A New Realm of Materials

A multidisciplinary approach to materials science provides insights on how solids respond to extreme dynamic stresses.

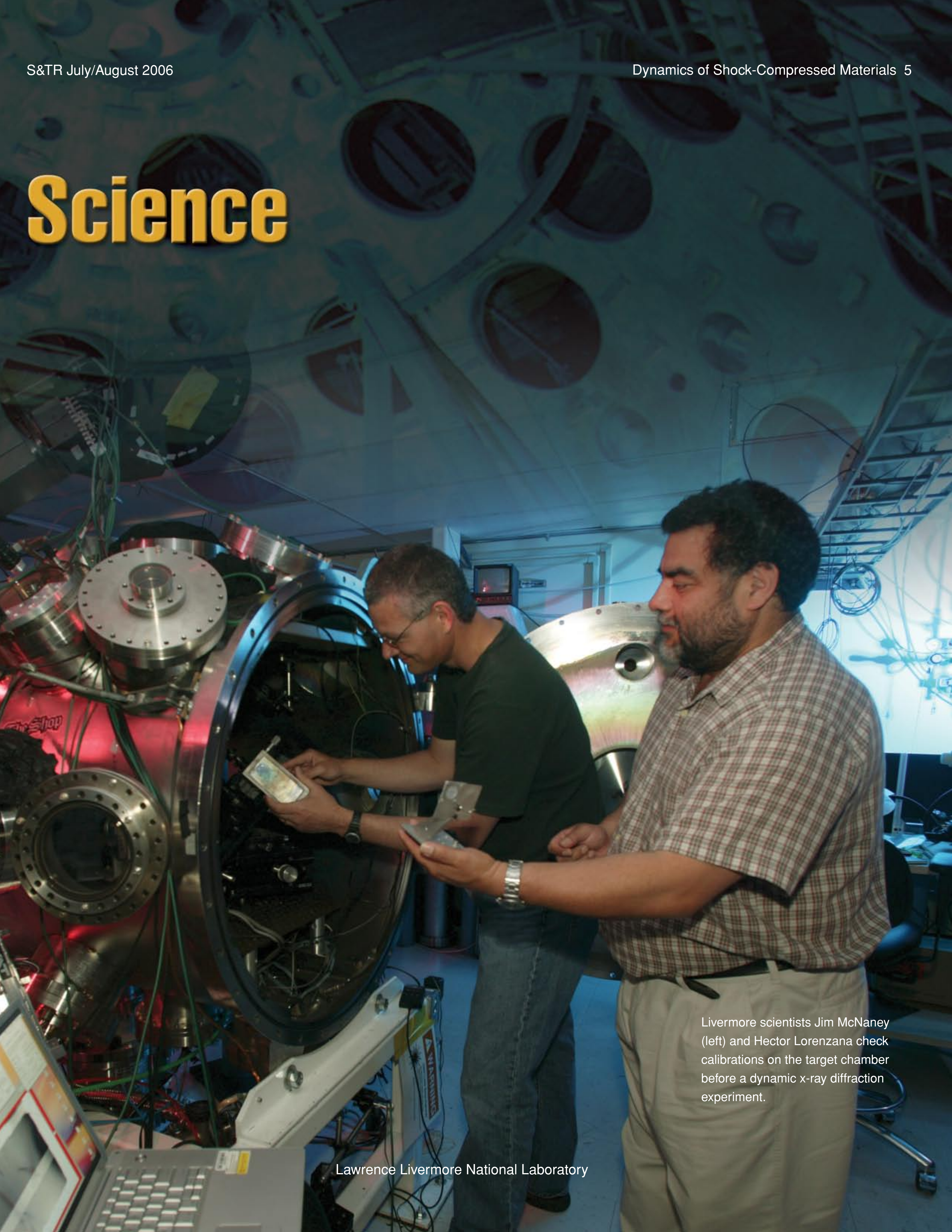
KNOWLEDGE is power, and for many scientists, understanding the dynamic lattice response of solids under extreme pressures, temperatures, and strain rates can be quite powerful, indeed. This quest to expand the fundamental knowledge of material behavior has spanned nearly a century, and it remains an exciting scientific frontier in high-energy-density materials science. Materials science is an essential part of Livermore's work in support of the National Nuclear Security Administration's Stockpile Stewardship Program to ensure the safety and reliability of the nation's nuclear weapons stockpile. In particular, scientists want to determine how extreme dynamic stress affects a material's phase, strength, and damage evolution.

