivity and selectivity are highly sought after. With the realization of an all-silicon-based laser, a host of new biological and chemical sensors could be integrated cheaply onto a compact-silicon chip. Developing an all-silicon laser would support the Laboratory's missions in counterproliferation and homeland security.

FY04 Accomplishments and Results

We conducted optical-characterization experiments on samples prepared by ion-implanting Si in fused silica substrates. A normal-incidence transmission spectrum showed useful (~10%) absorption at the 355-nm pump wavelength. Photoluminescence spectra in the 600- to 900-nm region were recorded. High-bandwidth recordings of the luminescence decay transients showed a "fast" component (~30 ns) and a "slow" (~1 ms) component. Waveguide characterization using a prism coupler showed single-mode guiding at 633 nm (in both the transverse electric and transverse magnetic modes) and at 800 nm. Assuming a rectangular refractive-index profile, we deduced an index of 1.61 and thickness of 0.5 µm for the Si-rich layer. The sharp guided-mode resonances showed promise for guided-wave pump-probe experiments.

Diode Laser Phase Conjugation

Ralph H. Page 03-FS-030

Abstract

This project investigated the feasibility of using optical phase conjugation (four-wave mixing) to achieve coherent beam combination using a plurality of high-power semiconductor laser diodes. Phase-conjugate reflectors undo variations in optical path length, in principle allowing coherent multiplexing. Net gain at each conjugate reflector is required to overcome system losses. Had this technique been demonstrated, it would have constituted an improved method of constructing a high-power, high-brightness, efficient, compact laser system. Based on our previous experience with solid-state-laser rate-equation modeling, we adapted theoretical models from low-power, single-mode diodes to high-power, broad-area (multimode) diodes and designed a laboratory experiment to test the concept.

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Our feasibility study aimed to investigate small-scale coherent operation of broad-area laser diodes. If feasible, large-scale coherent multiplexing of powerful, compact, efficient, inexpensive laser diodes would be a major development in laser technology, and could, in principle, displace alternative means of "radiance conditioning" (e.g., diode-pumping of solid-state lasers). High-power-laser applications (e.g., materials processing, missile defense, power beaming) would become more feasible. Diode laser phase conjugation may enable powerful systems to be more easily deployed in remote situations, such as aboard aircraft. Although not completely successful, the project identified key issues and obstacles to developing the technology.

Mission Relevance

High-power, high-brightness laser systems are applicable to missile defense and materials processing for the Laboratory's stockpile stewardship in support of the Laboratory's national security missions. Many other applications exist, as well.

FY04 Accomplishments and Results

In FY04, we conducted laboratory experiments using watt-level 800-nm broad-area diodes. We performed line-narrowing (important for four-wave mixing) using a Ti:sapphire laser for injection locking, and also tried a grating-tuned external cavity. We concluded that the many technical challenges make coherent beam combining of presently available broad-area diodes unfeasible. Continuing this work would require diode lasers that have inherently narrow line widths and large gains per pass, such as alpha distributed-feedback lasers.

A High-Efficiency Grazing-Incidence-Pumped X-Ray Laser

James Dunn 03-LW-001

Abstract

Our objective is to demonstrate a new high-efficiency, short-wavelength (<20 nm) x-ray laser operating at unprecedented 10-Hz rates and scalable to 1 kHz. This high-average-power tabletop x-ray laser would complement the waveband of current third- and future fourth-generation light sources, such as the Linac Coherent Light Source. The latter are large, expensive, synchrotron-based facilities, whereas our source will be compact and inexpensive, driven by a high-power laser. We will improve the laser-pumping efficiency by ten times and produce a proof-of-principle grazing-incidence-pumped (GRIP) 10-Hz x-ray laser. Plasma characterization, simulations of laser coupling, x-ray laser production and propagation are required. The work will be conducted on the LLNL JanUSP and COMET high-power lasers.

With our new idea for improving x-ray laser efficiency, we expect to achieve a high-output, 10-Hz x-ray laser operating at 13 to 20 nm with ~1 ps pulse duration equivalent to near-synchrotron-level output on a tabletop. The goal is to have a proof-of-principle demonstration but with useable output for applications relevant to LLNL programs, in particular an application that can utilize the high repetition rate and high output capability of the source. This methodology for generating x-ray lasers is also applicable to many other x-ray laser

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applications. In addition, with the single-shot high-energy capability of the JanUSP or Janus Intense Short Pulse (ISP) lasers, we will create the shortest wavelength (<4.5 nm) laboratory x-ray laser operating in the water window.

Mission Relevance

The development of a high average brightness x-ray source with ~100 fs to 1 ps pulse duration is relevant to various LLNL missions. X-ray probing of ultrafast processes in materials such as actinides is relevant to stockpile stewardship. A tabletop source would be ideal for at-wavelength, ~13-nm metrology of extreme ultraviolet lithography optics for microchip development, in support of LLNL's mission in fundamental science and technology. This x-ray laser could potentially operate below 4.5 nm, giving a single-shot capability lasting 100 fs for microscopy and holography of biological cells, in support of bioscience for our missions both in homeland security and in human health.

FY04 Accomplishments and Results

We demonstrated an 18.9-nm, 10-Hz laser output using the JanUSP laser at 150 mJ/pulse and performed simulations and experiments to optimize the output and characterize the pump conditions. achieving gain saturation. A beamline was implemented using Mo:Si multilayer coated optics to image the x-ray laser and characterize the gain region. This was the first step towards establishing the x-ray source for use in actual applications. We successfully repeated the GRIP x-ray laser on the COMET facility, where more energy is available to pump at a shorter wavelength. We submitted a Record of Invention and patent application and are preparing several papers for publication. Finally, we focused on further source optimization and a target design capable of running at up to 1 kHz.

Proposed Work for FY05

In FY05, we will complete the preparatory work and move from a proof-of-principle demonstration to a strong application that can use the full capability of the high-repetition-rate source. We will establish the laser pump energy parameters for a 13- to 20-nm x-ray source; the chosen wavelength will be strongly dependent on the laser pump available at LLNL or elsewhere. We will pursue collaboration with the National Science Foundation (NSF) Engineering Research Center for Extreme Ultraviolet Science and Technology at Colorado State University to work on an application based on interferometry, microscopy, or metrology, and the NSF Center for Biophotonics at UC Davis to investigate biological cell microscopy using high-power laser shots on a 4.5-nm x-ray laser at the Janus ISP facility.

Publications

Dunn J. et al. (in press). "Grazing incidence pumping for efficient x-ray lasers." *X-ray Lasers 2004: Proc. 9th Intl. Conf.*, Beijing, China, 24–28 May 2004. UCRL-CONF-204478.

Dunn, J. et al. (2004). *Recent developments in tabletop x-ray laser sources and applications.* Presented at the 28th Eur. Conf. on Laser Interaction with Matter, Rome, Italy, Sept. 6–10, 2004. UCRL-ABS-204479.

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Diode-Pumped Alkali Atom Lasers

Raymond J. Beach 03-LW-024

Abstract

One objective of our project was the first-ever scientific demonstration of a diode-pumpable laser using atomic alkali vapors as gain media. This new class of laser could potentially scale to extreme powers and might be better than diode-pumped solid-state lasers for machining special materials. The project demonstrated near-infrared first-resonance-line lasing in Rb and Cs using a bright Ti:sapphire laser as a surrogate diode pump. Diode-pumped operation was also attempted. Another project objective was the first demonstration of second-resonance-line (i.e., blue) laser action in alkali vapors. Stepwise pumping with Ti:sapphire lasers would improve the chance of reaching laser threshold, and the possibility of diode pumping would then be addressed.

The project aimed to demonstrate the viability of diode-pumpable alkali-based lasers in the blue region of the spectrum, which would have significant applications in materials processing, bioanalysis, and industrial uses.

Mission Relevance

High-average-power, high-beam-quality laser systems have particular relevance to the Laboratory's stockpile stewardship mission involving the machining of special materials, as well as scientific studies focused on the interaction of matter with intense resonant fields.

FY04 Accomplishments and Results

Using a cascaded two-pump sequence with Ti:sapphire lasers at 852.3 and 876.4 nm, we planned to demonstrate 455.7-nm lasing in Cs vapor. Our modeling codes, which reproduced the near-infrared laser behavior, predicted an easily reached laser threshold. However, without key highly-excited-state data for alkali atoms, the blue-laser code was based on important assumptions. Through experiment, we found that the second-step pump transition was badly pressure broadened, reducing its strength. In addition, the blue-laser transition was unexpectedly weak. We concluded that the laser would not work as originally proposed. However, by reducing the perturber-gas pressure, we raised the transition cross sections and obtained super-radiant blue emission in a "pencil beam." This indicates that blue lasing should still be possible.

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Publications

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Broadband Optical Parametric Amplification in Microstructured Devices

Igor Jovanovic 03-LW-040

Abstract

Optical parametric chirped-pulse amplification (OPCPA) is a technique for generating high-energy, high-average-power, ultrashort pulses. This project studied quasi-phase-matched (QPM) materials as nonlinear crystals for use with OPCPA. Quasi-phase-matched materials have many advantages over birefringent phase matching: higher gain, better beam quality, and greater wavelength flexibility. The project developed a theoretical framework for extending QPM OPCPA to a broad range of important wavelengths, and, for the first time, scaled QPM experimentally to multimillijoule energies by increasing the crystal aperture. The poling station developed in this project was used to design, fabricate, and test a novel, multilayer photonic structure.

Developing a multilayer photonic structure for OPCPA results in much improved beam quality and stability of the front-end high-gain preamplifier for petawatt (PW) laser systems and will enhance research at LLNL in the field of high-contrast, high-peak-power lasers.

Mission Relevance

This work is of critical importance for developing broad-bandwidth, short-pulse technology for PW high-peak-power lasers. The use of PW lasers in high-energy-density facilities will enable high-resolution x-ray backlighting and fast-ignition studies for the Stockpile Stewardship Program.

FY04 Accomplishments and Results

In FY04, we developed a computer code to optimize the spectral bandwidth in any nonlinear crystal configured for QPM. To our knowledge, the result of our modeling provided the first complete framework for amplifying broad-bandwidth pulses centered at an arbitrary center wavelength. Energy scalability of QPM OPCPA to >1 mJ was demonstrated using a novel double-pass architecture, meeting our energy milestone. Furthermore, we produced microstructured materials in several crystals using the new poling station. Finally, the first $BaZnF_4$ crystals were grown and tested for second-harmonic generation, resulting in improved prospects for direct amplification of ultraviolet pulses. This project produced two records of invention and a number of publications.

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Publications

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Ebbers, C., J. R. Schmidt, and I. Jovanovic. (2004). *High-beam-quality optical parametric chirped-pulse amplification in periodically-poled $KTiOPO_4$*. Presented at the Advanced Solid State Photonics, Santa Fe, NM, Feb. 1–4, 2004. UCRL-JC-I 55478.

