Materials Scientists Discover the Power of ParaDiS

Livermore researchers have developed a code to better understand dislocation lines and how materials gain strength.

SCIENTISTS know that the strength of most crystalline materials, including metals, derives from the motion, multiplication, and interaction of defects called dislocation lines. Dislocation dynamics—the interaction among dislocation lines—is believed to be responsible for strain hardening, a property of metals in which a material's strength increases as deformation increases. For example, the more one twists and bends a paper clip, the stronger it becomes due to the formation and interaction of dislocation lines.

However, scientists lack a detailed understanding of the mechanisms that confer strength, based on changes to a metal's crystalline microstructure. This knowledge is vital to predicting a metal's performance under extreme conditions, such as in the detonation of a nuclear weapon system. Achieving this knowledge requires obtaining a quantitative understanding of how plastic deformation arises from the dynamics of dislocations. This understanding will help in constructing a comprehensive numerical model that will provide accurate predictions of how materials respond under extreme conditions.

Livermore scientists are providing new insight on dislocation dynamics and how materials deform and fail by combining advances in supercomputing and materials experiments and characterization. The research, funded by the National Nuclear Security Administration's Advanced Simulation and Computing (ASC) Program and Livermore's Laboratory Directed Research and Development (LDRD) Program, involves materials scientists, chemists, physicists, computer scientists, and engineers. Their focus is on body-centered-cubic (bcc) metals, such as molybdenum and tantalum, because these metals are similar to some materials in the nuclear stockpile. In bcc crystals, an atom at the center of a hypothetical cube is surrounded by identical atoms at each corner of the cube. Other types of metals have different repeating arrangements of atoms.

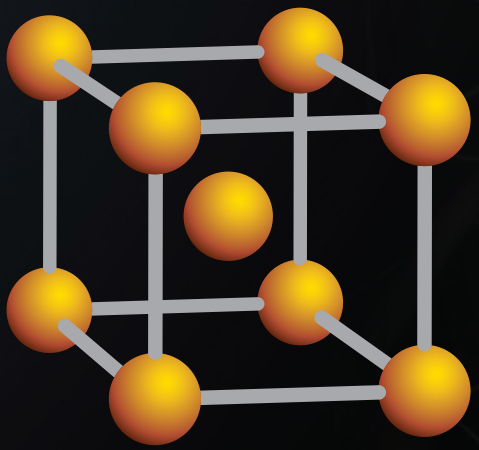
Materials scientist Vasily Bulatov of the Chemistry and Materials Science (C&MS) Directorate has been leading the effort to develop a code, called Parallel Dislocation Simulator (ParaDiS), that models in unprecedented detail the mechanisms of dislocation motion. Bulatov's team includes Gregg Hommes from the Computation Directorate, Moon Rhee from the Engineering Directorate, Meijie Tang from the Physics and Advanced Technologies Directorate, and Masato Hiratani and Tom Arsenlis from C&MS. Wei Cai, an assistant professor from Stanford University, is also on the team. Cai received a 2004 Presidential Early Career Award for Scientists and Engineers for work on ParaDiS while serving as an Ernest O. Lawrence Fellow at Livermore.

The team works closely with engineer Jeff Florando, who is the principal investigator on an LDRD project that conducts complementary experiments on well-characterized, high-purity crystals. Slices from the experimental samples are examined for dislocation microstructures with transmission electron microscopes and are further analyzed at the Advanced Light Source at Lawrence Berkeley National Laboratory. The results form a more complete model of the way crystalline materials behave on the microscale. Together with the ParaDiS simulations, the experiments contribute to multiscale modeling, a process that links data from codes operating on different time and length scales. (See the box on p. 6.)

Modeling Dislocation Evolution

ParaDiS models the evolution of dislocation lines into highly complex microstructures in response to an applied stress. Designed to run on massively parallel supercomputers, ParaDiS allows researchers to follow tens of thousands of simulated dislocations in a 10-micrometer-long cube over several seconds. "Before ParaDiS, the extreme computational cost of dislocation dynamics simulations made it unfeasible to simulate the motion and interactions of many dislocation lines over an adequate amount of time," says Bulatov. ParaDiS can simulate hundreds of times more dislocation lines than other codes. Also, previous codes could only simulate 0.1 percent strain (the change in length of a material under stress). At this point, the number of dislocations begins to grow rapidly. ParaDiS can simulate 3 percent and should be able to simulate far greater strain in updated versions.

