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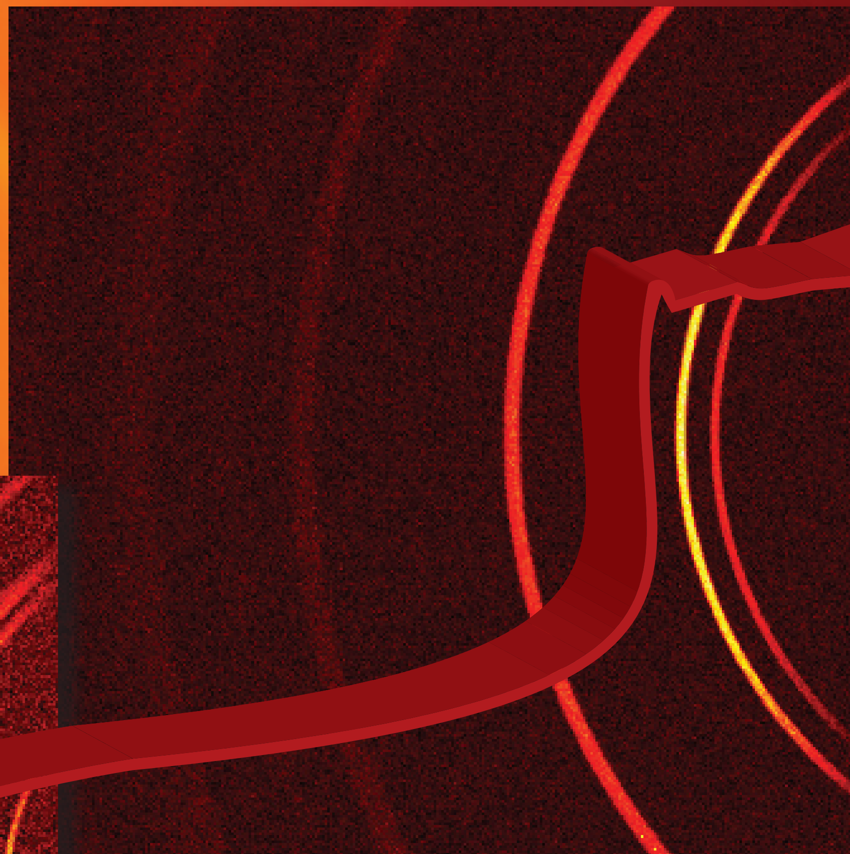
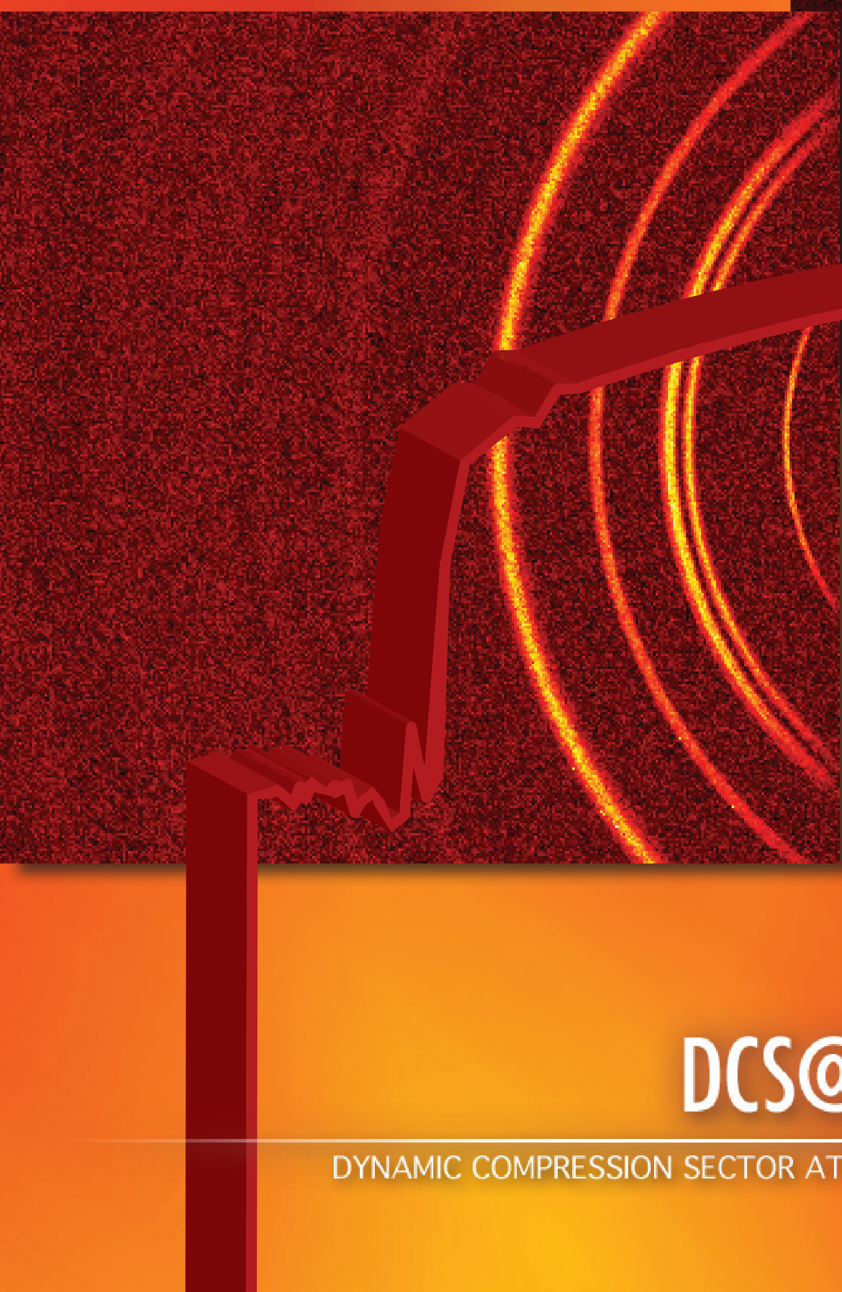
New Research Opportunities in

DYNAMIC COMPRESSION SCIENCE





Report on the DCS User Workshop

DCS@APS

DYNAMIC COMPRESSION SECTOR AT THE ADVANCED PHOTON SOURCE

NEW RESEARCH OPPORTUNITIES IN DYNAMIC COMPRESSION SCIENCE

Report on the Dynamic Compression Sector User Workshop
June, 2012

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About the cover:

For more information about the images on the cover, please see Sidebar 4: High Pressure Polymorphs of Silicon (page 35.)

Acknowledgement:

The workshop was supported by the Department of Energy [National Nuclear Security Administration] under Award Number DE-NA0001304 to Washington State University.