Jovanovic, I. et al. (2004). *Millijoule pulses from quasi-phase-matched optical parametric chirped-pulse amplification.* Presented at the Intl. Conf. on Ultrahigh Intensity Lasers, North Lake Tahoe, CA, October 3–7, 2004. UCRL-ABS-206996.

ICE: The Image Content Engine

James M. Brase

03-SI-003

Abstract

Advancements in imaging-sensor technologies (particularly for remotely sensed images) is resulting in volumes of data that overwhelm human analysts. The goal of this project is to develop a framework, the Image Content Engine (ICE), that will allow human analysts to interpret massive volumes of imagery in a timely fashion. The ICE approach focuses the attention of humans on relatively few small areas with specified attributes. This is achieved by capturing content extracted from images as nodes in semantic graphs whose links define relationships between pieces of image content and information obtained from non-image sources.

The ICE framework will allow content extracted from imagery, the most voluminous data source, to be seamlessly integrated with information from other sources. This will help break down the compartmentalized nature of analyst-based interpretation in the defense and intelligence communities. ICE will greatly increase the productivity of image analysts by very efficiently focusing attention on potential objects of interest. The ICE approach also applies to other areas of experimental science (e.g., physics, biology, and environmental science) in that mining massive archives of complex measurement data for new patterns and relationships is an emerging model for discovery in modern science.

Mission Relevance

The work supports LLNL's mission in national security by developing the technology to allow content extracted from images to be seamlessly integrated with information from other sources, thus enabling analysts to submit sophisticated queries relevant to defense, intelligence, counterterrorism, deployment strategies, determining the functions of enigma facilities, and detecting the production of weapons of mass destruction.

FY04 Accomplishments and Results

An infrastructure for a configurable feature-extraction pipeline was implemented for regions and model-based objects, allowing these feature-extraction tools to operate on

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multiple images of arbitrary size. Phase-sensitive detection for arbitrary 2-D object signatures was demonstrated. Algorithms for projecting 3-D target models into images have been developed and integrated with the phase-sensitive detector. This capability has been used to demonstrate a broad-area search capability for important national-security program targets. We put a baseline hierarchical image-feature model in place and demonstrated tile-based queries. We also implemented a baseline hierarchical data representation scheme for the semantic graph and metric-based matching schemes.

Proposed Work for FY05

We plan to complete the development and integration of tile-based feature extraction and query tools, extending them to multispectral imagery. Partial model matching will be implemented and evaluated for object detection. The hierarchical representation of graph data will be extended to include relevant link types and full search capabilities that employ metrics and morphological matching. Detection and query applications will be extended accordingly, with confidence estimation and relevance feedback incorporated, and performance analysis will be performed using real data where feasible. A parallel version of the pipeline will be implemented in a cluster environment for analysis of actual data streams from the Large-Area Synoptic Survey Telescope.

Publications

Abdulla, G. et al. (2004). S*imulation of the future LSST data pipelines.* Presented at the Astronomical Data Analysis Software & Systems XIV, Pasadena, CA, Oct. 24–27, 2004. UCRL-CONF-205562.

Grant, C. W. (2004). *Data structures and algorithms for graph based remote sensed image content and retrieval.* Presented at the IEEE Intl. Geoscience and Remote Sensing Symp., Anchorage, AK, Sept. 20–24, 2004. UCRL-PROC-204929.

Paglieroni, D. W. (2004). *Convergent coarseness regulation for segmented images.* Presented at the IEEE Intl. Geoscience and Remote Sensing Symp., Anchorage, AK, Sept. 20–24, 2004. UCRL-CONF-204537.

Sengupta, S. K., Lopez, A.S., Poland, D.N. (2004). *Class label statistics: A basis for fusing information from multi-spectral imagery with an application to unsupervised detection of human settlement.* Presented at the IEEE Intl. Geoscience and Remote Sensing Symp., Anchorage, AK, Sept. 20–24, 2004. UCRL-ABS-202833.

Sengupta, S. K. et al. (2004). *Phased-based road detection in multi-source images.* Presented at the IEEE Intl. Geoscience and Remote Sensing Symp., Anchorage, AK, Sept. 20–24, 2004. UCRL-CONF-204778.

Weinert, G., J. M. Brase, and D. W. Paglieroni. (2004). *Computer-aided content-based cueing of remotely sensed images with the Image Content Engine.* Presented at the IEEE Intl. Geoscience and Remote Sensing Symp., Anchorage, AK, Sept. 20–24, 2004. UCRL-ABS-202618.

Kinetic Simulation of Baoundary Plasma Turbulent Transport

William M. Nevins 04-SI-003

Abstract

In this project, we will develop a kinetic code to model the boundary region of tokamak plasmas, allowing first-principles predictions of the edge transport barrier, which is the greatest source of uncertainty in projecting the fusion power of next-generation tokamak reactors such as the International Thermonuclear Experimental Reactor (ITER). This code will include a nonlinear kinetic description of the edge plasma in realistic 3-D magnetic geometry. Molecular dynamics simulations will provide improved models of plasma contamination by hydrocarbons. The code will use efficient numerical algorithms to solve the coupled equations describing the edge plasma in a 5-D phase-space on high-end, massively parallel computers; and will provide the software structure that facilitates continual development of the physics by multi-institution collaborations.

This project will deliver the world's first ab initio integrated model of the boundary region of fusion plasmas, which is essential to predicting the fusion power of ITER. A full-fidelity ITER calculation will be within the capabilities of our emerging petaclass computers, such as BlueGene. A code architecture compliant with the goals of the DOE Fusion Simulation Project (FSP) will facilitate continued development of the physics by multi-institution collaborations, allowing the models developed under this project to be extended into a simulation of the full tokamak as part of the FSP. This project will also position the Laboratory to be a major participant in the FSP.

Mission Relevance

This project supports the energy-security mission of the Laboratory by advancing the physics required for magnetic fusion energy through the use of high-performance computing, advanced software technology, and fundamental simulation of the properties of materials. Fusion has been a core mission for LLNL since its founding.

FY04 Accomplishments and Results

We developed a 3-D code (two velocity and one spatial coordinate), including free streaming and Fokker-Planck collisions. We developed a parallelized Python framework, adapted a parallel data manager using LLNL SAMRAI software, formulated a field-aligned coordinate scheme, identified visualization tools, and developed numerical methods for solving the equations. After refining a molecular dynamics model, we used the model to produce a table of sputtering yields. Working with UC Berkeley researcher J. Verboncoeur and the XOOPIC particle code, we began modeling the near-surface sheath region and its chemistry. In collaboration with Princeton physicist H. Qin, the project team developed a new 5-D gyrokinetic formalism for the tokamak edge.

Proposed Work for FY05

We plan to develop and apply methods for solving the field equations in the 4-D code. To make the field-solver efficient and scalable to large numbers of processors, we will explore

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multigrid methods and continue to develop the underlying code infrastructure, test suite, and visualization capabilities. We plan to export SAMRAI data to the VisIt visualization system. A model for the turbulent transport and viscosity will be added to calculate flow-shear in the 4-D code and we will compare computed plasma flow profiles with the DIII-D experiment. We also will compute the edge bootstrap current and validate it against new diagnostic measurements on the DIII-D tokamak. Modeling of the near-surface sheath, its chemistry, and the molecular dynamics calculations of sputtering will continue.

Publications

LoDestro, L. L., A.M. Dimits, and W.M. Nevins. (2003). "Alternate treatment of the polarization drift for gyrokinetic PIC simulations." *Proc. Am. Phys. Soc. Div. Plasma Phys. Ann. Mtg. 2003*, CP1.074. UCRL-PRES-200549.

Development of Absolute Spectroscopic Diagnostics for Nonlocal-Thermodynamic-Equilibrium Plasmas

Christina A. Back 04-ERD-019

Abstract

The goal of this project is to develop a quantitative understanding of nonlocal-thermodynamic-equilibrium (NLTE) plasmas. The physical conditions present in many important stockpile-stewardship applications and virtually all radiation-dominated laboratory experiments are in the NLTE regime, which is complex and difficult to simulate. We will conduct experiments using low-density aerogel foams to measure the temporal evolution of the K- and L-shell emission of highly-ionized species. The data will be sufficiently accurate to enable the creation of benchmarks that can be used to refine and potentially validate NLTE codes such as CRETIN. Our experiments will address the recombination processes in the density regime of 10^{19} to 10^{22} cm^{-3} of mid-Z elements. The experiment will provide temporally resolved, absolute measurements of the emission over a spectral range of 200 to 1000 eV. Data will be compared with calculations at each stage of the project.

The data obtained in this project will resolve long-standing discrepancies in the study of laser-produced plasmas and enable better modeling of physical processes relevant to stockpile stewardship. These experiments investigate the NLTE phenomena that impact high-powered laser experiments. Deliverables will include absolute spectra from a NLTE plasma, temporal correlation of K- and L-shell spectra, and a new capability to perform absolute emission measurements in the soft x-ray regime that provide an important new constraint for simulations.

Mission Relevance

Accurate understanding of the mechanisms of x-ray production is an essential component of energy balance and x-ray transport in stockpile stewardship applications, and for the design and production of x-ray sources used for backlighting and material testing.

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FY04 Accomplishments and Results

The first experiments involving 30 full laser shots and 10 supporting shots were performed in March at the NIKE laser in Washington D.C. where the prototype spectrometer was installed. The plasma under study was created by directly irradiating the face of a cylindrical Ti-doped SiO_2 low-density, 3mg/cc, aerogel with 1.6 kJ in a 37-beam, 4-ns pulse. The principal aim was to assess the range of accessible plasma conditions and the uniformity of the emission. Preliminary analysis shows that we can successfully create low-temperature L-shell emission at intensities of 1×10^{12} W/cm^2 to K-shell emission at 4×10^{14} W/cm^2. The spectral diodes show that there is a shift in the emission produced from F-like to He-like ions, which span plasma temperatures from 200 to 1000 eV.

Proposed Work for FY05

Work planned for FY05 includes improving the spectral resolution to adequately determine spectral features and continuing with the next steps for characterizing the plasma for radiation studies. The present experimental data show that we can constrain, but not independently determine, the average *Z*. We plan to improve the spectral resolution by a factor of 3, which will enable us to use the gross spectral features to constrain the calculations. To provide adequate data for validating simulations, calculations and analysis of the absolute value of the peak emission will be correlated with the average *Z* in steady-state and time-dependent cases. We also expect to improve sample size and quality.

Publications

Chung, H.-K. et al. (2003). *Ionization balance of laser-produced underdense titanium plasmas.* Presented at the APS DPP Meeting, Albuquerque, NM, Oct.2003. UCRL-POST-200628.

Constantin, C. et al. (2004). *Supersonic heat-wave propagation in laser-produced underdense plasmas.* UCRL-JRNL-203465.

