December 2003

Science & Technology Review

National Nuclear Security Administration's
Lawrence Livermore National Laboratory

A New Generation of X-Ray Light Sources

Also in this issue:

- Proton Beams That Create Plasmas
- Faster Solutions to Complex Math
- Finding a More Stable Nitrogen Fullerene

About the Cover

Livermore researchers are part of the design team for the Linac Coherent Light Source, which is being installed at the Stanford Linear Accelerator Center. This fourth-generation light source will be powerful enough to reveal the structure and dynamic behavior of a single molecule. The article beginning on p. 4 describes the new light source and the experiments planned for it. The cover shows an example diffraction pattern, in this case, for the lethal-factor protein of an anthrax spore.

Cover design: Amy Henke

About the Review

Lawrence Livermore National Laboratory is operated by the University of California for the Department of Energy's National Nuclear Security Administration. At Livermore, we focus science and technology on assuring our nation's security. We also apply that expertise to solve other important national problems in energy, bioscience, and the environment. *Science & Technology Review* is published 10 times a year to communicate, to a broad audience, the Laboratory's scientific and technological accomplishments in fulfilling its primary missions. The publication's goal is to help readers understand these accomplishments and appreciate their value to the individual citizen, the nation, and the world.

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Science & Technology REVIEW

December 2003

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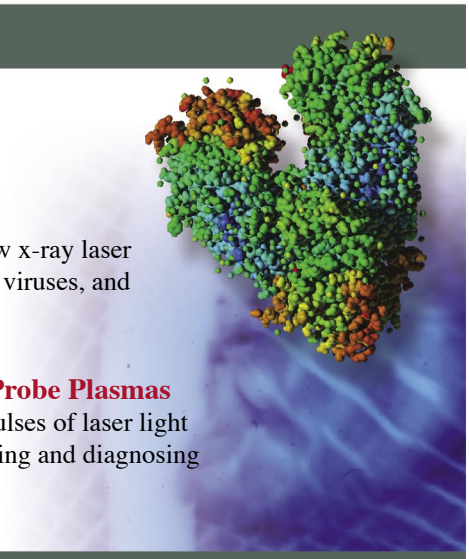

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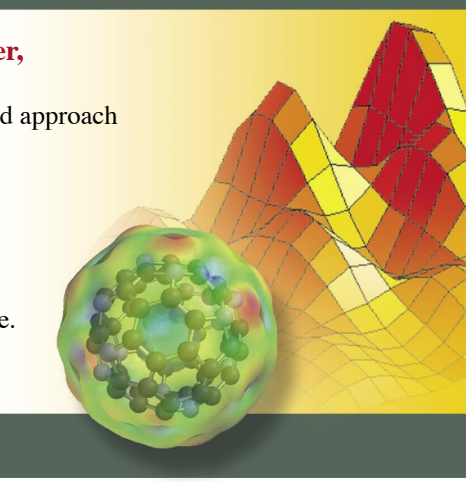

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Carbon dioxide release threatens oceans

Livermore researchers have found that continued release of carbon dioxide (CO_2) during the next several centuries would make the oceans more acidic than they have been during the past 300 million years, resulting in damage to marine life.

The burning of coal, oil, and gasoline releases CO_2 into the atmosphere. Since the Industrial Revolution, CO_2 emissions have contributed to global climate change, notable by an increase in overall temperatures worldwide. Eventually, the ocean absorbs most of the CO_2.

This absorption has been viewed as beneficial because it removed greenhouse gases from the atmosphere. However, recent research by Livermore scientists Kenneth Caldeira and Michael Wickett shows that continued CO_2 emission to the atmosphere from the burning of fossil fuel may make the oceans more acidic than they have been for millions of years, except following extreme events in Earth's remote past such as when the dinosaurs became extinct.

Caldeira and Wickett's results appear in "Anthropogenic CO_2 and Ocean pH" in the September 25, 2003, issue of *Nature*.

Their work complements other carbon cycle research at Livermore sponsored by the Department of Energy's Office of Biological and Environmental Research. These investigations include experiments to understand how ocean acidity can be neutralized; studies of the interactions between the carbon cycle and climate, leading to a more systematic evaluation of climate models; and research on ways of storing CO_2 underground so that it does not contaminate the atmosphere or the oceans.

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Milky Way and neighboring galaxy formed in similar way

Astronomer Kem Cook of Livermore's Institute of Geophysics and Planetary Physics and a collaboration of international researchers from South America, Australia, and Europe have discovered that a neighboring galaxy—the Large Magellanic Cloud (LMC)—and the Milky Way galaxy have similar early formation histories. The LMC is 160,000 light years away from our larger and more regular galaxy.

In their research, Cook and his collaborators identified a spherical halo in the LMC made of the oldest and most metal-poor stars moving like atoms in a hot gas. That halo is similar to a spherical halo in the Milky Way. They made this discovery by measuring the movement of 43 RR Lyrae stars in the inner regions of the LMC. RR Lyrae stars, which are found in both the Milky Way and the LMC, are excellent tracers of old, metal-poor star populations. The presence of spherical halos in both galaxies suggests that they had similar early formation scenarios: extended hierarchical accretion and rapid collapse.

Models of halo formation by accretion indicate that the old, metal-poor stars formed in small satellite galaxies, which were subsequently eaten up by the Milky Way. Models of halo formation by dissipational collapse indicate that the halo formed rapidly before the disk collapsed. When Cook and his collaborators applied these models to smaller galaxies such as the LMC, they found a halo was formed and populated by the LMC's oldest objects.

Results from their research are featured in the September 12, 2003, issue of *Science*, in an article titled "Kinematic Evidence for an Old Stellar Halo in the Large Magellanic Cloud."

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A sharper view of the universe

A major milestone in astronomical history took place on September 20, 2003, at the W. M. Keck Observatory on Mauna Kea, Hawaii. That night, scientists used a laser to create an artificial guide star on the Keck II 10-meter telescope, which allowed them to correct the blurring of a star with the telescope's adaptive optics systems. Laser guide stars have been used on smaller telescopes, but this is the first successful use on the current generation of large telescopes.

Installed in 1999, the Keck adaptive optics system allows astronomers to minimize the blurring effects of Earth's atmosphere, producing images with unprecedented detail and resolution. The system uses light from a relatively bright star to measure the atmospheric distortions and then correct for them, but only about 1 percent of the sky contains stars sufficiently bright to be of use. By using a laser to create a virtual star, astronomers can study much fainter objects, increasing coverage to more than 80 percent of the objects in the sky.

On September 20, the system locked on a 15th magnitude star—a member of a T Tauri binary called HK Tau—and revealed details of the circumstellar disk of the companion star. Throughout the evening, the laser guide star held steady and bright, shining at a magnitude of about 9.5. Although that magnitude is 25 times fainter than what the human eye can see, it is ideal for the Keck adaptive optics system to measure and correct for atmospheric distortions.

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Let There Be Light Sources

LAWRENCE Livermore continues to be a leader and innovator in the development and application of light sources, building the most powerful lasers in the world and demonstrating the first x-ray laser. As light sources become brighter, faster, and more energetic, their role in the Laboratory's future is as important as ever. In fact, the Laboratory's long-range science and technology plan identifies new light sources as crucial to progress in many of our core science and technology areas: stockpile stewardship; high-energy-density physics; nuclear and radiative science; and chemical, biological, and materials research.

Two articles in this issue look at applications of the new generation of intense light sources. The first, which begins on p. 4, discusses the Linac Coherent Light Source (LCLS), an x-ray laser being built at the Stanford Linear Accelerator Center by a consortium of institutions that includes Livermore. The second article, which begins on p. 11, describes exciting developments using the Laboratory's Janus-pumped ultrashort-pulse (JanUSP) laser.

The LCLS is what is called a single-pass, free-electron laser. A very short bunch of high-energy electrons is injected into an undulating magnetic field where they emit bremsstrahlung—literally, braking radiation—as they are accelerated. Under carefully designed conditions, the emitted radiation interacts with the electron bunch and builds in intensity. The resulting x-ray beam is 10 billion times brighter than currently available light sources. Its copious photons are coherent, with energies more than 10 times that needed to ionize any atom. Interactions between this beam and atoms are different from those produced by even the most intense optical lasers. X-ray pulses are tunable from 0.8 to 8 kiloelectronvolts, may be less than 100 femtoseconds long, and may have wavelengths as small as 0.1 nanometer. These photon intensities, pulse lengths, and wavelengths will allow scientists to make measurements on atomic scales.

One particularly exciting use of the LCLS will be to examine the structure and function of such large biomolecules as proteins. With current x-ray light sources, structure can be determined only for those molecules that can be formed into a crystal pattern, a process that invariably destroys the protein's functionality. With its ultrabright, ultrashort pulses, the LCLS will be used to image single molecules, without the need to crystallize or immobilize them. Many challenges remain to meet this goal, but the payoffs for understanding the mechanisms of life are enormous.

We will also be able to use LCLS's x-ray pulses to heat material to conditions replicating those inside weapons, stars, and planets. By splitting the x-ray beam, we can use part of it to heat a material and the other part to take measurements. Once the beam is split, one or more of its parts can be delayed with respect to the others, allowing us to look at what happens as a function of time. Using this technique, we can perform dynamic studies of materials fast enough to see molecular motion taking place during chemical reactions. We will also be able to measure the interactions of complex systems, such as protein folding and crystalline phase transitions.

JanUSP produces laser pulses as short as those produced by the LCLS but in visible light. When JanUSP's laser energy is focused onto a thin metal target, a plasma forms. Electrons in this plasma are accelerated and escape, which sets up an intense electric field that pulls a short-pulse, high-energy proton beam out of the target. This effect, discovered at Livermore, has created an entirely new field of science at laser laboratories around the world.

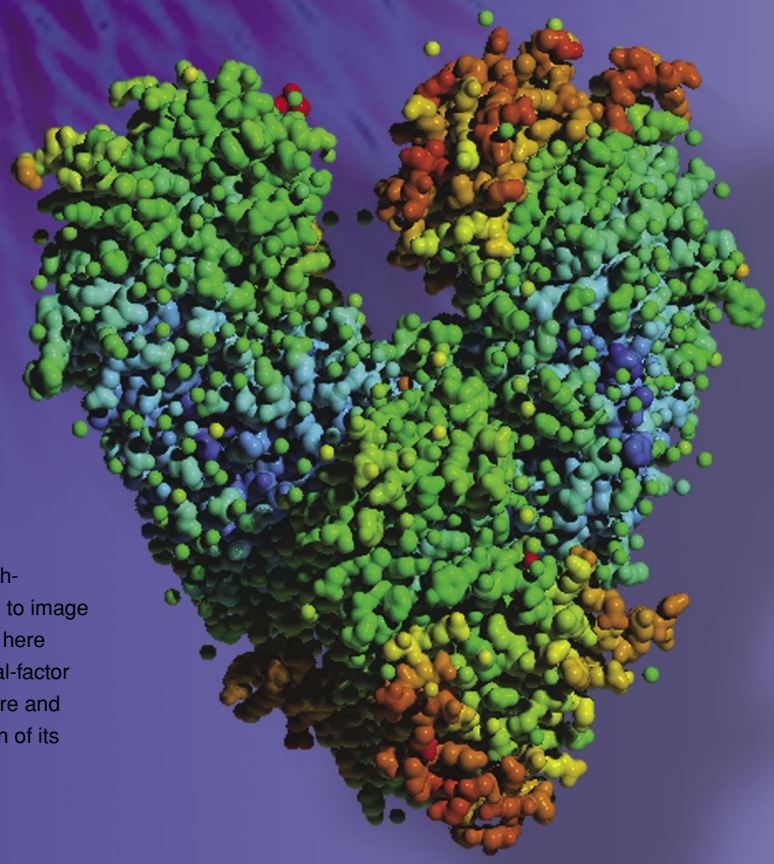
The JanUSP proton beam can be focused to heat material just as the x rays from the LCLS will be. The proton beam also can be used for radiography, providing time-frozen pictures with spatial resolution of 1 micrometer. Because the protons are charged, they respond to electric and magnetic fields, so they can be used to measure these fields on very small time and space scales.

This new, exciting proton beam may even find its way into the National Ignition Facility, providing a novel way to ignite inertial confinement fusion capsules.

■ William H. Goldstein is associate director of Physics and Advanced Technologies.

An Extraordinarily

Livermore researchers are part of the design team for the first large-scale x-ray laser.



The goal of the new fourth-generation light source is to image single molecules. Shown here are (foreground) the lethal-factor protein of an anthrax spore and (background) a simulation of its diffraction pattern.

Bright Idea

ITS short, intense pulses of x rays will reveal for the first time the structure and dynamic behavior of many proteins and viruses at atomic resolution and in three dimensions. It will unlock the secrets of high-energy-density plasmas, which are of interest to the nation's Stockpile Stewardship Program. And it will create the hot, dense matter believed to exist in the center of large planets.

It's the Linac Coherent Light Source (LCLS), the world's first large-scale x-ray laser, being designed for installation at the Stanford Linear Accelerator Center (SLAC). "The immense power of its short-pulse, laserlike x rays will create a revolution in science," says Alan Wootton, chief scientist for Livermore's Physics and Advanced Technologies Directorate.

