Spring 2018

# Mesoscale Connections

Science and technology on the roadmap to MaRIE

INSIDE



Dana Dattelbaum inspects a liquid cell containing an electromagnetic gauge membrane for use in shock initiation experiments on liquid explosives.

## Dana Dattelbaum

### *A dynamic proponent for light source–focused mesoscale materials research*

*By H. Kris Fronzak, ADEPS Communications*

*"... by getting more details at the mesoscale, we could construct more predictive, physics-based models that improve predictability in weapons regimes and off-normal conditions."*

Leveraging her background as a prolific experimentalist and principal investigator, Dana Dattelbaum is setting the technical direction of the Dynamic Materials Properties Campaign (C2) at Los Alamos National Laboratory.

C2 is part of an NNSA program that conducts experimental science in support of the nation's Stockpile Stewardship Program, which ensures the safety, security, and reliability of the nuclear stockpile in the absence of weapons testing. As the Lab's C2 program manager, Dattelbaum oversees research to develop enhanced predictive physics-based weapons models. Dattelbaum, who has almost two decades of weapons-relevant experience, intends to hone these efforts by exploring the potential for new x-ray free-electron light sources to illuminate mesoscale materials dynamics.

The mesoscale is the spatial scale where a materials structure strongly influences its macroscopic behaviors and properties—including strength, stability under heat and pressure, compressibility, and durability in use over time. However, the

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*Dattelbaum cont.*

complex nature of a material's microstructure at the mesoscale is difficult to probe with traditional techniques. Even more difficult is the ability to observe changes over time when materials are subjected to extreme, dynamic forces. New, high-brilliance x-ray sources, like those at the Linac Coherent Light Source at SLAC National Accelerator Laboratory and the Dynamic Compression Sector at the Advanced Photon Source have already provided mesoscale discoveries that challenge many model assumptions when combined with improved modeling and exascale computing capabilities. MaRIE, the Lab's proposed Matter-Radiation Interactions in Extremes concept, will provide future mesoscale discoveries. "Many of our nuclear weapons models are calibrated to data at the bulk or continuum-scale," Dattelbaum said. "But by getting more details at the mesoscale, we could construct more predictive, physics-based models that improve predictability in weapons regimes and off-normal conditions."

To deliver such results, Dattelbaum is gathering input from Los Alamos weapons scientists about their data and diagnostic needs. She is organizing seminars at DOE headquarters in Washington, D.C. that showcase C2 research. She engages with members of the scientific community to develop new x-ray techniques, new mesoscale probes, and ways to combine those experimental techniques with models. Dattelbaum is also eager to hear new ideas and to engage people from different technical backgrounds and disciplines in tackling C2 challenges.

**From applied science to influencing scientific directions**

Dattelbaum (Explosive Science and Shock Physics, M-DO) began conducting research as a student in her Maryland high school, which was partnered with a chemical company. She spent her senior year conducting chemical analysis on the catalytic cracking of hydrocarbons for renewable fuels.

That early exposure to chemistry in action, compounded by academic research positions and a PhD in organic chemistry from the University of North Carolina at Chapel Hill, brought Dattelbaum to Los Alamos in 2001 as a Director's Funded Postdoctoral Fellow. A desire to connect to the Laboratory's mission and explore how materials

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## Dana Dattelbaum's favorite experiment

**What:** Measuring carbon clustering dynamics in detonations using x-ray scattering at the Advanced Photon Source.

**Why:** These measurements of carbon segregation and temporal evolution behind a detonation front address physics questions about late-time energy release and explosive product compositions. They were the first of their kind on the explosive PBX 9502 in the United States.

**When:** November 2014-2017

**Where:** Dynamic Compression Sector, Advanced Photon Source, Argonne National Laboratory

**Who:** Erik Watkins, Millie Firestone, Rick Gustavsen, David Podlesak, Brian Jensen, Kirill Velizhanin, Bryan Ringstrand, Rachel Huber (Los Alamos National Laboratory), Trevor Willey, Michael Bagge-Hansen, Lisa Lauderbach, Ralph Hodgin, Tony van Buuren (Lawrence Livermore National Laboratory), Nick Sinclair, Tim Graber, Soenke Seifert, and Tom Gog (Argonne)

**How:** We assembled a multidisciplinary team of experts in detonation physics, high explosives, x-ray scattering, and time-resolved x-ray measurements at third-generation synchrotron light sources.

**The "a-ha" moment:** We could watch the scattering patterns evolve in real time from those of the nascent explosive to those associated with products. We could also tell that different explosives had different particle morphologies in the carbon product mixture. The team also figured out how best to run the experiments for timing and alignment so that we could obtain scattering data at many "snapshots" behind the detonation front. This was the kind of data that was considered a "holy grail" 5-10 years ago. Penetrating x-rays are making these measurements possible today at light sources like the Advanced Photon Source and Linac Coherent Light Source at SLAC.

**Mesoscale Connections**
is published by the Associate Directorate for Experimental Physical Sciences, which is the Laboratory's champion for the Materials for the Future science pillar. The goal of *Mesoscale Connections* is to promote awareness of mesoscale materials research relevant to the NNSA, advances in mesoscale science capabilities at user facilities, and modeling challenges and needs for big data sets in service of materials co-design. For information about mesoscale materials science at Los Alamos, please contact materials@lanl.gov. For information about the publication, contact adeps-comm@lanl.gov.

*The **mesoscale** is the spatial scale that exists between atomic structures and the engineering continuum—critical to controlling macroscopic behaviors and properties.*

MaRIE is an experimental facility concept that could address an NNSA capability gap identified in a 2016 CD-0 for simultaneous characterization of microstructure and response at the mesoscale.

