May 2015
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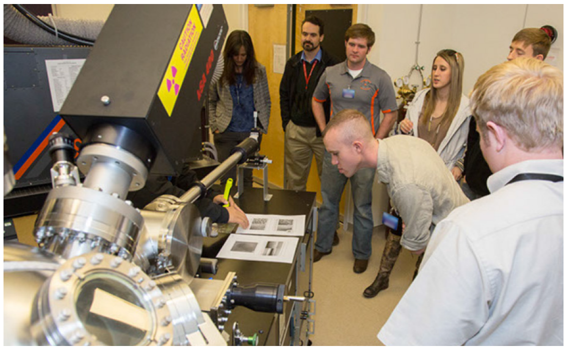
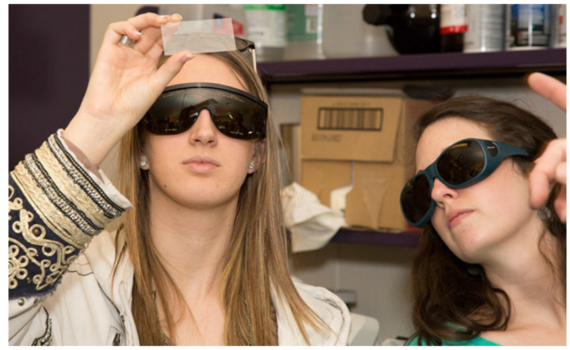
Roadmap to MaRIE

Inside





Photos by Richard Robinson, NIE-CS

Clockwise from top left: SSAP attendees visit CINT's physical vapor deposition lab and view 100-micron-wide microfluidic channels that have been femtosecond laser machined into the surface of a glass slide. At pRad, Matthew Murray (Subatomic Physics, P-25) describes proton radiography and gives a tour of the pRad dome. With a display in the symposium's main hall, the MaRIE project delivered examples of current and future research opportunities available at Los Alamos National Laboratory.

SSAP participants explore Los Alamos national user facilities

Stewardship Science Academic Programs Symposium featured pRad and CINT tours

Designed to foster ties between early career scientists, sponsors, and the National Nuclear Security Administration national laboratories, the 2015 Stewardship Science Academic Programs (SSAP) Symposium was held recently at Buffalo Thunder Resort in Santa Fe, New Mexico.

Presenting highlights of their research were recipients of grants from the Stewardship Science Academic Alliances, the High Energy Density Laboratory Plasmas, National Laser Users' Facility, and Predictive Science Academic Alliance programs.

The symposium also featured a keynote presentation by Scott Doebling (Verification and Analysis, XCP-8) on "Verification & Validation: Uniting Simulations, Theory, and Experiments" and talks by Amy Clarke (Materials Technology, MST-6) on the "Visualization and Control of

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SSAP cont.

Metal Solidification to Achieve Advanced Manufacturing" and Brian Jensen (Shock and Detonation Physics, M-8), and colleagues, on "Dynamic Compression Sector at the Advanced Photon Source: Overview and First Results."

Symposium attendees also had the chance to visit Los Alamos National Laboratory's Proton Radiography Facility (pRad) and the Center for Integrated Nanotechnologies (CINT). There, they met with Laboratory scientists, discussed recent research being performed in these national user facilities, and learned about opportunities for future experiments.

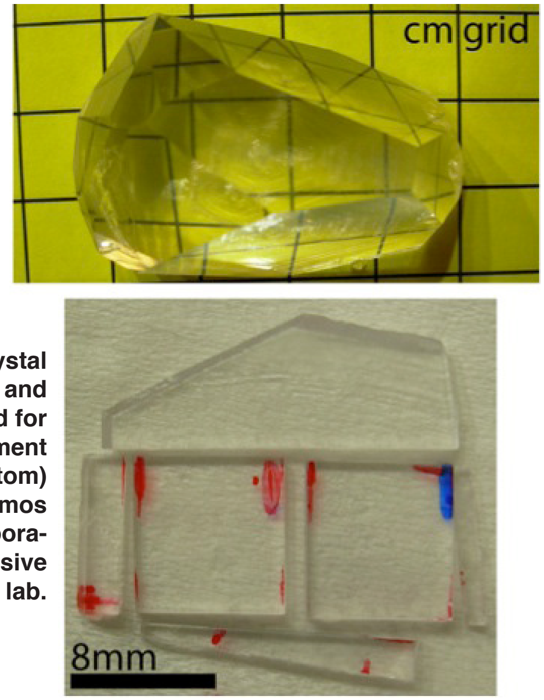
The MaRIE (Matter-Radiation Interactions in Extremes) project sponsored a booth in the symposium's main hall, attracting students and postdoctoral researchers interested in how the proposed Los Alamos facility for time-dependent materials science at the mesoscale will enable next-generation experiments and provide them with the tools needed to support national security missions.