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TABLE OF CONTENTS

Figures

Tables

Sidebars

EXECUTIVE SUMMARY

Understanding the atomistic-level response of condensed matter systems under extreme conditions is of central importance to fundamental science frontiers in numerous fields and to advanced technology. In particular, dynamic compression experiments are both unique and versatile in their ability to produce and probe a broad range of extreme conditions on very short time scales. State-of-the-art dynamic compression platforms have the ability and the flexibility to generate extreme thermo-mechanical conditions (very large compressions, high temperatures, and large deformations) on very short time scales (ps to μs) in a controllable manner. At the same time, advances in X-ray capabilities such as those provided by modern synchrotron sources enable the generation of bright, high-energy, and tunable X-rays that can be used to probe dynamic compression phenomena in real time and with unprecedented temporal and spatial resolutions. A key scientific feature of dynamic compression experiments coupled to high-energy, tunable X-ray probes is their ability to provide time-resolved, atomistic-scale investigations of condensed matter phenomena "on-the-fly" or as they occur.

In parallel with these experimental advances, modern theoretical approaches coupled with ever-increasing computational power now enable simulations of larger physical systems for longer times and with an unsurpassed level of physics fidelity. While advances in high-performance computing continue to extend the range of length scales available for numerical simulations, extending the time scales of such simulations to experimentally observable processes in materials is generally challenging and remains very much an active area of research. Consequently, an important overarching goal of time- and space-resolved investigations of dynamically compressed condensed matter is to perform experiments on the time and length scales of numerical simulations and to bridge this knowledge gap. **This is the frontier of dynamic compression science**. Recent community-based workshops have concluded that *in-situ, time-resolved measurements at microscopic length scales constitute the overarching science need for achieving a fundamental understanding of the mechanisms governing time-dependent condensed matter phenomena (structural transformations, inelastic deformation and fracture, and chemical reactions) under dynamic loading*.

As an important step in meeting this need, the Department of Energy's National Nuclear Security Administration (NNSA) is supporting the establishment of the Dynamic Compression Sector (DCS) at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL). By integrating a broad range of dynamic compression platforms and tunable high-energy X-rays, the DCS@APS will make significant contributions to advance the state-of-the-art in the field of dynamic compression science.

Within this context of unique emerging capabilities and significant scientific potential, a user workshop of approximately 110 international leaders spanning the frontiers of dynamic compression science was convened at the Advanced Photon Source on January 19-20, 2012. The objective of the workshop was to explore "Basic Research Directions for Dynamic Compression Science" and to identify the broad spectrum of scientific challenges and opportunities afforded by the integration of dynamic compression platforms and advanced X-ray capabilities. Topics of discussions included structural changes and phase transformations in condensed matter, deformation processes and fracture dynamics in materials, dynamics of chemical reactions, and time-resolved dynamic processes in materials beyond those generated by compression. In addition to exploring these frontiers of dynamic compression science, the workshop emphasized instrumentation opportunities, including advanced diagnostics and X-ray optics, and governance models for the DCS facility that would maximize the impact of DCS@APS on dynamic compression science.

Workshop attendees enthusiastically concluded that DCS would enable a broad suite of frontier dynamic compression science opportunities and would further fuel the development of additional techniques and capabilities for advancing this challenging scientific field. This report documents these opportunities and research directions.

INTRODUCTION AND SUMMARY

The Dynamic Compression Sector (DCS) at the Advanced Photon Source (APS) will focus on time-resolved X-ray diffraction and imaging measurements in materials subjected to dynamic compression. The X-ray energies (hard X-rays) and the time structure (ns-separated pulses) of the APS are well suited to examine time-dependent changes in materials subject to a broad range of peak stresses (~ 5 GPa to above 100 GPa) and time durations (tens to several hundred ns). DCS will emphasize dynamic compression science of condensed phase materials and will complement other user facilities that emphasize static high pressure studies of materials and warm dense matter/plasma physics regimes.

Approximately 110 researchers spanning the frontiers of dynamic compression science gathered at the Advanced Photon Source in January, 2012, to explore “Basic Research Directions for Dynamic Compression Science” and to identify the broad spectrum of scientific challenges and opportunities afforded by the integration of dynamic compression platforms and advanced X-ray capabilities. Topics of discussions included structural changes and phase transformations in condensed matter, deformation processes and fracture dynamics in materials, dynamics of chemical reactions, and time-resolved dynamic processes in materials beyond those generated by compression. In addition to exploring these frontiers of dynamic compression science, the workshop emphasized instrumentation opportunities, including advanced diagnostics and X-ray optics, as well as governance models for the DCS facility that would maximize the impact of DCS@APS on dynamic compression science.

Understanding the atomistic-level response of condensed matter systems under extreme conditions is of central importance to of fundamental science frontier in numerous disciplines and to the development of many advanced technologies. In particular, dynamic compression experiments are both unique and versatile in their ability to produce and probe a broad range of extreme conditions on very short time scales. State-of-the-art dynamic compression platforms have the ability and the flexibility to generate extreme thermo-mechanical conditions (very large compressions, high temperatures, and large deformations) on very short time scales (ps to μs) in a controllable manner. At the same time, advances in X-ray capabilities such as those provided by modern synchrotron sources enable the generation of bright, high-energy, and tunable X-rays that can be used to probe dynamic compression induced materials phenomena in real time and with unprecedented temporal and spatial resolutions. A key scientific feature of dynamic compression experiments coupled to high-energy, tunable X-ray probes is their ability to afford time-resolved, atomistic-scale investigations of condensed matter phenomena “on-the-fly” or as they occur. This experimental ability is central to achieving a mechanistic understanding of condensed matter dynamics.

In parallel with these experimental advances, modern theoretical approaches coupled with ever-increasing computational power now enable simulations of larger physical systems for longer times and with an unsurpassed level of physics fidelity. While advances in high-performance computing continue to extend the range of length scales available for numerical simulations, extending the time scales of such simulations to experimentally observable processes in materials is generally challenging and remains very much an active area of research. Consequently, an important overarching goal of time- and space-resolved investigations of dynamically compressed condensed matter is to perform experiments on time and length scales that are closer to numerical simulations to bridge this knowledge gap. **This is the frontier of dynamic compression science**. Recent community-based workshops [1-3] have concluded that *in-situ, time-resolved measurements at microscopic length scales constitute the overarching science need for achieving a fundamental understanding of the mechanisms governing time-dependent condensed matter phenomena (structural transformations, inelastic deformation and fracture, and chemical reactions) under dynamic loading*.

The workshop began with plenary talks that reviewed the current state of research in materials under extreme environments and high energy density science. The current capabilities and future plans of the Advanced Photon Source in general and the Dynamic Compression Sector in specific were overviewed. NNSA leadership provided a perspective on the importance of these science areas for national security as a motivation for its investment in DCS@APS. The important work of defining scientific challenges for which DCS is particularly well suited was completed through many hours of discussion and debate within panels spanning the frontiers of dynamic compression science. The resulting Priority Research Directions are summarized in Table 1 and are discussed in detail in the body of this report.