## Theory, modeling, and computation: the linchpin for mesoscale science

*By Turab Lookman,*
*Physics of Condensed Matter and Complex Systems, T-4*

Theory, modeling, and computation (TMC) at Los Alamos National Laboratory is an integral part of mesoscale science, and the Lab's Theoretical Division has a rich history associated with this concept. The concept of mesoscale dates back to the theoretical underpinnings provided by Lev. D. Landau in the late 1930s.

Today we have modeling and computational tools to perform large-scale simulations of many equilibrium properties as we have witnessed enormous progress in our understanding and control of single-crystal or homogeneous systems over the last 50 years. However, the role of defects, heterogeneities, and disorder in materials is becoming increasingly important as they affect mesoscale microstructure and ultimately material performance.

In contrast to quantum theory, which has been relatively well developed for atomistics, new theoretical efforts at the mesoscale are required to capture phenomena, such as nucleation, growth, and coalescence of new phases and voids, that lead to the collective and self-organized behavior seen at this scale.

Similarly, challenges abound for future TMC efforts to understand and model highly nonequilibrium and driven systems due to shocks. Materials driven out of equilibrium and subjected to extreme conditions can experience large fluctuations placing them into an amorphous state resembling neither a solid nor a liquid. The system may undergo fluctuations with highly skewed or nonstationary distributions with long tails reflecting extreme events. Rapid transient behavior may be followed by the development of extremely long-lived metastable responses, making it necessary to quantify how far the system is from equilibrium and how long these metastable states can persist as they will influence material history.

In addition, concomitant progress in computation is aiding our advances in these areas. Petascale ($10^{15}$ FLOPS) computation is becoming more common and future power and

*"From the era of the 1930s that led to fundamental advances in understanding the mesoscale to today's developments in computation and data science, theory, modeling, and computation continues its pivotal role in mesoscale science to solve problems of national interest."*

communication bottlenecks are already putting constraints on the integration of architecture and software in the design of exascale computers ($10^{18}$ FLOPS).

But more importantly, it is changing the way we will solve scientific problems. The paradigm of codesign, bringing various requisite theory, simulation, and experimental expertise together in a feedback loop to seek the optimal solution, is already starting to mature since the creation of codesign centers in the United States. The need to accelerate our multiscale modeling codes and find new materials and processing environments with targeted properties and desired microstructures within prescribed uncertainties is forcing us to construct emerging codesign paradigms. These adaptively integrate materials theory and knowledge, known data from experiments and calculations, and information theoretic and machine learning methods to "learn" to guide the next experiment or large-scale calculation to be performed. The application of these methods to materials problems promises to have similar impact as the development of bio-informatics has had in cancer genomics, drug discovery, and medicine.

From the era of the 1930s that led to fundamental advances in understanding the mesoscale to today's developments in computation and data science, TMC continues its pivotal role in mesoscale science to solve problems of national interest.

*Turab Lookman is an expert in the computational physics of materials, complex fluids, and nonlinear dynamics and a Los Alamos National Laboratory Fellow.*

*Dattelbaum cont.*

perform in extreme environments led her to become a principal investigator, project lead, and team leader on several static high pressure and shock physics projects for national security sponsors.

She is the recipient of a Los Alamos Fellows Prize for Outstanding Leadership and nearly a dozen NNSA Defense Programs Awards of Excellence. She is an American Physical Society Fellow, nominated by the Shock Compression of Condensed Matter Topical Group—a recognition even more noteworthy considering her degree is not in shock physics, or physics in general.

Dattelbaum researches shock and detonation physics in her limited spare time, and her favorite experiment—which made the first measurement of its kind in the United States—is detailed on the previous page. Her scientific expertise and role as C2 program manager allow her to advance understanding of dynamic material properties and enrich the Laboratory's stockpile stewardship mission for years to come.

# Momentum projects catalyze Lab capabilities for evolving mission needs

*LDRD competition nets many strong proposals, six selected for funding*

Laboratory Directed Research and Development (LDRD), a program that builds Lab capabilities for future mission challenges, is funding six research projects through its FY18 reserve Momentum proposal call.

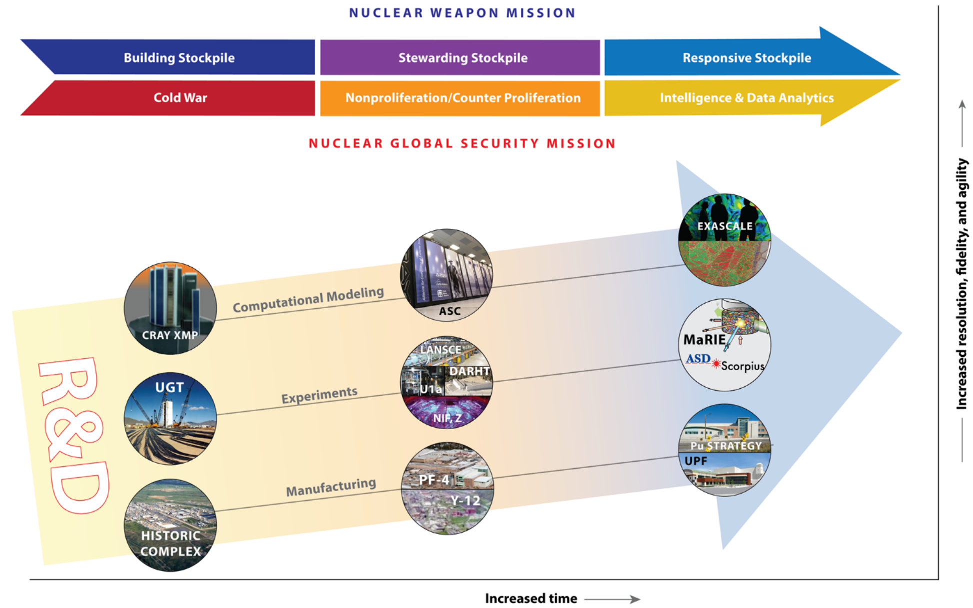
The call aimed to enable work in two essential capability thrusts foundational to the Lab's future:

- exploration of mesoscale materials phenomena using state-of-the-art light source user facilities, and
- accelerator capability enhancement and development of advanced accelerators.