First in situ x-ray diffraction recorded during impact at APS paves way for transformative models of explosives

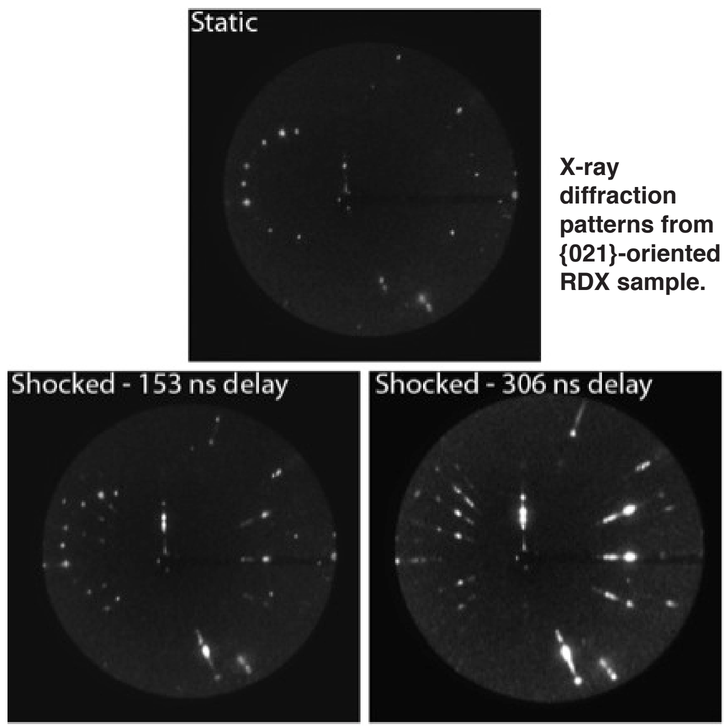
By capturing the first x-ray diffraction patterns of explosives in situ during gas-gun driven impact loading using synchrotron radiation, Los Alamos researchers and collaborators demonstrated a crucial diagnostic for studying how crystalline explosives deform under shock loading. These in situ data are one of the first experimental steps (second-ever x-ray diffraction from a molecular explosive during shock loading) toward developing next-generation, physics-based mesoscale models with predictive capability for high explosives to support associated Science Campaign 2 milestones scheduled through FY2023.

Crystalline deformation in high explosives is thought to control the size, temperature, and possibly also the spatial distribution of "hot spots," which lead to shock-induced reaction and detonation initiation. In this study, the researchers used the synchrotron at Argonne National Laboratory's Advanced Photon Source (APS) to measure x-ray diffraction in situ during shock loading to investigate high-rate crystalline mechanics in single crystal RDX (cyclotrimethylene trinitramine, an explosive).

Next steps include analyzing the x-ray diffraction patterns and comparing them to results from recently developed finite element model simulations of the crystalline deformation during shock loading. The analysis will enable the extraction of previously unattainable equation-of-state information and the development and validation of an anisotropic plasticity model crucial for predicting thermomechanical localization of deformation and associated heating.



RDX single crystal (top) grown and prepared for experiment samples (bottom) at the Los Alamos National Laboratory high explosive crystal lab.



X-ray diffraction patterns from {021}-oriented RDX sample.