Panel	Priority Research Directions (PRDs)
Structural Changes and Phase Transformations in Condensed Matter	Structural Changes During Loading
	Kinetics and Dynamics of Phase Transitions
	Non-Crystalline Diffraction for DCS
Deformation Processes and Fracture Dynamics in Materials	Examining Microscopic Response to Dynamic Compression
	Examining Dynamic Tensile Damage and Spall
Dynamics of Chemical Reactions	Mechanics Leading to Chemical Change
	Chemical Reaction Mechanisms In Extreme Conditions
Time Resolved Dynamic Processes in Materials Beyond Those Generated by Compression	Deformation and Fracture of Materials
	Phase Transition Dynamics
	Chemical Reaction Dynamics

Table 1. Dynamic Compression Science Priority Research Directions

In the area of structural changes and phase transformations in condensed matter, much of our understanding of phase transformations and structural changes under dynamic compression comes from careful wave propagation analysis of continuum measurements. In most cases, the identification of the phases comes from auxiliary experiments, e.g., with diamond anvil cells and diffraction analysis, or from molecular dynamics simulations, both classical and quantum. DCS will provide the dynamic materials community with a dedicated facility for performing research on phase transitions and structural changes using X-ray diagnostics, and open up these research areas to a broader scientific community. Time resolved diffraction measurements open a window on the crystal distortions, lattice rotations, and dislocation densities that emerge during the loading process. These insights will provide a wealth of data to inform better models of material strength under dynamic loading and extreme conditions. Research on kinetics and dynamics of phase transitions will provide much needed information on the role of loading rates in phase transitions.

In the area of deformation processes and fracture dynamics in materials, our current understanding is derived largely from real-time continuum measurements (velocity interferometry, stress gauges, etc…) and detailed microstructural examination of shock recovered samples. DCS will provide the opportunity to obtain new real-time information regarding the time evolution of microstructural heterogeneities: defects need to be identified; time evolution of defect densities and defect spatial arrangements need to monitored during the dynamic compression event; the time evolution of phase fractions needs to be monitored for materials that undergo phase transformation during dynamic compression; void initiation, growth, and coalescence need to be monitored in real-time as dynamic tension leads to failure. Such information will lead to more realistic, physically-based models for the response of dynamically compressed materials.

In the area of dynamics of chemical reactions, the X-ray diagnostics offered by DCS promise to generate a revolutionary increase in our understanding of chemical processes during dynamic compression. Bragg X-ray scattering can probe the creation and destruction of crystalline phases in chemically reacting systems. Time-resolved small-angle X-ray scattering can monitor the kinetics of nanoparticle formation or consumption during detonation. For instance, the precipitation of graphite and diamond-like nanocarbon has long been recognized as essential to understanding the product Equation of State (EOS) generated from the explosive material triamino-trinitro-benzene (TATB). Finally, dynamic X-ray spectroscopies, a longer-term undertaking, could offer revolutionary information on chemical bond changes induced by dynamic compression. This suite of new experimental techniques promises to deliver unprecedented insight into fascinating chemical processes occurring at extreme conditions.

In the area of time resolved dynamic processes, the anticipated lower repetition rate of DCS' primary compression drivers creates an opportunity for complementary experiments. Three broad classes of complementarity are envisioned: (i) detailed three-dimensional microstructural characterization of materials to be examined in the single event experiments, (ii) dynamic experiments under thermo-mechanical conditions complementary to the primary single-shot dynamic compression experiments and (iii) technique and diagnostic development including the development of new techniques such as stereoscopic imaging.

The science of dynamic compression will be enabled by its instrumentation. The primary challenges for dynamic compression science instrumentation are: (1) beam optics, (2) gating and synchronization, (3) detector technologies, and (4) loading platforms. Also important will be (5) complementary diagnostics, both coupled to dynamic experiments and to the efficient use of other beam and characterization resources to support desired pre- and post-characterization of targets. The final challenge is (6) offline support needs unique to DCS and specific to the precision assembly and modification of experimental targets. Of primary note, the current dedicated detector developments at synchrotrons will still fall short in their capabilities to fully exploit this unique science sector. A short-term and long-term detector strategy is needed to develop a full set of desired and minimum specifications for each area of dynamic compression science.

Lastly, building on this breadth of science and instrumentation opportunity, *the panel on Governance Models and User Experience Considerations was motivated by four guiding principles*: i) maximize the quality of science emerging from DCS, ii) develop the unique attributes of DCS, iii) provide effective leadership in the field, and iv) consistency with operating principles of DOE SC user facilities. As a result of the workshop, suggestions and recommendations in the areas of policy and governance, operations, user experience, and outreach and education emerged as topics that the panel believes will contribute directly to the successful realization of dynamic compression science opportunities through DCS@APS.

In the end, workshop attendees enthusiastically concluded that DCS would enable a broad suite of frontier dynamic compression science opportunities and would further fuel the development of additional techniques and capabilities for advancing this science. Finally, workshop attendees recognized that DCS should be the first, not the last, of a next generation of advanced measurement capabilities that coupled e.g., advanced X-ray sources to relevant extreme environments with measurement resolution designed to validate and stretch state-of-the-art modeling capabilities to advance our predictive understanding of materials in extreme environments.

Dynamic Compression Sector at the Advanced Photon Source
Innovation, Discoveries, and Education to Understand Materials at Extreme Conditions

Dynamic compression experiments have two unique attributes: (1) the ability to achieve extreme thermo-mechanical states of matter, and (2) the opportunity to probe the temporal evolution of these states, or materials dynamics, in real time.

To develop a fundamental understanding of the materials phenomena under dynamic compression (structural changes and phase transformations; deformation and fracture; and chemical reactions) and their relationship to the evolving atomistic structure, it is necessary to examine the time-dependent response of materials at microscopic length scales with nanosecond time resolution. Such measurements are essential for gaining scientific insights and for evaluating multi-scale simulations of key phenomena under dynamic compression.

To address the overarching scientific need indicated above, the DOE/NNSA is establishing a first-of-a-kind user facility at the Advanced Photon Source (APS). Washington State University (WSU), representing the dynamic compression science community, is partnering with the APS to establish the Dynamic Compression Sector (DCS), an experimental capability dedicated to multi-scale measurements under dynamic compression. DCS will couple a variety of dynamic compression platforms to a state-of-the-art synchrotron beamline to routinely obtain time-resolved X-ray measurements with nanosecond resolution. DCS will permit a range of tunable incident energies (hard X-rays) and time-structures (ns-separated pulses) to observe time-dependent changes in materials subjected to a range of peak stresses (~5 GPa to above 100 GPa) and time-durations (tens to several hundred nanoseconds).