The code is available to qualified researchers, who are free to modify it. Fifteen research groups worldwide are



Livermore's ParaDiS code has been used to model simulations in body-centered-cubic crystals, in which an atom at the center of a hypothetical cube is surrounded by identical atoms at each corner of the cube.

efficiently on two of the world's most powerful supercomputers: Livermore's Thunder and Blue Gene/L machines. ParaDiS simulations require at least 500 processors. Today, the code runs on 16,000 of BlueGene/L's 100,000 microprocessors.

The code incorporates a new theory of dislocation that is both mathematically rigorous and analytically simple. The theory is an extension of classical dislocation theory, which provides useful solutions for the behavior of dislocations associated with stress but makes calculations about individual dislocations difficult. ParaDiS removes these

Understanding Materials at Different Length and Time Scales

Materials are at the heart of most issues associated with stockpile stewardship, the National Nuclear Security Administration's program to ensure the safety and reliability of the nation's nuclear weapons. In particular, scientists want to improve their ability to predict the effects of aging on weapon parts or the likely performance of remanufactured parts. Material properties are inherently multiscale, depending on phenomena that occur at all length and time scales. These scales range from a fraction of a nanometer to meters and from nanoseconds to decades.

Multiscale modeling involves a wide array of disciplines and specialists at Livermore from the Engineering, Chemistry and Materials Science, Defense and Nuclear Technologies (DNT), Physics and Advanced Technologies, Energy and Environment, and Computation directorates. Research teams model dislocations at atomistic (nanometers), microscopic (micrometers), and meso (millimeters and above) scales. In multiscale modeling, data are passed from the smallest scale to the next scale up.

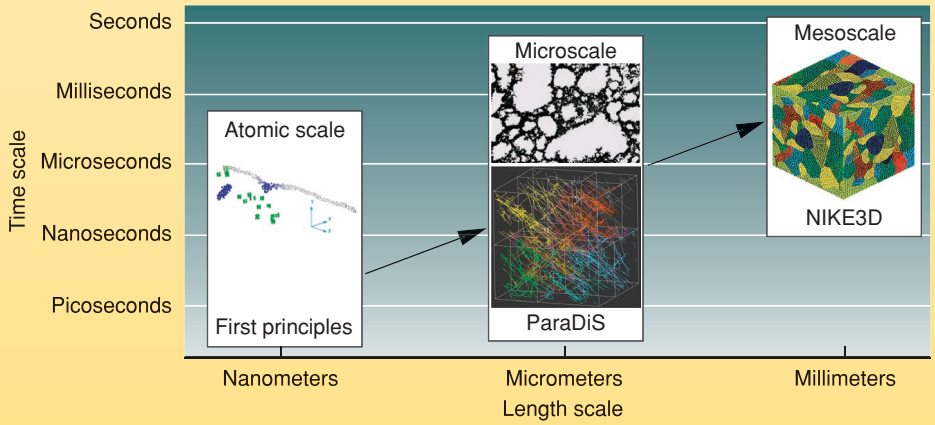
Dislocation activities (the formation, movement, and interaction of dislocation lines of misaligned atoms) occur across a range of length and time scales that must be accounted for to describe crystal plasticity accurately. Although single dislocations and reactions between a few dislocations are controlled by mechanisms at the atomistic scale, dislocations also spontaneously organize themselves into highly complex microstructures over distances of micrometers or longer during plastic deformation. The massively parallel supercomputing resources at Livermore allow researchers to connect all three scales.

Researchers begin with fundamental atomic properties to develop atomistic simulations involving many thousands of atoms. These simulation codes show the movement and interaction of individual dislocations in a perfect crystal under an applied stress.

At the microscale, scientists analyze the motion, multiplication, and interaction of dislocations, a phenomenon called dislocation dynamics. The simulations show the movement and interaction of millions of dislocations and how materials deform plastically—that is, how they change shape without breaking. The most advanced microscale code for dislocations is ParaDiS, developed by a team of Livermore researchers led by materials scientist Vasily Bulatov. By simulating thousands of dislocations and their intersecting behavior, ParaDiS bridges fundamental understanding at the atomic scale and the mesoscale.