Fournier, K. B. et al. (2004). "Efficient multi-keV sources from Ti-doped aerogel targets." *Phys. Rev. Lett.* **92**, 165005. UCRL-JC-151219.

Electronic Transitions and Phonons in f-Band Metals at High Pressures

Choong-Shik Yoo 04-ERD-020

Abstract

Unusual phase transitions driven by electron correlation effects occur in many f-band metals. The exact nature of these transitions has not been well understood, including the short-range correlation effects themselves, their relation to long-range crystalline order, the role of magnetic moments and order, and the critical behavior, among other issues. Many of these questions represent forefront physics challenges central to stockpile materials. In this study, we propose to investigate electronic phase transitions occurring in solid and liquid f-band metals at high pressures and temperatures using the nation's brightest third-generation synchrotron x-ray source, the Advanced Photon Source (APS). This work will be complemented by new theoretical activities.

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The success of the proposed study will result in significant advances of our current understanding in two forefront scientific areas of condensed matter physics—electron correlations and phonons of f-band metals. The study will deliver fundamental data to challenge and validate theoretical understanding of f-electron calculation and interatomic forces and will also develop and apply cutting-edge experimental and theoretical tools.

Mission Relevance

This project should result in better understanding of forefront physics challenges central to stockpile actinides, such as electron-correlation effects, phonons, and liquid equations of state for stockpile stewardship. These studies on strongly correlated systems additionally provide opportunities to develop collaborations with leading scientists in the field and attract talented young physicists to the Laboratory.

FY04 Accomplishments and Results

Major progress in FY04 included investigating the f-band electronic structure of Gd at high pressures. Utilizing an intense, third-generation synchrotron x-ray, we resolved the low-lying x-ray band of Gd at high pressures for the first time. The 3d-4f resonance at 7.241 keV was nicely separated from the 3d–4d transition at 7.249 keV. In studying the Mott insulator–metal transition in MnO, we determined the change in magnetic moment in MnO across this volume-collapse transition by using high-resolution x-ray emission spectroscopy and angle-dispersive x-ray diffraction at the APS. The pressure-induced structural and spectral changes suggest that Mott transition in MnO occurs above 100 GPa.

Proposed Work for FY05

In FY05, we will continue resonant inelastic x-ray scattering (RIXS) measurements of Gd at pressures above 60 GPa, where volume collapse occurs. Nonresonant high-resolution inelastic x-ray diffraction will be conducted on single-crystal U to determine phonon dispersion at high pressures. In addition, we will begin developing (1) in situ angle-resolved x-ray diffraction to study volume-collapse transitions in several d- and f-band transition metals in both solid and liquid form at high pressures and temperatures and (2) nuclear RIXS to determine the phonon density of states in Fe-doped transition metals and the rare earth Dy at high pressures.

Publications

Yoo, C. S. et al. F*irst-order isostructural Mott transition of highly compressed MnO.* UCRL-JRNL-20481.

Short-Pulse Laser Absorption and Energy Partition at Relativistic Laser Intensities

Ronnie Shepherd 04-ERD-023

Abstract

This project is conducting detailed and integrated experiments to perform the first comprehensive measurements of laser absorption and energy partitioning in solid targets

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heated with an ultrashort laser pulse focused to relativistic laser intensities (>10^{17} W/cm^2). The measurements will include a determination of the density scale length, a critical parameter in determining the contribution of absorption processes dominant at low laser intensities. In parallel, we propose to benchmark LSP, a new collisional particle-in-cell (PIC) model that is planned as the primary modeling tool for future petawatt and high-energy-laser experiments. The resulting information will be used to produce high-brightness, short-pulse-laser-produced broadband x-ray sources for these experiments.

We expect to measure scale length versus laser intensity, scale length versus absorption, relative hot electron and thermal electron production, magnetic field strength versus laser intensity, and particle energy distribution versus laser intensity; perform energy-scaling absorption experiments; design and test a time-resolved particle diagnostic; perform time-dependent broadband x-ray measurements at high-energy petawatt (HEPW) conditions; and measure the time history of particles at HEPW conditions. These experimental results will provide insights into the relativistic laser–matter interaction regime and act as a foundation for designing and performing HEPW experiments.

Mission Relevance

This work will provide data and examine phenomena in an area of relevance to weapons physics (e.g., proton generation and x-ray back illuminators) and experiments at future large fusion-class lasers (e.g., hot electrons for fast-ignitor schemes) by acquiring high-quality physics data. This research will also promote the recruiting of scientists in high-energy-density physics, in support of stockpile stewardship.

FY04 Accomplishments and Results

In FY04, we performed an experiment on short-pulse-laser-heated enclosures for use as broadband x-ray sources at HEPW lasers. This experiment determined the upper boundary for the density scale length and measured radiation and hot-electron temperatures. The data suggest the interaction of high-intensity, short-pulse laser with short-scale-length plasmas results in two sources of hot electrons: (1) ponderomotive heating, which generates a moderate electron temperature (~3 MeV), and (2) stimulated Raman scattering, which generates a higher electron temperature (~17 MeV). The radiation temperature was estimated between 75 eV and 150 eV. Slightly ahead of schedule, we also performed the first experiment to characterize the nonthermal/thermal electron energy partition using x-ray spectroscopy.

Proposed Work for FY05

In FY05, we will complete our hot/thermal electron study to determine absorption into each channel and the subsequent characteristic time for the hot-electron energy to be converted to other forms of energy (e.g., radiation and hydrodynamic expansion) via collisional damping. We will also measure the absorption at laser intensities exceeding 10^{17} W/cm^2 to determine the total amount of energy deposited in the target. This is essential to modeling laser–solid interactions at such intensities. Finally, we will perform our second short-pulse-laser-generated broadband x-ray source experiment to improve our previous results by applying the information from the previous experiments.

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The Creation of a Neutron Star Atmosphere

Richard I. Klein 04-ERD-028

Abstract

Extreme conditions of density and temperature that are relevant to stockpile stewardship are similar to those of low-altitude atmospheres of magnetized neutron stars. This project will design an experiment for future petawatt lasers that reproduces aspects of a neutron star atmosphere, and will assess the possibility of using time-dependent two-dimensional radiation-hydrodynamics codes to experimentally produce and detect photon-bubble instabilities observed by x-ray satellite and predicted by LLNL. We will test key components of the design on existing lasers; perform simulations to estimate temperatures, radiation fields, and magnetic fields; conduct analytic studies of instability timescales; and perform experiments on existing ultraintense lasers.

The experimental design developed in this project will advance the study of extreme physics generated by strongly radiative flows and laser–plasma interactions for the laboratory study of both distinct astrophysical phenomena and the physics of extreme conditions relevant to stockpile stewardship. Establishing a laboratory capability to probe the physics of accreting, magnetized compact objects will enhance LLNL's international reputation in high-energy-density (HED) physics.

Mission Relevance

This project has direct relevance to understanding the extreme conditions of HED, ultrashort-pulse-laser matter interactions that are relevant to LLNL's stockpile stewardship mission and will be a driver for advanced scientific applications at future petawatt lasers where Laboratory personnel will carry out their work. The world-class science that this project represents will improve recruiting and forge links with the U.S. astrophysical community.

FY04 Accomplishments and Results

In FY04 we (1) uncovered key aspects of the scaling properties of the equations of radiation magneto-hydrodynamics, enabling us to begin to design a scaled "cold" experiment that will mimic photon-bubble instability; (2) developed a new simulation analysis methodology that couples radiation hydrodynamic simulation code with the PIC and LSP simulation codes and atomic physics codes to produce synthetic spectra; (3) designed experiments for the interaction of an ultraintense petawatt beam having the realistic intensity of the Rutherford Appleton Laboratory (RAL) petawatt laser with copper slabs and disks; and (4) carried out the first set of experiments at the RAL petawatt facility on our copper slab designs, obtaining data from several shots.

Proposed Work for FY05

In FY05 we will (1) complete the design of a target for a scaled "cold" experiment to assess the best use of the diagnostics available at Omega and JanUSP lasers; (2) evaluate various techniques for adding magnetic fields to a "cold" scaled experiment; (3) assess experiments directed towards simulating a strongly nonlinear stage of photon-bubble instability, including propagation of short pulses of intense radiation through various bubblelike

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structures; (4) design an experiment to achieve a radiation-dominated environment in an optically thick medium; (5) conduct petawatt laser experiments at RAL to achieve high radiation temperatures in a small (~10- to 50-μm) copper sphere; and (6) develop the extreme ultraviolet polarimetry concept to measure high magnetic field strengths at JanUSP.

Publications

Moon, S. J. et al. (2004). *A neutron star atmosphere in the laboratory with petawatt lasers.* Presented at the 5th Intl. Conf. High-Energy-Density Laboratory Astrophysics, Tucson, AZ, Mar. 3, 2004. UCRL-CONF-204495.

New Generation X-Ray Optics: Focusing Hard X Rays

Regina Soufli 04-ERD-032

Abstract

We propose to design, build, and deploy a 30- to 70-keV reflective system for dynamic characterization of shocked materials at the single-grain scale. The first system we are building will collect the emergent beam from a Thomson x-ray source, then monochromatize and refocus it on the target to increase the power by 2 orders of magnitude. Using hard x rays, the focusing system will significantly increase the flux on target and will provide a smaller spot size than conventional methods. The optic will be constructed using a combination of technologies that will be integrated into a single system for the first time, thus providing a new x-ray diagnostic capability.

Studying material properties at the single-grain scale will require a new x-ray diagnostic capability. The homogeneity provided by such a small sample will lead to a dramatic increase in understanding dynamical processes in matter. Beam monochromatization by our reflective element will eliminate background and thus significantly improve the qualitative and quantitative content of dynamic diffraction results. By increasing the power and resolution of the currently available sources and techniques by orders of magnitude, this new diagnostic will enable dynamical studies of homogeneous material samples and propagating shocks, and detailed studies of physical properties of micrometer-scale objects.

Mission Relevance

The demonstration of this entirely new technological capability will have immediate impact on x-ray diagnostics for target and plasma characterization at future, large, fusion-class lasers, in support of the stockpile stewardship mission. In addition, the new capability will find application for basic science missions at other next-generation lasers and light sources (e.g., the Linac Coherent Light Source), in major NASA programs (e.g., the Nustar and Constellation-X telescopes), and for medical applications to improve human health.

FY04 Accomplishments and Results

We built a 30-keV optical system that enables monochromatization (1.7-keV full-width at half maximum) and focusing of the Picosecond Laser Electron Interaction for Dynamic Evaluation

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of Structures (PLEIADES) source x-ray beam onto a target. The optic consists of segmented, multilayer-coated, grazing-incidence glass shells with 40-arcsecond resolution, and will be used in diffraction experiments. To further improve angular resolution, we used a novel method of smoothing the glass substrates. To further increase spatial, spectral, and temporal resolving power, we designed hybrid systems using this reflective optic in conjunction with large-scale, ultraprecise, multilayer diffractive elements. We also demonstrated three multilayer elements for hard x-ray (20- to 40-keV) backlighting experiments.