The LDRD Program invests in high-risk, potentially high-payoff projects at the discretion of the Laboratory Director. To maintain agility in the face of evolving mission needs, it holds a strategic reserve to support emerging needs at any point throughout the fiscal year.

"The Momentum call produced some very exciting ideas that were directly responsive to the two thrusts of the call," said LDRD Deputy Program Director Jeanne Robinson, who issued the call.

The following projects were selected for their technical strength and impact on the strategic goals for the thrust.




**As the nation has moved from building the stockpile to supporting a responsive stockpile, it requires tools, facilities, and expertise with increased resolution, fidelity, and agility. The illustration above provides the strategic context for the Lab's vision for meeting these evolving mission needs. The LDRD Momentum proposal call aimed to enable work essential to capability thrusts foundational to the Lab's future.**

### Accelerated microstructure reconstruction from high-energy x-ray diffraction data collected at light sources

NNSA missions rely heavily on the use of advanced light sources for material science studies. For example, facilities such as the Advanced Photon Source, Cornell High Energy Synchrotron Source, and Linac Coherent Light Source can probe in situ polycrystalline materials at the mesoscale under various thermomechanical conditions and at multiple length and time scales. As a result, experimentalists are collecting large amounts of data at unprecedented rates. This project, led by Reeju Pokharel (Materials Science in Radiation and Dynamics Extremes, MST-8) and Turab Lookman (Physics of Condensed Matter and Complex Systems, T-4), is designing a data analysis framework for accelerating mesoscale microstructure reconstructions from diffraction datasets toward the goal of enabling real-time feedback to drive experiments.

### In situ measurements of strain at x-ray light sources within irradiated microstructures

Understanding how radiation damages materials and ultimately leads to their failure is crucial to many national security challenges including nuclear energy, infrastructure lifetimes, and stockpile stewardship. Richard Sandberg (Center for Integrated Nanotechnologies, MPA-CINT) and Saryu Fensin (MST-8) are creating a technique that will develop the tools necessary to first image at near atomic resolution the effects of radiation damage that will lead to the understanding necessary to design better materials. The results will be a revolutionary step toward filling the gap in understanding micro-scale and mesoscale materials phenomena and will provide a critical advance toward the proposed MaRIE (Matter-Radiation Interactions in Extremes) capability.

### Multi-branch x-ray split and delay

Advancements in x-ray probes coupled with high-repetition laser-driven shock waves have enabled the condensed matter physics community to begin phase transition kinetics studies for material transformations during dynamic compression. However, the multiple target requirement and dependence on the driver laser shot-to-shot reproducibility are not adequate for high-resolution kinetics studies. At the Matter in Extreme Conditions endstation at the

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*Momentum cont.*

Linac Coherent Light Source, researchers led by Arianna Gleason Holbrook (Shock and Detonation Physics, M-9) are creating a single-shot/single-target platform that will yield time-resolved data at higher resolution than current methods and therefore gather more accurate kinetics information and stereoscopic imaging, from which improved material models and predictive capabilities will follow.

**Adaptive feedback and machine learning for tuning electron beam phase space**

NNSA missions increasingly rely on particle accelerators, especially free-electron lasers, for conducting research and experiments. Increasing complexity, more stringent beam requirements, and a wider user base of these instruments present extreme challenges in quickly tuning between various user-required setups. Alexander Scheinker (RF Engineering, AOT-RFE) and collaborators at SLAC are developing a novel hybrid feedback control algorithm that combines model-independent adaptive feedback with machine learning techniques to enable automated, fast, optical tuning between different user experiments. The algorithms developed will be directly applicable to the Linac Coherent Light Source and are applicable to particle accelerators in general. For example, the algorithms could be modified to optimize the performance of the Los Alamos Neutron Science Center linear accelerator or to benefit MaRIE.

**Enabling physics-based design of next-generation photocathode guns: an integrated first-principle approach**

To meet the requirements of high-brightness electron/x-ray sources to support DOE mission areas and to enable frontier material science, next-generation photocathode guns are needed. Chengkun Huang (Applied Mathematics and Plasma Physics, T-5) and Thomas Kwan (Plasma Theory and Applications, XCP-6) and collaborators aim to develop an end-to-end modeling capability for electron beam extraction from a semiconductor photocathode gun. This capability, when validated, will form the physics basis for future improvement of semiconductor photocathodes and gun designs.

**Hybrid cryogenic accelerators**

Many national security challenges, ranging from stockpile stewardship to border protection and special nuclear material detection, can be addressed with particle accelerators. However, limitations inherent in existing accelerator designs often force the application to adapt to the needs of the accelerator. A team headed by John Lewellen (Accelerators and Electrodynamics, AOT-AE) and Ghanshyam Pilania (MST-8) aims to develop a hybrid ceramic-copper accelerator structure that expands performance tradespace beyond what is possible with existing design approaches. The eventual goal is development of a type of accelerator that fills mission-critical needs in a smaller footprint, at lower cost, and with fewer compromises than possible with existing designs.