Developing capabilities for making, measuring, and modeling materials and demonstrating the extent of mission relevant information that can be extracted from in situ experiments envisioned at the proposed MaRIE (Matter-Radiation Interactions in Extremes) flagship science facility is important and timely for the Laboratory. The Materials In Extremes exploratory research funding for this Laboratory Directed Research and Development (LDRD) project enabled the experimental development of the x-ray diffraction and complementary multi-scale modeling capabilities and benefited from the unique national resource for single

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First cont.

crystal growth at the Los Alamos National Laboratory high explosive crystal lab. The x-ray diffraction capabilities developed using the Laboratory's IMPact system for ULtrafast Synchrotron Experiments (IMPULSE) and applied here to explosives will be used to study other materials, including metals, within the fiscal year.

The research team includes Kyle Ramos, Brian Jensen, Timothy Pierce, Virginia "Tate" Hamilton, Claudine Armenta, Charles Owens (Shock and Detonation Physics, M-9), Marc Cawkwell, John Barber (Physics and Chemistry of Materials, T-1), D.J. Luscher, Frank Addessio (Fluid Dynamics and Solid Mechanics, T-3), with collaborators Adam Iverson (National Security Technologies LLC, NSTEC) and Nick Sinclair and Thomas Gog (Dynamic Compression Sector, APS).

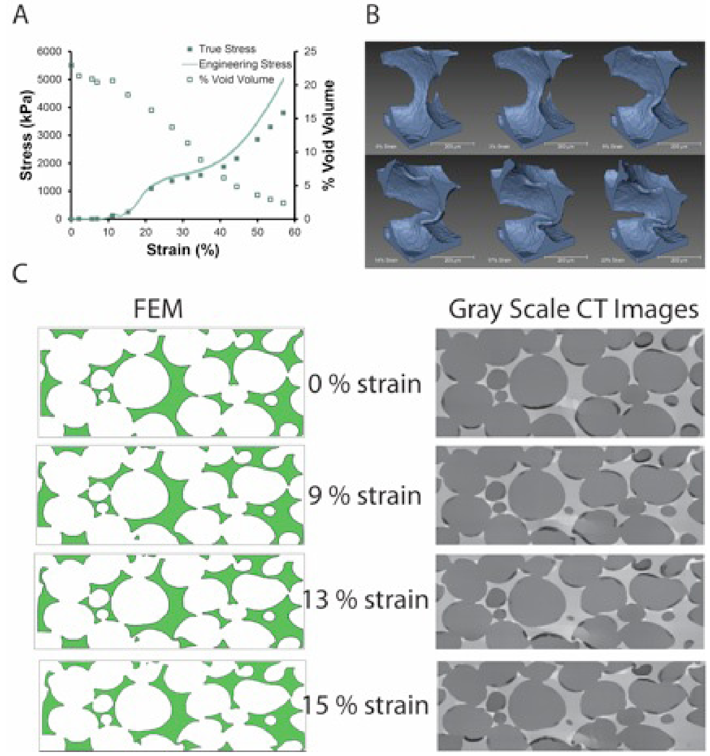
The LDRD program funds the works through the "In situ x-ray imaging and diffraction to understand the mechanics of initiation mechanisms in explosive single crystals," (LDRD, 20140643ER) project. The research supports the Stockpile Stewardship program and Materials for the Future science pillar with emphasis on Los Alamos leadership in extreme environments

Technical contact: Kyle Ramos

In situ synchrotron imaging and modeling of polymer foam deformation

Collaborative research between Los Alamos National Laboratory, Arizona State University, and the Advanced Photon Source (Argonne National Laboratory) has demonstrated the collection of full 3-D tomographic data sets within 1 second and 20 3-D data sets within 100 seconds while continuously compressing soft polymer foams and other cellular materials at a 10^{-2} s^{-1} strain rate. This information can be used in mechanical performance models of these materials.

Methods to characterize the mechanical performance of soft materials must account for their elastic nature. For example, after placing an object on a marshmallow, the flow of marshmallow material continues for several minutes in response to this mechanical load. Engineered soft materials, such as polymer foams, exhibit this same flow during compression performance. Studying the progression of this visco-elastic flow is difficult because the experiment cannot be stopped for data collection. X-ray computed tomography is often used to image this flow because it provides a nondestructive 3-D representation of the real external and internal structure. The challenge in observing this visco-elastic flow is that images must be captured with a high enough frame rate to prevent motion blurring. The team's experiments into the hyper-elastic performance of polymer foams at the Advanced Photon Source demonstrate that it is possible to capture this flow during compression. The data could be used to model the mechanical performance directly.