The heart of the LCLS is a free-electron laser that produces beams of coherent, high-energy x rays. Coherence—the phenomenon of all photons in a beam acting together in perfect lockstep—makes laser light far brighter than ordinary light. Think of a 10-watt night light; then compare its brightness with that from a 10-watt laser—a beam so bright it can cut metal. Because x-ray photons at the LCLS will be coherent, the resulting beam of light will be as much as 10 billion times brighter than any other x-ray light source available today.

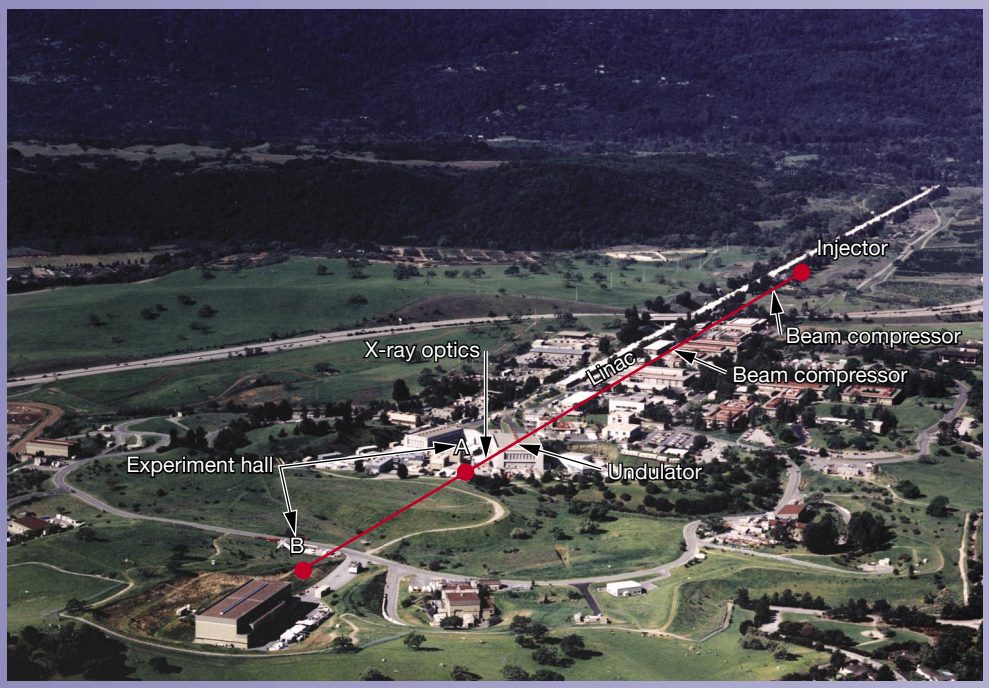
The LCLS, and a cousin planned in Germany, will improve on so-called third-generation light sources. The third-generation sources are circular, stadium-size synchrotrons, and they produce streams of incoherent x-ray photons. Because their pulses are long compared to the motion of electrons around an atom, synchrotron light sources cannot begin to explore the dynamic motion of molecules. The light from the fourth-generation LCLS will last for quadrillionths of a second, allowing its beam to capture such dynamic behavior.

Even determining the static structure of proteins and molecules will be easier and faster with the LCLS. Today, proteins and other macromolecules must be crystallized before their structure can be probed with synchrotron radiation. But not all proteins can be crystallized, and the crystallization process is long and involved. With the LCLS, a single powerful pulse will image one molecule with no prior crystallization required.

Lawrence Livermore is part of a SLAC-led consortium to plan, design, and build the LCLS. Other partners include the University of California at Los Angeles (UCLA) and Los Alamos, Brookhaven, and Argonne national laboratories.

Livermore's primary responsibility, under physicist Richard Bionta, is to design and fabricate the optics that will transport the x-ray beam to experimental chambers and to measure, or diagnose, the beam's condition. The extreme brilliance and ultrashort duration of the beam's pulses will give the beam a peak power of as much as 10 gigawatts. These features make designing optics a challenge because, says Bionta, "The energy of the beam can melt many materials in a single pulse."

Meanwhile, physicist Henry Chapman and other Livermore scientists are planning the first experiments at the LCLS and



The Linac Coherent Light Source will use the linear accelerator (linac) at the Stanford Linear Accelerator Center to create very bright, ultrashort pulses of laser radiation.

establishing the x-ray pulse parameters that are needed for various measurements.

"At the LCLS, we'll use lens-less imaging to determine the three-dimensional arrangement of atoms in a molecule," Chapman says. "We'll detect x rays scattered by a sample when the beam hits it and then examine the diffraction pattern." Radiation from the powerful beam will destroy each sample, but the beam's ultrashort pulse will generate diffraction data before that happens. "Every molecule has a unique diffraction pattern," says Chapman, "and that pattern depends on the molecule's structure."

Experiments at the LCLS will reveal protein structure, which determines protein function. Hence, the LCLS is expected to profoundly benefit structural biology and medical research. It could eventually help scientists solve the proteome—the entire system of proteins in the human genome.

A Single Straight Shot

When the LCLS becomes operational, sometime in 2008, the free-electron laser's photoinjector will shoot electrons down part of the SLAC linear accelerator, or linac. The photoinjector will produce tiny bunches of electrons that travel in a narrow, bright beam at almost the speed of light. After the electrons enter the kilometer-long linac, compressors along the accelerator path reduce the length of each bunch by a factor of 30, which increases their peak current. Their energies may be pushed as high as 14 gigaelectronvolts, a value that will be adjusted from experiment to experiment to produce the desired range of x-ray frequencies.

The electrons then enter an undulator—a vacuum chamber just 5 millimeters across and about 125 meters long and lined with 7,000 magnets arranged in alternating poles. As the electron bunches move down this narrow channel, the magnetic fields push and pull on them, causing the bunches to emit x rays. The LCLS undulator is so tightly focused that x rays emitted by one electron interact with the electrons in front of it. This interaction causes the electrons to bunch more tightly, which generates more x rays.

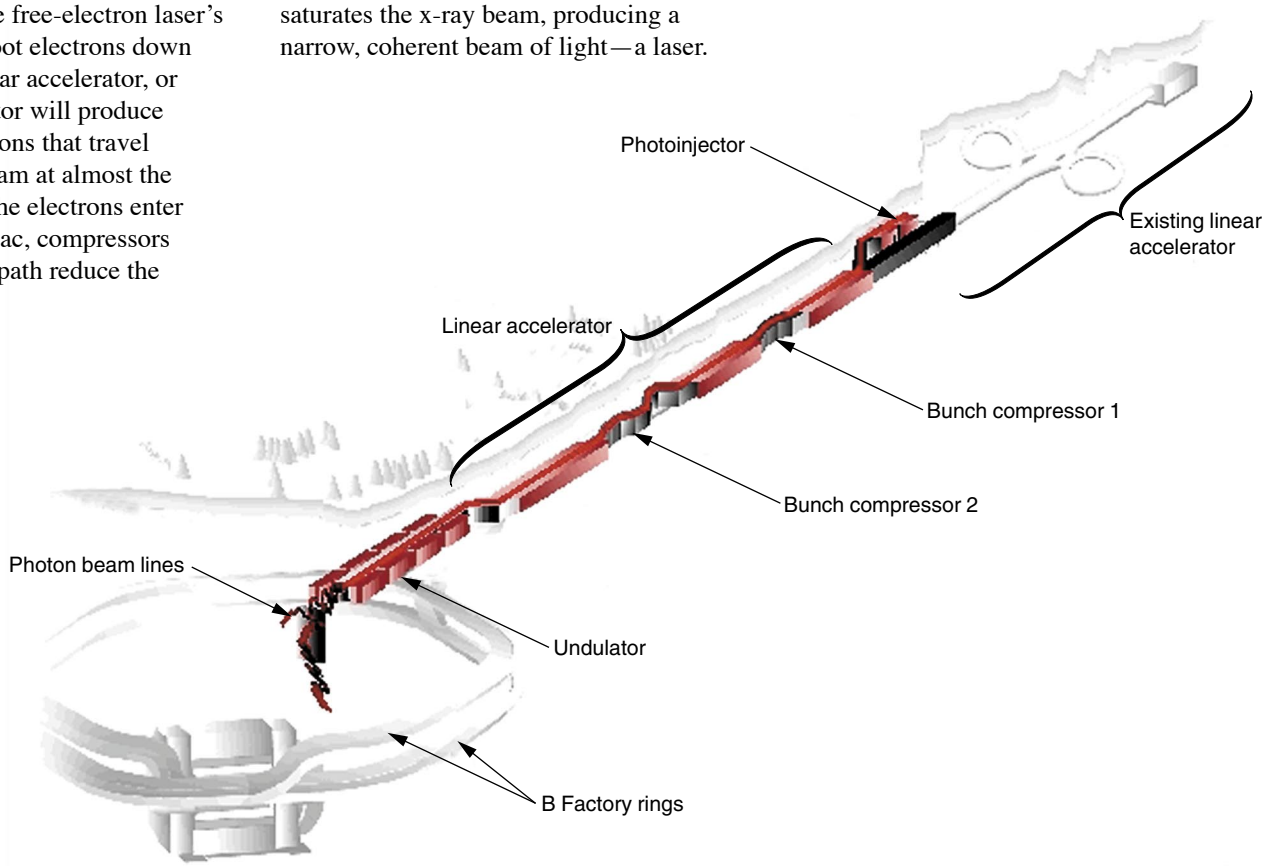
As the process repeats, the bunches become smaller and smaller. This chain reaction is called self-amplification of spontaneous emission, or SASE (pronounced "sassy"). SASE eventually saturates the x-ray beam, producing a narrow, coherent beam of light—a laser. Broadband spontaneous (not coherent) radiation about 10,000 times brighter than that from any other light source emerges from the undulator as well.

Livermore-designed optical devices, placed beyond the undulator, will manipulate the direction, size, energy spread, and duration of the x-ray beam. They also will diagnose the beam and direct x rays to one of two halls for use in experiments.

The experimental halls, A and B, are located 50 and 400 meters downstream from the end of the undulator. Experiments requiring a very narrow, high-energy-density beam will use facilities in Hall A, while Hall B will house experiments that require lower energy densities.

Optics Bear the Brunt

All the diagnostic equipment on the LCLS is designed to minimize interference with the beam. "Because the beam is so



Layout of the Linac Coherent Light Source at the Stanford Linear Accelerator Center. Experimental halls A and B will be to the left, beyond the photon beam lines.

powerful," says Bionta, "our goal is to not put anything in its path, except a gas attenuator."

A mask, valves, and movable jawlike slits just beyond the undulator intercept most of the spontaneous radiation that accompanies the beam. These devices are designed so that they do not block the narrow, intensely coherent x-ray beam.

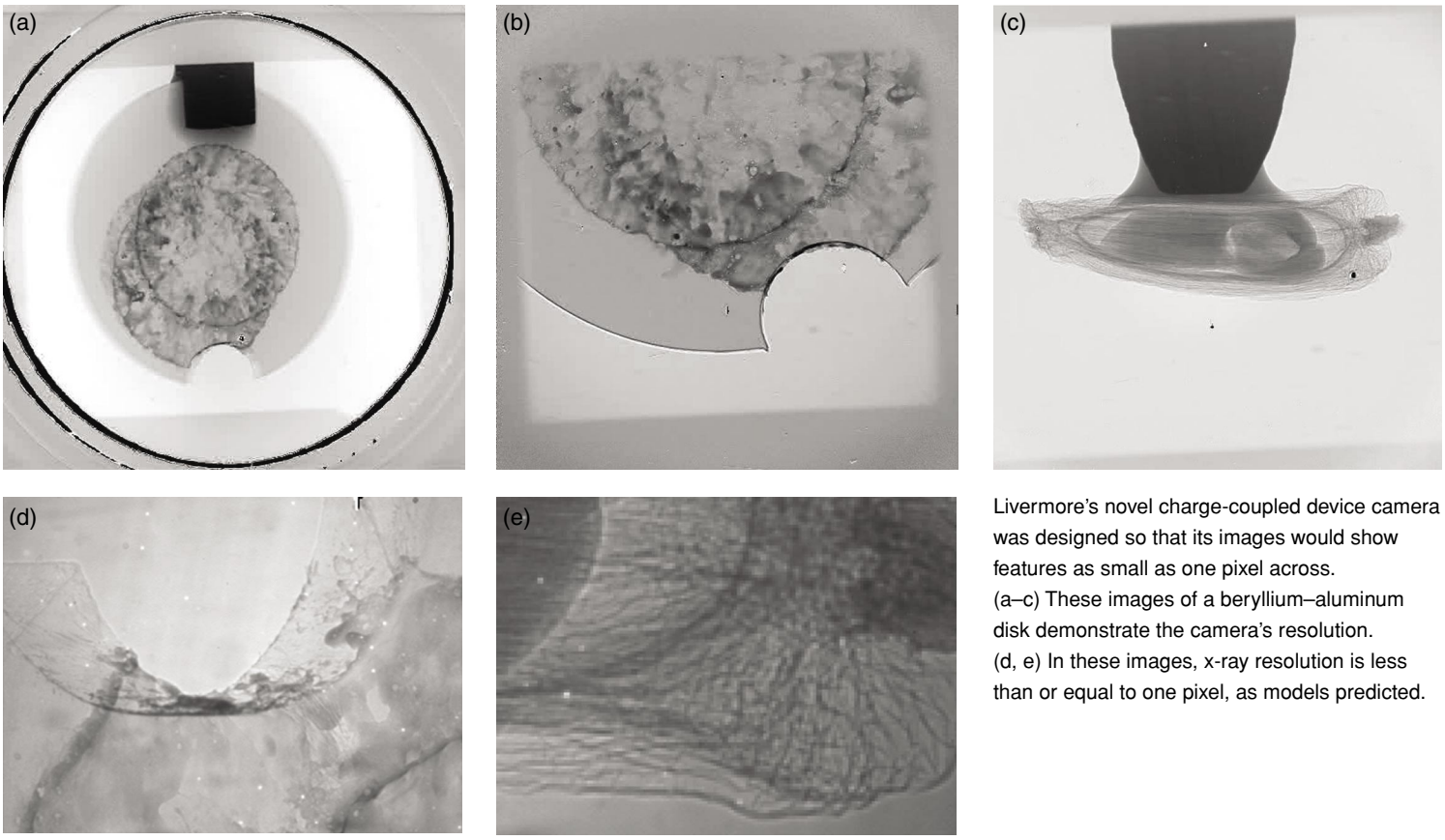
Livermore researchers have been working for several years to understand the damage that occurs when an LCLS beam encounters optics, diagnostics, and targets. Several types of simulations, including Monte Carlo and wave models, helped them fully characterize the x-ray beam. Armed with these data, Wootton, Bionta, and others began to develop schemes for imaging such a bright beam.