## WWG meeting illuminates use of light source data for weapons mesoscale R&D

In a recent gathering at the Laboratory's 607th Weapons Working Group (WWG), participants discussed how x-ray light source data might be used to answer key national security questions about weapons-relevant materials at the mesoscale.

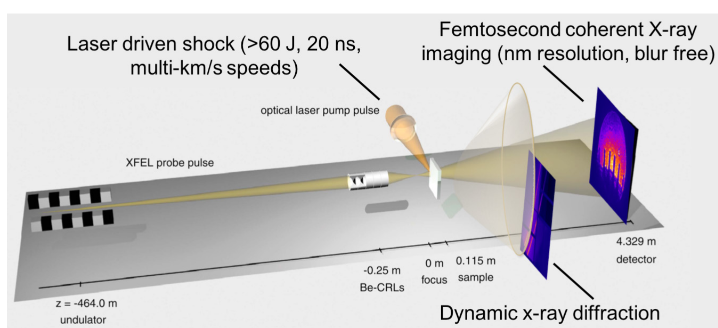
Future predictive capabilities of materials performance under conditions relevant to weapons physics will be increasingly phase and microstructure aware. New high-brilliance x-ray sources offer unprecedented insights into mesoscale materials dynamics, which dominate the continuum-level response, and are challenging many model assumptions.

The WWG session, "Materials R&D at the mesoscale: relevance to primary physics," featured talks by Dana Dattelbaum (Explosive Science and Shock Physics, M-DO), Garry Maskaly (Verification and Analysis, XCP-8), Brian Jensen (Shock and Detonation Physics, M-9), and Richard Sandberg (Center for Integrated Nanotechnologies, MPA-CINT) reviewing several materials-physics uncertainties and discussing data needs for improving predictive capabilities.

Examples of recent Los Alamos experiments at high-brilliance x-ray light sources were presented as well as future developments that can be applied to mesoscale materials dynamics. The topics were framed in the context of the LANL Mesoscale Materials Initiative, which seeks to provide better understanding of materials at the spatial dimension bridging the nano- and macroscopic that is essential to controlling their performance.

WWG meetings provide an interactive setting where experimental plans and results can be shared with a diverse, knowledgeable audience. The goal is to inform the weapons community and capture feedback from people with different technical backgrounds and viewpoints.

*Technical contacts: Richard Sandberg or Dana Dattelbaum*




**An illustration of the power of an x-ray free-electron laser to reveal details in materials driven by a high-speed shock as compared to the traditional method of dynamic x-ray diffraction.**

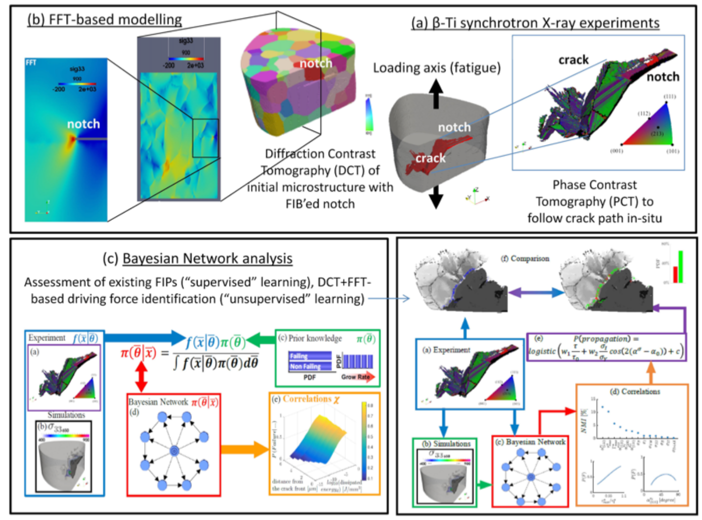
## Toward improving fatigue life predictions

In a recently published paper in the *Journal of the Mechanics and Physics and Solids* (JMPS), a team of researchers from Purdue University, Ecole de Mines de Paris, Los Alamos National Laboratory, and the European Synchrotron Research Facility (ESRF) reported a combination of three-dimensional (3D) in situ synchrotron x-ray experiments using a technique known as diffraction contrast tomography (DCT), LANL's fast Fourier transform (FFT)-based micromechanical modeling, and Bayesian network (BN) analysis. BN enabled machine learning from a fusion of experimental DCT and simulated FFT-based data of a new material-specific indicator to predict the propagation path and growth-rate of a fatigue crack in a beta-Ti alloy.

Microstructurally small crack propagation accounts for most of the fatigue life of engineering structures subject to high-cycle fatigue. Determining the crack path and growth rate of small cracks propagating in engineering alloys is a critical step toward improving fatigue life predictions, thus lowering cost and increasing safety. Cycle-by-cycle measurements of a small crack propagating in a beta metastable Ti alloy using phase contrast tomography (PCT) and DCT measured at ESRF (Figure 1a) were used as input to LANL's micromechanical FFT-based simulations (Figure 1b) to supplement the experimental data by including the micromechanical fields ahead of the crack tip. It was first demonstrated using supervised BN analysis (Figure 1c) that existing fatigue indicator parameters, proposed in the literature for other materials and loading conditions, were not predictive (correct less than 50% of the time). Next, unsupervised BN analysis, driven by the PCT/DCT/FFT multimodal dataset, was used for machine learning of a newly postulated crack propagation driving force. The spatial correlation of the identified driving force showed much better agreement (correct 60-80% of the time) with the experimentally determined crack path.