A) The changes in the percent void volume within the syntactic silicone foam material correlates with the engineering stress (measured from the foam starting diameter) and the true stress, adjusted directly from the cross-sectional area changes seen within the CT images. B) Dynamic changes within single ligaments such as bending, buckling, and tension before breaking are directly observed in real time. C) The direct comparison of the finite element modeling and the gray scale CT images for these first experiments directly capture the robustness of the model. Notice the single ligament bending at the bottom center of the FEM and CT data that is highlighted in B.

The full 3-D data provided by this technique enabled the researchers to demonstrate that changes in the void volume correlate to inflections within the stress-strain response (A). The team directly measured the true stress. The in situ stress-strain curves exhibit identical compressive performance to the acceptance testing at the National Security Campus where these materials are manufactured. Imaging of the bending, buckling, and tension (which leads to fracture) of ligaments (B) can be compared between foam types with this technique. Along with the 3-D structure data, the stress-strain and Poisson's ratio data could be entered directly into the finite element model (FEM). Direct comparisons between the imaged compression and the finite element model would better calibrate and improve the robustness of the model (C).

These experiments are an example of science on the roadmap to MaRIE (Matter-Radiation Interactions In Extremes). They enable researchers to better understand material fail-

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In situ cont.

ure through time-resolved characterization techniques and integrate measuring and modeling. The time resolution in these experiments is such that the team can collect the 3-D images before the material has sufficient time to relax as a result of the applied stress. The Poisson's ratio is dependent upon the strain rate and relaxation time after compression. The ability to reconstruct and process 3-D data while collecting other in situ information, such as mechanical performance, adds several layers of detail for the modeling community that was never possible previously. Integrating these experiments with modeling enables a comparison of the modeling and experimental results and provides a direct fiducial to measure the robustness of the model.

The team includes Brian M. Patterson, Kevin Henderson, and Nikolaus Cordes (Polymers and Coatings, MST-7); Nikhilesh Chawla, Sudhanshu Sing, Angel Ovejero, Jason Williams, and Tyler Stannard (Arizona State University); and Xianghui Xiao (Advanced Photon Source). The NNSA Enhanced Surveillance Campaign (Tom Zocco, LANL Program Manager) and the NNSA Engineering Campaign (Eric Mas, LANL Program Manager) funded the work, which supports the Laboratory's Nuclear Deterrence mission area and the Materials of the Future and Science of Signatures science pillars.

Technical contact: Brian Patterson

Neutron diffraction reveals temperature-dependent crystallographic properties and texture evolution of high explosive powder during compaction

In work relevant to Los Alamos's role as the design agency for extending the reliability and safety of B61 bombs that have exceeded their lifespan, Laboratory researchers have obtained experimental results about the high explosive TATB. By collecting first-ever data on a scale useful to developing constitutive mesoscale models, the researchers will aid in benchmarking and improving modeling efforts that incorporate how TATB responds to temperature fluctuations and compaction. Literature reports on thermal dependence of TATB lattice parameters and crystal structure do not always agree, and in situ measurements of texture evolution during a pressing process have never been made. These data are critical for development of mesoscale models that can inform system-level models, and can be used to interpret and identify underlying mechanisms for other experimental findings (e.g., ratchet growth).

Many explosives detonate when exposed to fire or a shock, and may exhibit partial reaction or take significant damage when a charge falls on the ground. Since TATB (2,4,6-triamino-1,3,5- trinitrobenzene) is difficult to detonate by

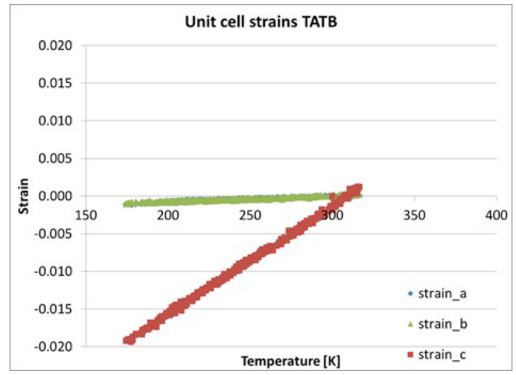

Figure 1: Relative change (strain) of the three crystallographic lattice parameters of TATB crystals as a function of temperature measured on the HIPPO neutron diffractometer.