Critical to this work are studies that probe material properties at the scale of the controlling physical processes—at length scales of 1 nanometer (10^{-9} meter) and time scales of less than 1 nanosecond (10^{-9} second). Investigations at these characteristic length and time scales were unthinkable a decade ago, but the technologies and facilities available today are bringing such studies within reach. Using ultrabright, ultrafast x-ray sources at high-energy laser and accelerator facilities, scientists can directly measure the x-ray diffraction and scattering that result from dynamic changes in a material's lattice.

A multidisciplinary team led by physicist Hector Lorenzana of the Laboratory's Defense and Nuclear Technologies Directorate is collaborating with researchers from other national laboratories and universities on a Laboratory Directed Research and Development (LDRD) project to probe the real-time lattice response of metals under high shock loads. The team's collaborators include researchers from Los Alamos, Argonne, Oak Ridge, and Lawrence Berkeley national laboratories as well as from Rochester University, Oxford University, Universidad Complutense de Madrid, and the University of Texas at Austin. The team's measurements, which have nanometer and subnanosecond resolutions, allow scientists to examine the fundamental mechanisms governing macroscopic behavior.

"Critical scientific questions about material response at the lattice level must be answered," says Lorenzana. "For our stockpile stewardship work, we are interested in characterizing the condensed-matter phase transformations and damage that occur in shocked solids. Working closely with the Laboratory's theoretical and computational experts, we are coupling the results of our experiments with first-principles simulations, which for the first time can explore physical phenomena at overlapping temporal and spatial scales."

Science



Livermore scientists Jim McNaney (left) and Hector Lorenzana check calibrations on the target chamber before a dynamic x-ray diffraction experiment.

Current Challenges and Obstacles

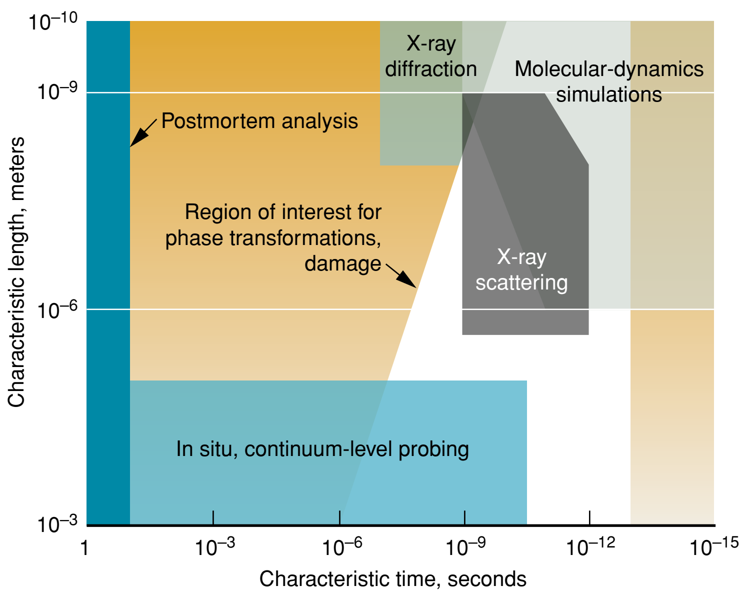
The measurement methodologies used to study dynamically compressed solids have been limited to large-scale probes such as time-resolved surface diagnostics or characterizations of recovered specimens. Such measurements limit scientists to three general time and length regimes: slow macro (slow time scales and macroscopic dimensions), slow micro, and fast macro. For example, slow macro techniques include Hopkinson bar mechanical testing. To study the slow micro regime, scientists can characterize the microstructure of shock-recovered samples or observe materials compressed by a diamond anvil cell. With surface velocimetry or optical spectroscopy, they can probe the fast macro regime.

However, using these techniques, scientists must infer how physical processes interact at the atomic level under extreme shock loading. "To improve our understanding of the shocked solid, we must access the fast micro regime," says Lorenzana. "For those studies, we need techniques that allow us to directly observe dynamic changes as they occur in a material's lattice structure."