In mesoscale modeling, individual crystals and their boundaries are resolved, but individual dislocations are not. At this scale, finite-element simulation codes such as NIKE3D examine how a system composed of multiple single crystals deforms in response to an applied stress.

When complete, comprehensive models of crystalline metals will help stockpile stewardship researchers predict with greater confidence the performance of nuclear weapons and the changes that might occur in the stockpile. In particular, multiscale modeling is important for DNT's metal dynamics research, the National Ignition Facility's target dynamics effort, and the Department of Energy's armor and antiarmor research. Advances in multiscale modeling of crystalline metals also provide data about material behavior that is of interest to industrial manufacturers.



In multiscale modeling, data are passed from the atomistic scale, to the microscopic scale, and on to the mesoscale. Different time scales are also associated with each modeling code.

calculational obstacles to allow simulations of individual dislocation lines.

For the first time, researchers can see how dislocations spontaneously organize into highly complex microstructures over distances of micrometers or greater during plastic deformation. "In a ParaDiS simulation, dislocation lines behave realistically under an applied stress," says Bulatov. "The dislocation lines multiply and move the same as they do in a physical sample. We just sit back and watch the lines advance and obtain a wealth of information."

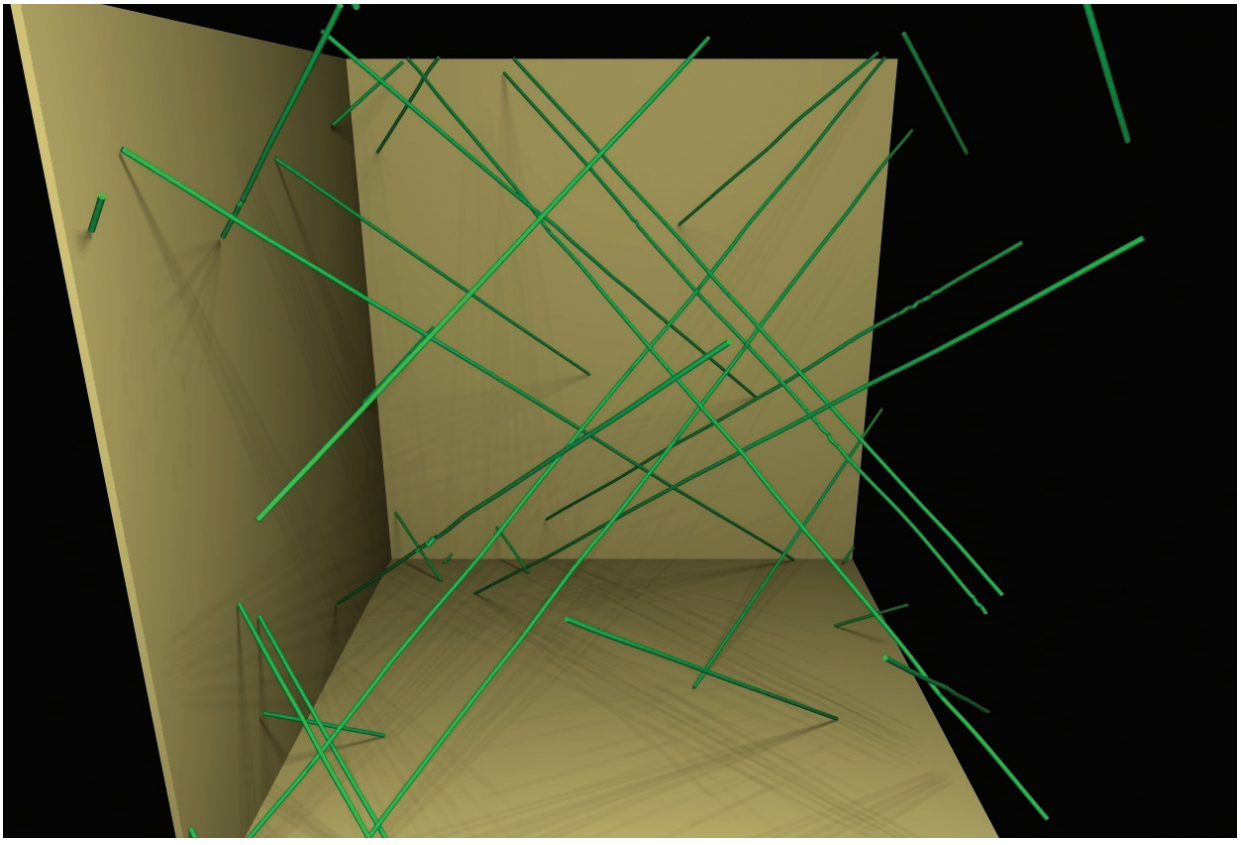
Code's Major Discovery

The power of ParaDiS was recently demonstrated when large-scale simulations revealed a new type of dislocation microstructure, one that current theory did not predict. The simulations showed that collisions among three or more dislocations result in the formation of distinctive elements previously unrecognized, called multijunctions and multinodes. "Multijunctions tie together three or more dislocation lines into a very tight knot," says Bulatov. Multinodes are located at each end of a multijunction. (See the figure on p. 8.)