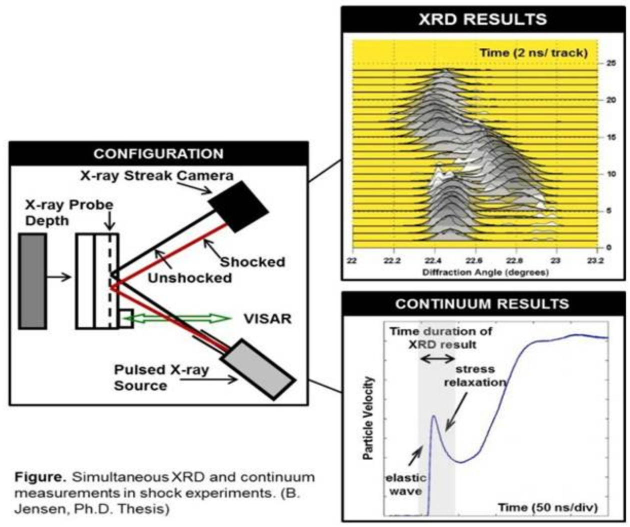


Figure. Simultaneous XRD and continuum measurements in shock experiments. (B. Jensen, Ph.D. Thesis)

Additionally, DCS will provide a rapid turnaround of experiments to optimize scientific throughput for the users. The NNSA sponsored user facility represents a new paradigm to undertake scientific discovery challenges related to dynamic compression of materials, and to train the next generation of scientists in this exciting and challenging field.

Sidebar 1. Dynamic Compression Sector at the Advanced Photon Source

References

[1] “Dynamic Response of Materials: Scientific Challenges and Opportunities,” prepared for the DOE/NNSA, June, 2007.

[2] Workshop on “Understanding Condensed Matter Dynamics at the Microscopic Level, APS, Argonne National Laboratory, IL, June 23-24, 2008.

[3] “21st Century Needs and Challenges of Compression Science Workshop,” Santa Fe, NM, September 22-25, 2009.

STRUCTURAL CHANGES AND PHASE TRANSFORMATIONS IN CONDENSED MATTER

Chairs

Michael P. Desjarlais, *Sandia National Laboratories*
Gilbert "Rip" Collins, *Lawrence Livermore National Laboratory*

Panel Contributors

Tom Duffy, Jon Eggert, Stefan Hau-Riege, Brian Jensen, and Reed Patterson

Introduction

The great preponderance of our knowledge concerning the structures and phases of materials come from ambient or near-ambient observations. With the advent of diamond anvil cells and structural diagnostics using X-rays, insights into structural changes and phase transformations have been extended to several Mbar stresses. We are now entering an era where the combination of X-ray diagnostics and dynamic compression of samples is providing a wealth of new insights into these processes under dynamic conditions. Along with dynamic loading, which has the potential of achieving much higher pressures than is currently possible with diamond anvil cells, there is also the added emphasis of time scales and kinetics. The shorter time scales and elevated thermal states under dynamic loading are, in many instances, more relevant to understanding the integrated physics of complex events, and the only approach to providing access to the necessary regions of phase space at present.

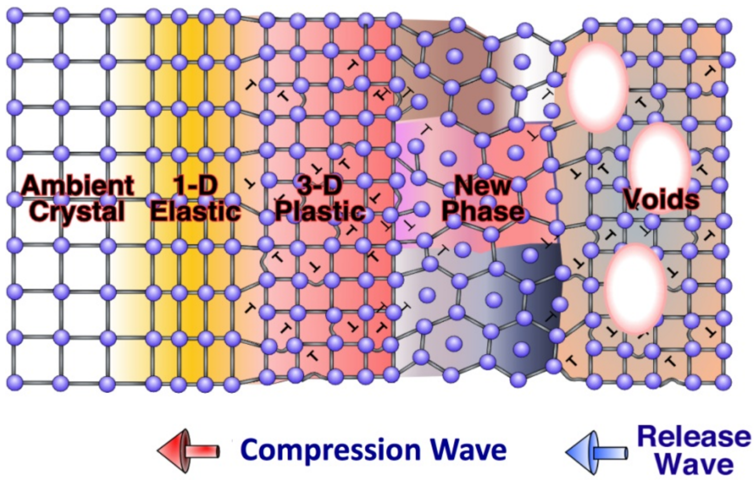


Figure 1. A schematic view of an ideal crystal undergoing elastic and plastic deformation, followed by a phase transformation, and finally structural changes associated with the release wave. (Lorenzana, LLNL).

The range of topics that fit under the heading of Structural Changes and Phase Transformations is very broad. The list includes the macroscopic phase transitions, such as solid-solid, solid-liquid (melting), liquid-liquid, and liquid-solid (freezing). More subtle are structural changes at the micro and meso scales within a given phase, for example, the changes associated with elastic to plastic deformation. Due to the nature of dynamic loading, the influence of loading direction relative to crystal orientation, and the production of non-isotropic stresses is an important area of study.

Adding to the challenge in unraveling the complex behavior of materials under dynamic loading is that all of these processes have kinetic rates associated with them, potentially complicating the landscape with metastable states and transient heterogeneity.

The advent of X-ray diagnostics fielded in concert with dynamic loading experiments has opened the door for tremendous growth in our understanding. In many, many cases, the depth of our ignorance is still quite great, which makes the future extremely interesting.

Status of the Field

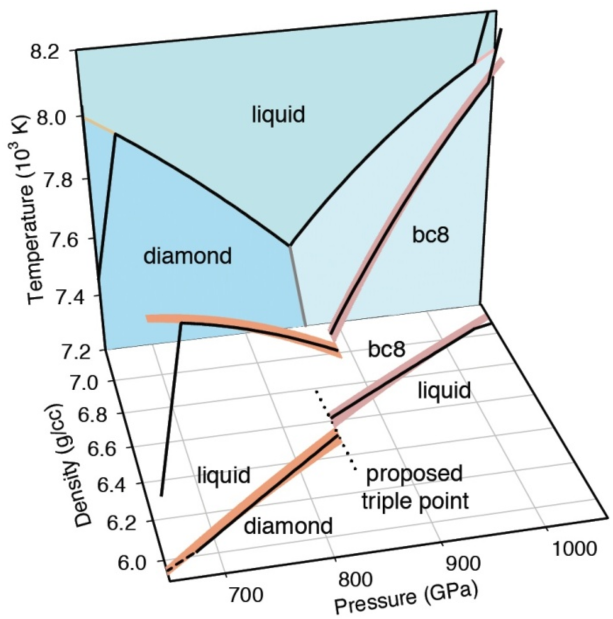


Figure 2. The high-pressure phase diagram of carbon. [Knudson, Desjarlais, and Dolan, Science (12/19/08)]

With the exception of a very small body of notable work on the application of X-ray diagnostics to dynamically loaded materials, much of our understanding of phase transformations and structural changes comes from careful wave analysis. In most cases, the identification of the phases comes from auxiliary experiments, e.g., with diamond anvil cells and diffraction analysis, or from molecular dynamics simulations, both classical and quantum.