Proposed Work for FY05

We will install, align, and apply our 30-keV reflective optic in ultrafast diffraction experiments at the PLEIADES x-ray source to study phonon excitation and nonthermal melting phenomena. This optic will enable a monochromatic, background-free, focused x-ray beam and thus should have a dramatic positive impact on experimental resolution. Enabling technologies (i.e., novel segmented substrate materials and smoothing) will be advanced to further improve the angular resolution of our next-generation hard x-ray reflective optics. In addition, we will finish designing and test a large-scale diffractive multilayer element for use in tandem with the hard x-ray reflector to further enhance the resolving power of our optical system for future experiments on shocked materials.

Publications

Kang, H. C. et al. (2004). "Synchrotron X-ray study of multilayers in Laue geometry." *Proc. SPIE* **5537**, 127. UCRL-ABS-202673.

Soufli, R. et al. (2004). "Smoothing of diamond-turned substrates for extreme ultraviolet illuminators." *Optical Engineering* **40**, 3089. UCRL-JRNL-201108.

Nonlinear Free-Electron Light Sources

Frederic V. Hartemann 04-ERD-038

Abstract

Nonlinear Compton scattering, including temporal laser-pulse shaping for spectral x-ray control, is a novel concept that offers the opportunity to develop very compact, extremely bright, tunable x-ray sources in the range of 10 keV to >1 MeV. As our experimental platform, this project leverages the Picosecond Laser-Electron Interaction for Dynamic Evaluation of Structures (PLEIADES) machine, a world-class Compton light source combining a terawatt laser and a high-brightness, 100-MeV electron linear accelerator. The main goal of the proposed research is to perform a dynamic diffraction experiment at PLEIADES on a high-*Z* metallic crystal using a new, high-contrast Bragg diffraction technique. The target will be pumped by a 25-mJ, 100-fs, 800-nm laser pulse to induce ultrafast, nonthermal melting. The diffraction technique will yield a direct measurement of the 2d-spacing.

The successful completion of the proposed research will provide the complete design and proof of concept for a transportable light source that could be used for in situ material probing at the JASPER facility at the Nevada Test Site or high-explosive facilities. It will also

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provide the approach to a nonlinear Compton x-ray backlighter that could be deployed at large lasers. The main deliverables for the proposed work are a high-visibility publication in a first-rate scientific journal and a complete experimental setup, including a pump laser and a probe x-ray beamline, target, and a gated windowless microchannel plate–intensified charged coupled device to enhance the signal-to-noise ratio. Once the experimental method is implemented and demonstrated, a number of important targets could be evaluated, including bismuth, lanthanides, and actinides.

Mission Relevance

Key programmatic responsibilities, including material studies for diagnostics for large lasers will drive our parameter requirements for an advanced x-ray source; therefore, this project is directly aligned with stockpile stewardship. These issues also have direct bearing upon LLNL's pioneering role in basic science projects such as the Linac Coherent Light Source and the Next Linear Collider.

FY04 Accomplishments and Results

In FY04, we developed two additional 3-D codes; extensively benchmarked them in the linear regime, which has resulted in five refereed publications; and studied the theory of high-energy Compton scattering sources, including a spectrally controlled, nonlinear source. The dynamic diffraction work included the development of a theoretical model for dynamic diffraction in high-*Z* metallic crystals, and modification of the PLEIADES interaction region for dynamic diffraction experiments. We met our major goals for FY04, including the determination of the PLEIADES x-ray spectrum using a dispersive crystal, the production of an Au (111) single crystal, and the evaluation of static diffraction from the crystal.

Publications

Anderson S. G. et al. (2004). "Short-pulse, high-brightness x-ray production with the PLEIADES Thomson-scattering source." *Appl. Phys. B* **78**, 891. UCRL-JC-154110.

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Modeling the Production of Beta-Delayed Gamma Rays for Detecting Special Nuclear Materials

James M. Hall

04-ERD-042

Abstract

The ability to screen maritime cargo containers for the presence of hidden special nuclear materials (SNM) is a high-priority goal for the Department of Homeland Security (DHS). Recent work at LLNL suggests that the relatively intense, high-energy (>2.5 MeV), beta-delayed gamma rays emitted from short-lived, neutron-induced fission products may be a useful signature for detection of SNM in the presence of thick hydrogenous and other cargos. The objective of this project is to evaluate this signature and develop detailed computational models of delayed gamma emission suitable for use in Monte Carlo transport codes. This will be done by mining existing nuclear databases and analyzing recent experimental measurements.

This project will develop working delayed-gamma-ray emission models that can be coupled into Monte Carlo radiation-transport calculations complex enough to determine SNM signature characteristics in a wide variety of cargo-loading and prototype-interrogation scenarios. We will also produce a set of very well defined model and database specifications for delayed gamma-ray emission from SNM fission products. These specifications will serve as the first-ever standards in this area.

Mission Relevance

This project supports a critical aspect of LLNL's national-security mission—countering the proliferation and use of weapons of mass destruction. This work will lay the theoretical groundwork for assessing a key signature for SNM. Our models will be offered to the scientific community as standards for use in other Monte Carlo radiation transport codes.

FY04 Results

In FY04, two different computational models for beta-delayed gamma emission from neutron-induced fission of SNM were developed. The first is a simple (low-resolution) model involving the sum of a series of exponential decay curves, similar to that already used to represent delayed neutron emission in the Evaluated Nuclear Data File (ENDF) database. The second is a more complex (high-resolution) model that tabulates the source function on a discrete time–energy grid and can provide detailed (keV-scale) spectral information. These models were computationally validated, benchmarked against experimental data, and incorporated into LLNL's radiation-transport code, COG. Descriptions were published in the open literature and submitted for inclusion in the ENDF database.

Publications

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Nanomechanics: Strength and Structure for Nanotechnology

Robert E. Rudd 04-ERD-043

Abstract

In this computational study, we are developing fundamental principles of nanomechanics, the science of the mechanical behavior of nanoscale structures. We focus on mechanical nanoscale processes driven by surface effects and how they lead to novel physical laws: in particular, how the stiffness and strength of nanostructures are affected by their relatively large surfaces, and how these effects can be used to control the properties of nanostructures through self-assembly. We are investigating two kinds of systems: (1) nano-electro-mechanical systems (NEMS) (i.e., nanorods) and (2) epitaxial nanostructures. The NEMS nanoresonators are ideal for studying the stiffness and strength effects. The epitaxial nanostructures exemplify nanomechanical self-assembly.

This project will develop the basic science of nanomechanics by advancing the theoretical understanding of novel mechanical phenomena at the nanoscale (e.g., through new mechanical laws and a deep scientific understanding of size effects and surface phenomena) and by developing and validating computational methodologies suitable for nanomechanical systems. In particular, we expect to develop a new theory of stiffness and strength of nanostructures and a deeper understanding of how structures are produced in nanomechanically driven self-assembly. This work will have extensive implications for nano- and biotechnology.

Mission Relevance

The scientific understanding and computational tools developed will potentially benefit efforts such as the control of interface features in fusion-class laser targets, development of next-generation biothreat detectors, and mechanical characterization of protein–ligand binding for homeland-security applications. This project has already begun to have impact in these areas through calculations in support of on-going research. In addition, issues associated with dislocation nucleation concern the Advanced Simulation and Computing Program, which supports stockpile stewardship.

FY04 Accomplishments and Results

In FY04, we simulated novel mechanical behavior of nanoscale mechanical rods using molecular dynamics enhanced with new algorithms to calculate finite temperature elastic moduli of beams. We also developed new algorithms to calculate the stress fields associated

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with dislocations terminating on a surface within dislocation dynamics and to incorporate nanomechanical strain effects in kinetic Monte Carlo codes. We also developed algorithms for concurrent multiscale simulation. We used these codes to calculate the elastic properties of nanorods, to study the plastic yield of nanoscale plates and to study the behavior of dislocations in thin films. We submitted four papers to peer-reviewed journals and presented four invited talks.

Proposed Work for FY05

In FY05, we will simulate the novel mechanical behavior of nanorods by using molecular dynamics, including the effect of stress-driven surface reconstructions and plastic yield, relating the classical molecular dynamics calculations to first principles wherever possible. We plan to perform the first large-scale dislocation-dynamics simulations of metallic nanostructures, extending the dislocation dynamics code to include surface nucleation of dislocations, and we will conduct the first epitaxial growth simulations that incorporate the effect of nanomechanical interaction between metallic nanostructures in kinetic Monte Carlo codes. We expect to achieve significant advances in nanomechanics and validation of new computational techniques.

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High-Average-Power, High-Energy, Short-Pulse Fiber Laser Systems

Jay W. Dawson 04-ERD-048

Abstract

Our objective is to develop the technology for high-energy (>10 mJ), high-average-power (>300 W) fiber laser systems. The primary limits to scaling the output energy in a fiber laser are nonlinear interactions and beam quality for larger core sizes. To increase the output energy of a fiber amplifier, we are developing a novel waveguide design that distributes the optical intensity of the light more evenly across the core. Consequently, smaller cores can produce pulses with higher output peak powers.

We will develop the key technology components for a high-energy, high-average-power, short-pulse fiber laser system to scale the pulse energy and average output power of short-pulse fiber laser systems by an order of magnitude beyond previously reported results. We will also demonstrate a prototype system. This should solve many longstanding research hurdles to make the technology suitable for applications in materials processing, high-resolution x-ray imaging, and short-pulse front ends for large-scale, high-energy laser systems. We expect this research to lead to a number of spin-offs.

Mission Relevance

High-average-power, high-energy, short-pulse fiber laser systems that are easy to use, reliable, and efficient are relevant to (1) high-energy petawatt laser front ends for future large laser systems; (2) cutting, drilling, and other materials-processing applications; and (3) small-spot-size, high-flux x-ray sources. All three of these areas support DOE and LLNL's national-security mission, specifically in stockpile stewardship.

FY04 Accomplishments and Results

In FY04 we redesigned a large, flattened-mode (LFM) fiber for higher output energy (up to 10 mJ). This included an extensive study of the design space to maximize the fiber's manufacturability and output beam quality. Testing of the fiber showed beam quality to be in excellent agreement with theoretical predictions. We also began work to demonstrate improved output energy, presented work to date at an international conference, and applied for a patent for the fiber and amplifier designs.

Proposed Work for FY05

In FY05 we will demonstrate scaling of the output energy of the fiber to 10 mJ, then begin work to recompress this high-energy pulse and demonstrate a good-quality pulse. We will also acquire additional pump diodes to scale the average power of the laser system towards the design goal of 300 W (10 mJ at 30 kHz). This will place us in a strong position to use the resulting beam in application demonstrations such as cutting and x-ray generation.