Bulatov explains that during strain, two colliding dislocations may partially merge, or zip, into a junction bounded by two nodes. Transmission electron micrographs indicated that these binary junctions tie separate dislocation lines, and this process seems to confer strength in metals. The ParaDiS simulations showed that as an applied stress gradually rises, many binary junctions previously formed at a lower stress unzip. However, the multijunctions, being much stronger, endure, and new multijunctions appear. The stress required to unzip a multijunction is about four times that for a binary junction.

Multijunctions present nearly indestructible obstacles to dislocation motion and furnish new sources for dislocation multiplication. In this way, they play an essential role in the strength evolution of deforming crystalline metals. The rate at which bcc metals harden during plastic straining is defined, to a large extent, by the presence or absence of multijunctions.

Multijunctions may account for phenomena unexplained by the widely accepted theory of strain hardening caused by pair-wise dislocation interactions. In particular, multijunctions are the reason for the large, directional variation of strength observed in bcc single crystals. Indeed, multijunctions form much more rapidly when stress is applied to a crystal at certain orientations. Bulatov explains that bcc crystals exhibit various degrees of symmetry depending on the vantage point



In this ParaDiS simulation, molybdenum atoms form dislocation lines that multiply and move in response to applied stress.

of the viewer. The orientation called [001] exhibits the greatest amount of symmetry and the greatest number of multijunctions. When stress is applied along other crystal orientations with less symmetry, few if any multijunctions appear. The greater the number of multijunctions formed, the greater the amount of stress required to deform a metal (strain) before it breaks, and the harder the resulting metal. The figure on p. 9 shows this relationship using a stress–strain curve.

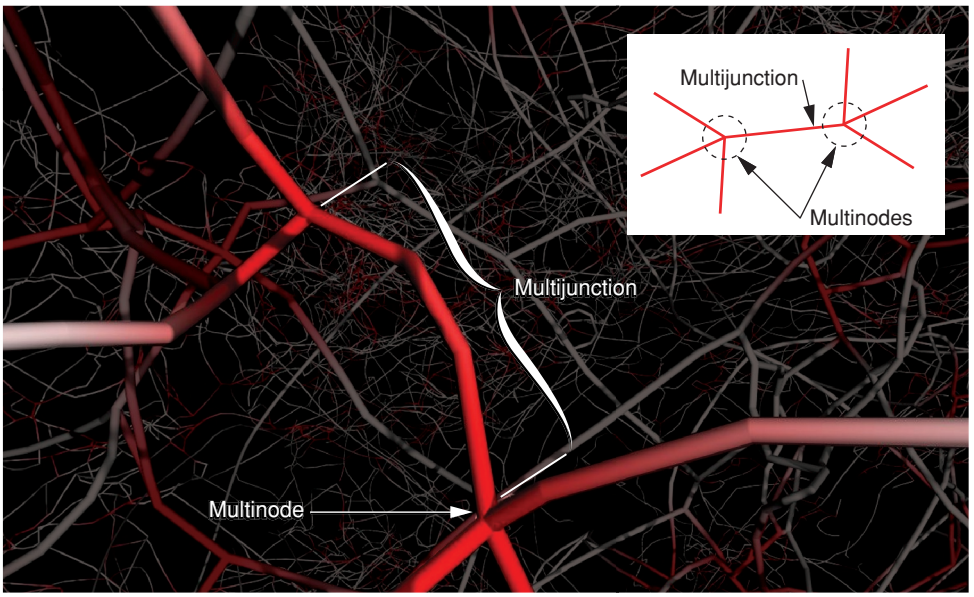
Experiments Verify Simulations

Bulatov's team was able to duplicate the surprising ParaDiS findings using atomistic calculations. These simulations created a cubic block of a perfect bcc single crystal measuring 17 nanometers on each side. The simulations showed discrete dislocation lines forming multijunctions.