The concept they selected uses a camera that will be one of the first diagnostic devices beyond the undulator. In this setup, a small fraction of the beam's light is directly reflected off a thin, polished beryllium foil. Beryllium was chosen for the foil because it has a low electron density and tends to absorb few x rays. Beryllium also will be used for many of the reflective optics at the front end of the system where photon densities are highest.

When the beam reflects off the foil, it strikes the surface of a 100-micrometer-thick lutetium oxyorthosilicate (LSO) crystal doped with a 5-micrometer-thick scintillating layer of cerium. "The LSO crystal is designed to reflect just one-ten-thousandth of the total light from the beam," says Bionta. Reflected visible light is collected by a microscope lens and forms a magnified image on a charge-coupled device (CCD) camera. The images of a beryllium–aluminum disk shown below demonstrate the fine resolution of the camera.

Using the CCD camera, the team has studied how photons from short-pulse lasers at various wavelengths interact with different materials. For example, silicon was irreversibly damaged even at low energy densities using a laser in the visible wavelength and pulse lengths similar to those of the LCLS.

Focusing the LCLS's high-energy beam will be a challenge. The Livermore researchers are developing a new class of tubular optical devices in which the x-ray beam reflects off the inside wall. The slight grazing incidence of the beam on the wall of the lens reduces the absorbed energy considerably. X rays enter the tube at one end and are reflected once by the highly reflective interior surface. They then exit from the other end of the tube, but now the



Livermore's novel charge-coupled device camera was designed so that its images would show features as small as one pixel across.
(a–c) These images of a beryllium–aluminum disk demonstrate the camera's resolution.
(d, e) In these images, x-ray resolution is less than or equal to one pixel, as models predicted.

x rays are traveling in a slightly different direction.

A special focusing element has also been designed for the warm, dense matter experiments that will take place in Hall A. Warm, dense matter is an energetic plasma whose density is almost that of a solid, but it may be as hot as 10,000 kelvins. Scientists believe this matter may exist in the centers of large planets, such as Jupiter, and its properties are important to astrophysics and relevant to the production of inertially confined fusion reactions. Warm, dense matter will be created in the laboratory by focusing the x-ray laser's beam to a 2-micrometer spot in the center of a sample of solid matter.

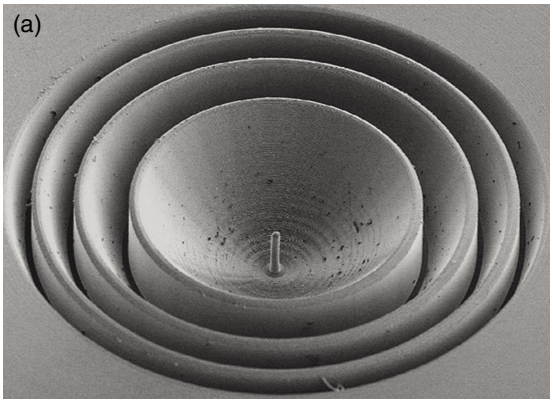
The focusing element will be a blazed phase lens, as shown in the top image below. The lens is made of carbon, which has low x-ray absorption characteristics. Although carbon is not as resistant as beryllium is to the intense power of the LCLS, it has a higher refractive power and is easier to machine precisely, allowing more interesting optical designs. To test the lens design, the research team had a prototype machined at Livermore's Large Optics Diamond Turning Machine (LODTM).

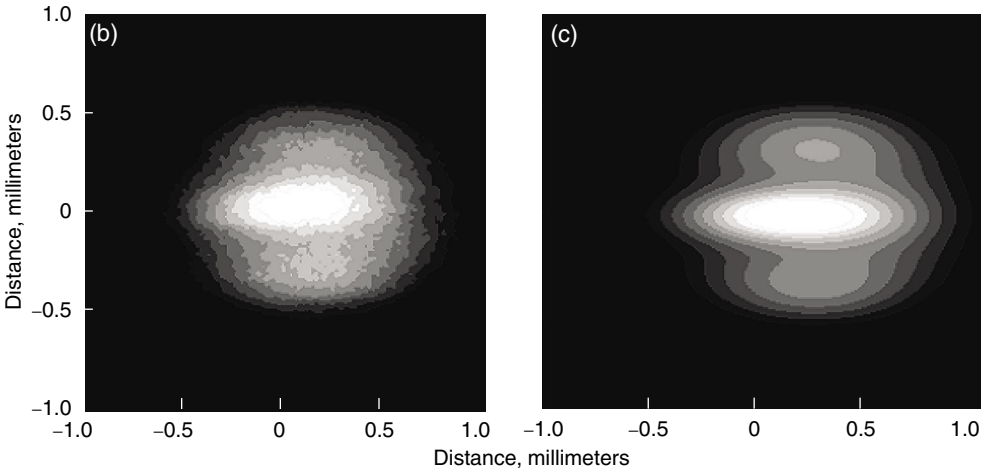
The prototype lens is made from a thin disk of aluminum, which has the same optical properties as carbon. The aluminum lens was tested at the Stanford Synchrotron Radiation Laboratory. Although lens performance was limited by the material chosen and the geometry of the experiment was not ideal, the measured performance closely matched predictions from simulations.

Precision machinists at the LODTM are trying to make lenses from blocks of pure beryllium. Beryllium is a challenge to machine because of its grain structure and because it's a hazardous material. In fact, Livermore's LODTM is one of the few facilities in the nation authorized to work with it.

Perhaps the most challenging LCLS diagnostic will be measuring the 230-femtosecond pulse length. Streak cameras are not an option because they measure down only to 500 femtoseconds. One potential device is a fiber-optic interferometer developed by Livermore photonics experts. The interferometer uses the beam from a continuous-wave laser to monitor the electronic state of a tiny waveguide inserted across its measurement arm.

"When we tested the interferometer at Stanford's synchrotron," says Bionta, "it was sensitive to x rays perturbing the waveguide. In fact, its response was faster than we could measure with the synchrotron beam." Further experiments will be conducted with shorter-pulse x-ray sources at Livermore and SLAC, to determine if the device is really fast enough to measure the 230-femtosecond LCLS pulse.





(a) A prototype blazed phase lens made of pure aluminum. The pattern was carved using Livermore's Large Optics Diamond Turning Machine. Each groove is 18.7 micrometers deep, and the final thickness of the disk is 79 micrometers. In tests of the lens at the Stanford Synchrotron Radiation Laboratory, (b) measured images closely matched (c) simulations.

Technologies for Experiments

Two general classes of experiments have been proposed for the LCLS. In the first class, the x-ray beam will be used to probe the sample without modifying it, which is the current practice for most experiments with synchrotron sources. For example, scientists can use the x-ray laser to determine the dynamic behavior of chemical interactions, essentially by watching the interaction occur on a femtosecond scale, which has never been possible before.

In the second class of experiments, the LCLS beam will induce nonlinear

photoprocesses, or it will create matter in extreme conditions. These experiments include creating warm, condensed matter, as previously described, and determining the structure of macromolecules, by recording crucial information about a molecule before it is vaporized.

It is in biology that the hard x rays of the free-electron laser are expected to have the biggest effect. No technique available today can image the interior of micrometer-size particles in three dimensions at high resolution. With the LCLS, scientists will be able to analyze very small samples, from tens of micrometers down to single molecules.

With third-generation synchrotrons, the low-intensity x rays can diffract to atomic resolution only when a molecule has been crystallized. Once a protein has been crystallized, scientists can't study its interactions with other biological molecules. Nuclear magnetic resonance spectroscopy is used to overcome these shortcomings of x-ray crystallography, but it does not work for larger proteins. With the LCLS, researchers can study proteins that can't be crystallized, such as proteins linked to lipids (fats) and embedded in cell membranes. The short pulses of the LCLS will also reveal how some molecules change shape in just a few femtoseconds.

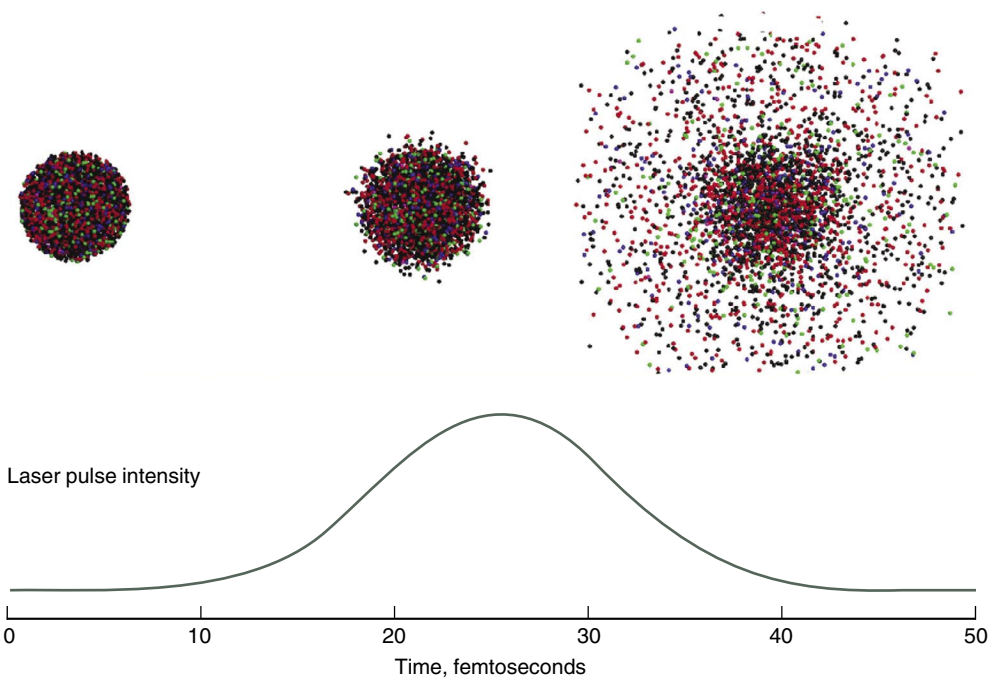
Recording the diffraction pattern before the molecules blow up is critical. X-ray pulses are diffracted by electrons orbiting the atoms in molecules. By studying the patterns made by these diffracted rays, biologists can deduce the structure of the molecule under analysis. Team members have developed a hydrodynamic model to understand the various interactions between the x-ray beam and the sample and to verify that the beam's pulse will end before the sample begins to be torn apart. The figure to the right shows results from simulations of a 20-nanometer protein molecule when it's hit by an x-ray free-electron laser. Models also indicate that a water tamper will suppress the explosion, extending the time range of the diffraction process. Researchers have used simulations to establish the minimum photon density required to classify diffraction and have determined that the necessary pulse durations range from 10 to 30 femtoseconds.

Experiments to verify the timing of the explosion will be performed at the Tesla Test Facility (TTF), the proving ground for the TESLA x-ray free-electron laser that is being designed in Germany. "The TTF is the only place we have now for testing any of these simulations experimentally," says Chapman. "Its wavelength is longer than the LCLS's will be, so we can't get to atomic resolution. But we can begin to understand how and when damage occurs."

The team is also exploring ways to get samples into the beam's path. "Molecules will be just a few billionths of a meter wide," says Wootton. "Somehow, we have to get them lined up with a beam that's only slightly larger, a few millionths of a meter wide and running at the speed of light."