Publications

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Development and Application of a Predictive Computational Tool for Short-Pulse, High-Intensity Target Interactions

Max Tabak 04-ERD-054

Abstract

The goal of this theory-and-computation effort is to produce and validate a computational tool that can model a high-density plasma driven by high-intensity laser light. Integral components of this model will be coupling between laser light and a dense plasma, production and transport of relativistic electrons, self-consistent production and transport of large-scale electric and magnetic fields, realistic equations of state (EOS), electrical and thermal transport properties of thermal plasmas, and coupling to a hydrodynamic (burn) code. This tool will be used to model past and ongoing experiments driven by high-intensity lasers to provide optimized designs for applications such as fast ignition.

If the project is successful, we will be able to model experiments driven with ultraintense lasers, from laser–plasma interaction to the heating of dense plasmas with intense beams of particles. This will enable the design and optimization of various applications in high-energy-density physics and fusion energy. We will also design experiments for future large laser systems to obtain EOS and opacity data of relevance to stockpile stewardship; improve the efficiency of radiography techniques; and improve fast-ignition target designs, reducing the short-pulse laser energy necessary for ignition.

Mission Relevance

Powerful short-pulse, high-intensity lasers are expected to enable a number of applications that are important to defense and fusion energy but difficult, if not impossible, to accomplish by other means. By providing theory and computational support for an integrated high-intensity, short-pulse laser effort, including laser construction and experiment, this project supports LLNL's missions in national security and energy security.

FY04 Accomplishments and Results

Using the electromagnetic code LSP, we achieved excellent computational agreement with measured backsurface temperatures. A saturation-model-based turbulence growing to marginal instability levels was devised, and realistic EOS and electrical conductivity models were identified. We used LSP to model electron focusing along the surface of full-density gold cones, and began installing code steering in LSP using the PYTHON interpreter. Using the particle-in-cell (PIC) code Z3, significant work on electron source models was accomplished that may explain an experimental anomaly in which the electron spot on the front surface appears significantly larger than the laser spot. We also began developing a model of electron stopping power and scattering based on plasma-corrected Moller scattering and organized an international workshop on computational physics of intense laser–plasma interactions.

Proposed Work for FY05

In FY05, we will (1) construct subscale models based on the marginal instability theory to provide an anomalous-resistivity model; (2) continue the comparison of LSP, theory, and

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explicit PIC; (3) develop the electron source function and couple it to LSP; (4) incorporate, into LSP, nonlocal thermodynamic equilibrium atomic physics data, improved stopping-power models for electrons and protons, a runaway electron model, and a field ionization model; (5) benchmark the models with cone geometry experiments; and (6) use LSP with hydrocodes to model short-pulse-laser-driven cans and derive radiation outputs.

Publications

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Surrogate Nuclear Reactions and the Origin of the Heavy Elements

Jutta E. Escher 04-ERD-057

Abstract

This work will establish an innovative method for indirectly determining reaction cross sections by surrogate nuclear reactions. This is important because a large number of nuclear reactions relevant to astrophysics and Laboratory programs cannot be measured with currently available techniques. We will provide a comprehensive framework for planning and analyzing experiments that will allow us to obtain the cross sections of interest. Our applications will focus particularly on reactions involving unstable nuclei that play a key role in the production of the elements between iron and uranium. We expect to achieve an improved understanding of the astrophysical s-process (slow neutron capture) and to provide significant new insights into the synthesis of heavy elements.

The proposed research promises to provide significant new insights into the synthesis of heavy elements between iron and uranium. In particular, we expect to achieve an improved understanding of the astrophysical s-process and more accurate s-process abundances. More generally, we anticipate providing a comprehensive framework for determining reaction cross sections on unstable nuclei by surrogate nuclear reactions.

Mission Relevance

Establishing a novel technique for determining reaction cross sections on unstable nuclei will satisfy important needs for stockpile stewardship and homeland-security applications that require reliable information about reactions involving unstable nuclei.

FY04 Accomplishments and Results

Our focus in FY04 has been the development of the theoretical tools to resolve the angular-momentum mismatch issue. We identified the relevant tools and acquired an appropriate set of computer codes: FRESCO, ECIS, and EMPIRE. A surrogate experiment was carried out at Yale University. The results from this experiment will allow us to infer cross sections that can be compared to directly measured cross sections. The comparison will provide an important benchmark for the surrogate method. In addition, we organized an international workshop that focused on the surrogate reaction technique, hired three new postdocs, and initiated a set of nuclear-reaction tutorials (http://www-phys-d.llnl.gov/Research/N_Tutorials/www/).

Proposed Work for FY05

We will finalize the analysis of our surrogate experiment for ^{90}Zr(n,x) and compare the extracted cross sections to available direct measurements. The results will provide an important benchmark for the surrogate approach. Upon verification and improvement of our models and codes, we will apply the surrogate approach to problems relevant to astrophysics. We will plan and analyze a surrogate reaction for ^{85}Kr(n,γ) ^{86}Kr and then ^{151}Sm(n,γ) ^{151}Sm, which involves the s-process branch-point nucleus ^{151}Sm . For the latter

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study, we will expand our models to describe deformed nuclei. To test the models and establish a benchmark for deformed nuclei, we will reconsider surrogate experiments for Gd.

Publications

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Stellar Astrophysics and a Fundamental Description of Thermonuclear Reactions

William E. Ormand 04-ERD-058

Abstract

This project will investigate new methods to provide a comprehensive understanding of reactions between light nuclei in hot, dense environments, such as stellar interiors and implosion capsules for future large fusion-class lasers. The project will develop a new theoretical framework that describes the dynamics of nuclear collisions based on the fundamental interactions of nature. In addition, the project will undertake a theoretical study of the quantum corrections to electron screening in thermal plasmas to resolve a discrepancy exhibited in previous theoretical approaches and investigate the viability of using fusion-laser facilities to observe a wide range of stellar thermonuclear reactions.

Achieving an accurate and predictive theoretical view of thermonuclear reactions in thermal plasmas based on fundamental physical properties will have far-reaching applications for astrophysics. Benefits include improved stellar modeling capabilities that could impact our knowledge of neutrino oscillations, better databases important to stockpile stewardship, and higher confidence in understanding the dynamics of fusion-class laser target capsules. The ability to perform stellar astrophysics experiments at such facilities could lead to an exciting new experimental program.

Mission Relevance

A more comprehensive understanding of nuclear reactions in hot, dense systems based on fundamental physical properties will be achieved. In particular, this project will support the nuclear data effort for the Laboratory's stockpile stewardship mission and provide insight for experiments to be performed at future fusion-class laser facilities. In addition, by advancing the understanding of stellar evolution, this project supports LLNL's basic science mission.

FY04 Accomplishments and Results

In FY04, codes were developed and tested to compute radial-cluster overlaps for an incoming cluster of up to three nucleons, a critical ingredient for the reaction formalism under development. Preliminary applications were performed for the $^4He(^3H,\gamma)^7Li$ and $^6Li(n,\gamma)^7Li$ systems. These first results correspond well with resonances observed in experimental data. Invited talks were given at meetings of the American Physical Society and at the 3rd International Conference on Inertia Fusion Science and Applications. A paper detailing the formalism to evaluate the radial-cluster overlaps was published in *Physical Review C*.

Proposed Work for FY05

In FY05, we will (1) apply existing formalism and shell-model overlaps to simple systems, e.g., ^4He+n, ^6Li+n, and compute cross sections; (2) write codes to compute radial-cluster overlaps, including renormalization effects due to truncated model spaces, a vital step for the viability of the entire formalism; (3) extend the Keldysh method to the specific case of two-temperature plasmas and make screening predictions for plasma screening in the case

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of runaway burn; (4) establish the capability to perform target capsule simulations and carry out a differential study of capsule designs to determine the viability of measuring S-factors; and (5) assess the usefulness of accurately computed or measured astrophysical S-factors as a diagnostic for future fusion-class laser facilities.

Publications

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High-Energy-Density Galaxy Jets

Willem Van Breugel 04-ERD-059

Abstract

Jets, ionizing radiation, and high-energy particles from active supermassive black holes and exploding stars profoundly affect the formation of new stars in their parent galaxies. At LLNL we are in a unique position to investigate this by combining our expertise in complex, multidimensional numerical simulations and high-energy experiments, and our access to world-class observatories. We propose to continue our multidisciplinary studies of the

interaction of jets with interstellar medium clouds and the energetic processing of astrophysically relevant materials using an ion beam accelerator.

We will (1) analyze astronomical observations at optical, x-ray, and radio wavelengths and determine input parameters for numerical jet–cloud collisions; (2) improve our numerical simulations code COSMOS to perform more realistic, 3-D simulations of jet-cloud collisions; (3) conduct further irradiation experiments of astrophysically relevant materials using the ion accelerator and analyze the results; and (4) obtain astronomical observations of galaxies to investigate the effects of active black holes on interstellar dust grains.

Mission Relevance

Our research will help validate computer codes used to simulate high-energy-physics phenomena important for nuclear physics research and to experiments at future large lasers for fusion studies. Our laboratory experiments, combined with astronomical observations, will provide new information about the effects of high-energy particles on materials under extreme conditions, which will further the national-security mission.

FY04 Accomplishments and Results

In FY04 we (1) completed numerical simulations of jet–cloud collisions, showing that jets can induce star formation; (2) obtained new observations of jet–cloud collisions, which we will use to constrain more detailed numerical simulations; and (3) irradiated an astrophysical material (forsterite), obtained transmission electron microscopy images, and determined that under conditions which might occur in the interstellar medium of galaxies, the material changes from a crystalline to an amorphous structure without changes in chemical composition.

Proposed Work for FY05

In FY05, we will (1) improve COSMOS by including the effects of magnetic fields and adaptive mesh refinement; (2) use COSMOS to perform 3-D simulations; (3) compare our observations with these new simulations and publish the results; (4) perform new laboratory irradiation experiments using a larger energy range and different types of materials; (5) complete a computer program that allows us to predict the fluences of high-energy particles in starbursts or black holes.

Publications

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High-Brightness, Laser-Driven, X-Ray Source for Nanoscale Metrology and Femtosecond Dynamics

John K. Crane

04-ERD-064

Abstract

This project will develop a new, bright, ultrafast x-ray source based on laser-driven K-alpha generation that will produce an x-ray flux 10 to 100 times greater than current microfocus x-ray tubes. The short-pulse (~100 fs) duration of this x-ray source also makes it ideal for observing time-resolved dynamics of atomic motion in solids and thin films. We plan to develop this K-alpha x-ray source using the existing Falcon terawatt laser driver to demonstrate its radiographic potential on surrogate high-energy laser cryotargets and its ability to resolve ultrafast dynamics using the Falcon driver. We will also develop a liquid Ga source or a high-speed Cu tape source and pair it with a 300-W fiber laser to produce a high-average-flux x-ray source for radiography and nanoscale microscopy.