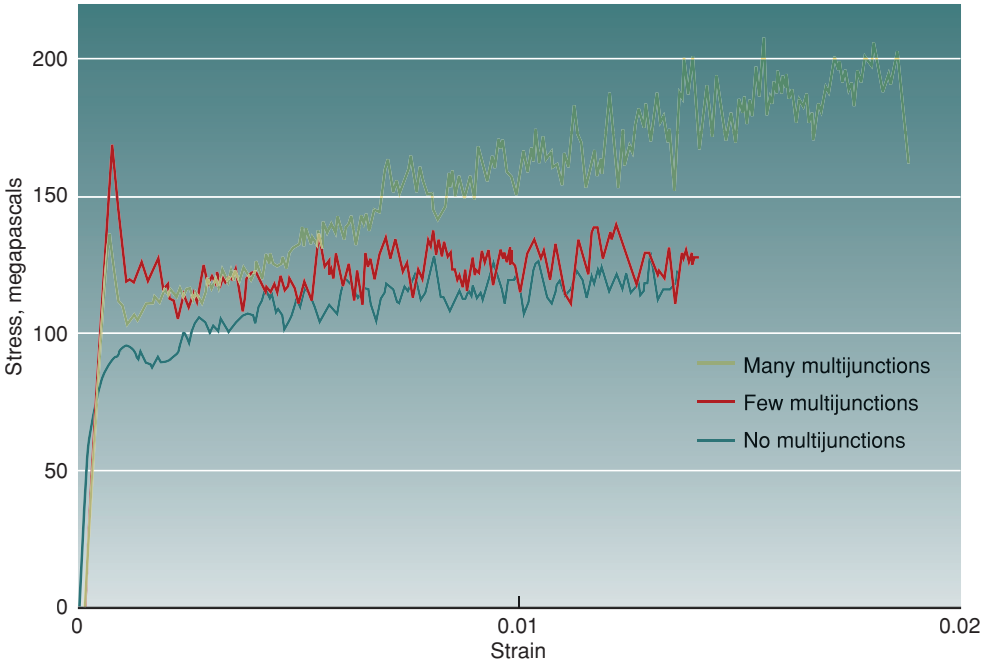
Taken together, the ParaDiS and atomistic simulations appear convincing. However, for verification, Bulatov looked to laboratory experiments and transmission electron micrographs of single-crystal molybdenum samples. In this effort, ParaDiS, like other Livermore codes, is linked closely both to theory and to data obtained from laboratory experiments. Bulatov worked with Florando and Mary LeBlanc, also of Engineering.

The engineers are conducting research on crystalline metals as part of an LDRD-funded initiative. The initiative provides large-strain data to validate ParaDiS simulations and to develop crystal-plasticity models, all as part of Livermore's multiscale modeling program on bcc metals.

The first definitive experimental work on crystals was done in the 1920s, when researchers began to understand atomic structure, crystal lattices, and the response of metals to deformation. This work was primarily one-dimensional (1D); today's research at Livermore is primarily 3D and provides the underlying physics to understand dislocation dynamics in more accurate



ParaDiS shows the eventual formation of a multijunction, which ties together three or more dislocation lines into tight knots. Multinodes are located at each end of a multijunction (see inset). Red indicates dislocations involved in multijunctions, whereas gray represents the more common binary junctions.



These stress–strain curves from a simulation of body-centered-cubic molybdenum show that the formation of multijunctions significantly increases the hardening rate (line slope). The yellow line represents stress applied along the [001] orientation, which exhibits the greatest amount of symmetry and, hence, the greatest number of multijunctions. Stress along crystal orientations with less symmetry results in few (red) or no (blue) multijunctions.

detail. Livermore researchers have been conducting experiments for more than 40 years to improve scientific understanding of crystalline solids and the way they deform. The most recent experimental series, funded by LDRD, began in the 1990s to verify advanced codes and was led by David Lassila, a materials scientist in Engineering.