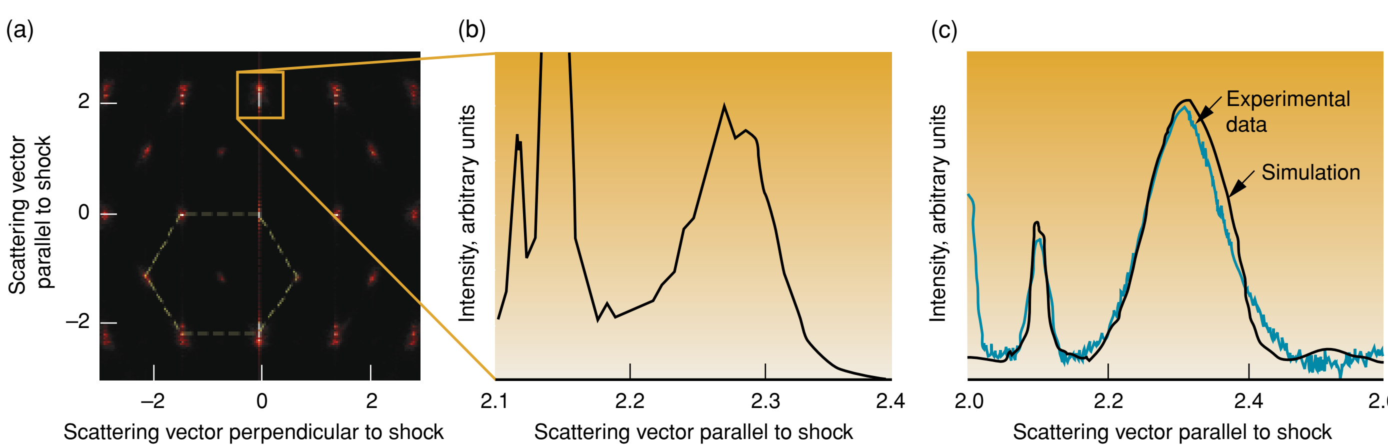
Measurements in the fast micro regime can be used to characterize materials as they change phases, for example, as a solid melts or a material plastically deforms by creating a defected state. With such information, scientists can determine how damage evolves in a material under extreme pressures and causes the material to fail. "This next generation of fast micro probing will push our fundamental knowledge of shocked solids forward," says physicist Bruce Remington, who works in Livermore's National Ignition Facility (NIF) Programs Directorate, "and it will help us understand the microscopic lattice response under extreme conditions of compression. Research in the fast micro regime is important for achieving ignition on NIF, for example, where the target capsule's response to the first shock can affect its subsequent implosion dynamics."

Blending Experiment and Theory

Lorenzana emphasizes the importance of bridging experiment and theory at comparable temporal and spatial scales in the pursuit of new scientific discoveries. The LDRD project is the first comprehensive lattice-level study of the behavior of shocked solids using both experiments and simulations at overlapping length and time scales. "Experimental data allow us to quantify and validate our theories," says Lorenzana. "Theoretical studies, in turn,



Few data are available to study the physical interactions that occur at very short time and length scales when materials are placed under extreme pressures. To examine this regime, scientists are combining new diagnostic techniques with computer simulations.



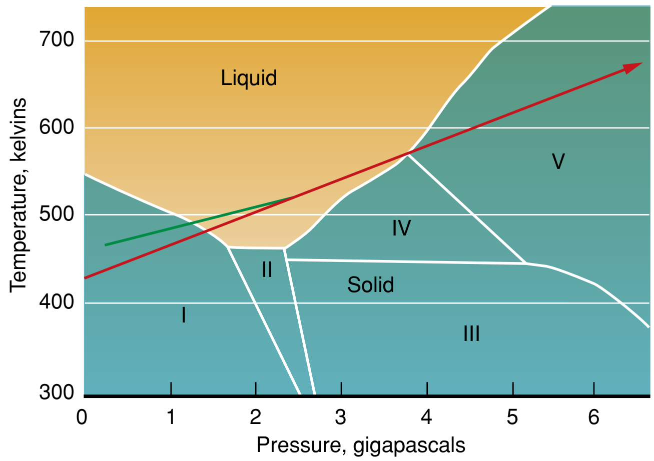
(a) Molecular-dynamics simulations of iron being transformed by shock predict the pattern of x-ray diffraction from many planes. (b) A closeup view of one lattice plane shows the predicted broadening and peak position of the diffraction signal. (c) The predicted response (black curve) from a simulation using a grain size of 8 × 8 × 8 cells agrees with the experimental data (blue curve).

help us interpret the data and better design subsequent experiments. The outcome of this cycle is enhanced experiments and theory that lead us to an understanding of new physics."