Figure 2 illustrates our best current understanding of the high pressure phase diagram of carbon along with the coexistence boundary for diamond, liquid carbon, and the bc8 phase. The diagram resulted from a series of shock experiments coupled with extensive analysis of first-principles calculations. The location of the triple point and the transition to the bc8 phase was inferred from comparisons between the observed shock speeds and the shock speeds calculated for the various phases. While the agreement between the observations and the calculations is quite good, a direct structural identification of the bc8 phase in carbon at these conditions would put our understanding on a much firmer foundation. Much of our insight on processes at the micro and meso scale has come from large scale molecular dynamics simulations, but a great deal of validation is needed.

With regard to the use of X-ray diagnostics in the study of the dynamic response of materials, we have entered an era of what promises to be an explosively growing phase. Much of this current interest – citations to papers on X-ray diffraction of shock loaded materials have been growing steadily since the mid-1990s – is driven by the early successes and the increasing availability of high quality X-ray sources.

In 1970, Johnson, Mitchell, Keeler and Evans reported the first observation of X-ray diffraction during shock compression. Explosively driven shocks were launched into samples of LiF and a Cu K-α source was used to probe the shocked LiF. These measurements provided the first direct evidence that the material remained crystalline during compression. One of the greatest challenges was synchronization of the X-ray pulse with the shock. Johnson and Mitchell reported the first X-ray diffraction evidence for a phase transition during shock compression in 1972. Their experiments on pyrolytic BN suggested a transformation from the graphite-like (pre-shock) structure to a wurtzite-like structure under stress. An interesting side note was the observation of differences in diffraction line intensities and sharpness between the measurement in the shocked state and post shock that could suggest a change to a smaller crystallite size during the release phase.

In the late 1980s, Wark and colleagues used the two beam JANUS laser at LLNL to demonstrate subnanosecond X-ray diffraction from laser-shocked crystals of (111) silicon. One laser pulse was used to drive the shock, the second, more highly focused and of much shorter duration, impinged on a solid target material to create highly stripped atoms and bright X-ray line radiation. This two-beam, pump-probe system provided a big step forward in the synchronization of the probe beam with the shock and time resolution.

In the early 1990s, Zaratskii and colleagues reported on the use of X-ray diffraction to observe the B1 to B2 transition in KCl, and concluded that the unit cell compression before the transition was uniaxial. This system was revisited in the late 1990s by d'Almeida and Gupta, who were able to conclusively show with a combination of diffraction data and analysis that the transition proceeded from an isotropic compression state. Using the X-ray diffraction results, d'Almeida and Gupta also worked out the atomistic pathway for the B1 and B2 transition in KCl.

Work on pressure induced phase transitions in iron provides a good example of the evolution in understanding afforded by X-ray diagnostics. Starting with the shock compression work of Bancroft, Peterson, and Minshall in the 1950s, a pressure induced phase transition in iron at 13 GPa was deduced from the onset of wave splitting in the shock profile at the transition pressure. The transformation was originally hypothesized to be α (bcc) to γ (fcc), due to the known existence of γ at elevated temperatures and ambient pressure. It was in the early 1960s that Jamieson and Lawson, using a pressure vessel and X-ray diffraction, ruled out the γ phase as the high pressure phase under static loading and showed that the diffraction lines were consistent with the ε (hcp) phase at the observed volume compression. Under planar shock loading the sample is under uniaxial strain, so the transformation observed under isotropic loading could well be different, or might occur through a different path.

Wang and Ingalls (PRB, 1998) used X-ray absorption fine structure (XAFS) measurements at the Stanford Synchrotron Radiation Laboratory (SSRL) to explore the α to ε transition of iron in a boron carbide anvil. They described three models for the transition, the first going back to Mao *et al.,* (JAP 1967) and two others attributed to Burgers (Physica, 1934).

The first direct observation of the α to ε transition in iron under shock loading is found in the work of Kalantar and colleagues (PRL, 2005). The experiments were performed using the OMEGA, Janus, and Vulcan lasers. This work is an excellent example of the insights possible with nanosecond resolution diffraction data. Through analysis of the non-α lines in both reflection and transmission Bragg diffraction, the transformation process was found to be in very good agreement with the mechanism proposed by Mao, and also found in the large-scale molecular dynamics simulations of Kadau *et al.*, (Science, 2002) and later explored in more detail in the post experiment analysis of Hawreliak et al., (PRB, 2006).

It should be pointed out that wave propagation and continuum analysis will continue to play an important role. In the context of the α to ε transition of iron, the paper by Jensen *et al.* (JAP, 2009) explores the transition kinetics and also highlights some of the difficulties faced by large scale molecular dynamics simulations when trying to reproduce plastic deformation on realistic time scales.

As an alternative approach to diffraction measurements to detect phase transformations under shock loading, the use of EXAFS is noteworthy. Yaakobi and colleagues used the broad spectrum of X-ray radiation from an imploding CH capsule on OMEGA to detect the bcc to hcp transformation in iron. The broad spectrum from APS and other synchrotron sources is well suited to this approach. An appealing aspect of the EXAFS approach is the material selectivity that derives from the choice of which absorption edge to probe.

Another material that has received considerable early attention with diffraction measurements is lithium fluoride (LiF). Whereas in iron the principle focus has been the α to ε phase transition, the work on LiF has focused on the elastic-plastic structural deformation under shock loading. Prior to the first diffraction work, there was already a considerable body of work using continuum measurements and wave analysis where it became clear that LiF demonstrated different elastic to plastic behavior depending on the orientation of the crystal relative to the loading direction. Rigg and Gupta (APL, 1998) used copper $K\alpha$ X-rays to perform diffraction measurements of single crystal LiF compressed along the [111] and [100] directions. Their diffraction data show clearly that for lattice compressions of around 1 to 2% in the loading direction, LiF compressed uniaxially when loaded along the [111] direction and underwent isotropic compression when loaded along the [100] direction. Subsequent work by Rigg and Gupta (PRB, 2001) employed multiple diffraction measurements to observe both transverse and longitudinal deformation for these same crystal orientations. Their addition of an X-ray streak camera permitted time-resolved diffraction measurements for loading in the [111] direction with 2 to 4 ns resolution (JAP, 2003), providing, in essence, a diffraction movie of the elastic compression. The work of Rigg and Gupta was followed by the work of Jensen and Gupta where the elastic-plastic transition in shocked magnesium-doped LiF was explored (JAP, 2006 and 2008). It was known from earlier continuum data that the elastic limit of LiF is considerably enhanced by magnesium impurities at the 100 to 200 ppm level (Asay, *et al.*, JAP, 1972). These experiments achieved 2 ns time resolution in time-resolved diffraction measurements and very good agreement was achieved between the diffraction measurements and continuum modeling. Of particular note is the insight gained on the uniaxial to isotropic (elastic to plastic) transition behind the shock front and the observation of a kinetic time scale of tens of nanoseconds for the transition to take place.

In the sections that follow we will amplify three priority research directions. The first two can be considered direct development and extensions of the pioneering work on iron and LiF discussed above. The third takes us into an area that is particularly important for understanding materials at more extreme conditions, in particular, the structure of liquids and complex fluids that result from shock loading of materials to very high pressures, often pressures much beyond what can be reached in any static experiment.