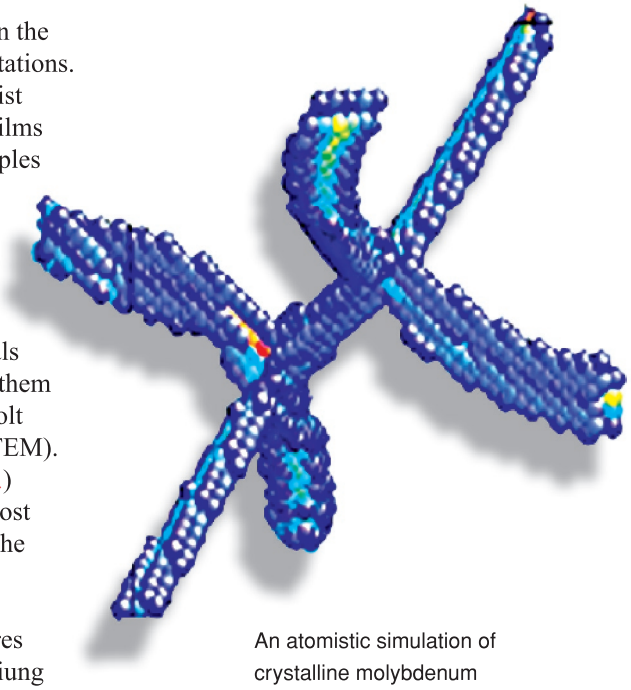
The current experiments focus on the behavior of dislocations in molybdenum and tantalum crystals. A recent series focused on whether the multijunctions seen with ParaDiS were indeed predictable by-products of bcc crystalline metals created in response to stress. The experiments matched the physical parameters simulated by the ParaDiS code.

The team obtained crystals of molybdenum and tantalum that measured about 15 centimeters tall and 1.2 centimeters in diameter. Although almost every material is composed of polycrystals—aggregates of single crystals in different orientations—researchers first need to examine how a single crystal behaves. Technologist Barry Olsen used a vacuum furnace to purify the crystals, because impurities affect a metal's strength and codes cannot yet simulate impurities. Samples of the crystal measuring 5 millimeters wide by 5 millimeters deep by 15 millimeters tall were then carefully cut at different angles using electric-discharge machining and, one by one, subjected to compression. "We needed to know how different orientations of the same crystals responded to the same amount of stress," says Florando.

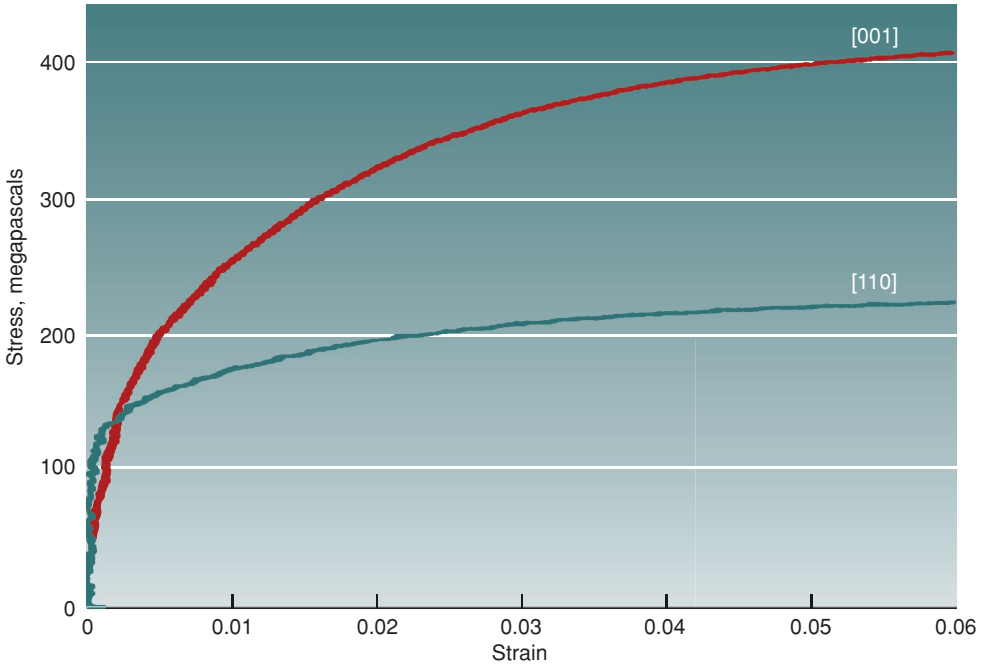
The experimenters compressed the samples to strains of 15 percent. The results showed that when stress was applied to purified crystals of different orientations, the crystals exhibited different strain-hardening behavior. The differences were probably due to multijunctions, which in the ParaDiS simulations formed preferentially in some orientations. For example, much greater stress is required to produce the same amount of strain in the [001] orientation than in other orientations.