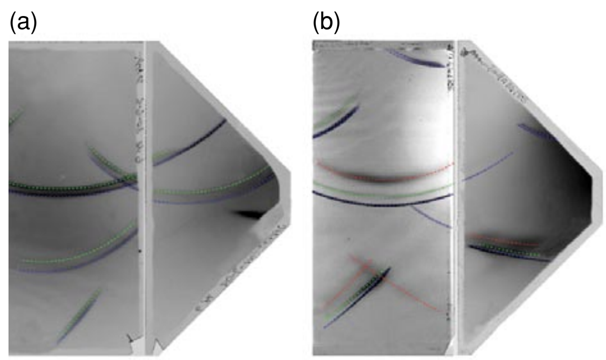
Also working on this project are Livermore physicists Eduardo Bringa, Babak Sadigh, and Jaime Marian of the Chemistry and Materials Science (CMS) Directorate. According to Bringa, the team's work is providing a building block to improve scientific understanding of material behavior under dynamic loading, which is critical to the success of stockpile stewardship, energy research, and other important national endeavors.

"Livermore has some of the best computers in the world," says Bringa. "With them, we can simulate systems atom by atom at the length and time scales used in our experiments and validate our materials models. In addition, the simulations provide detailed information about interactions that experiments cannot yet probe. Until we can develop experiments to study the evolution of shock-induced dislocation density with nanosecond resolution, we must rely on atomistic simulations to access such processes. Relevant information is lost in current recovered samples, where the unloading and thermal history greatly modify dislocation content."

The metal bismuth illustrates the importance of unraveling material processes such as phase transformations. Bismuth exhibits a complex static phase diagram that presents challenges to researchers who study its solid-to-solid and solid-to-melt phase transformations during shock loading. At about 420 kelvins, shocked bismuth may melt and eventually resolidify as pressure is increased or released. According to Lorenzana, scientists want to predict how such phase transformations affect the material properties of the shocked solid, but they must first acquire accurate data on the changes that occur at the lattice level. Bismuth could serve as a test bed to study the kinetics of such transformations, allowing scientists to determine the rates of phase changes and the location of phase boundaries.



A phase diagram of bismuth shows the material states that can occur as pressure and temperature vary. At about 420 kelvins, shocked bismuth may melt and resolidify as compression increases (red curve) or is released (green curve).



Micrographs from a dynamic x-ray diffraction experiment show the phase transformation in iron under extreme pressure: (a) at low pressure before transition and (b) at high pressure after the transition. Blue lines indicate the unshocked material. Green lines show the compressed phase of a material that is not transformed. Red lines show a new phase beginning to emerge.

Because damage prediction depends on a material's microstructure and its phase history, scientists cannot predict a material's response to shock loading until more questions are answered. "We must first address a broader series of open, fundamental physics questions," says James McNaney, a materials scientist in the CMS Directorate. "For example, how is the microstructure modified under shock conditions? What are the dynamic high-pressure phases? How do molten and solid phases and the kinetics of the transitions affect the final state? And, perhaps most importantly, how does the resolidified material behave during subsequent loading?"

Directly measuring and simulating the response of an ordered crystalline solid to shock loading will go a long way toward answering these questions. "We are studying key atomistic processes such as relaxation, phase transformation, and kinetics," says Lorenzana. In addition, his team is looking at defect dynamics and evolution, including dislocations and the nucleation and growth of voids. (See the box on p. 8.)

Dynamic X-Ray Diffraction

A new diagnostic technique, called dynamic x-ray diffraction (DXRD), is providing great insight about the structure and spacing of the crystalline lattice in a shocked material. DXRD uses high-intensity lasers to generate both a shock in a solid and a precisely timed flash of x rays to image the lattice in motion. The x-ray flash can produce 100 to 1,000 times more photons per pulse than are produced by synchrotron accelerators. When the x rays interact with the crystal's atoms, they are diffracted in a pattern that characterizes the lattice. This image is captured on x-ray film. By resolving this time-dependent diffracted signal, scientists can develop a fundamental understanding of the kinetics of transformation and lattice relaxation.