We expect to develop a high-average-brightness x-ray source with subpicosecond temporal and submicron spatial resolution. This source could be used for the in situ radiography of Be shells and cryotargets, dynamic diffraction experiments with actinides, and microscopic imaging of chemical and biological systems, nanostructures, and advanced semiconductor devices. Careful measurements of K-alpha x-ray generation versus laser and target parameters will help verify codes being developed for high-energy-density physics and backlighters. When combined with high-average-power, ultrafast fiber-laser technology, this x-ray source could serve as a multiple-application facility.

Mission Relevance

An improved, high-average-power x-ray source will be useful to many applications that support the national-security mission, such as the in situ characterization of cryotargets for fusion ignition, three-dimensional imaging of actinides under dynamic stress (when used with ultrafast electron diffraction technology), advanced streak cameras, x-ray detectors, and x-ray optics.

FY04 Accomplishments and Results

In FY04, we assembled the experimental chamber, developed the Cu-tape drive for the x-ray source, and set up detectors, diagnostics, and shielding for x-ray experiments. We then performed a series of single-shot experiments to optimize the production of K-alpha x rays as a function of laser energy, focal spot size, and laser pedestal energy, and measured the ratio of K-alpha to Bremsstrahlung. From the results of these measurements, we determined the best operating conditions for K-alpha x-ray generation in Cu. In addition, we developed a code to predict K-alpha generation in thin Cu foil that combines the atomic physics code LSP and a Monte Carlo transport code, ITS. We designed a Mylar film barrier to protect the parabola and permit continuous source operation.

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Proposed Work for FY05

In FY05, we will finish any remaining source characterization work from FY04, then begin using the source to radiograph high-power-laser targets and compare our results with those from other x-ray sources and determine the image-capture rate and how this will scale to the high-average-power fiber-based x-ray source. We plan to design a 20× x-ray microscope using our source and an x-ray optic, then fabricate and begin testing the x-ray microscope, if possible. We will design a high-speed Cu tape drive or liquid Ga source for a higher-repetition-rate, fiber-based source.

Quantifying Sea-Level Cosmogenic Neutron/Gamma Backgrounds and Their Effects on Large-Volume Sea-Level Detectors

Leslie J. Rosenberg 04-ERD-080

Abstract

Detecting the energetic gamma rays emitted by fissile materials after muon capture is only one of many proposed methods for finding fissile material in cargo. An issue affecting these methods is the correlated cosmogenic background, which can be significant relative to signal, varies little globally at a fixed height above sea level, and is irreducible without significant overburden. Yet this background is only imperfectly understood and considered in these applications.

In this project, we focus our attention on the muon-capture method for detection by characterizing cosmogenic backgrounds that limit this and other approaches to fissile-material detection. We will use Monte Carlo modeling, which can be later validated with actual detectors.

If successful, the project will provide an estimate of important classes of irreducible cosmogenic backgrounds that are certain to plague the muon-capture approach or any method using a large scintillator detector operating at sea level. Our characterization of these backgrounds will produce a useful tool for evaluating new approaches to cargo inspection and will aid basic- or applied-physics projects that depend on sea-level, large-scintillator detectors to produce a signal.

Mission Relevance

The DHS and NNSA are embarking on a variety of large-scale programs for enabling detection of fissile material in cargo. The baseline evaluation of the backgrounds provided by this project will be a useful resource for these homeland-security mission areas. This project also supports many basic-science efforts involving detection of rare correlated events at sea level. An accurate, predictive tool for cosmogenic gamma-rays will contribute to the Laboratory's mission in breakthroughs in basic science.

FY04 Accomplishments and Results

In FY04, we inventoried the various mechanisms of cosmogenic background, then focused our attention on the neutrons associated with these cosmic rays. These neutrons are time-

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correlated with the proposed signals and are therefore especially troublesome. These neutrons induce recoils in the large-volume hydrocarbon scintillators and mimic in-time signals. We estimated the neutron "halo" surrounding cosmic rays as they traverse a volume of scintillator, and convoluted the yield with the proton-recoil reaction, arriving at the background rate. We found that this single cosmogenic mechanism is a significant background in cases for any large liquid scintillator operating at sea level, where the expected signal rate is low.

Nonequilibrium Phase Transitions

Andrew Ng 04-ERD-108

Abstract

Exploring nonequilibrium phase transitions is a scientific frontier that holds promise for discovering new phases and metastable states, chemical reaction pathways, and biological functioning processes. This project will conduct the first systematic study of phase transitions in an extreme, nonequilibrium regime to correlate nonthermal melting with lattice disordering, quantify the role of electronic excitation on phase-transition kinetics, and develop approaches in finite-temperature condensed matter for constructing an equation of state (EOS). The project will use simultaneous measurements of optical and structural properties under ultrafast laser excitation to help develop density functional theory approaches. Work will be conducted in collaboration with the University of Toronto.

We expect to obtain time-correlated data on optical and structural properties by tracking solid–liquid to liquid–plasma transitions under ultrafast excitation conditions. These data will benchmark quantum simulations based on the density functional theory approach. If successful, the project will achieve new understanding of the connection between electronic (optical) and atomistic (structural) behavior, opening up possibilities of manipulating phase stability and boundary while validating new developments in theory to improve predictive power. Success in this area will also help describe the convergence of condensed matter and plasma physics, a critical 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 EOS data development in support of the stockpile stewardship mission.

FY04 Accomplishments and Results

After a late-year start, we identified a potential candidate for the experimental postdoctoral position on our research team and began discussion with a theory group at Los Alamos National Laboratory to plan collaboration in calculating the electrical conductivity of the high-energy-density, nonequilibrium states of gold. We also acquired a charge-coupled camera system, set up the equipment, and began designing our experiments.

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Proposed Work for FY05

Work in FY05 will focus on experimental and theoretical tasks. To attain the first data on nonequilibrium optical and structural properties, we will begin by implementing the tunable, supercontinuum source and the ultrafast electron gun and performing proof-of-principle experiments of interband transition threshold reflectivity and transmissivity and ultrafast electron diffraction. In parallel, the theory work will begin with a study to yield equilibrium properties as a basis for testing nonequilibrium calculations. The supercomputers Qbox and Thunder will be used to test convergence (e.g., system size and pseudopotential), examine the microscopic properties of equilibrium liquid metal surface, and perform finite electron temperature molecular dynamics.

Development of Insulating Liquids for Detecting and Imaging Low-Energy Particles

Adam Bernstein 04-LW-017

Abstract

Noncryogenic detectors that are capable of providing high-efficiency, high-resolution spatial imaging and good energy resolution could help solve a variety of long-standing questions in the area of detecting low-energy (keV to MeV) particles. Ion drift within insulating liquids could allow the development of new imaging detectors with wide application to outstanding problems in fundamental physics, medical imaging, and fissile-material detection. We propose a staged research program aimed at developing and exploiting this largely unexplored detection technique.

If successful, this work will enable the development of a wide class of imaging, energy-resolving detectors capable of measuring the properties of 10-keV to 10-MeV particles with high efficiency at room temperatures. This will help solve problems for detection in low-rate applications such as the detection of weakly radioactive materials and rare particle decays, as well as medical imaging applications in which low doses are imperative.

Mission Relevance

This work is broadly applicable to NNSA and LLNL's national-security mission, through the possibility of improved detection of fissile materials. It has the potential to make a significant contribution toward the development of a new technological capability for biomedical imaging and may also considerably improve sensitivity for a rare process known as neutrinoless double beta decay, of central interest to DOE and LLNL's mission in breakthroughs in fundamental physics in the 21st century.

FY04 Accomplishments and Results

Our goal was to establish the viability of room-temperature insulating liquids for high-resolution tracking of MeV-scale particles. We confirmed the existence of three essential features of the method, and found no obstacles to its continued development. The three features are a signal-to-background ratio of ten or greater, generation of ion–electron pairs in a candidate liquid in response to MeV-scale particles, and the existence of low-noise-

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junction field-effect transistors as an existing readout solution that met our noise requirements. The first two properties were measured experimentally using a test cell of our own design. We also prepared, but have not yet tested, a well-collimated x-ray source for use in measuring ion mobility, a decisive property of the system.

Proposed Work for FY05

In FY05, we will continue using our detector cell to test promising liquids for use as ion-drift media. As results and progress dictate, we will increase the pixelization of the detector readout, which we will modularize to allow exchange of different readout systems in the cell. We will also seek to measure individual pulses from beta or x-ray sources. By adding a transparent window to our test cell, we will explore the possibility of using scintillation light to create a clean start pulse for true 3-D tracking and improved energy resolution. If resources permit, we will develop a simulation package to estimate the background rejection capability in a double-beta-decay experiment.

Relativistic Antimatter Plasmas Created by Ultra-Intense Lasers

Scott C. Wilks

04-LW-020

Abstract

The goal of this project is to generate and characterize relativistic electron–positron (e^+e^-) plasma jets. These relativistic antimatter plasmas, when generated near the boundaries of black holes, are thought to play a key role in the production of gamma-ray bursts, which are the most energetic objects in the universe. We propose to study the formation of this peculiar state of matter in scaled laboratory astrophysics experiments by using ultrashort lasers to heat small amounts of high-*Z* material to extremely high temperatures and densities, surpassing conventional laser–solid interactions. We will build the first-ever electron–positron spectrometer for laser–plasma physics and use it to characterize the e^+e^- jet that is generated in ultraintense laser–solid interactions. We plan to model the experimental results and test novel target configurations with 3-D computer simulation codes.

If successful, we expect to achieve the highest density relativistic electron–positron plasma ever generated in a controlled fashion. This, combined with the design and deployment of a new diagnostic for these extreme experiments, will open up an entirely new window to understanding this exotic state of matter, thought to exist only in energetic astrophysical objects. We also expect to build the first electron–positron spectrometer for laboratory astrophysics and to produce a point design for one of the first laboratory astrophysics experiments to be fielded on future large laser facilities with petawatt capability. Developing a laboratory astrophysics capability is an exciting new tool that will attract talented young researchers to LLNL.

Mission Relevance

The knowledge about hot, dense matter gained from this project, as well as the

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development of a laboratory capability using ultraintense lasers, support the Laboratory's stockpile stewardship mission.

FY04 Accomplishments and Results

Our work in FY04 began with designing and simulating simple gold-foil experiments to increase the hot electron temperature and thereby maximize the number of pairs produced. In addition, we obtained excellent data using the JanUSP laser, compared the results to those predicted by simulation, and wrote two journal articles based on this work. This was the first direct experimental evidence that hotter electron temperatures are obtained from thinner foils—an important result for maximizing the number of positrons. The agreement between theory and the simulation predictions was impressive and led to a clear physical picture of the interaction. In addition, we built and began testing the new electron–positron spectrometer at JanUSP.