Each pulse of the x-ray beam can hit just one sample in its path. Complete three-dimensional information about a molecule will then be collected by examining multiple, identical samples, one by one. One method for acquiring such data is to use some kind of "molecule gun" to feed samples into the beam path.

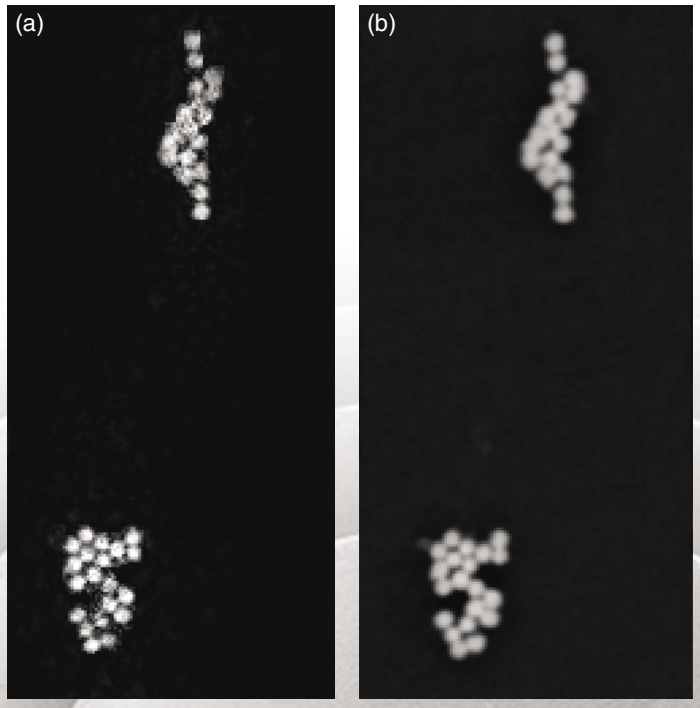
An alternative method is to tether several protein molecules to a membrane positioned in the beam's path. This option, in which the molecules are oriented the same way, requires a lower photon density. Says Chapman, "Because in this case we can now use a lower photon density, which will cause overall less damage, models show we can use longer pulses where the rate of damage is reduced." The team is



Atom trajectories computed by a hydrodynamic model show a 2-nanometer protein exploding after it is hit by a 20-femtosecond, 12-kiloelectronvolt x-ray pulse that is 0.1 micrometer wide. Models indicate that atomic-resolution imaging can be achieved with pulses shorter than 20 femtoseconds. They also show that a water tamper on the protein slows its destruction so that longer pulses could be used.

exploring whether dip-pen nanolithography can be used to produce this carefully oriented pattern of molecules. In dip-pen nanolithography, molecules of a protein or other organic material are deposited on a substrate in a regular pattern.

The Livermore researchers who are developing the algorithms to reconstruct diffraction patterns have been aided by experiments at the Advanced Light Source, a third-generation synchrotron at Lawrence Berkeley National Laboratory. One recent experiment, in collaboration with colleagues from Berkeley and Arizona State University, used a silicon nitride pyramid decorated with 50-nanometer gold spheres. These spheres were chosen because they could be well characterized by other, independent means. As shown below, a reconstructed image of gold ball clusters compares extremely well with an image obtained using a scanning electron microscope. "This reconstructed image is the first true lens-less x-ray image," says Chapman.



Computer algorithms are being developed to use diffraction pattern data to reconstruct an image of the molecule under study. In experiments at Lawrence Berkeley National Laboratory's Advanced Light Source, (a) a reconstructed x-ray image of gold ball clusters compares extremely well with (b) an image obtained using a field-emission scanning electron microscope.

Beyond the Tip of the Iceberg

Chapman's team will also conduct experiments at the TTF in Germany. Those single-shot diffraction experiments will use samples mounted on a substrate and samples shot across the beam. Samples will include lithographic test patterns, diatoms, and wet cells.

"With these experiments, we'll be able to achieve the long-sought goal of x-ray imaging at resolutions beyond the radiation-damage limit," says Chapman. "We hope to get spectacular images. But they will be just the tip of the iceberg compared to what we will be able to achieve at the LCLS."

—Katie Walter

Key Words: free-electron laser; Linac Coherent Light Source (LCLS); linear accelerator (linac); protein structure; Stanford Linear Accelerator Center (SLAC); x-ray laser; warm, condensed matter.

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Using Proton Beams to Create and Probe Plasmas

Proton beams generated by ultrashort-pulse lasers will help advance our understanding of plasmas.

PROTONS, the positively charged, subatomic particles discovered by Lord Rutherford nearly 100 years ago, are still surprising scientists. Lawrence Livermore researchers are discovering that proton beams created by powerful, ultrashort pulses of laser light can be used to create and even diagnose plasmas, the superhot state of matter that exists in the cores of stars and in detonating nuclear weapons. The proton-beam experiments promise new techniques for maintaining the nation's nuclear arsenal and for better understanding how stars function.

The proton beams used in the Laboratory's experiments are produced by pulses of laser light lasting only about 100 femtoseconds (a femtosecond is 10^{-15} seconds, or one-quadrillionth of a second) and having a brightness, or irradiance, up to 5×10^{20} watts per square centimeter. When such fleeting pulses are focused onto thin foil targets, as many as 100 billion protons are emitted, with energies up to 25 megaelectronvolts. The protons come from a spot on the foil about 200 micrometers in diameter, and the beam's duration is a few times longer than the laser pulse. The highest-energy protons diverge 1 to 2 degrees from the perpendicular, while the lowest-energy protons form a cone about 20 degrees from perpendicular.

Funded by the Laboratory Directed Research and Development Program, the Livermore experiments are led by physicists Pravesh Patel and Andrew Mackinnon. Patel, who works in the Laboratory's Physics and Advanced

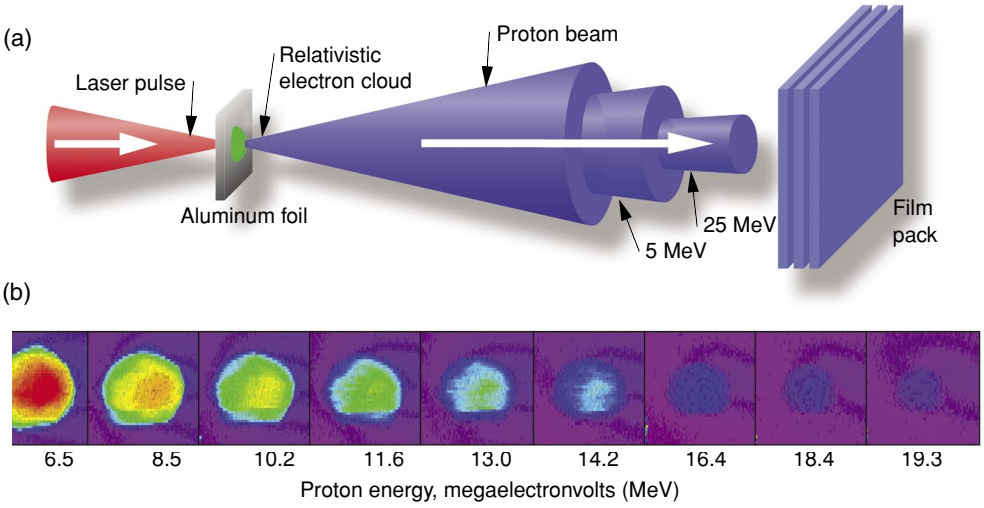
Technologies Directorate, is researching new ways to create and better understand plasmas. Mackinnon, from Livermore's National Ignition Facility (NIF) Programs Directorate, is developing new ways to measure the plasmas created in NIF experiments. Both physicists are collaborating with colleagues from Queen's University in Belfast, Northern Ireland; Heinrich-Heine-Universität in Düsseldorf, Germany; the LULI laser facility at l'Ecole Polytechnique in France; Rutherford Appleton Laboratory in the United Kingdom; and the University of California (UC) at Davis.

"Plasmas are often referred to as the fourth state of matter," says Patel. "They are abundant in the universe but relatively uncommon on Earth. Plasmas are extremely hot, highly transient objects and thus are difficult to control or to accurately probe."

The team wants to develop new methods for creating plasmas in the laboratory, so they can study them at temperatures ranging from a few electronvolts to hundreds of electronvolts and at the high energy densities (more than 100,000 joules per gram) that exist in stars. The current generation of high-power lasers makes such studies possible because they can compress and heat matter to these extreme states.

Ideally, scientists want to measure plasmas in a uniform-density, single-temperature state. As a material is heated to several electronvolts, the pressure in it increases to more than a million times atmospheric pressure. This increased pressure causes the plasma to expand hydrodynamically, as in a violent explosion. Under these conditions, measuring plasma properties is extremely difficult.

One way to overcome these problems is to use what scientists call isochoric heating—heating at constant volume. With isochoric heating, plasmas don't expand during the time they are heated, and their energy can be relatively uniform. Established methods of isochoric heating, such as laser-driven shock heating, x-ray heating, and ion heating, are relatively fast (10^{-6} to 10^{-9} seconds), but these timescales are still longer than those during which significant hydrodynamic expansion can occur (10^{-11} to 10^{-12} seconds).



(a) Diagnostic setup for imaging proton beams produced by a 10-joule pulse of laser light that lasts only 100 femtoseconds. The pulse irradiates a thin aluminum foil. The generated protons are then "pulled" from the back side of the foil by electrons traveling close to the speed of light. Protons with the highest energy diverge the least from an angle perpendicular to the back foil surface. (b) Images from a multilayer film pack taken over a broad energy range show that the number of protons decreases with increasing energy.

Another method, direct heating with intense subpicosecond (10^{-12} seconds) laser pulses, creates a highly nonuniform heating pattern. The laser energy is absorbed within less than 100 nanometers of material, and the heat localization creates a large temperature and density gradient.

A novel approach to isochoric heating, discovered at Livermore, uses laser-produced proton beams to generate fleeting, dense plasma states at constant volume and density. The heating period is shorter than the time needed for significant hydrodynamic expansion to occur, so the material is heated to a plasma state in a few picoseconds. In effect, says Patel, the proton beam dumps a huge amount of energy almost instantaneously and suddenly increases a target's temperature to millions of degrees.

JanUSP Makes It Possible

In their experiments, the researchers rely on Livermore's Janus ultrashort-pulse (JanUSP) laser, one of the brightest lasers in the world. (See *S&TR*, May 2000, pp. 25–27.) JanUSP produces a beam with an average intensity of 5×10^{20} watts per square centimeter that lasts about 100 femtoseconds. The laser operates at a wavelength of 800 nanometers and delivers 10 joules of energy.

In one set of experiments, the laser pulse produced a proton beam from a 10-micrometer-thick sheet of aluminum foil. The proton beam then heated a second 10-micrometer-thick aluminum foil that was placed 250 micrometers directly behind the first. Within a few picoseconds, the heating created a 4-electronvolt plasma almost 200 micrometers in diameter—too short for much hydrodynamic expansion to occur.

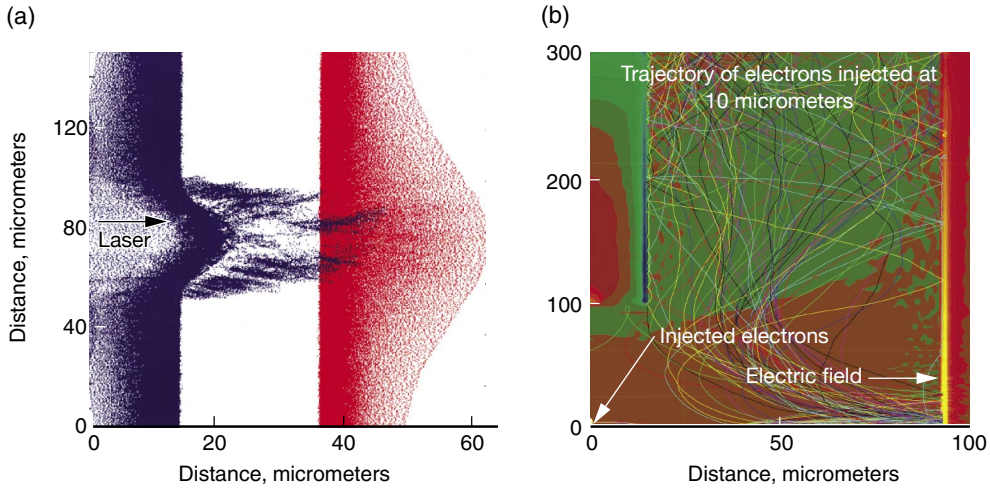
The discovery that intense, highly directional proton beams could be generated from an ultrashort laser pulse heating a solid target was made by Livermore researchers several years ago while conducting experiments with the Laboratory's Petawatt laser. The Petawatt laser operated on 1 of the 10 beam lines

of Livermore's Nova laser, which was decommissioned in 1999. (See *S&TR*, March 2000, pp. 4–12.) Experiments by Livermore physicist Richard Snavely and others to characterize the proton beams revealed a unique combination of properties, including peak proton energies of 55 megaelectronvolts and conversion efficiencies (of laser energy to proton energy) up to 7 percent.