In an effort to establish the effectiveness of the DXRD technique, a research team led by Livermore physicist Daniel Kalantar examined phase transformations in shocked iron. Materials scientists have frequently studied shock-compressed iron because the metal plays a key role in many important technologies and in the geophysical properties of Earth's formation. Despite the mature body of work associated with iron, no experiments had directly probed its lattice structure during shock loading. "In our DXRD experiments, we made the first direct observation of a transition in iron," says Kalantar. "Now, we're experimenting with other materials and other configurations."

Lorenzana's team is expanding on these initial studies. "The feasibility studies with iron established the experimental and theoretical foundation for implementing DXRD methodologies," he says. "We look forward to investigating other important phenomena such as melting in bismuth and plasticity in vanadium."

To date, DXRD experiments have been conducted using the high-energy Janus laser at Livermore, the Trident laser at Los Alamos, the Omega laser at Rochester University, and the Vulcan laser at Rutherford Appleton Laboratory in the United Kingdom. "From a materials science point of view, being able to study samples at the atomic level while they are being shocked opens up a new realm for us to investigate," says physicist James Hawreliak, a postdoctoral researcher working on Lorenzana's team. "Such studies under shock conditions are just scratching the surface of what we can learn."

X-Ray Scattering

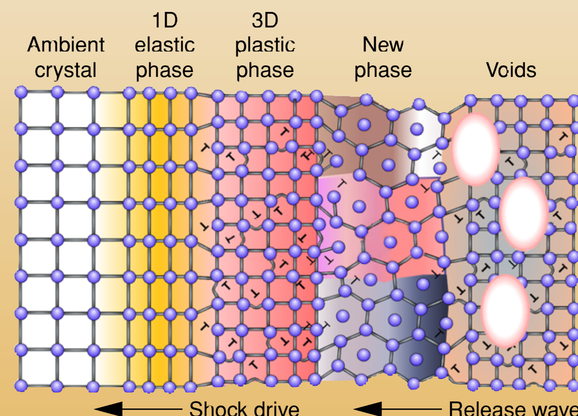
An even newer technique is in the pipeline to probe these crystalline structures at superfast speeds and with high collimation. As with x-ray diffraction, scattering experiments require a high-intensity x-ray source directed at the crystal sample. Defects in the crystal lattice will cause the x rays to scatter, generating two types of

How Do Materials Behave under Shock?

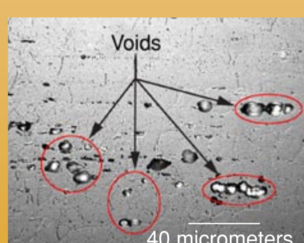
Most metals are crystalline in nature—that is, they are solids composed of atoms arranged in a regularly ordered repeating pattern. When crystals form, they may solidify into either a polycrystalline solid or a single crystal. In a single crystal, all the atoms are arranged into one lattice or crystal structure. The structure of single crystals makes them ideal for studies of material response to shock loading.

When a highly ordered material, such as a metal crystal, is put under a planar shock, the crystal is compressed along the direction of the shock propagation. This uniaxial response can remain elastic so that, once the disturbance is removed, the lattice will relax back to its original configuration. However, under high-stress conditions, the configuration of atoms in the lattice may be changed irreversibly. Irreversible changes in phase and the development of defects at the atomic level lead to macroscopic changes, such as plasticity, melting, or solid-to-solid phase transformations. When the dynamic compression is removed, the shock-modified microstructure may influence the formation and growth of voids, cracks, and other processes that may cause the material to fail.

These atomistic changes can dramatically affect a material's behavior, such as its thermodynamic state, strength, and fracture toughness. Few data are available on the phase transformations that occur under highly dynamic stress conditions or on the defects and voids that may form and grow as a result.



A crystal placed under shock loading will initially compress uniaxially and then relax plastically through defects (labeled with T's), a process known as the one-dimensional (1D) to three-dimensional (3D) transition. The material may also undergo a structural transformation, represented here as a cubic to hexagonal change. The transformation occurs over a characteristic time scale. The new phase may be a polycrystalline solid or melt. Once pressure is released, the microvoids that formed may grow, leading to macroscopic damage that causes the solid to fail.