Proposed Work for FY05

We will field our newly built spectrometer on the Rutherford Appleton Laboratory Petawatt laser in England in December, where we will make the first simultaneous electron–positron energy spectra measurements. The experimental results will be modeled using a 3-D collisional particle-in-cell code and the high-performance computing facilities at LLNL. We will continue the hot electron experiments on the JanUSP after it is upgraded, then finish the design of an electron–positron astrophysics experiment to be conducted with the petawatt beam of a future fusion-class laser.

Publications

Chen, H. and S. C. Wilks. (in press). "Experimental observation of enhanced heating in ultraintense laser-solid interactions." *Phys. Rev. E.* UCRL-JRNL-203377.

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Wilks, S. C. et al. (in press). "Electron-positron plasmas created by ultra-intense laser pulses interacting with solid targets." *Astrophysics and Space Sciences.* UCRL-JRNL-203467.

Wilks, S. C. et al. (2004). *Laboratory astrophysics.* Presented at the Short-Pulse Laser–Matter Cmputational Workshop, Pleasanton, CA, Aug. 25–27, 2004. UCRL-PRES-206188.

Generation and Advanced Diagnosis of Femtosecond, High-Brightness Electron Beams

Scott G. Anderson 04-LW-031

Abstract

This project created kiloampere-peak-current electron beams of femtosecond duration with the 100-MeV electron/positron linear accelerator (linac), using a novel technique to produce ultrashort bunch lengths while maintaining the high brightness of the injector. In addition, advanced diagnostics were developed to measure the temporal distribution of the beam and

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characterize its longitudinal phase space. The creation and diagnosis of beams with both femtosecond length and high brightness is of major concern to advanced radiation production experiments. This project leverages investments in, and will considerably enhance, the Picosecond Laser Electron Interaction for Dynamic Evaluation of Structures (PLEIADES) Thomson scattering source and its ability to produce high-brightness electron beams.

This work evaluated the potential of the velocity-bunching method to produce not only ultrashort bunches, but also the extremely high-brightness beams important to the fields of radiation production and advanced accelerators. Our temporal diagnostic enabled single-shot phase-space measurements, providing valuable information about high-brightness beam physics. In addition, the work expanded the capability of the 100-MeV linac to produce and maintain femtosecond beams with very high brightness, making it a more attractive facility for radiation production, including free-electron laser harmonics, nonlinear Compton scattering, and laser acceleration experiments.

Mission Relevance

This work addresses the requirements of advanced x-ray sources such as the PLEIADES Thomson scattering source. By furthering materials studies within the Physical Data Research Program and developing diagnostics for advanced radiation production experiments, this project supports LLNL's stockpile stewardship mission. It also supports LLNL's mission in basic science by contributing to LLNL collaborations such as the Linac Coherent Light Source and the Next Linear Collider.

FY04 Accomplishments and Results

In FY04, we utilized the velocity bunching compression scheme to create bunches with root-mean-square durations under 300 fs and peak currents approaching 1 kA. These results have been found to agree well with simulations. The effect of this bunching technique on the transverse beam quality and final energy spread were measured to evaluate the potential of this method to produce ultrashort, high-brightness beams. This compression technique has been applied in the PLEIADES project, resulting in a subpicosecond x-ray source with a peak brightness over 50% greater than that produced without compression. Finally, these efforts produced a manuscript that has been accepted for publication.

Publications

Anderson, S. G. et al. (in press). "Velocity bunching of high-brightness electron beams." *Phys. Rev. Special Topics: Accelerators & Beams.* UCRL-JRNL-207372.

Fractality of Fracture Surfaces in Polycrystalline Materials

Eira T. Seppala

04-LW-036

Abstract

We propose to combine statistical mechanical models of fracture in random media with detailed experimental data that characterize certain polycrystalline materials and their

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fracture behavior. The computer model we have developed provides a fast way to find the weakest path through a random medium as an estimate for the fracture path typically showing a self-affine fractal behavior. To date, this model has been applied only to simplified model systems. In this project, electron backscatter diffraction data and molecular dynamics (MD) simulations of various grain boundaries will provide input for the model, including the topology of the grain boundary network and the strength properties of the boundaries. The results will then be compared with the experimental fracture of the system.

This project will link statistical physics models to the fracture mechanics of real materials. The models predict certain scaling exponents that are related to the degree of autocorrelation in fracture paths, which when experimentally validated can be used to help predict how a material will fail as a function of its microstructural parameters. For example, grain-boundary-engineered materials that have intrinsic correlations in their boundary networks tend to have somewhat different failure properties than those of ordinary materials, and this seems to affect the experimentally derived exponents. A basis will thereby be developed for extrapolation of "universal" behavior to specific engineering cases.

Mission Relevance

The statistical mechanics model for fracture in random media and the analysis of polycrystals developed in this project enhance the Laboratory's core competence in dynamic fracture of metals, which supports the Laboratory's national security mission, specifically stockpile stewardship. The applicability to stress-corrosion fracture also makes this project relevant to understanding failure mechanisms in reactor systems.

FY04 Accomplishments and Results

Algorithms were developed for generating microstructures consistent with conventional and grain-boundary-engineered material and were validated against statistics from real materials. A system was developed for efficiently and automatically generating structures and finding the weakest paths through them. The resulting roughness scaling curves were shown to exhibit fractal-like behavior, which was explored as a function of the special boundary fraction. Bismuth-embrittled copper samples were obtained, machined into tensile samples, and broken, after which we quantified the resulting fracture surfaces using laser-scanning confocal microscopy and power spectrum and wavelet analysis. Pre-existing stress-corrosion fracture surfaces were analyzed with the same methods.

Proposed Work for FY05

We will (1) continue analysis of the fracture surfaces and make detailed comparisons with predictions made with the model; (2) extend our models to three dimensions (3-D), which will also involve validating a new stereological procedure for interpreting 2-D electron backscatter maps in terms of 3-D structure; (3) address some minor issues associated with material processing and the production of sufficient brittle-fracture surface area for accurate quantification; and (4) continue MD calculations of the strengths of relevant grain boundaries.

Publications

Reed, B. W. and M. Kumar. (2004). "Stereological corrections for grain boundary number

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fractions in three dimensions." *MRS Proc.* **819**, N7.6. UCRL-CONF-203247

Reed, B. W. et al. (2004). "The structure of the cubic coincident site lattice rotation group." *Acta Crystallographica A* **60**, 263. UCRL-JRNL-202141.

Seppala, E. T. et al. (2004). "Roughness scaling of fracture surfaces in polycrystalline materials." *MRS Proc.* **819**, N1.4. UCRL-CONF-203930.

Renewable Reflective Optics Based on Thermoelelastic Effects: Concept and Design Equations

Dmitri D. Ryutov 04-LW-062

Abstract

This project theoretically analyzed a novel concept of renewable optics for ultrahigh-intensity, pulsed, high-repetition-rate x-ray beams, such as those that will be produced at the Linac (Linear Accelerator) Coherent Light Source (LCLS) facility. Our approach is to use an auxiliary, low-intensity optical laser, with a 1- to 100-ms pulse width, to heat a flat slab and, by thermal expansion, create the desired surface relief, i.e., a focusing grazing-incidence mirror. After every pulse of the main laser, the slab can be shifted to expose a "fresh," undamaged surface. This approach can also be used to make renewable optics for optical range. The project has developed a set of engineering design equations and formulated design specifications to pave the way for the first prototype.

The design equations and specifications derived in this project make it possible to build the first prototype—a system for creating, within milliseconds, an optical element with desired properties, without having any direct contact with the surface. Potential applications include controlling and focusing ultraintense, high-repetition-rate x-ray beams, high-power pulsed optical beams, and low-fluence systems.

Mission Relevance

Ultraintense, high-repetition-rate x-ray beams will open new horizons in a number of research areas directly related to LLNL national security missions: experiments on warm, dense matter, focusing and defocusing mirrors with variable properties, and the x-ray imaging of biological objects. In addition, such single-shot disposable optical elements will expand the experimental capabilities of fusion-class lasers and other facilities.

FY04 Accomplishments and Results

In this project, we assessed thermoelastic effects to determine (1) the range of deformations that can be produced without reaching the yield strength, (2) accessible mirror shapes, (3) optimum thickness of the initial slab, (4) advantages of initially nonplanar slabs, (4) optimum configuration for illuminating the slab, (5) uniformity requirements for mechanical and thermal properties of the slab, and (6) maximum possible repetition rate. Several candidate slab materials were identified, and constraints on auxiliary-beam uniformity were found. A set of engineering design equations and specifications was also developed.

Publications

Ryutov, D. D. (2005). "Thermoelastic effects as a way of creating transient renewable reflective optics." *Rev. Sci. Instr.* **76**, 023113. UCRL-JRNL-206120.

Ryutov, D. D. (2004). "Multipulse effects in the damage to the LCLS reflective optics." *Proc. SPIE* **5534**, 58. UCRL-CONF-205647.

Laser Pulse Compression by Stimulated Raman Scattering in a Plasma

Robert Kirkwood 04-FS-011

Abstract

In this project, we propose to develop a new technique to increase the peak power of a laser by using three-wave stimulated mixing and pulse compression in a plasma. This work will determine the nonlinear wave response needed to compress a laser pulse of approximately 1 ns in duration into a 1-ps pulse, and builds on recent successes with the Janus laser in which a 1-ps seed pulse was amplified and acquired ~1% of the pump power during the duration of the seed pulse. Our results will demonstrate the feasibility of operating existing laser facilities at an output as much as 1000 times higher than is currently possible. This will be achievable largely because the power-handling limits of a plasma (>10^{14} W/cm^2) are much higher than those of conventional solid-state optics (<10^{11} W/cm^2).

We expect to demonstrate, for the first time, pulse compression in a plasma by compressing a several-picosecond portion of a 1-ns pulse beam to 1 ps in duration. The results will be used to benchmark laser–plasma interaction (LPI) simulations and design a compression scheme for a ~1-ns pump pulse with the best achievable efficiency. These results can lead to a low-risk plan for attaining 10 to 100 kJ of energy in a short pulse at future, fusion-class lasers. This work will also study laser–plasma interactions on a time scale short enough to identify saturation mechanisms and will result in high-quality scientific papers.

Mission Relevance

If successful, these experiments will provide great leverage to existing and planned fusion-class lasers and will have broad application in inertial fusion, radiation sources, and particle accelerators. All these topics are of primary importance to NNSA and Laboratory missions in national security, particularly stockpile stewardship.