This synergistic combination of state-of-the-art experimental, modeling, and machine-learning capabilities—contributing to the discovery of material-specific correlations between microstructure and fatigue life—illustrates the Laboratory's progress in developing new techniques to model materials at the mesoscale. Future light sources such as MaRIE would enable in situ 3D characterization as materials are processed and/or dynamically deformed and the use of such data to inform and refine predictive models. The highlighted work presents a viable avenue towards this goal.

Laboratory Directed Research and Development funded the work (FY14 LDRD-DR project: "Mesoscale materials science of ductile damage in four dimensions"), which supports the Lab's Energy Security mission area and its Materials for the Future science pillar toward the creation of materials with controlled functionality, a central vision of the Laboratory's materials strategy.




**(a) In situ synchrotron x-ray experiments, used to characterize polycrystalline microstructure (by DCT) and fatigue crack path (by PCT) in β-Ti notched sample. (b) FFT-based polycrystal plasticity model operating directly on DCT images, used to obtain micromechanical fields. c) Supervised Bayesian network (BN) analysis, used to assess existing fatigue indicator parameters (poor correlation), and unsupervised BN analysis, used to learn from multimodal data a new material-specific driving-force for fatigue crack propagation, highly correlated with measured crack path.**

Reference: A. Rovinelli, M.D. Sangid, H. Proudhon, Y. Guilhem, R.A. Lebensohn and W. Ludwig: "Predicting the 3D fatigue crack growth rate of short cracks using multimodal data via Bayesian network: in situ experiments and crystal plasticity simulations," *Journal of the Mechanics and Physics of Solids* **115**, (2018).

*Technical contact: Ricardo Lebensohn*

## Probing mesoscale features to improve predictions of additively manufactured materials

### *Residual stress determination of AM metallic components for validating AM computational models*

Additive manufacturing (AM) of metals brings new possibilities to the production of low-cost industrial applications. One of the main AM processes for the production of metals is the selective laser melting (SLM) process. SLM is based on powder bed fusion technology and uses a high power-density laser to selectively melt and fuse the metallic particles together.

During fabrication, the rapid solidification and large thermal gradient cause the formation of unwanted and, in some cases, detrimental residual stresses. For example, high tensile residual stresses at a surface can cause crack initiation and consequent failure of the structure. Up to now, there has

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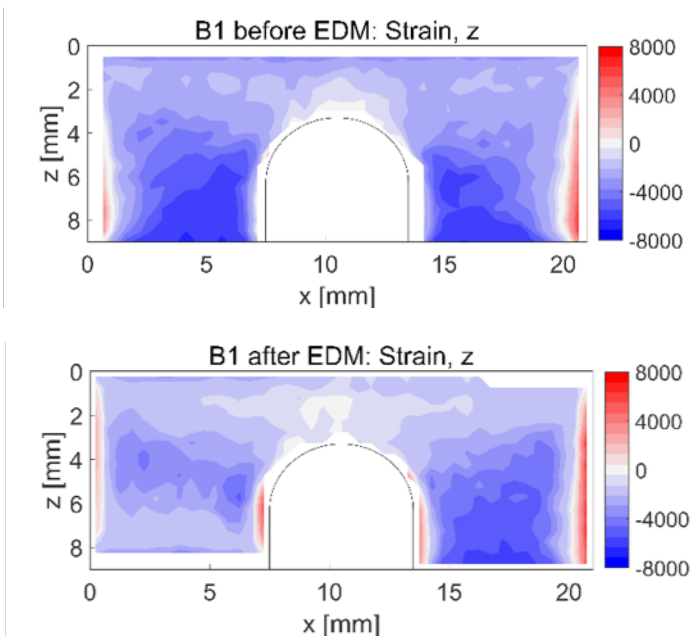
*Residual stress cont.*

been a limited understanding of the process-structure-property-performance relationship of the AM components. This limitation has a critical impact on the usage of the AM metallic components, while it restrains the unique ability of process models to predict the final behavior of the AM structures.

Lawrence Livermore National Laboratory (LLNL) has adapted Diablo, a predictive computational code, for residual stresses in AM structures. In an effort to provide valuable experimental validation data for Diablo, a Los Alamos team (Maria Strantza, Bjorn Clausen, and Don Brown, Materials Science in Radiation and Dynamic Extremes, MST-8) performed a residual stress investigation on AM metallic components in collaboration with researchers from the National Institute of Standards and Technology (NIST).

In this work, Ti-6Al-4V bridge-shaped specimens were built via SLM using four different scan strategies: continuous scan aligned with x-axis, continuous scan at 45° to x-axis, island scan aligned with x-axis, and island scan at 45° to x-axis. A 90° rotation in scan orientation was performed after each layer for all four cases. Energy dispersive diffraction measurements (white beam: 35-150 keV) were performed on all four SLM Ti-6Al-4V components at the A2 instrument of the Cornell High Energy Synchrotron Source (CHESS). X-ray diffraction was used to accurately measure lattice parameter and then determine the elastic strains present in these bridges while still attached to the base plate and after one leg had been cut off the base plate.




**Contour plots of the calculated strain (×10[6]) in z direction, which corresponds to the building direction of the additively manufactured Ti-6Al-4V sample. In (a) the sample is still attached on the baseplate and in (b) the left leg of the "bridge" is detached from the baseplate.**

The figure shows the strain on a cross section of one of the bridge-shaped samples before and after cutting. Relaxation in the cut leg is apparent. In contrast to expectations, the diffraction results showed higher strains present in the bridges built via the island scan strategies, particularly near the edges of the parts. The thermomechanical simulations of these bridge builds exhibited good qualitative agreement with the experimental results. However, the effects of varying scan strategy were not captured with the current modeling technique. Validation of the computational models allows for process optimization as well as establishes limits for off-normal processes. These are necessary for qualification of AM components in high-value, safety-critical applications, such as in the aerospace industry.