This micrograph shows the voids that occur when a polycrystalline aluminum alloy is shocked and recovered. As the shock wave releases, the voids grow and may coalesce, resulting in material failure.

signatures: diffuse and small-angle scattering. Diffuse scattering can be used to examine atomic-scale defects in the crystal. Small-angle scattering allows scientists to characterize large-scale defects, particles, and voids.

"X-ray scattering studies have not been conducted under shock loading," says McNaney. The team plans to extend existing methodologies and analysis tools so scientists can use x-ray scattering to quantify damage in dynamically compressed materials. "Small-angle x-ray scattering experiments with subnanosecond resolution are quite challenging because they combine extreme pressures, short time scales, and low signal-to-noise ratios," says Livermore physicist Anthony Van Buuren of the CMS Directorate. "Success in developing these techniques will allow us to study processes that must be understood to predict macroscopic behavior."

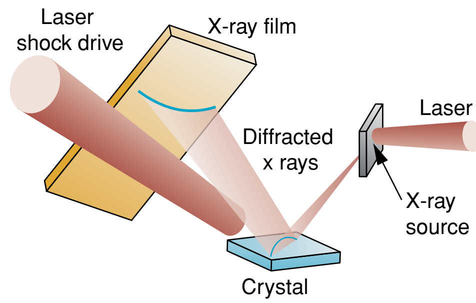
To bring this new experimental scheme into the regime needed to probe the shocked lattice response in situ, Lorenzana's team proposes to couple laser-based shock-generation techniques with the high brightness of an accelerator light source, or synchrotron, which can generate x-ray pulses that last about 100 picoseconds. X-ray scattering of the highly collimated and monochromatic beams produced by a synchrotron will provide data that can be analyzed to determine feature size, spacing, and morphology in the fast micro regime.

"Synchrotron radiation includes a large part of the electromagnetic spectrum—from infrared light to x rays," says team member Art Nelson, who works in the CMS Directorate. "In our measurements, we need a narrow band of x-ray wavelength, and on average, the synchrotron sources give more x-ray photons per pulse than conventional tabletop x-ray sources, such as diffractometers." In addition, these superbright, ultrafast x-ray pulses—as short as 70 femtoseconds—are highly collimated and can be focused to very small dimensions, making them ideal for looking at minute fluctuations in material structures in the ultrafast time regimes. (See the box on p. 10.)

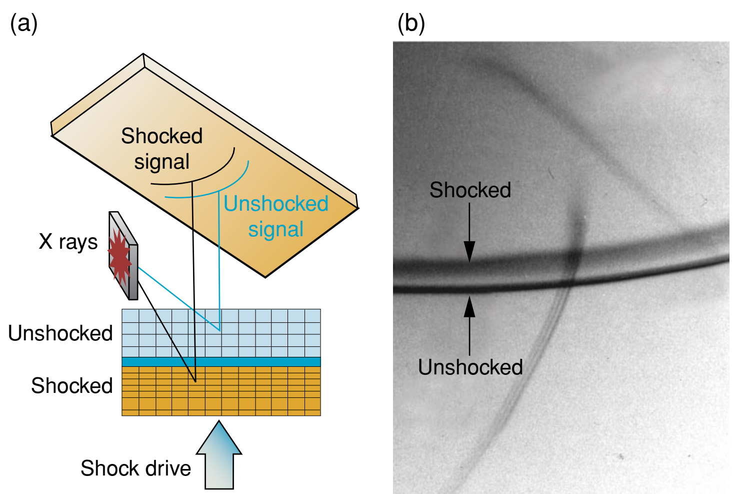
"Probing ultrafast phenomena is at the forefront of science," says Nelson, "not only in physics, but also in areas as diverse as chemistry and biology." For example, ultrafast biochemical processes such as photosynthesis are a potential area of inquiry that could benefit from superfast microscale probing. Says Nelson, "With these emerging methodologies, we can observe processes as they happen in nature."