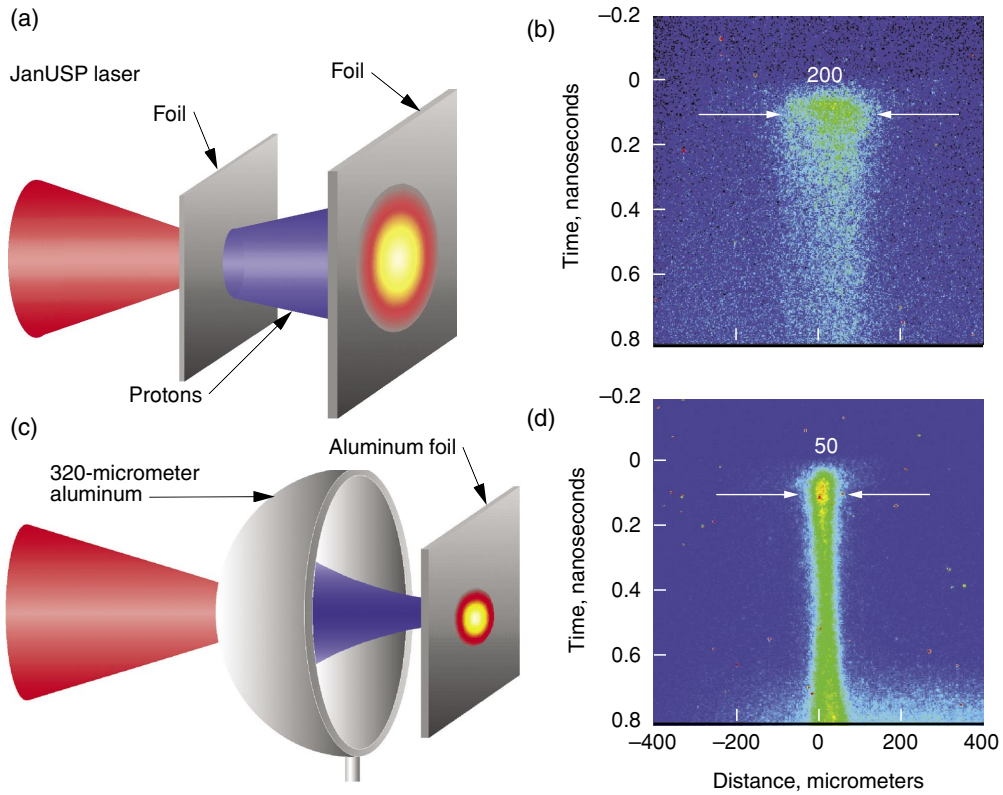
The scientists also discovered that the protons in the beam originated in hydrocarbons found in surface contamination on the foil's back surface. Livermore theoretical physicists, led by Steve Hatchett and Scott Wilks, used computer simulations to study this behavior. They found that the pulse from an ultrashort laser accelerates electrons from the interaction region at the front of the target with relativistic energies; that is, the electrons travel close to the speed of light. The electrons emerging at the foil's rear surface induce a large electrostatic charge field, which in turn accelerates protons from hydrocarbon contaminants on the rear surface. The protons accelerate from 0 to 20 megaelectronvolts at 20 percent the speed of light and travel in a well-defined, highly directional beam perpendicular to the target. X rays, in contrast, are emitted at random angles.

Simulations by Wilks showed that by curving the laser target's rear surface, the proton beam could be focused to a far higher state of energy density. To test this design, the team asked General Atomics in San Diego, California, to manufacture aluminum hemispheres that are 10 micrometers thick, 320 micrometers in diameter, and almost perfectly smooth on the inside to ensure a high-quality proton beam. With the shaped targets, the proton beam was almost 10 times more powerful than the beam produced from flat targets. The proton beam was focused on a 50-micrometer-diameter area of a foil placed behind the target, which was then heated to 23 electronvolts.

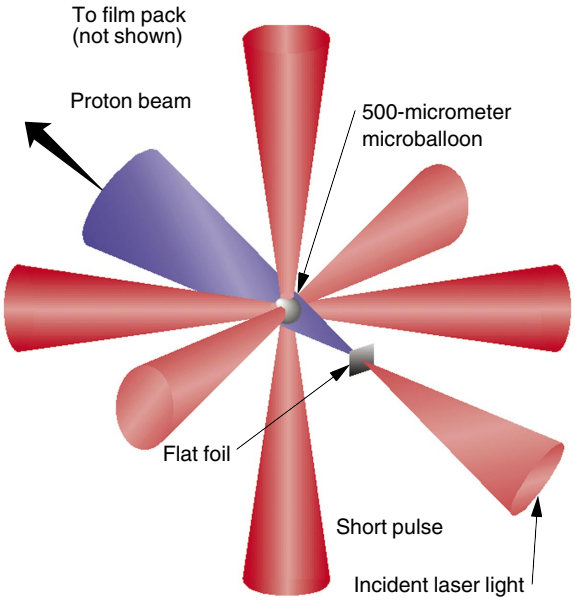
"For the first time, the experiments showed that we can focus proton beams," says Patel. He notes that when the



(a) Particle-in-cell code showing how a thin foil target expands after laser irradiation. (b) Hybrid code calculation of the transport of high-energy electrons through a solid density foil. The calculation shows the complex electron trajectories and large electric fields at the rear surface of the foil that create the protons.



(a) Layout for the proton-beam experiments using the Janus ultrashort-pulse (JanUSP) laser. When JanUSP irradiates an aluminum foil, it creates a proton beam that then heats a second foil. Both foils are 10 micrometers thick, and the second foil is placed 250 micrometers behind the first. (b) The plasma created in the second foil is almost 200 micrometers in diameter. (c) Layout for the experiments with a curved target. (d) When the target's rear surface is curved, the laser produces a focused proton beam that is 50 micrometers in diameter and almost 10 times more powerful than the beam produced with a flat target.



The first proton probing experiments of laser-driven implosions were conducted using the 100-terawatt Vulcan laser system at Rutherford Appleton Laboratory in the United Kingdom. The targets were plastic microballoons, 500 micrometers in diameter. Six beams from the Vulcan laser were focused onto a microballoon while an ultrashort-pulse laser beam was used to make either the diagnostic proton or x-ray beam.

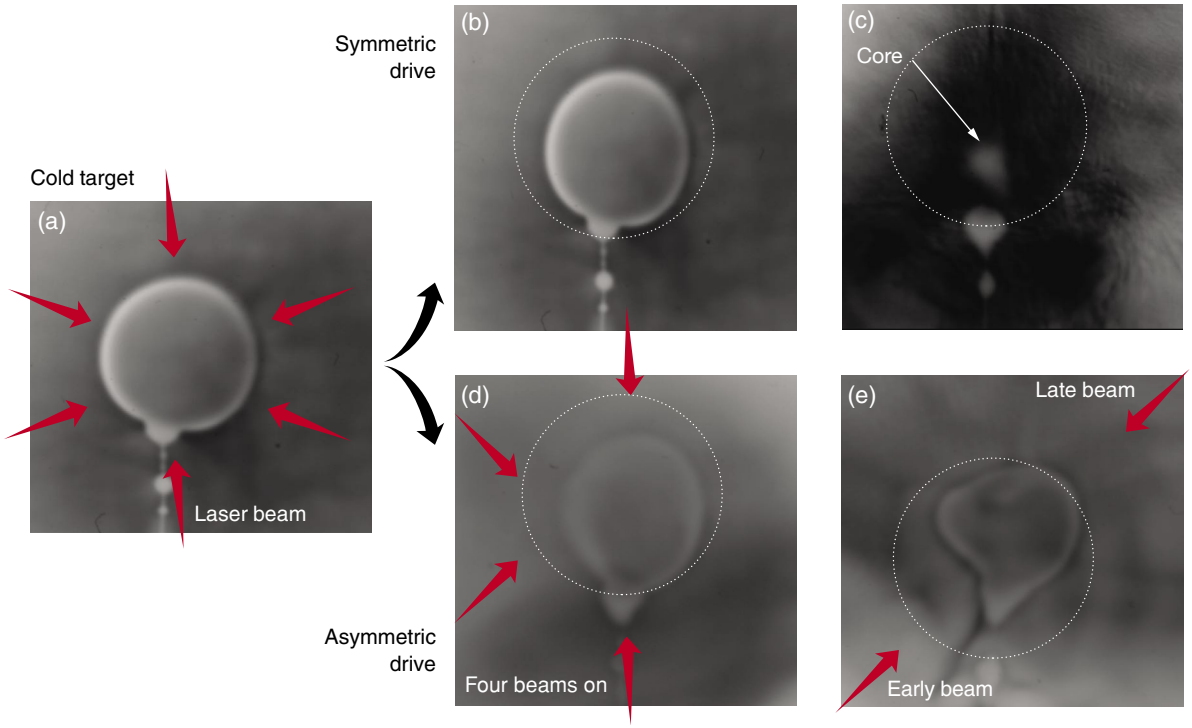
techniques of proton heating and focusing can be applied with more powerful lasers, scientists may be able to isochorically heat plasmas to much higher temperatures and pressures. This advance would provide many opportunities in high-energy-density physics and fusion energy research.

Using Protons for Radiography

The team is also using proton beams for radiographic applications to diagnose plasma conditions generated by high-power lasers at picosecond timescales. The first proton probing experiments of a laser-driven implosion were conducted by Mackinnon in 2002 using the 100-terawatt Vulcan laser at Rutherford Appleton Laboratory. This experiment was conducted in collaboration with scientists at Queen's University and UC Davis. "We wanted to investigate the suitability of proton radiographs to diagnose an implosion capsule in inertial confinement fusion experiments," says Mackinnon.

Plastic microballoons, 500 micrometers in diameter—or about one-fourth the size of the targets planned for NIF—were used as targets. Each of the Vulcan laser's six long-pulse beams was fired for 1 nanosecond at a wavelength of 1 micrometer and an irradiance of 10 terawatts per centimeter. Each beam's energy was 100 to 150 joules, so the maximum energy on the target was up to 900 joules. The six laser beams illuminating the target arrived from six orthogonal directions, a setup designed to provide the best symmetry for this number of beams.

In addition, an ultrashort laser beam was used to make either a diagnostic proton beam of about 7 megaelectronvolts or a diagnostic x-ray beam of about 4.5 kiloelectronvolts. The proton beam was obtained by focusing a 100-joule laser pulse with an irradiance of about 5×10^{19} watts per square centimeter for 1 picosecond onto a tungsten foil 25 micrometers thick. To image the implosion, the team used a multilayer pack of dosimetry film in which each piece of film was filtered by the preceding piece. In this way, the film pack gave a series of images from each shot with an energy ranging from 3 to 15 megaelectronvolts.



Proton radiographs showing the evolution of a laser-driven implosion of a 500-micrometer-diameter balloon: (a) prior to implosion and at (b) 2 nanoseconds and (c) 3 nanoseconds after the laser pulse. When the laser beams are slightly mistimed, the radiographs show asymmetries in the target: (d) Only four beams (shown by red arrows) strike the target, all at 4 nanoseconds after the laser pulse. (e) Beam arrival is staggered from 1 to 5 nanoseconds after the laser pulse.

The team took radiographs of microballoons both before and during implosion. One image, of a 500-micrometer-diameter microballoon with a 7-micrometer wall thickness, showed good contrast at a resolution of 5 to 10 micrometers. A series of radiographs (shown on p. 14), which were taken by varying the delay between the implosion beams and the beam used to produce the proton or x-ray beam, revealed how the implosion process evolved.

In one experiment, the beams were set to converge on the target asymmetrically—that is, the six beams arrived at the target at slightly different times. The laser beams on the left-hand side arrived 1 to 2 nanoseconds before the laser beams on the right-hand side. This asymmetry led to significant distortions. For example, the shell traveled much farther inward on the left-hand side than it did on the right.

Under more symmetric drive conditions, the target remained nearly spherical during the implosion. However, even when the beams arrived at the same time, the proton radiographs revealed some plasma asymmetries. For example, in one experiment, the upper part of the shell traveled almost twice the distance traveled by the lower part of the shell.

These proton radiographs were the first taken of a laser-driven implosion with picosecond resolution. The team found that the temporal and spatial resolution remained high throughout all stages of the implosion.

"The images show the promise of proton radiography for diagnosing early time distortions in the implosion process with high resolution and very good image contrast," says Mackinnon. "The x-radiographs also had good resolution, but the image contrast was high only when the density was high."