High-energy x-ray light sources such as CHESS provide a novel method to probe the mesoscale, the "middle" scale where imperfections, defects, and heterogeneities are critical to controlling a material's macroscopic behaviors and properties. The experiment is an example of science that could be furthered with MaRIE, Los Alamos's proposed Matter-Radiation Interactions in Extremes capability for in situ time-dependent materials science at the mesoscale. MaRIE would allow researchers to observe in real time how the residual stresses develop as the part is built, thus yielding information that could be used to enable design of processes to achieve tailored mechanical properties.

This research is being performed by a Los Alamos team from MST-8 (Brown, Clausen, and Strantza), collaborators from NIST (Lyle Levine and Thien Phan), and collaborators from LLNL (Wayne King, Neil Hodge, and Rishi Ganeriwala). The work was funded by C1 and the Gas Bottle Qualification program and benefited from the use of the A2 beamline at CHESS. The work supports the Lab's Stockpile Stewardship mission area and Materials for the Future science pillar.

*Technical contact: Maria Strantza*

## *In situ diffraction measurements of mesoscale features at unprecedented time scales during metal additive manufacturing*

Additive manufacturing (AM) is an innovative fabrication and repair technology that can build metallic components in a layer-by-layer manner. Currently, there is a limited understanding of the process-structure-property-performance relationship of metal AM materials, which severely limits the ability of process models to predict the final behavior of the components. This is the prime obstacle to widespread adoption of metal AM components in property-critical applications. Insight into the dynamics of AM processing through in situ measurements is imperative for providing the mesoscale information required to develop physics-based predictive models for properties and performance. With these developed relationships in hand, AM can begin to support

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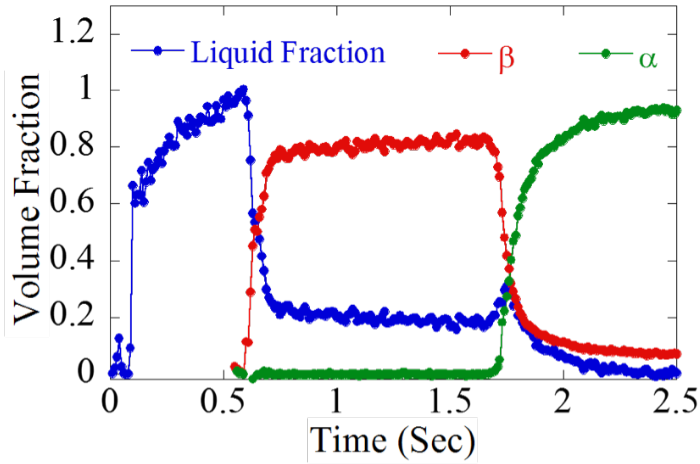


**Phase evolution in point deposited Ti64 as a function of time. Note that the alloy initially solidifies as beta before transformation to the alpha phase. Through the use of the APS, researchers obtained high time resolution data that shows the rate and time of these phase transformations. This mesoscale information forms the critical data for linking process to microstructure in AM. With this linkage in hand, a future can be envisioned where the amount and locality of the beta phase can be controlled in order to tailor the properties of AM Ti64 and impact the Materials for the Future science pillar.**

*In situ diffraction cont.*

Los Alamos's Stockpile Stewardship mission through providing materials with controlled functionality and predictable performance.

The unique capability of the Advanced Photon Source (APS) at Argonne National Laboratory provides the required photon flux and state-of-the-art detector technology to enable in situ AM measurements. Recently, a team of Los Alamos researchers with Lawrence Livermore collaborators performed in situ experiments at APS to capture microstructure evolution information during metallic wire-feed AM process. They successfully obtained phase evolution information during the solidification of Ti-6Al-4V (Ti64) and 304L stainless steel (SS). In the work, metallic wires of SS and Ti64 were deposited onto a base material of SS and Ti64, respectively, using the relatively new cold metal transfer process in an additive configuration.

High-energy x-rays ($E$=71 keV) were used to continuously record diffraction data at 200 Hz with a spot size of 50 µm to capture rapid solidification and microstructure evolution in highly localized areas of the bulk material during the AM process. Capturing data at every 0.005 seconds provided the high temporal resolution at the appropriate length scale (microns) to validate and inform microstructural models and to support LANL's mesoscale strategy. In addition to the high-energy x-rays, high-speed optical images were recorded at frame rates between 2 kHz and 12.5 kHz to complement the diffraction information acquired during the deposition and solidification process.

Two types of experiments were performed while collecting diffraction data and optical imaging data. In the first case, molten beads were deposited in a stationary setup to investigate solidification during additive repair of small surface defects. In the second case, the material was deposited in a linear fashion while collecting data at positions throughout. This experiment simulated AM as a fabrication technology and enabled the researchers to capture the dynamics of the system in the time-critical process of solidification. Coupled diffraction and optical imaging provided both process diagnostic and microstructure evolution information during deposition of material.

The figure shows the phase fraction evolution as a function of time for the Ti64 of the first experimental case (repair of small surface defects). The results indicate that beta phase forms first in Ti64 while at the end of the solidification (2-2.5 sec after the deposition) Ti64 mainly consists of alpha phase. Although the transformation was expected, the rate at which the transformation occurred provided the new, needed information to validate models that predicted microstructure based on process parameters. From a performance perspective, the amount and position of the beta phase has a significant impact on the strength and ductility of the overall component, making predictive capability in this area a priority. Developing these predictive capabilities is consistent with Los Alamos's Materials for the Future science pillar goals.