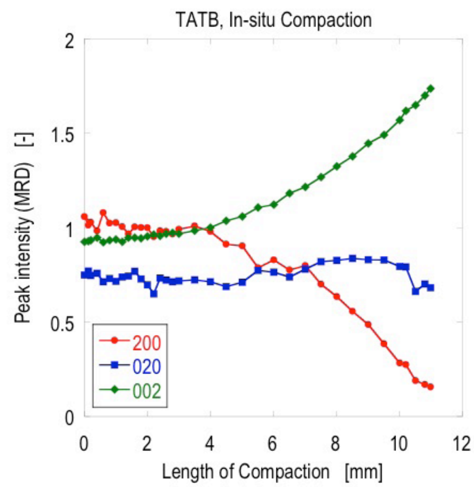

Figure 2: Relative change of the peak intensity during compaction of a TATB powder measured on the SMARTS diffractometer. The (200), (020), (002) reflections correspond to the a, b, and c crystallographic axes, respectively. "Length of Compaction" refers to the plunger location as it presses into TATB powder in a plunger-and-die setup, so that higher length means a higher degree of compaction in the powder.

accident, it is a prime choice for applications where extreme safety is required. However, TATB has undesirable properties; the most problematic is the anisotropy of the crystal. A detailed understanding of TATB's response to temperature changes and the compaction process is of great importance to mitigate or at least accommodate the problem.

The crystal structure of TATB is triclinic, the lowest possible crystal symmetry, and it expands about 10 times more on its crystallographic c-axis than on the a- and b-axes. When a pellet of compacted TATB is heated and cooled a few times, this anisotropic thermal expansion leads to a permanent change in shape and size of the bulk material, an effect called ratchet growth. The temperature change experienced outdoors over the course of a year is sufficient for this effect to happen. When the platelet-shaped crystals of TATB powder are compacted, the crystals become preferentially oriented due to their shape, similar to a deck of cards falling on the floor—it is very unlikely to see a card standing up.

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Neutron cont.

This preferred orientation, or texture, makes the thermal anisotropy even worse as more crystals are oriented with their c-axis parallel to the compaction direction, thus amplifying the effect of the thermal anisotropy.

Researchers Don Brown, Bjørn Clausen, Sven Vogel (Materials Science in Radiation & Dynamics Extremes, MST-8), John Yeager (Shock and Detonation Physics, WX-9), and D.J. Luscher (Fluid Dynamics & Solid Mechanics, T-3) designed and conducted experiments to probe these TATB crystal effects on an atomic and microstructural level. Using Lujan Center instruments at the Los Alamos Neutron Science Center, they employed diffraction techniques to measure lattice parameters as well as orientations of the crystals as a function of temperature and compaction level, thus simulating the temperature changes and the compaction process experienced by a "real" component. The neutrons probed a large volume of crystals, enabling diffraction on a quantity representative of the bulk material. Comparable experiments using a synchrotron x-ray source, for example, would sample a much smaller amount of material and so would require many more tests to gather equivalent information.

Figure 1 shows the relative change of the lattice parameters of TATB powder as a function of temperature, measured on the high-pressure/preferred orientation (HIPPO) neutron diffractometer at LANSCE. The data shown results from multiple heating and cooling cycles, finding that the unit cell dimensions do not change with the number of cycles. The temperature-dependence of the dimensions strongly agrees with one particular literature report and somewhat disagrees with another, enabling the model developers to identify the correct data to use.