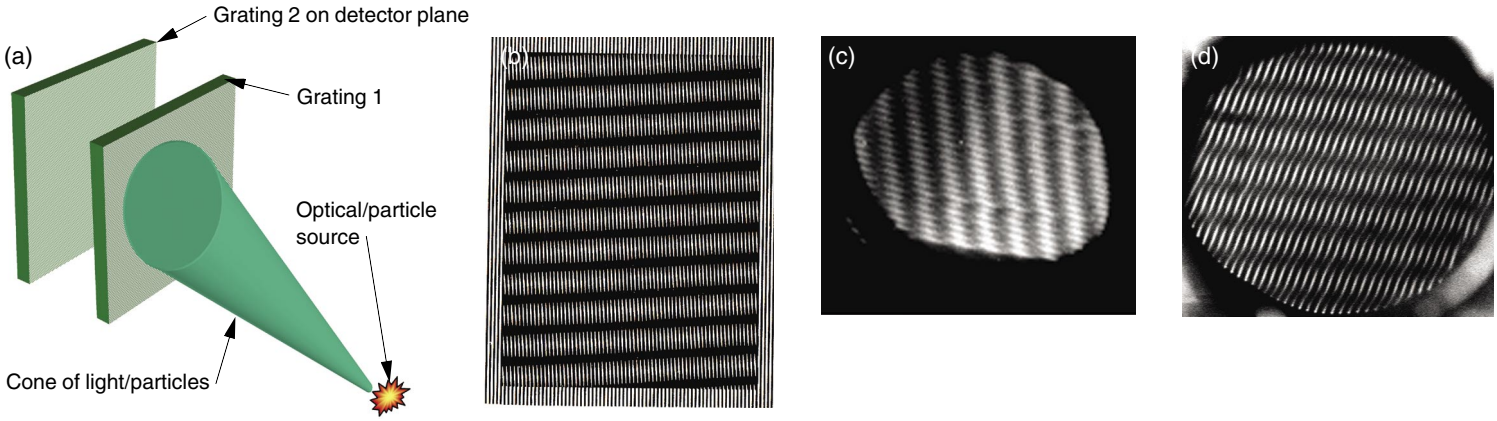
According to Mackinnon, proton beams with energies from 50 to 100 megaelectronvolts, produced by an ultrashort-pulse laser, could one day be used to probe the cores of NIF targets as they are compressed by laser light. Lower-energy protons also could be useful, for example, to diagnose electric and magnetic fields inside hohlraums, the metal cases that enclose many NIF targets. More experimental and theoretical work is under way to fully investigate this promising technique.

Mackinnon notes that another kind of proton radiography is being studied by researchers at Lawrence Livermore and Los Alamos national laboratories. But the protons created in those studies are much more energetic—about 800 megaelectronvolts. (See *S&TR*, November 2000, pp. 12–18.) That research centers on beams of extremely high-energy protons focused with magnetic lenses and is designed to image deep inside larger exploding objects.

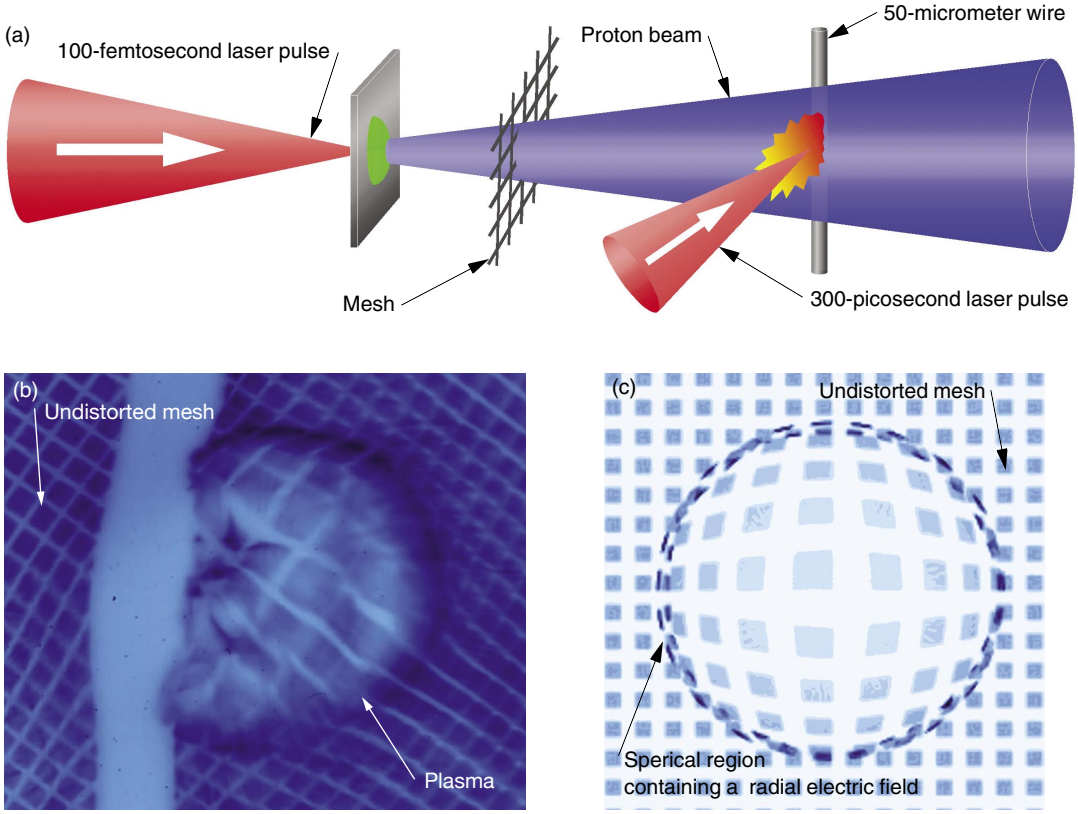
Livermore physicists Mike Key and Richard Town are also studying whether proton beams, instead of electron beams, can be used to drive fast ignition on NIF. (See *S&TR*, March 2000, p. 4–12.) In fast ignition, at the moment of maximum compression, a laser pulse plows through the plasma to make a path for another very short, high-intensity pulse (presumably, of electrons) to ignite the compressed fuel. In theory, fast ignition reduces both the laser energy and the precision requirements for achieving ignition.

Field Strength and Geometry

In collaboration with Marco Borghesi from Queen's University and Oswald Willi and G. Pretzler from Heinrich-Heine-Universität, the Livermore team is investigating another aspect of proton radiography: diagnosing the transient electric and magnetic fields directly through particle-deflection measurements. Unlike x rays, protons are electrically charged, so they interact with electric and magnetic fields in plasmas. Proton probing would provide a new method to visualize and measure fields in laser plasma experiments, which are not well understood.



(a) In one particle-deflection technique for diagnosing the transient electric and magnetic fields, a proton beam passes through two identical gratings. The gratings are separated by a small distance, and their rulings are rotated at slight angles to each other. (b) The proton beam is imprinted with a grating pattern called proton moiré. A proton beam passing through the plasma causes a shift in the moiré pattern, which can be used to infer the strength of the electric and magnetic fields. (c) Proton moiré is similar to (d) the more common optical moiré.



(a) Another particle-deflection technique uses a single grid to subdivide protons into hundreds of small proton beamlets. (b) Proton radiograph of plasma created by a 300-picosecond laser pulse on a 125-micrometer-thick wire shows the plasma magnified and distorted because of the plasma's electric and magnetic fields. The pattern is similar to (c) a hybrid code simulation of protons propagating through a spherical region that contains a radial electric field, which reproduces the essential features of the experiment.

For these experiments, the researchers are using the JanUSP, Vulcan, and LULI lasers. Developing such proton radiography diagnostics supports the Laboratory's stockpile stewardship mission by helping scientists better understand hot, dense plasmas.

In one technique, the proton beam passes through two identical gratings. The gratings are separated by a small distance, and their rulings are rotated at slight angles to each other. In effect, the proton beam is imprinted with a pattern of the gratings, called proton moiré. When the beam passes through the plasma, the electric and magnetic fields can cause shifts in the moiré pattern. The change in pattern can then be used to infer the strength of the electric and magnetic fields.

A related technique uses a single, two-dimensional grid to subdivide protons into hundreds of small proton beamlets. A hybrid code that simulated proton propagation through a plasma containing a radial electric field essentially reproduced the main features of the experimental observations.

The Livermore team expects protons to complement x rays as a diagnostics tool, not replace them. The team is confident that its pioneering use of protons to create and diagnose plasmas will advance a host of research projects, both at Livermore and at plasma research centers worldwide.

—Arnie Heller

Key Words: Janus ultrashort-pulse (JanUSP) laser, National Ignition Facility (NIF), Petawatt laser, plasma, protons.

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Multigrid Solvers Do the Math Faster, More Efficiently

TAKE a simulation. A big simulation, in which you want to view a fairly high level of detail. Maybe you want to study inertial confinement fusion as part of the nation's Stockpile Stewardship Program or find out the effects of global climate change in a specific area or explore the mechanism by which stars collapse and then explode in supernovae. To model those events in three dimensions and at high resolution, you'll need immense computer codes. And processing these codes will require the computational speeds and large memories of massively parallel supercomputers, such as those developed for the National Nuclear Security Administration's (NNSA's) Advanced Simulation and Computing (ASC) Program.

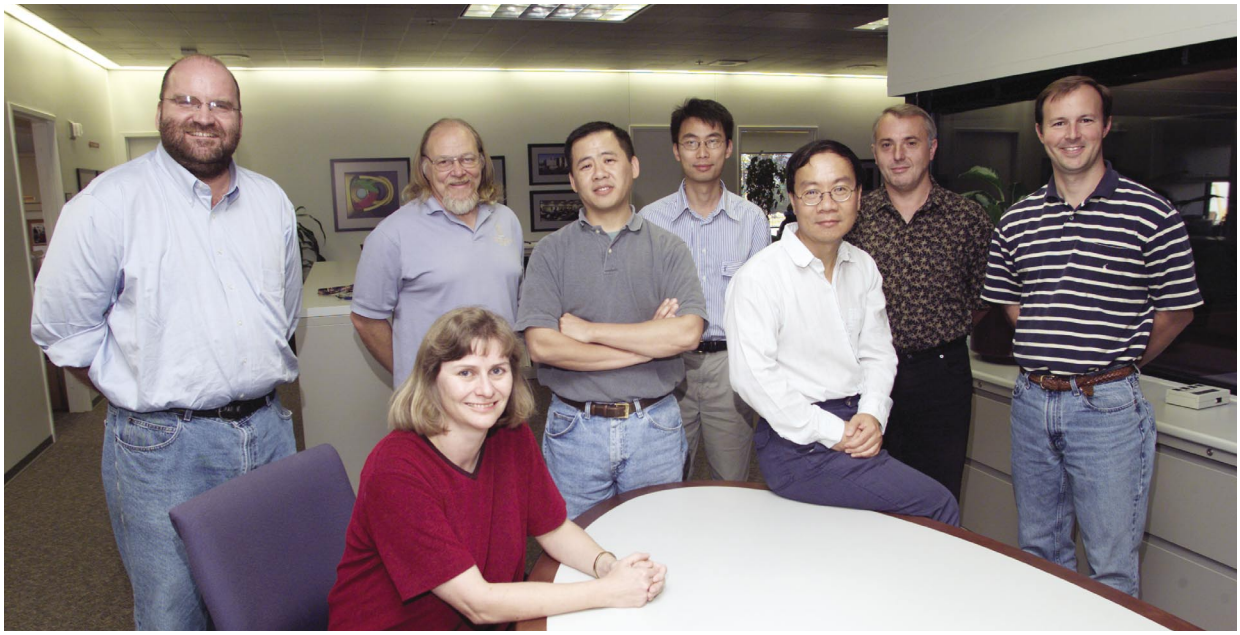
Ideally, you would want the computing time to remain the same even though the size of the problem increased. But unfortunately, as simulations become more lifelike and detailed and more processors are added to handle the calculation, the run times to solve a calculation may become even longer.

At least, that was the situation until scientists developed software codes called scalable linear solvers. These codes, including those developed at Lawrence Livermore, help keep simulations running fast as they grow larger and more processors are added to attack the problem.

Flowing from Groundwater Research

According to computational mathematician Rob Falgout, Livermore researchers began to develop scalable linear solvers in the early 1990s as part of a project funded by the Laboratory Directed Research and Development (LDRD) Program. "Steven Ashby and I were working with a team to develop a code called ParFlow, which simulates the flow of groundwater through different kinds of materials underground," says Falgout. "As a part of that LDRD project, we began developing solvers—special algorithms or processes for solving specific problems—to significantly speed up the solution of the mathematical equations generated by ParFlow."

In that particular application, the sites being modeled were several square kilometers, and the calculations had to resolve differences in subsurface media of a few meters. As a result, the



The Scalable Linear Solvers project team: (seated) Ulrike Yang; (standing, left to right) Jim Jones, Van Emden Henson, Barry Lee, Edmond Chow, Charles Tong, Panayot Vassilevski, and Rob Falgout. Not pictured: Jeff Painter and Tom Treadway.

computational grids had up to 100 million spatial zones. For ParFlow to handle such detailed simulations, the researchers had to design the code so it would effectively harness the power of massively parallel processing.

ParFlow did all that and more. It proved to be portable and scalable—that is, it can run on a variety of computing platforms, and it efficiently uses the processors that are added to handle larger problems. It's also very fast, reducing the time for some simulations from 30 minutes to only 13 seconds.