Although the results of these experiments provide information specific to the two alloys, the long-term impact of this project is largely centered on the methodology of using cutting-edge beamline science to capture mesoscale science at temporal and length scales unreachable in the past. Volume fractions of liquid and solid, including different phases that formed during the solidification process, were captured every 0.005 seconds. Limitations in detector and photon flux did not permit measurements of other microstructural parameters, such as texture and elastic strain, without compromising the temporal resolution.

Being able to capture phase evolution as well as texture and elastic strain with high temporal resolution would further improve the models that link processing to microstructure. In order to achieve this, however, future facilities like MaRIE coupled with improvements in detector technology are necessary. As AM continues to grow, developing the platforms (MaRIE, APS-U) and tools for in situ experiments that can capture the full range of mesoscale data (phase, texture, strain) during solidification in materials that undergo the rapid cooling rates of AM ($10^3$ and $10^6$ K/s) will be important for the ability to insert components into property-critical applications.

This research is being performed by a team from Materials Science in Radiation and Dynamics Extremes (MST-8) (Don Brown, Maria Strantza, Reeju Pokharel and Adrian Losko), Sigma Division (Sigma-DO) (John Carpenter [principal investigator] and Jason Cooley), and Materials Synthe-

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*In situ diffraction cont.*

sis and Integrated Devices (MPA-11) (Erik Watkins), and collaborators from Lawrence Livermore National Laboratory (Ibo Matthews and Nick Calta). The work was funded by a Laboratory Directed Research and Development Exploratory Research Momentum award and benefited from the use of the APS at Argonne National Laboratory, which is funded by the DOE's Office of Basic Energy Sciences.

*Technical contact: John Carpenter*

## X-ray laser provides atomic-level observations of material deformation during high-pressure shock loading

Pressure-driven shock waves in solid materials cause extreme damage and deformation, and understanding that damage is important to phenomena including planetary formation, asteroid impact sites, spacecraft shielding, and ballistic penetrators. However, studies of crystal lattice-level plastic deformation are complicated by the limits of post-shock analysis and modest pressures of less than 100 GPa. In situ x-ray diffraction measurements can help explain a material's dynamic behavior, but have only recently been applied to plasticity during shock compression and until now have not provided detailed insight into competing deformation mechanisms.

Using the Matter in Extreme Conditions (MEC) endstation at the Linac Coherent Light Source (LCLS), researchers performed x-ray diffraction experiments with femtosecond resolution on shocked polycrystalline tantalum and captured, for the first time, in situ lattice-level information that quantifies the contributions of the competing microstructural processes of dislocation slip and twinning during shock-wave-driven deformation. The technique will be useful for studying shock waves and other high-strain-rate phenomena, as well as a broad range of processes induced by plasticity.

The team's recent publication in a *Nature* letter detailed twinning and related lattice rotation that occurred on the timescale of tens of picoseconds. Despite the common attribution of deformation under strong shock to twinning, the data showed that the material's dominant deformation mechanism changed to dislocation-slip mediated plasticity at more than 150 GPa, or about 1.5 million times atmospheric pressure, which is a regime that recovery experiments cannot accurately access.

Another significant breakthrough is that the experiment occurred on timescales similar to those probed by molecular dynamics simulations at high-pressure regimes. This is enabling direct comparison of lattice-level simulation and experimental results on commensurate length- and timescales in an unprecedented manner, further highlighting the transformative insights to materials under extreme conditions provided by in situ measurements by x-ray lasers. Combining ultrafast light pulses with atomic resolution, next-generation light sources such as the LCLS allows researchers to explore the mesoscale, the "middle" scale where imperfections, defects, and heterogeneities are critical to controlling macroscopic behaviors and properties.

The experiment is an example of science that could be further advanced with MaRIE, LANL's proposed Matter-Radiation Interactions in Extremes capability for time-dependent materials science at the mesoscale. Two of the main aspirations for MaRIE are to provide multiple x-ray measurements per experiment to record the time evolution of deformation processes and to simultaneously bring to bear other particle probes, such as electrons or protons, that sample very different material volumes.

The work was funded by the Department of Energy: its Office of Basic Energy Sciences supports the LCLS at the SLAC National Accelerator Laboratory, while the Office of Fusion Energy Sciences supports the MEC instrument. The research supports the Laboratory's Nuclear Deterrence mission area and its Nuclear and Particle Futures science pillar. Los Alamos helped perform the experiments at the LCLS MEC. The team intends to use these experimental techniques in conjunction with work at Lawrence Livermore National Laboratory's National Ignition Facility to study such phenomena at higher pressures.

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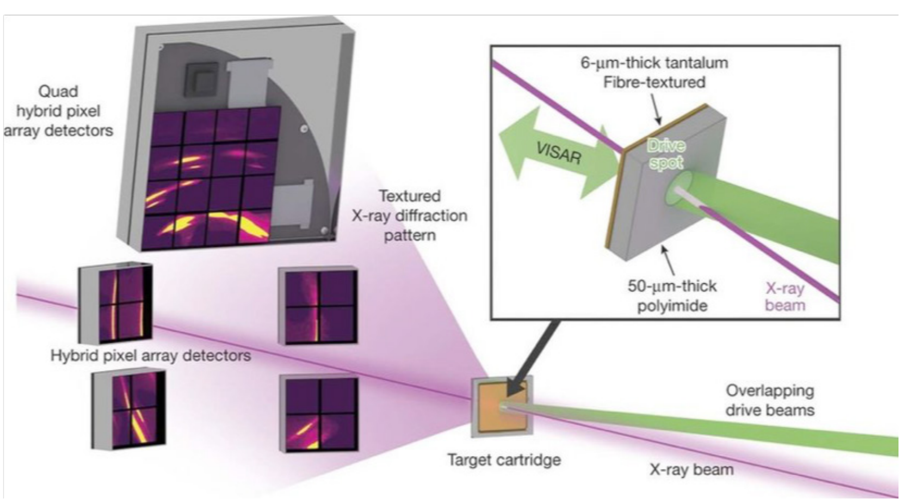