Success So Far

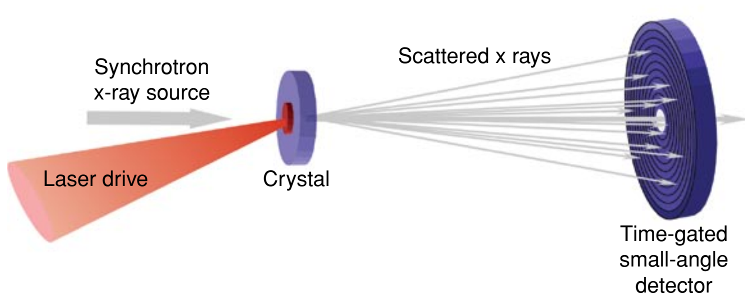
To date, the first-phase experiments have been performed on laser-based platforms at Livermore. Synchrotron-based investigations of shocked solids are planned for the latter part of 2006. Lorenzana acknowledges that much work is left to do. So far, the team is laying a technical foundation and demonstrating proof of principle for in situ studies of lattice-level processes in shocked solids. "We have demonstrated that our approach to probing highly dynamic shocked systems produces quantitative results at the physically relevant temporal and spatial scales," he says. "Success in this endeavor using laser- and synchrotron-based platforms will



Dynamic x-ray diffraction uses a high-intensity laser to shock a crystal sample. The x-ray diffraction pattern is recorded for later analysis.



Lattice compression shifts the x-ray diffracted signal in characteristic ways. (a) The change in lattice spacing will shift the angle of the diffracted arc, displacing the signal on the detector. (b) Measurements on titanium with Livermore's Janus laser clearly show the diffraction pattern.



X-ray scattering experiments using synchrotron accelerator light sources will allow scientists to measure the bulk distribution of material defects between 1 nanometer and 0.1 micrometer.

In Materials Science, Bright Is Might

Accelerator, or synchrotron, light sources, such as the Advanced Light Source at Lawrence Berkeley National Laboratory, provide the most brilliant x-ray beams available for scientific research. In these machines, electrons traveling at nearly the speed of light are forced into a circular path by magnets and emit bright radiation, from the infrared to the hard x ray, that shines down beam lines to experiment end stations. The x-ray light produced is one billion times brighter than that from the Sun.

The process begins by heating a cathode to more than 1,000°C. Electrons emitted from the cathode are accelerated first in a linear accelerator and then in a booster synchrotron. When the electrons reach the target speed, they are injected into a storage ring about the size of a football field. This ring uses a powerful electromagnetic field to focus the electrons into a narrow beam. During its orbit in the vacuum chamber, the electron beam is bent on a circular path. As the electrons circle the ring, they give off light that is called synchrotron radiation.

Synchrotron x rays have unique properties that make them ideal for dynamic materials science research, which involves fast time scales, microscopic length scales, and high collimation. Synchrotron sources produce more photons per pulse and brighter light than other x-ray sources. The light generated is a hundred million times brighter than that from the most powerful conventional x-ray tube.

An even brighter fourth-generation light source, the Linac Coherent Light Source, is under construction at the Stanford Linear Accelerator Center. Once it is operational, this x-ray free-electron laser will produce light that is hundreds to thousands of times brighter and shorter than the light from existing synchrotron-based x-ray sources. Single pulses will have enough photons to image diffraction patterns with femtosecond (10^{-15}-second) time resolution, providing an unmatched capability for investigating the material dynamics of shocked solids.

A multidisciplinary team of scientists drawn from several Livermore directorates is working together to better understand shock-compressed materials. Team members include (standing, from left to right) Daniel Kalantar, George Gilmer, Anthony Van Buuren, Jaime Marian, Bassem El-Dasher, James McNaney, Bruce Remington, and Santiago Casado and (sitting, from left to right) James Hawreliak, Babak Sadigh, Ben Torralva, Hector Lorenzana (team leader), and Eduardo Bringa.

position Livermore at the forefront of high-energy-density materials science."

The successes from the shocked iron DXRD experiments have gone a long way in demonstrating a new ability to measure the lattice under shock conditions in previously unexplored regimes. This work, along with the real-time observation of phase transformations in situ, is breaking new ground beyond materials science and applications to nuclear weapons research. "The possibilities in these ultrafast, micro regimes are nearly limitless," says Nelson. "Potential applications range from the genesis and characterization of new materials to probing any ultrafast process that occurs in nature, such as biochemical processes that underlie human disease."

In this case, accessing the fast micro regime is likely to mean huge scientific returns for a long time to come.

—Maurina S. Sherman

Key Words: crystalline lattice, dynamic x-ray diffraction (DXRD), materials science, phase transformations, shocked solids, ultrafast lattice response, x-ray scattering.

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