After the experiments, technologist Ann Bliss prepared extremely thin films of the deformed crystals. Small samples measuring about 3 millimeters in diameter were cut using a diamond saw and then polished until they were transparent to high-energy electrons for imaging. The thin sections were turned over to materials scientist Luke Hsiung, who imaged them with Livermore's 300-kiloelectronvolt transmission electron microscope (TEM). (See *S&TR,* March 2001, pp. 23–25.) TEMs are special because, unlike most electron microscopes, which probe the surface of materials, they examine a material's internal structure.

Finding specific material structures using a TEM is not an easy task. Hsiung spent two weeks slowly tilting the samples to view them from slightly different angles and thereby distinguish the



An atomistic simulation of crystalline molybdenum atoms shows the formation of multijunctions and multinodes.



This stress–strain curve obtained from experiments on purified crystal samples shows that a [001] crystal orientation, which forms the most multijunctions, requires about double the stress to achieve the same amount of strain as a [110] orientation.

dislocation lines forming the multijunction. Eventually, with the help of electron diffraction experiments, Hsiung found multijunctions that looked strikingly similar to the structures seen in ParaDiS simulations.

Samples of the deformed crystals are also being analyzed at the Advanced Light Source (ALS). Lassila and Florando are collaborating with University of California, Berkeley professor William Morris, Jr., and graduate student Karen Magid, who are using the ALS for a 3D study of dislocation structures. The ALS data can provide maps of the dislocation structure that are several hundred micrometers square, a much greater scale than the TEM observations.

Simulation Is a Full Partner

Bulatov notes it is quite unusual for a code to predict a phenomenon in a material that is later verified by experiment. "ParaDiS predicted something that no one before had seen, or at least recognized." Although dislocation theory is more than seven decades old, previously no one had discovered the connection between multijunctions and metal hardening. "We're sure other investigators have seen multijunctions in electron microscopes, but they didn't understand what they were seeing," says Bulatov.

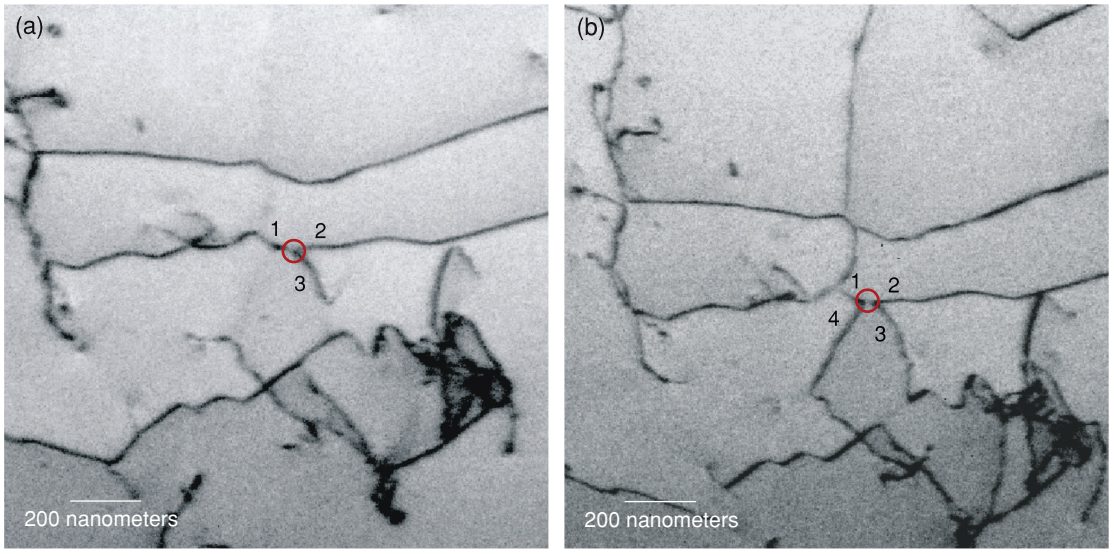
ParaDiS underscores the role of simulation as a full partner with theory and experiment. Bulatov notes that some scientists are skeptical about a code's ability to predict new phenomena. "We watch the simulations with no preconceived notions; we just observe and let the model teach us about the structure of crystals. Others often use codes to confirm what they intuit."