PRD: Structural Changes During Loading

Introduction

The ability to directly examine structural deformation and phase transitions, at the atomic length scales, in materials subjected to dynamic loading has been a long-standing scientific challenge. Traditional experiments have relied on continuum wave propagation measurements to indirectly examine the underlying physical mechanisms responsible for the deformation. These measurements have been largely successful and have been able to provide information about elastic-plastic deformation, equation-of-state, and phase transitions for a number of materials. Although there has been significant progress made over many decades, experiments are needed that can access the atomic length scales directly to: (1) uniquely identify atomic structure in new high pressure phases, (2) obtain information on a material's microstructure (grain size, dislocation densities, etc.), and (3) examine the evolution of a materials microstructure during both the loading process and during transformations (structural and phase).

Science and Technology Opportunities with DCS

There are diagnostics currently in use at the APS that have potential to provide information on structural deformation, including phase contrast imaging and monochromatic/ polychromatic Laue diffraction. Work by WSU scientists has demonstrated the use of monochromatic diffraction to examine lattice compressions and line-broadening in an exploratory series of experiments at the APS using a small-bore powder gun. More recently, scientists from Los Alamos and Argonne have developed a mobile impact facility (IMPULSE) and demonstrated dynamic Laue diffraction and phase contrast imaging using a single 60-ps width X-ray bunch synchronized to the dynamic event. These two independent efforts show clearly that diagnostic capabilities exist that could revolutionize the dynamic compression field although significant effort remains to adapt these methods to the dynamic experiments. Refinement of target designs, identification and/or development of suitable detectors, development of synchronization schemes, and loading platform that are compatible with the current synchrotron facilities are areas that should receive attention in the near-term.

Research Directions and Needs

To examine structural changes in materials, well-defined dynamic experiments are needed that can:

- Obtain line-broadening and diffraction data to estimate dislocation densities (including generation and nucleation), to determine crystallite size and orientation (mosaicity), to measure unit cell compression, and relate all these to the microstructure for a bulk material

- Uniquely identify crystal structures of dynamically created states to provide a basis for studying strength, elastic-plastic deformation, etc. for various regions of a materials phase diagram
- Pursue fundamental studies on single crystal materials followed by a transition to real-world polycrystalline and/or heterogeneous materials
- Develop the analysis tools to analyze the data and generate an understanding of the dynamic material properties.

To accomplish these objectives, the following development must begin now to ensure success of DCS when the facility is on-line:

- Detector development is KEY and essential for the success of DCS as well as future efforts such as MaRIE (LANL). Detector development requires: (1) more sensitive detectors utilizing better efficiency and/or amplification, (2) phosphor and scintillator development with faster decay times and better light conversion, (3) large-format detectors for Laue diffraction, (4) multi-frame detectors for Laue and phase contrast imaging, and (5) trigger and gate features to allow for synchronization. Ultimately, multi-frame images are required to generate "shock movies" of the events in the ps-μs regimes.
- Platform development is required to scale traditional loading platforms to accommodate the synchrotron environment and to provide synchronization schemes as needed. For some platforms such as two-stage guns, considerable engineering will be required for use at the APS.
- Diagnostic development is required to adapt the currently available synchrotron methods to dynamic events. Diagnostics include phase contrast imaging, monochromatic diffraction, and polychromatic Laue diffraction.
- Development of analysis methods and tools to facilitate analysis of the data.

Potential Impact

The potential impact of DCS is clear. The ability to obtain time-resolved diffraction data and microstructure information using a combination of imaging and diffraction techniques is essential to an in-depth understanding of dynamic phenomena including structural transitions. Except in a few cases, information regarding dislocation densities, microstructures, etc. and other details that exist *during* the shock event have been inferred either from static testing, indirect wave profile measurements, or post-mortem recovery/analysis.

PRD: Kinetics and Dynamics of Phase Transitions

Introduction

Our knowledge of the kinetics of structural phase transitions influences our understanding of the behavior of materials over a broad span of physical sciences; including geophysics (the composition and behavior of the interior of the Earth), high-pressure states of condensed and warm dense matter (transitions rates, barriers, and regions of metastability or co-existence), materials science (phase nucleation, growth, and synthesis), and experimental design (decoupling the effects of drivers from intrinsic material physics). For example, recent quasi-static measurements on geophysical compositions of Fe-Ni compounds[1] show the influence of kinetics on similarly performed measurements and highlight the need to examine such phenomena at high compressions and rates. To understand the nature of the kinetics of phase transformations during dynamic compression experiments, a detailed and accurate understanding of the structures of materials at these conditions is essential. While dynamic compression studies can span timescales from milliseconds to fractions of a nanosecond, time-resolved structural measurements remain some of the most difficult to perform. Experiments typically produce indirect measurements, thus dynamic properties are often inferred and interpretations can be strongly model dependent. At DCS, direct measurements of crystal structures and the kinetics of phase transitions will be possible, providing the opportunity to rapidly further our knowledge of the dynamic behavior of materials under a wide range of loading conditions.

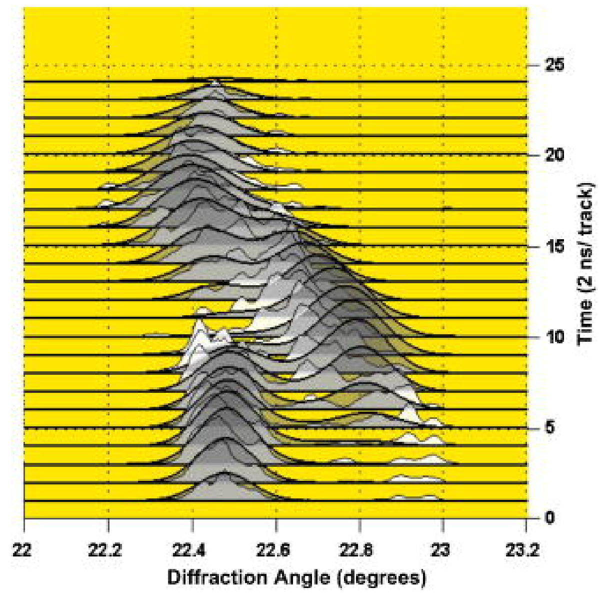


Figure 3. Streak camera images and calculations of time-resolved X-ray diffraction of shocked LiF [7].