Part of ParFlow's success sprang from the multigrid approach it used, which solved problems 100 times faster than other solvers. Since that initial success, interest in multigrid linear solvers for parallel computers has grown not only at Livermore but throughout the scientific computing community. As a result, with support from the ASC Program and the Department of Energy Office of Science's Mathematical, Information, and Computational Sciences Program, the Scalable Linear Solvers project was formed at the Laboratory's Center for Applied Scientific Computing. Led by Falgout, the project now employs 10 people and is considered a premier group in this esoteric area where mathematics and computational sciences mesh.

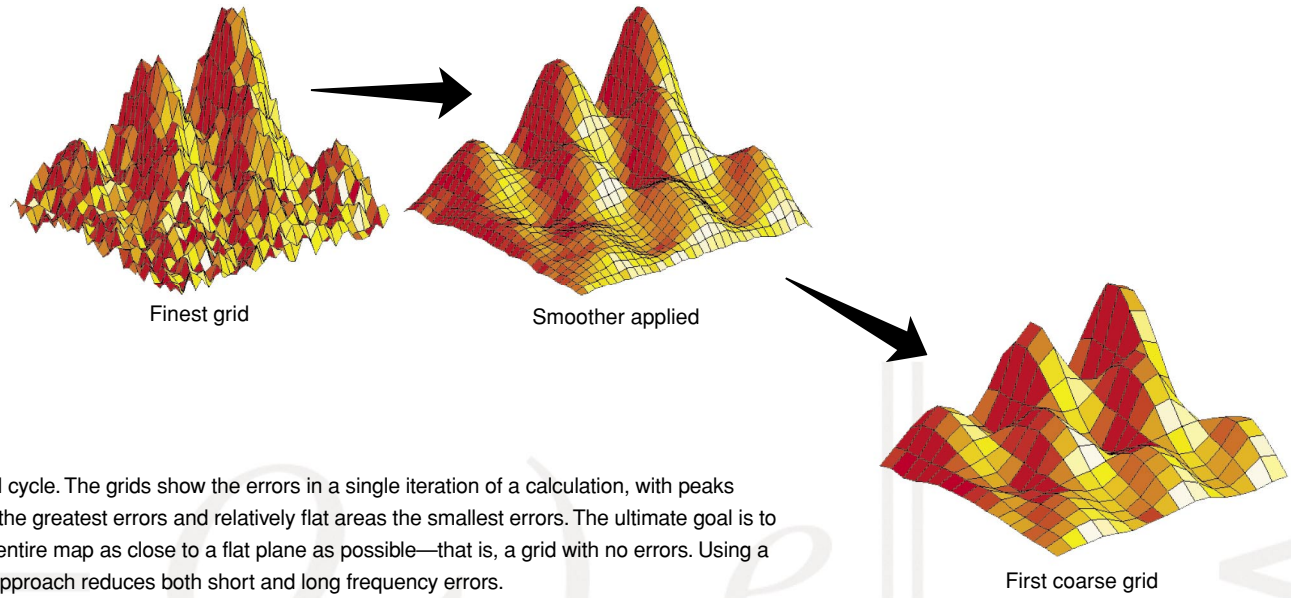
Solvers R Us

In general, a solver is an algorithm for calculating the solution to a set of mathematical equations. A linear solver calculates the solution to a linear system of equations—a set of equations just like those found in high school algebra, except that the set has millions or billions of equations to solve. The unknowns in these linear systems of equations can represent a variety of physical quantities, such as the pressure at a particular location underground or the new location of a piece from an automobile frame after a crash. In most simulation codes at Livermore, these unknowns are also associated with points on a grid. For example, in the groundwater-flow application, the grid points represent underground locations.

In many large-scale scientific simulation codes, a large fraction of the overall run time on the computer is spent in linear solvers. Cut the processing time for these solvers, and the total run time shrinks. Thus, says Falgout, much of the research and development in scalable algorithms is aimed at solving these large, linear systems faster and more efficiently on parallel computers.

In the multigrid approach, code developers can reduce this time by making clever use of a sequence of smaller linear systems, each associated with a coarser grid—hence, the term multigrid. Computational mathematician Jim Jones, who also works on the project, says, "Probably the simplest way to think about it is to imagine a two-dimensional problem, where you have some initial guess of what your answer should be. The idea is to generate a new or better guess through some simple procedure or algorithm, then repeat the procedure until your guess converges with the solution of the linear system."

Initially, each unknown in the guess is incorrect, and it may differ significantly from the correct value. When these errors are plotted, the plot has lots of peaks and valleys, as shown on p. 18. The goal is to generate a new guess that matches the correct value, or has zero error. Standard solver algorithms generate new guesses



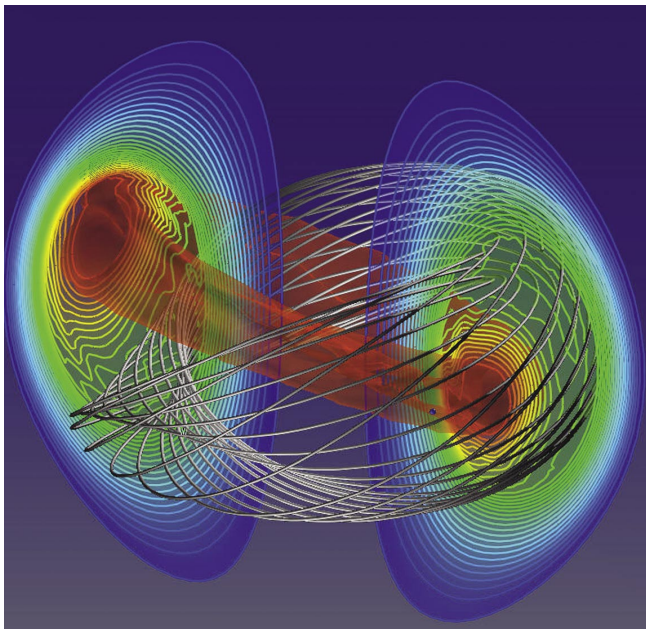
A multigrid cycle. The grids show the errors in a single iteration of a calculation, with peaks indicating the greatest errors and relatively flat areas the smallest errors. The ultimate goal is to make the entire map as close to a flat plane as possible—that is, a grid with no errors. Using a multigrid approach reduces both short and long frequency errors.

with a "smoother," a special method that smooths the errors when they are plotted. With the multigrid approach, code developers take advantage of the smoother by recognizing that low-frequency (or long-length-scale) errors can be accurately and efficiently resolved on a coarser, or smaller, grid.

The Livermore researchers are creating scalable linear solvers based on this multigrid technology. The main ingredients of a multigrid solver are the smoother, the coarse-grid linear systems, and the procedures for transferring data back and forth between the algorithms. "If all the components are properly defined," says Jones, "the method will uniformly damp error frequencies, and the computational cost will depend only linearly on the problem size. In other words, multigrid algorithms are scalable."

The Laboratory's multigrid solvers are based on two methods: geometric and algebraic. Geometric multigrid methods are used for linear systems defined on rectangular grids or meshes; algebraic multigrid methods are for linear systems defined on unstructured, or nonrectangular, grids. The geometric methods are used more often, but constructing the solver requires geometric information about the physical problem. Algebraic methods require no geometric information but are more difficult to design.

The Livermore team implements its solver algorithms in a software library called *hypre*, which runs on simple laptops and workstations as well as on massively parallel computers such as ASCI White. The solver codes in *hypre*—including the algebraic multigrid code BoomerAMG and the geometric multigrid solvers SMG and PFMG—are maintained by the project team and are available to the scientific community worldwide.



The Department of Energy's Scientific Discovery through Advanced Computing (SciDAC) Program is funding multigrid research and development for various application areas. For example, algebraic multigrid techniques are being applied to tokamak fusion applications in collaboration with the SciDAC Center for Extended MHD Modeling (CEMM). This example from a CEMM simulation is of a tokamak sawtooth instability showing pressure contours and surface with some magnetic-field lines.

Making the Impossible Possible

Scalable linear solvers are allowing scientists to both pose and answer new questions. For example, consider a simulation at a particular resolution that would take several days to run. Increasing the resolution to make the model more accurate and lifelike means that the simulation will take even longer to run. In the world of simulations, run time is money. Because the Livermore multigrid solvers reduce the run time, scientists can push their simulations to the next level of detail.

At Livermore, for example, researchers integrated a parallel algebraic multigrid code into the hydrodynamics code ALE3D to solve difficult elasticity problems for modeling the deformation of materials. Researchers in Germany have also used *hypre* solvers to predict results from operations to correct facial deformities—another use of elasticity equations, but this time for medical applications.

"For those of us on this project," says Falgout, "our work is mostly about mathematics. But it's important to remember that the equations we're trying to solve are not just abstract symbols and numbers. They describe real physical processes that researchers are trying to better understand—whether it's radiation flow in a supernova, the flow of groundwater through the subsurface, or the behavior of a plasma in a complex magnetic field. Today, computer simulations are increasingly important in scientific investigations, aiding or even taking the place of traditional experiments. So any method we can find to make detailed, three-dimensional problems run more quickly and efficiently opens new doors to our understanding of the world."

—Ann Parker

Key Words: Advanced Simulation and Computing (ASC) Program; algebraic multigrid; ASCI supercomputers; geometric multigrid; *hypre*; Laboratory Directed Research and Development (LDRD) Program; linear equations; Mathematical, Information, and Computational Sciences Program; scalable linear solvers; simulation codes; Scientific Discovery through Advanced Computing (SciDac) Program.

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Predicting Stability for the High-Energy Buckyball

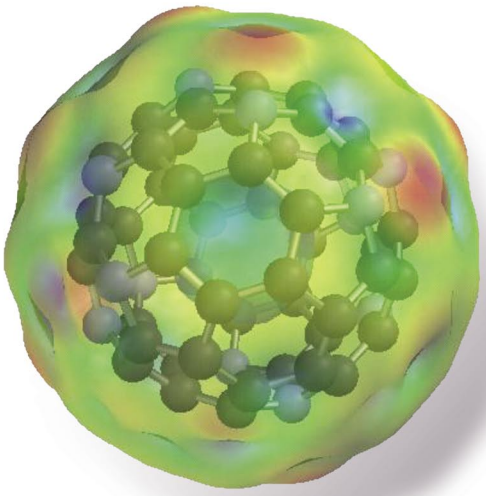
WHY do atoms have a propensity to bond in certain configurations? That mystery continues to puzzle the scientists who study molecules and their isomers—molecules that have the same atomic weight as the original but a different structure. A better understanding of the rules governing molecular structure would help them predict which forms of a substance would be most useful.

In 1985, scientists were excited by the discovery of a new form of carbon. This molecule, called a buckyball or a fullerene, contains 60 carbon atoms (C_{60}). Its molecular structure resembles a soccer ball or the geodesic dome designed by architect Buckminster Fuller, for whom the molecule is named.

Building on that discovery, Livermore scientists are developing computer models to study buckyballs that feature other atoms, such as nitrogen and boron, in place of some of the carbon atoms in C_{60}. (See *S&TR*, June 2001, pp. 22–23.) In particular, the research has focused on how to predict the most stable forms of these new compounds and on how other atoms bond to one another to create unique structures.

One team, led by theoretical chemist Riad Manaa, is studying nitrogen fullerenes, especially $C_{48}N_{12}$. Nitrogen-doped fullerenes offer an impressive range of potential applications, from orthopedic implants to new pharmaceuticals to high explosives.

"These fullerenes are interesting to study," says Manaa, who works for the Chemical Engineering Division in Livermore's Chemistry and Materials Science Directorate. "Their hollow, cagelike shape and their extreme stability at high temperature and pressure allow them to retain their spherical structure when they interact with other atoms and molecules. By understanding how the carbon and nitrogen bonds come together, we can study the properties and predict how other atoms will interact with the fullerene to form new compounds."



The molecular structure of $C_{48}N_{12}$ with its electronic cloud.

The nitrogen fullerene has properties that differ from the more commonly known carbon fullerene. All electron shells in the C_{60} molecule are filled, so C_{60} is inert. However, when some of the carbon atoms are replaced with nitrogen atoms, the new molecule acts as an electron donor. Nitrogen also carries much more energy than carbon, so nitrogen fullerenes might be useful in developing new high-explosive formulations. Computer simulations indicate that other elements could be added to the molecule to form compounds for a range of applications.

Computing the Possibilities

The team's search to find the most stable forms of nitrogen fullerenes began as a teaching project between Manaa and a group of summer interns. Manaa, who investigates energetic materials and conducts simulations of these materials in extreme conditions, worked with the interns to study various forms of the $C_{48}N_{12}$ molecule, which has a high energy content.

When $C_{48}N_{12}$ was first synthesized several years ago, the electron microscopy and energy-loss spectroscopic analysis showed that its structure corresponds to that of a buckyball. At that time, researchers believed the most stable form of this nitrogen-substituted fullerene had 12 pentagons with evenly spaced nitrogen atoms, one in each pentagon. Nitrogen atoms tend to repel each other and destabilize the structure, so if the molecule is stable, they must be separated.