Figure 2 shows the change of certain peak intensities as a function of compaction of TATB powder, measured on the Spectrometer for Materials Research at Temperature and Stress (SMARTS). During compaction, the grains gradually rotate such that the c-axis, as measured by the (002) reflection intensity, is increasingly aligned with the compaction axis. Thus the platelet TATB crystals are shifting during compaction so that the largest face is perpendicular to the loading. This is confirmed with the full orientation data from a measurement after final compaction, Figure 3. While this final configuration was generally known from previous experiments, the evolution of the texture during compaction had never been observed. One particularly interesting finding about the evolution of the texture is that the c-orientation grows at the expense of the a-orientation, while the b-orientation remains approximately constant. The researchers expected a more balanced reduction of both axes, and this surprising finding will have direct impact on the mesoscale model. Such experimental observations are uniquely provided by in situ diffraction measurements and can be compared to proposed deformation modes from TATB molecular dynamics simulations (e.g. Mathew & Sewell, *Philosophical Magazine* **95**, 2015, p. 424-440).

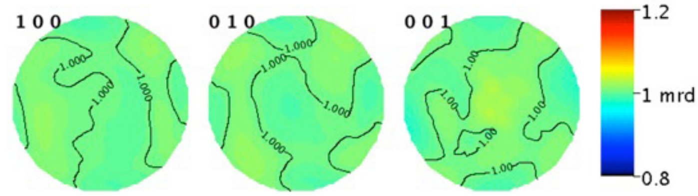
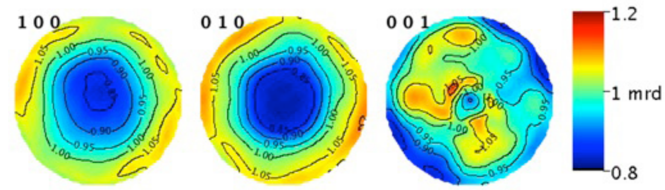


Figure 3: Preferred orientation of TATB crystals for loose powder (above) and a compacted pellet (below) as measured on HIPPO and displayed as pole figures for the a (100), b (010), and c (001) axes. The sample cylinder axis is out of the plane of the paper. The contours are in multiples of random orientation (mrd), i.e., a perfectly random orientation would be 1 in all directions. The powder shows completely random orientation, while the pellet shows strong (001) orientation.



The data will inform a new mesoscale model described in "Self-consistent modeling of the influence of texture on thermal expansion in polycrystalline TATB," *Modelling and Simulation in Materials Science and Engineering* **22**, 7 (2014). The modified model will then be used to predict mesoscale effects in other geometries/experiments, which can be verified with further Lujan Center diffraction experiments in an iterative fashion. The work is an example of science on the roadmap to MaRIE, the Laboratory's proposed experimental facility for control of time-dependent material performance. MaRIE's combination of x-ray and neutron scattering methods will provide unprecedented, time-resolved access to structural properties of materials from atomic- to mesoscales.

The B61 Life Extension Program (Project Realization Team Lead Daniel Trujillo) and Campaign 1: Primary Assessment Technologies (Program Manager Stephen Sterbenz) funded the work, which supports the Laboratory's Nuclear Deterrence mission through lifetime extension research on TATB and PBX 9502. With Sandia National Laboratories, the U.S. Air Force, and the Department of Defense, Los Alamos is using science-based R&D to find specific solutions to the need for certifying the lifetime of each component, as well as the functionality of the system as a whole. If a component is too old and cannot be recertified for a longer time period, then it can be rebuilt as designed or completely redesigned.

Technical contact: Sven Vogel (neutron diffraction) and John Yeager (TATB)

Los Alamos-developed new spectrometer contributes to advances in high energy density physics research

Using their expertise in x-ray imaging, Los Alamos researchers, in collaboration with University of Michigan researchers, have created an imaging x-ray Thomson spectrometer (iXTS) for experiments at Los Alamos National Laboratory's Trident Laser Facility and the University of Rochester's Omega Laser Facility[1,2].

This instrument, developed, constructed, and tested with funding from the NNSA Science Campaigns (Steve Batha Campaign 10, Steve Sterbenz Campaign 1), has been mainly used to provide high-resolution imaging x-ray scattering measurements for warm dense matter and high energy density experiments[3 – 6], yet has also found use in dynamic x-ray fluorescence imaging[7] as well as x-ray emission. The iXTS's sophisticated capability has led to advances in understanding of warm dense matter and strong shock propagation in the high energy density (HED) regime. Future experiments are planned to measure plasma conditions of turbulence in HED regimes.