Research Directions

Observations of phase transitions in dynamic compression experiments are frequently made via inferred techniques, e.g. wave profile analysis, and compared with structural measurements made under static loading conditions. Comparison with models of sufficient complexity to include time-dependent effects allow some conclusions to be drawn about transition kinetics [see e.g., 2, 3 or 4]. To accompany these methods, additional measurements are commonly made to provide complementary information and to further constrain the high-pressure state of the material. Such measurements may include optical spectra, temperatures, sound velocities, and radiographs. However, these measurements do not provide direct information on the crystal structure, thus even time-resolved measurements still need to be correlated with a known phase. State of the art X-ray diffraction measurements on gas gun[5] and laser[6] compressed samples demonstrate the capability to directly measure a material's phase as well as providing insight into the nature of the dynamics of the observed phase transitions, Figure 3 [7]. Considering the complexity of these experiments, there is clearly a need to stimulate and make accessible such experiments. It is evident that there are many, largely unexplored, opportunities to better understand the evolution of crystal structures under dynamic loading conditions. The observed differences in shock versus shockless or ramp compression experiments are one example that highlight the importance of kinetics in material behavior under these conditions. Measurements of the crystal structures themselves, the paths and/or mechanisms from one structure to another, the relevant timescales associated with particular transitions, the possibility of different end-states for a given (or slight variants on a) set of experimental conditions, and even the potential for creating or recovering metastable or novel materials from high-pressure states are all areas with the potential for significant growth.

Coupling with DCS

The integration of dynamic compression drivers with synchrotron radiation and the associated detector technologies will provide a platform to explore the kinetics of phase transformations that is unparalleled in its' combination of temporal and spatial resolution, photon energy, and brilliance. Single-bunch X-ray diffraction has the capability of producing time-resolved "snapshots" of structural data at intervals of several ns (depending on the APS operating mode). Furthermore, samples can be interrogated over length scales on the order of 100 microns or better, permitting observation of the dynamics of single- or poly-crystal-like behavior. Taking advantage of the high brilliance and spectral range of the APS source, a large number of new materials can be studied that were previously inaccessible due to lower symmetry structures, lower scattering factors, etc. In addition, complimentary measurements such as X-ray absorption and imaging can provide temperature dependences, local order parameters, and dislocation nucleation, growth, and motion. All of these measurements will both constrain and enhance our mechanistic understanding of the dynamic behavior of materials at extreme conditions.

In order to implement these sophisticated experiments, DCS will draw strongly from the experience and support/collaboration of the APS. For example, to take advantage of the time structure of the electron bunches, new synchronization schemes will need to be developed to ensure that the X-ray measurements are acquired during the desired part of the sample loading. Scheduling experiments for measurements of kinetics may strongly depend on the inter-bunch spacing (which can be from 10 ns to 150 ns e.g.), which is dependent upon the APS mode of operation. Both of these considerations are also dependent on the driver used for a given experiment, implying that the logistics for laser vs. gun driven experiments need not be exclusive with respect to kinetics. In addition, the wealth of experience available at the APS in the fields of X-ray diagnostics, detectors and recorders will facilitate the next generation of these tools that will be required to meet the needs of dynamic compression experiments.

Potential Impact

For a limited number of materials (e.g. Fe or LiF), there exist excellent data on phase transitions and the kinetics of these under dynamic compression. However, any generalized understanding of the kinetics of phase transitions is severely hampered by the lack of available information. Given this general paucity of experiments, the potential for significant advances in our understanding of the evolution of materials subject to these conditions is great. Considering the explosion of phase transition studies and the impact on our understanding of material behavior that synchrotron diffraction studies of statically compressed materials have had, one could imagine a similarly large impact when synchrotron X-ray diffraction studies are paired with dynamic compression experiments. DCS is the realization of such experiments and it will vastly increase the accessibility of structural measurements to the dynamic compression community. Its unique capabilities will provide new insights into the dynamic processes of phase transitions, further constrain the models we use to understand these phase transitions, and potentially reveal new or unexpected phenomena. Furthermore, the desire to examine in greater detail the structure of materials on sub-ns timescales and with few-micron spatial resolution (to examine phase transformations *in-situ*) will drive the next generation of diagnostics, requiring e.g. innovation in fast and/or large-area detectors, as well as high-dynamic range and high-efficiency devices.

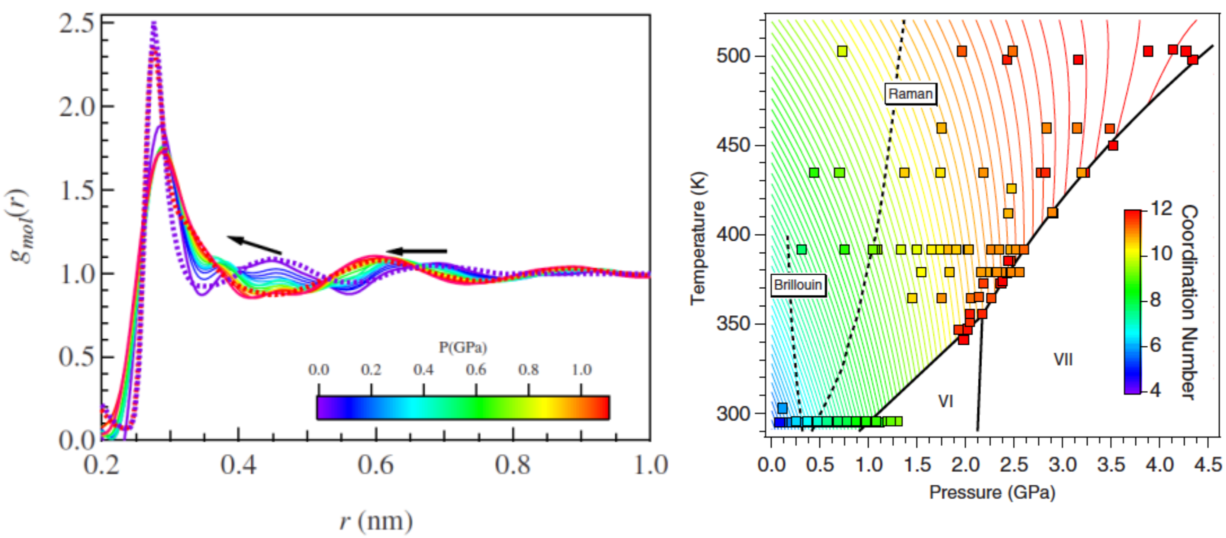


Figure 4. Molecular radial-distribution function, $g_{mol}(r)$ and coordination of water at high pressure and temperature measured by X-ray diffraction in diamond-anvil-cell experiments. [Weck, et al., Phys. Rev. B 80, 180202 (2009)]

PRD: Non-crystalline Diffraction for DCS

Introduction

While pressure, volume, energy, and even temperature measurements have long been a staple of shock-compression experiments, structural information has been much more elusive. Thus, crystal-structure determination is a major goal of the Dynamic-Compression Sector (DCS) for shock and ramp compression. Sufficiently strong shock compression will melt any material, so it is important to consider the possibility of structure determination in the fluid phase. Recent advances in diffraction techniques for small samples with large background scattering in diamond-anvil cells (DACs) have made high-pressure structural determination of liquids a reality, as illustrated in Figure 4. It is a goal of DCS to achieve comparable results in dynamic-compression experiments.