Not surprisingly, the Livermore team is planning additional ParaDiS simulations of bcc single crystals. Bulatov anticipates that by 2006, deformation simulations with 10-percent strain will become standard using BlueGene/L. The result will be greater understanding of multijunctions and multinodes. More complex multijunctions and multinodes appear to exist; some ParaDiS simulations have produced multinodes with five, six, and more arms, and TEM observations have located six-armed nodes.

Once again, experiments on different orientations of purified crystals will be compared to the simulations, as part of the continuing interaction between simulation and experiment. In addition, Bulatov predicts the existence of multinodes and multijunctions in face-centered-cubic crystals (a different form of crystalline organization) and believes that confirmation of their existence by simulation, experiment, and TEM observations will validate the predictions.

Multijunctions represent a potentially outstanding hardening mechanism. If a method to create multijunctions efficiently were developed, it could be important to industry as a new mechanism to harden metals.

According to Florando, new experimental techniques are needed to accurately measure the deformation behavior of single crystals subjected to



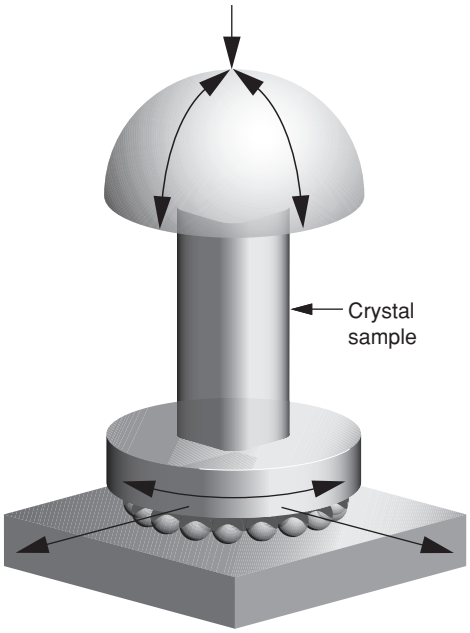
These images of a molybdenum crystal sample were taken with a transmission electron microscope at different viewing angles. (a) Three dislocation lines appear to form a multijunction (circled), but when the crystal sample is examined with a slightly different tilt, (b) an additional fourth line can be seen.

large extents of strain. In response, the experimental team is performing six-degrees-of-freedom (6DOF) experiments, unique to Livermore, which provide data on single crystals as they advance from elastic (reversible) deformation to plastic (nonreversible) deformation, the point when dislocations start to move. The experiments allow the crystals to deform essentially unconstrained in six directions because the bottom of the sample can move as the sample is being compressed. This unconstrained motion prevents the internal crystal planes from rotating during deformation.

The new experimental setup includes a 3D imaging method to measure strains with high-resolution video cameras taking images at 20 frames per second. So-called full-field optical techniques are becoming popular as a method to measure deformation, especially in materials that exhibit nonuniform behavior. The imaging technique measures the deformation that occurs in single crystals better than conventional strain gauges. ParaDiS will eventually include the data derived from the 6DOF experiments.

"We're helping researchers understand simulations. In turn, simulations are helping us understand our experiments," says Lassila. "It's the synergy between simulation and experiment that allows progress to be made." For Bulatov, the rapid acceptance of ParaDiS in the materials science community shows that researchers are well on their way to unraveling the mysteries of how metals harden.

—Arnie Heller



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Key Words: Advanced Light Source (ALS), Advanced Simulation and Computing (ASC) Program, BlueGene/L, body-centered-cubic (bcc) metals, dislocation dynamics, multijunction, multinode, Parallel Dislocation Simulator (ParaDiS), six degrees of freedom (6DOF), stockpile stewardship, Thunder, transmission electron microscope (TEM).

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