Los Alamos researcher David Montgomery (Plasma Physics, P-24) developed the concept for the instrument and designed and built the first prototype. He also led development of the final instrument design for Trident and Omega.

iXTS also constitutes a contribution in kind pledged by Los Alamos National Laboratory to the Helmholtz International Beamline for Extreme Fields (HIBEF) of the European XFEL, under the aegis of the MOU (LANL MOU 0081 signed 2014) between Los Alamos and the Helmholtz Zentrum Dresden Rossendorf, the lead institution for HIBEF.

The iXTS instrument consists of a toroidally curved ideal crystal (such as germanium), and a CCD camera for measurement readout. The instrument in its present configuration has a spatial resolution dx = 25-μm and spectral resolution δE = 4 eV at 8-keV with an imaging magnification

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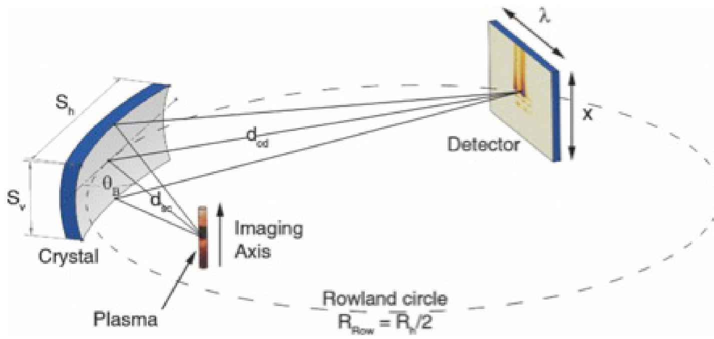
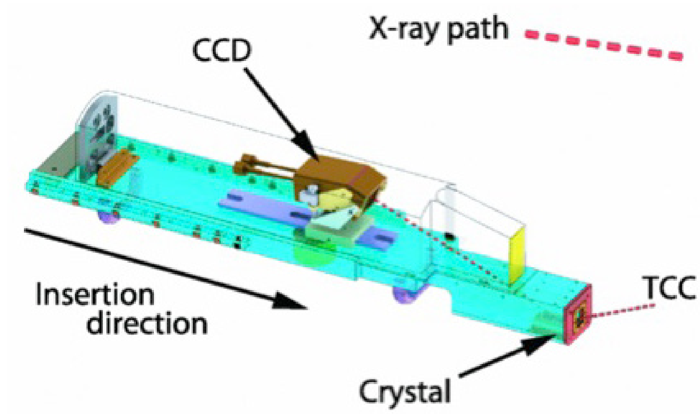
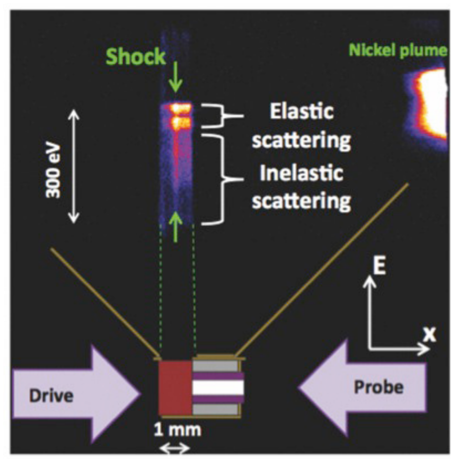


A schematic of the toroidal imaging crystal spectrometer arrangement (from Ref. 2).



Layout of the instrument as implemented for Trident and Omega as a 10-inch manipulator (TIM) insertion diagnostic (from Ref. 2).

Example of using the iXTS to diagnose a strong shock driven in a carbon foam. The x-ray source in this experiment was generated from a laser-heated nickel plasma producing Ni He-α x-rays at 7.8-keV (from Ref. 2).



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Los Alamos-developed cont.

of 2, and throughput of 4e-5 sR. In principle, higher spatial resolution can be achieved with higher magnification without sacrificing throughput, and conceptual designs exist for these configurations.