**Laser ablation (green beams) drives a shock wave into the target. The shock-compressed tantalum is probed by the x-ray beam (purple), and the resulting textured diffraction patterns are collected on hybrid pixel array detectors.**

*X-ray laser cont.*
Researchers: C. E. Wehrenberg, A. Lazicki, H.S. Park, B. A. Remington, R. E. Rudd, D. Swift, and L. Zepeda-Ruiz (Lawrence Livermore National Laboratory); D. McGonegle, M. Sliwa, M. Suggit, and J. S. Wark (University of Oxford, UK); Cindy Bolme (Shock and Detonation Physics, M-9); A. Higginbotham (University of York); and H. J. Lee, B. Nagler, and F. Tavella (SLAC). Reference: "In situ x-ray diffraction measurement of shock-wave-driven twinning and lattice dynamics," *Nature* **550**, 496-499 (2017).

*Technical contact: Cindy Bolme*

## State-of-the-art photocathodes grown on atomically thin layers of graphene

*Configuration a promising step in quest for transformational advances needed for new light sources*

Many grand challenges, including sustainable energy, scaling of computational power, detecting and mitigating pathogens, and studying the building blocks of life, require better access and control of matter on the timescale of electronic motion and the spatial scale of atomic bonds.

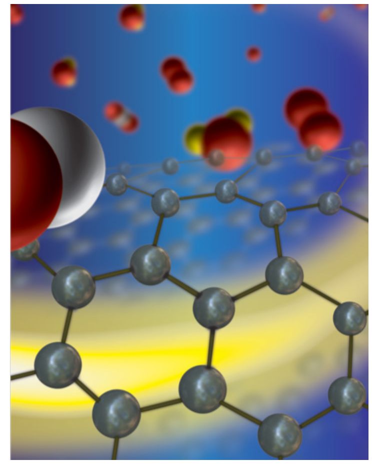
Such insight can only be gleaned with future coherent x-ray sources and advanced colliders, which demand increasingly high-performance electron beams far outstripping present state-of-the-art technologies. DOE-commissioned studies have repeatedly pointed to electron sources as one of the highest accelerator R&D priorities for the next decade, requiring a transformational advance in cold cathode performance in particular.

To that end, Los Alamos researchers and their collaborators created a unique approach that decouples two competing physical mechanisms that have prevented researchers from improving cold cathode efficiency and lifetime. This breakthrough was achieved by integrating atomically thin materials with high-performing existing photocathode technologies for better protection.

The study relied on the bialkali antimonide semiconductor $K_2CsSb$, which has one of the highest quantum efficiencies of any photocathodes, combined with thin layers of graphene, a form of carbon possessing an exceptionally high gas barrier property, ultrahigh electrical and thermal conductivity, optical transparency, high charge mobility, and the ability to sustain extreme current densities.

The research showed significant progress in growing state-of-the-art photocathodes (traditionally grown on thick substrates) on super thin, transparent graphene substrates that have a quantum efficiency comparable to those deposited on rigid substrates. This configuration marks progress toward eventually encapsulating high performance, environmentally susceptible photocathodes using graphene as a passivating barrier. It is a promising step toward fabricating photocathodes with enhanced lifetimes and on optically transparent yet electrically conductive substrates.

**An illustration depicting accelerator technology-relevant photocathodes that were successfully deposited on freestanding atomically thin substrates of graphene. This advances an ultimate goal of enhancing the lifetime of photocathodes without sacrificing the high quantum efficiency by using an atomically thin protection layer.**



Los Alamos capabilities that were critical to the project's success were chemical vapor deposition to synthesize high-quality graphene and nanomaterial processing to transfer graphene onto challenging substrates. The progress provides several advances to MaRIE, the Laboratory's proposed Matter-Radiation Interactions in Extremes capability, including the option to switch from metal cathodes to higher performance semiconductor cathodes that reduce emittance by 50%. This reduces risk by creating "headroom" throughout the design and would decrease system complexity and beam energy for immediate cost savings.

The work was supported by the Laboratory Directed Research and Development Program and partially conducted at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated by Los Alamos and Sandia national laboratories. The research supports the Lab's Energy Security mission area and its Materials for the Future science pillar, including its defects and interfaces and emergent phenomena science themes by pursuing materials with controlled functionality and predictive performance, the central vision of the Laboratory's materials strategy.

Researchers: H. Yamaguchi, F. Liu, C.W. Narvaez Villarrubia, A.D. Mohite (Materials Synthesis and Integrated Devices, MPA-11); J. DeFazio (Photonis USA Pennsylvania Inc.); M. Gaowei, J. Sinsheimer, and J. Smedley (Brookhaven National Laboratory); J. Xie (Argonne National Laboratory); D. Strom (Max Planck Institute for Physics); V. Pavlenko, N.A. Moody (Accelerators and Electrodynamics, AOT-AE); and K.L. Jensen (Naval Research Laboratory). Reference: Free-standing bialkali photocathodes using atomically-thin substrates," *Advanced Materials Interfaces* (2018).

*Technical contact: Nathan Moody*