In the 12-pentagon model, every nitrogen atom is separated by two carbon atoms. The remainder of each pentagon is composed of carbon atoms. That molecule also has two all-carbon hexagons, or benzenelike rings. Benzene rings are very stable, so having two benzene rings and at least two carbon atoms between each nitrogen atom provided the molecule's stability.

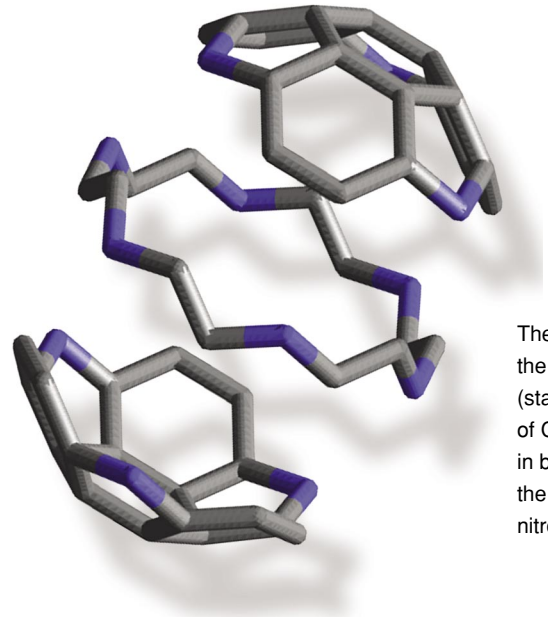
Predicting the most energetically stable structure of a molecule is a formidable task for computational scientists, especially when they must determine the various configurations for as many as 60 atoms. It's time-consuming work to examine the many possibilities of distributing the 12 nitrogen atoms in $C_{48}N_{12}$ among the 20 hexagons and 12 pentagons of a buckyball structure. Even the Laboratory's supercomputers, such as ASCI Blue, must process calculations day and night for weeks to model all the configurations.

According to Manaa, the team's original goal was to find stable molecular structures with subunits of nitrogen–nitrogen

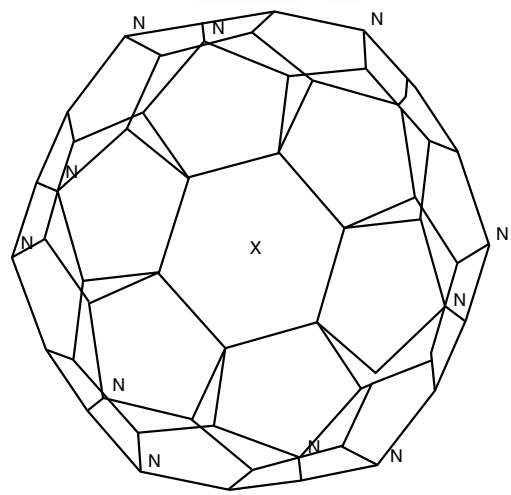
(N–N) bonding, which have high energy content. The team used quantum-chemical methods to predict the stable structures of these fullerenes, which have a radius of 0.35 nanometer. The computer code calculates the distribution of electrons around each atom, which then determines the chemical property of a molecule.

"While we were studying the high-energy, fullerene-analog structures of $C_{48}N_{12}$ with $6N_2$, $4N_3$, and $2N_6$ subunits," says Manaa, "we also found the energetically most stable structure of this molecule. This finding allows us to predict the chemical and physical properties of the material."

The new molecule has eight highly stable all-carbon hexagons. Although the nitrogen atoms are separated by only one carbon atom, it has six additional benzenelike rings, which more than make up for any possible repulsion between the nitrogen atoms. Thus, the new $C_{48}N_{12}$ structure is more stable because the molecule's resonance energy is maximized and the repulsive force from the N–N bonds is minimized. In fact, the team's calculations showed that this structure is much more stable (as much as 13.1 kilocalories per mole) than the most stable structure reported for the first $C_{48}N_{12}$ molecule.



The geometry of the minimum energy (stable) structure of $C_{48}N_{12}$ (nitrogen in blue), indicating the distribution of nitrogen atoms.



Front view of the symmetric $C_{48}N_{12}$ structure, showing the position of the nitrogen atoms. Seven all-carbon hexagons are visible, and the eighth is superimposed on the central ring.

Designing New Molecules

Doping the C_{60} molecule—that is, substituting some of the molecule's carbon atoms with other atoms—changes the structural, electronic, chemical, and physical properties of the parent fullerene. For example, when some of the carbon atoms on the buckyball cage are replaced with nitrogen, the molecule's electronic properties change to match those of a semiconductor. Other doped fullerenes are ideal candidates for phototonic devices, such as optical switches, eye protectors, and sensors, and some are being considered as therapeutic agents.

Manaa's group also examined a fullerene molecule doped with boron. Their results showed that $C_{48}B_{12}$ has the same stability as $C_{48}N_{12}$. This same molecular structure as $C_{48}N_{12}$ decisively confirmed the overall stability of the $C_{48}X_{12}$ molecules (where X can be boron, nitrogen, or silicon). Recent calculations show that, even though $C_{48}N_{12}$ and $C_{48}B_{12}$ have similar structures, they have opposite properties. While $C_{48}N_{12}$ acts as an electron donor, the charge distribution in $C_{48}B_{12}$ makes it an electron acceptor. Thus, when these molecules are combined, $C_{48}N_{12}$ and $C_{48}B_{12}$ become a donor–acceptor pair for molecular electronic building blocks. Potential applications combining the two molecules include circuits for electronic switches and nanocircuits for data exchange.

The team's efforts are now directed toward building carbon structures with other combinations of nitrogen and boron, such as $C_{48}B_6N_6$. Materials made from this combination would be harder than C_{60}.

Most of the fullerene research to date has been conducted as part of the hardware tests and code development work for the National Nuclear Security Administration's (NNSA's) Advanced Simulation and Computing Program, which is an integral component of the NNSA's Stockpile Stewardship Program. However, according to Manaa, the team is also interested in collaborating with groups outside the Laboratory to build on Livermore's expertise in the computational research of fullerenes.

"So much of the progress in synthesizing new forms of nitrogen fullerenes and other molecules requires a thorough understanding of their structure and properties," he says. "Understanding why molecules take the form they do adds to the predictive possibilities scientists can make about new molecules for all kinds of applications."

—Gabriele Rennie

Key Words: Advanced Simulation and Computing Program, ASCI Blue, buckyball, carbon, fullerenes, nitrogen.

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Each month in this space we report on the patents issued to and/or the awards received by Laboratory employees. Our goal is to showcase the distinguished scientific and technical achievements of our employees as well as to indicate the scale and scope of the work done at the Laboratory.

Patents

Apparatus and Method for Electrical Insulation in Plasma Discharge Systems

Mark A. Rhodes, Scott N. Fochs
U.S. Patent 6,605,901 B1
August 12, 2003
An apparatus and method to contain plasma at optimal fill capacity of a metallic container. The invention uses anodized layers to form the internal surfaces of the container volume. Bias resistors are calibrated to provide constant current at variable voltage. The voltages of the metallic container can be adjusted relative to the voltage of an anode by choosing the appropriate values of the bias resistors. In this way, the optimal plasma fill can be achieved while minimizing the chance that the breakdown voltage of the anodized layer is reached.

Method for Enhancing the Solubility of Dopants in Silicon

Babak Sadigh, Thomas J. Lenosky, Tomas Diaz de la Rubia
U.S. Patent 6,627,522 B2
September 30, 2003
A method for enhancing the equilibrium solid solubility of dopants in silicon, germanium, and silicon–germanium alloys. The method involves subjecting silicon-based substrate to biaxial or compression strain. It has been determined that boron solubility was largely enhanced (more than 100 percent) by a compressive biaxial strain, based on a size-mismatch theory, since the boron atoms are smaller than the silicon atoms. The large enhancement or mixing properties of dopants in silicon and germanium substrates is primarily governed by their charge and, to second order, by their size mismatch with the substrate. Furthermore, the dopant solubility enhancement with strain is most effective when the impurity's charge and the size mismatch favor the same type of strain. Thus, the solid solubility of small p-type dopants (such as boron) and large n-type dopants (such as arsenic) can be raised most dramatically by appropriate biaxial (compressive) strain. The solubility of a large p-type dopant (such as indium) in silicon will be raised because of its size mismatch with silicon, which favors tensile strain, while its negative charge prefers compressive strain. Thus, the two effects counteract each other.

Interrogation Cradle and Insertable Containment Fixture for Detecting Birefringent Microcrystals in Bile

Chris Darrow, Tino Seger
U.S. Patent 6,628,387 B2
September 30, 2003
A transparent flow channel fluidly communicates a fluid source and a collection reservoir. An interrogating light beam passes through a first polarizer having a first plane of polarization. The flow channel is orthogonal to the light beam. The light beam passes through a fluid sample as it flows through the flow channel. The beam is then filtered through a second polarizer that has a second plane of polarization rotated 90 degrees from the first plane of polarization. An electronic photodetector aligned with the light beam signals the presence of birefringent microcrystals in the fluid sample by generating voltage pulses.

A disposable containment fixture includes the flow channel and the collection reservoir. The fixture is adapted for removable insertion into an interrogation cradle that includes optical and data-processing components. The cradle rigidly positions the centerline of the flow channel orthogonal to the light beam.

Inductrack Configuration

Richard Freeman Post
U.S. Patent 6,629,503 B2
October 7, 2003
A simple permanent-magnet-excited maglev geometry provides levitation forces; it is stable against vertical displacements from equilibrium but is unstable against horizontal displacements. An Inductrack system is then used with this system to effect stabilization against horizontal displacements. The Inductrack system also provides centering forces to overcome centrifugal forces when the vehicle is traversing curved sections of a track or when another transient horizontal force is present. In some proposed embodiments, the Inductrack track elements are also used as the stator of a linear induction-motor drive and braking system.

Inductrack Magnet Configuration

Richard Freeman Post
U.S. Patent 6,633,217 B2
October 14, 2003
A magnet configuration comprising a pair of Halbach arrays magnetically and structurally connected together. The Halbach arrays are positioned with respect to each other so that a first component of their fields substantially cancels at a first plane between them and a second component of their fields substantially adds at this first plane. A track of windings is located between the pair of Halbach arrays, and a propulsion mechanism is provided for moving them along the track. When the arrays move along the track and the track is not located at the first plane, a current is induced in the windings, which then exerts a restoring force on the pair.

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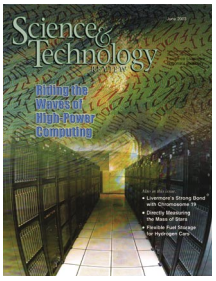

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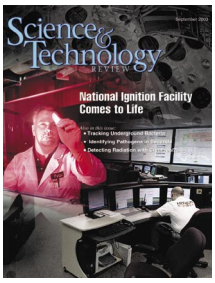

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Abstracts

An Extraordinarily Bright Idea

Livermore researchers are part of the design team for the Linac Coherent Light Source (LCLS), the world's first full-scale x-ray laser. This fourth-generation light source will be built at the Stanford Linear Accelerator Center in Palo Alto, California. The LCLS uses a free-electron laser to produce beams of coherent, high-energy x rays that will be as much as 10 billion times brighter than third-generation (synchrotron) light sources. The very short pulses of this extraordinarily bright beam will be used to make major advances in biological research, astrophysics, and stewardship of the nation's nuclear stockpile. The Livermore team is using computer simulations and laboratory experiments to design the optics that transport the beam to two experimental chambers and the diagnostics to measure the x rays. The team is also establishing x-ray pulse parameters for the first biological experiments.

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Henry Chapman (925) 423-1580 (chapman9@llnl.gov).

Using Proton Beams to Create and Probe Plasmas

Lawrence Livermore researchers are investigating the utility of proton beams created by powerful, ultrashort pulses of laser light to create and diagnose plasmas. The proton-beam experiments, conducted with researchers from Europe and the University of California at Davis, promise new techniques to help scientists maintain the nation's nuclear arsenal and better understand how stars function. The proton beams are produced by pulses of laser light lasting about 100 femtoseconds and with brightness of up to 5×10^{20} watts per square centimeter. When the laser pulses are focused onto thin foil targets, protons with energies up to 25 megaelectronvolts are emitted in a beam that is nearly perpendicular to the foil's rear surface. The team has discovered that the proton beams are suitable for radiographic applications to diagnose plasma conditions generated by high-power lasers. The team's proton radiographs were the first taken of a laser-driven implosion with picosecond resolution. The researchers are investigating another aspect of proton radiography—diagnosing the transient electric and magnetic fields directly through particle-deflection measurements.

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U.S. Government Printing Office: 2003/683-056-70053

Hunting the Elusive Axion

A Livermore-designed experiment to observe the axion is poised to answer key questions in the realms of particle physics and astrophysics.

Also in January/February

- *For the first time, scientists have fully mapped the phonons in gallium-stabilized delta plutonium.*
- *Plant and wildlife monitoring and research programs ensure a safe haven for rare flora and fauna at Lawrence Livermore.*
- *Common buoys outfitted with radiation detectors could soon play an important role in safeguarding marine environments.*

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