FY04 Accomplishments and Results

In FY04, we carried out target and beam-transport system designs that will allow us to continue pulse-compression experiments with the increased beam intensity that is now available on the Janus Upgrade facility. Simulations, benchmarked against earlier experiments, showed that the higher Janus power will require a larger, phase-plate smoothed spot to keep the plasma uniform for 700 ps. A phase plate was designed for the pump beam. A beam-transport system for the seed beam was also designed and purchased that will allow higher beam intensity and longer seed wavelength for use with increased plasma density. The system uses high-damage-threshold components to allow higher pump and probe energies.

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Proposed Work for FY05

We will use the improved target and beam transport system to perform experiments to demonstrate the compression of a several-picosecond portion of a 1-ns pulse to a duration of less than 1 ps that can occur at high laser intensity. Power scattered from the pump will be increased a hundredfold by (1) a tenfold increase in pump intensity (by means of the recent Janus upgrade), as well as the associated increase in the gain exponent for the simulated scattering, and (2) an increase of the energy of the 1-ps pulse seed provided by the improved beam transport system.

Publication

Kirkwood, R. K. et al. (2004). "Studies of linear and non-linear beam transmission in plasmas driven with multiple laser beams." *Bull. Amer. Phys. Soc.* **48**. UCRL-JC-154232.

Demonstration of Silicon Nanocrystalline Lasers and Amplifiers

June Yu 04-FS-016

Abstract

The goal of this project is to demonstrate the world's first Si-nanocrystal-based laser. Such a laser, fabricated using conventional technologies, would be the first completely Si-based photonic system. This laser would be both small (a few tens of micrometers) and compatible with conventional complementary metal oxide semiconductor fabrication technology, enabling the integration of arrays of such lasers with standard integrated-circuit chip technology. We propose to conduct a complete set of laser-gain measurements of Si nanocrystal material using a 355-nm Nd:yttrium-aluminum-garnet (YAG) pump laser, which delivers pulses powerful enough to saturate the samples and has a pulse width comparable with the luminescence decay time. We will work with samples that exhibited luminescence at room temperature and whose waveguiding properties we understand.

The project aims for a "clean" demonstration of optical gain in our samples. This would lead to a systematic study to determine the conditions that optimize optical gain with a variety of samples made using different techniques, e.g., implantation, sputtering, and carbon-vapor deposition. Constructing a working laser would enable us to measure its quantum efficiency and thermal properties, for instance. Knowledge of fundamental laser (and amplifier) properties would enable assessment of the potential applications.

Mission Relevance

An Si laser would enable significant advances in computing, high-speed communications, instrumentation, and diagnostics, such as remote sensing and countermeasures, all central to LLNL's national security mission. Silicon lasers also have applications in support of LLNL's mission in bioscience to improve human health.

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FY04 Accomplishments and Results

We used a beam homogenizer to provide a 1-mm "ribbon" of 355-nm pump light with less-severe "hot spots," raising the average pump fluence applied to our damage-prone samples. Our pump-delivery optics allowed the pump spot to be translated, and we recorded data on variable stripe length and shifting excitation spots, which unfortunately did not show clear evidence of gain. However, by looking in detail at the emission spectra and luminescence transients as a function of pump fluence, we saw spectral shifts and changes in the time evolution. These were interpreted as evidence of pump-induced bleaching (or some other energy-transfer process) in the samples. In a low-power 355-nm-pump, 670-nm-probe experiment, our inconclusive results suggested that higher pump fluence is needed.

Small-Sample Heat Capacity under High Pressure

Scott K. McCall 04-FS-020

Abstract

Measurements of specific heat provide information on thermodynamic properties and the electron density of states. The ability to measure specific heat as a function of pressure also makes it possible to construct a detailed equation of state. Previous measurements of specific heat under pressure done by adiabatic and alternating-current methods were difficult due to coupling of the sample heat capacity to the pressure cell. A different technique, referred to as 3ω, uses a two-dimensional heat-flow model to determine heat capacity and thermal conductivity, but does not require that the sample be thermally isolated from the heat bath. We propose to develop this technique for eventual use with samples at low temperatures and pressures of up to 1.6 GPa.

We expect to measure the specific heat of a material using the 3ω technique, then use the technique to measure specific heat as a function of pressure using a piston–cylinder pressure clamp. Refining this technique on a well-studied material such as Gd will enable us to use the 3ω technique to measure the specific heat of a wide variety of materials. This would represent a significant experimental breakthrough in science. An immediate benefit would be an improved understanding of Pu at low temperatures under pressure.

Mission Relevance

Obtaining information about the specific heat of materials as a function of pressure is important to stockpile stewardship. This work will also have broad importance for the scientific community, thus contributing to the Laboratory's mission in breakthroughs in fundamental science.

FY04 Accomplishments and Results

In FY04, we developed a system to measure and extract the third-harmonic signal under ambient conditions. This work included constructing a reliable temperature-dependant linear heater, appropriate bridge circuitry, and associated instrumentation. Software was developed to sweep several variables, providing an accurate and dependable measure of the

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desired signal. Data were obtained on Gd metal near its Curie temperature, where the heat capacity changes sharply with temperature. Finally, we designed a suitable Cu–Be piston–cylinder pressure cell and completed the necessary technical drawings. This pressure cell was designed specifically for the 3ω technique and will be suitable for measurements at pressures of up to 16 Kbar and temperatures between ~2K and 400K.

Two-Phase Noble Liquid-Gas Detectors for Detection of Coherent Elastic Neutrino Scattering

Adam Bernstein 04-FS-032

Abstract

Researchers have recently converged on a design path for a new class of very-low-noise and -background particle detectors, known as two-phase noble gas ionization detectors, which may prove capable of detecting the faint but high-rate coherent neutrino-nucleus scattering signal. Coherent scattering is predicted by and calculable within the Standard Model but has never been measured. This feasibility study will build and test a gas phase detector from which data will be analyzed to establish or reject the feasibility of a larger-scale experiment to pursue the detection of coherent elastic neutrino-nucleus scattering. Initial Monte Carlo and reactor simulations are encouraging.

If successful, we expect to determine the likelihood of being able to detect between 1 and 10 electrons produced at rest in a noble gas (Xe) through the use of their luminescent signal under an accelerating voltage. This work is significant because it will allow actual experimental determination of the prospects for detecting coherent scattering of neutrinos, a long predicted yet unmeasured feature of weak interactions.

Mission Relevance

This world-class measurement would have immediate impact on the Standard Model of particle physics and influence cosmology and astrophysics by confirming or correcting widely used star- and supernovae-opacity calculations. One practical application of this technology is a compact device for reactor monitoring, in support of the national-security mission area of nonproliferation. This research will also help attract top-ranked young scientists to LLNL.

FY04 Accomplishments and Results

After a late-year start, we acquired the equipment needed, began assembly of the detector to enable the collection of data, and developed a design for an initial test chamber.

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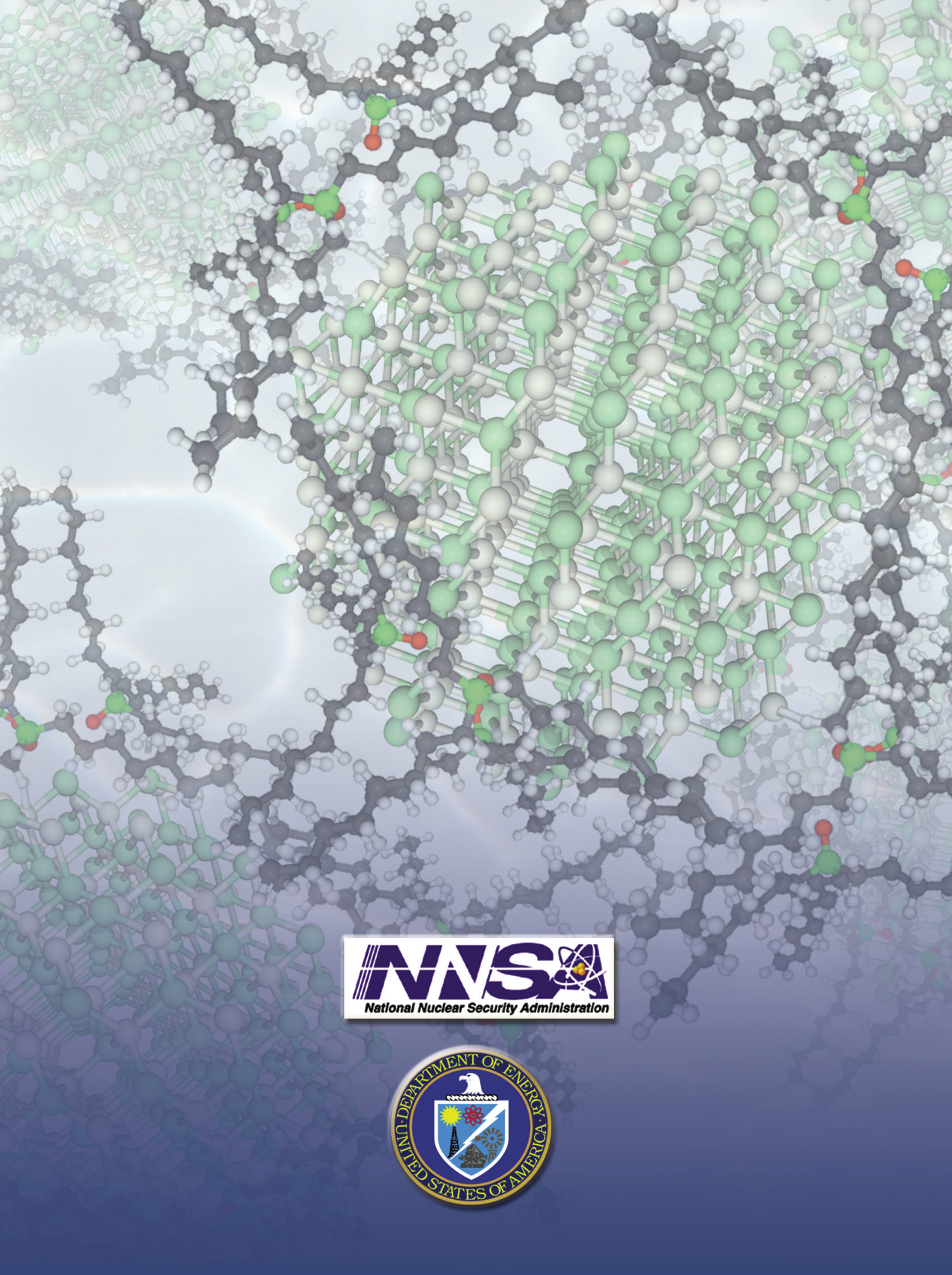
Proposed Work for FY05

In FY05, we will test the noise properties of the system and observe the effects of gas purity and drift field intensity on signal and background; verify the luminescence relations in the drift gas and study the light-collection efficiency of our optical system; and work towards establishing the feasibility of detection of few electron signals in the gas phase, an important part of the overall goal of assessing the feasibility of coherent scatter detection.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