Its development is also an example of progress on the path to diagnostic capabilities needed for MaRIE (Matter-Radiation Interactions in Extremes), the Laboratory's proposed experimental facility for control of time-dependent material performance. MaRIE will introduce the world's highest energy hard x-ray free electron laser, which will help accelerate discovery and design of the advanced materials needed to meet 21st-century national security and energy security challenges. Thomson scattering is one of the diagnostic techniques to be applied for MaRIE experiments, and a future version of this spectrometer will enable such measurements there.

Technical contact: David Montgomery

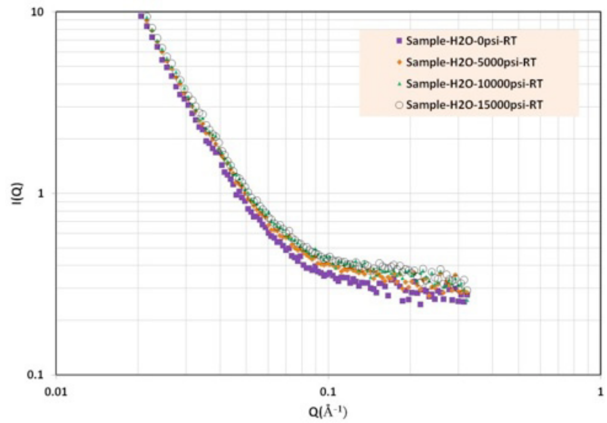
SANS of pore morphology in shale reveals nanoscale pore structure and gas behavior

Research benefits gas and oil extraction

In an effort to maximize unconventional oil and gas recovery, Los Alamos researchers probed pore structure and water-methane fluid behavior in nanoporous shale rock at reservoir pressure and temperature conditions using LQD, the small-angle neutron scattering (SANS) Low-Q Diffractometer at the Los Alamos Neutron Science Center. Their results reveal information key to understanding how nanopore structure determines the distribution of fluids in reservoirs and its impact on oil and gas recovery.

Using a pressure sample environment capable of 414 MPa and 250°C, and a gas-fluid mixing apparatus to deliver fluid mixtures to the pressure cell, the researchers performed in situ SANS measurements at various elevated pressures up to 172 MPa and at temperature up to 66°C with and without the presence of injected fluid. They discovered that the pores in shale are highly ramified interconnected sheets, accessible to water and methane, which flatten with increasing pressure, thus squeezing out the fluid. Emphasis in hydraulic fracture is to open cracks allowing the fluid to flow. This result may give insight into the distribution of fluid in situ with pore size and how this distribution will change with hydraulic fracture.

The development of an oedeometer to examine the hydrocarbon flow behavior in nanoporous shale rocks under uniaxial stress, representing in situ reservoir conditions, is planned, which will provide the first experimental results on hydrocarbon phase behavior and flow properties in shale formations where nanopore size, geometry and connectivity are sensitive to pressure, strain, temperature, water content and hydrocarbon species.



SANS of shale at room temperature and various pressure: small length scale effects.

The study is preliminary to time-dependent nanoporous fluid flow studies, examples of science on the roadmap to MaRIE, LANL's proposed facility for time-dependent materials science at the mesoscale. With MaRIE, studies on time dependent in situ geomechanical and fluid flow with extremely high time and spatial resolution will be possible to emulate gas and oil extraction procedures.

Participants include Mei Ding (Earth Systems Observations, EES-14), Erik Watkins (Materials Synthesis and Integrated Devices, MPA-11), Mel Borrego (LANSCE Weapons Physics P-27), Daniel Olds (Materials Science in Radiation and Dynamics Extremes, MST-8), Honwu Xu (EES-14) and Rex Hjelm (MST-8). The work, which supports the Laboratory's Energy Security mission and Materials for the Future science pillar, is funded by Energy, Oil & Gas (LANL program manager George Guthrie).

Technical contact: R. Hjelm

To learn more about MaRIE, please see marie.lanl.gov, or contact Cris Barnes, capture manager, at cbarnes@lanl.gov.

Roadmap to MaRIE, featuring science and technology highlights related to Los Alamos National Laboratory's proposed experimental facility, is published by the Experimental Physical Sciences Directorate. For information about the publication, please contact adeps-comm@lanl.gov.

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