Science and Technology Opportunities with DCS

While all samples become hot fluids at sufficiently high shock stresses, there is experimental evidence suggesting that structural changes may still occur. As shown in Figure 5, recent Hugoniot experiments on $MgSiO_3$ suggest a major structural change, with a 6% volume discontinuity, at 300-400 GPa and 10,000-16,000 K. It is possible that many liquid-liquid transitions exist, and diffraction experiments on DCS offer unprecedented potential to study them.

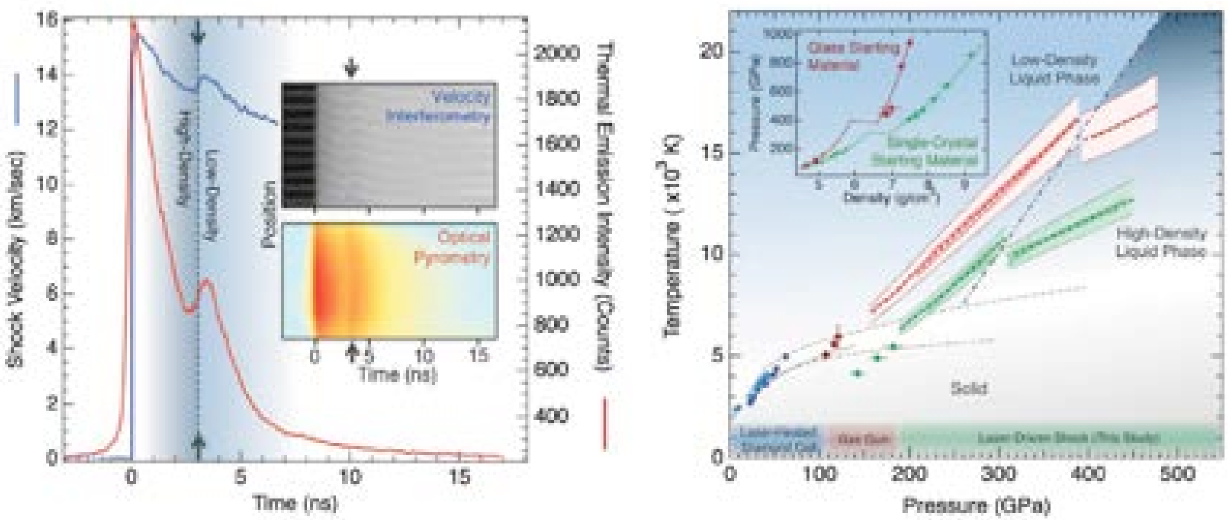


Figure 5. Phase transition in $MgSiO_3$ identified by observed volume and temperature discontinuities on the Hugoniot. The transition is striking due to its location well within the liquid stability region of phase space. [Spaulding, et al., Phys. Rev. Lett., 108, 065701 (2012)]

Research Directions Opportunities and Needs

Achieving fluid diffraction data on DCS will be a major challenge. The total scattering efficiency for a material is independent of structure, but the directionality of fluid diffraction is more spread out, and harder to measure, than for crystalline diffraction. Even so, a recent proof-of-principle run on BIOCARS at the APS demonstrated diffraction from 24.5 µm-thick Kapton (a common low-scattering-intensity semi-crystalline polymer [Virk, et al., Bull. Mater. Sci., **24**, 529 (2001)] yielded high-quality signal with a single bunch of pink-beam irradiation (Figure 6). With a high-repetition-rate (1 Hz) laser drive we can repeat the shot many times (Figure 7). Soller-slit spatial filtering [M. Mezouar, et al., Rev. Sci. Instrum., **73**, 3570 (2002)] is also a possibility to isolate the sample volume from the ablation front.

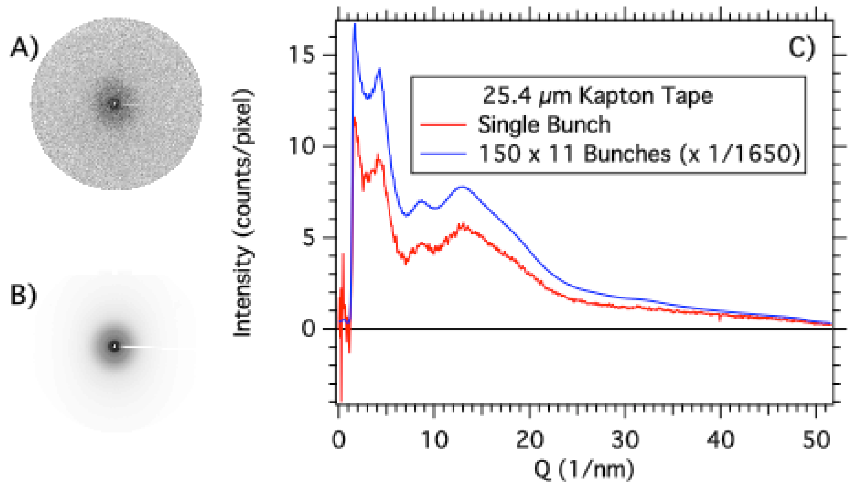


Figure 6. Preliminary experiments done at BIOCARS showing pink-beam 25.4 μm Kapton tape diffraction for A) a single bunch and B) 150 x 11 bunches. C) The integrated intensity is shown for the single bunch and the scaled multi-bunch images (background diffraction from air was subtracted).

Potential Impact on Discovery and Innovation

Solid electride structures (ionic crystals where the anion lattice sites are occupied by localized electrons with no associated nuclei) have been observed in high-pressure solid Li and Na, and predicted for many other elements. At this time no electride phase has been melted experimentally, and the structure of the adjacent liquid phases is an open question. DCS opens the possibility of routinely studying electride-forming materials in the liquid phase. Such studies have the possibility of dramatically affecting our understanding of high-density materials.

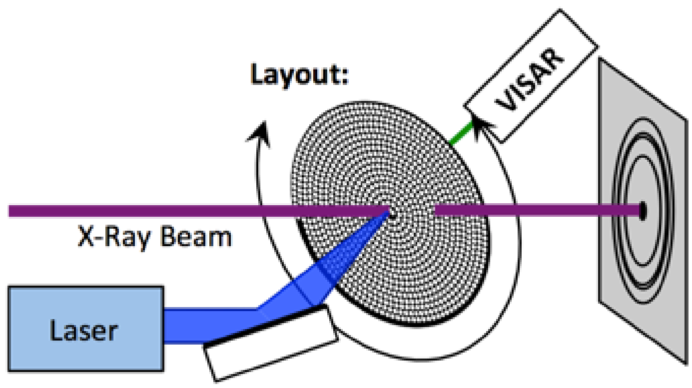


Figure 7. Potential setup for multiple repeated laser driven experiments at DCS. Repeated shots will improve the photon counting statistics dramatically.

Conclusion

The use of X-ray diffraction to study structural changes and phase transitions has grown rapidly over the last several decades but work in this area has largely been limited to a few pioneering groups. The work on the α to ε transition in iron and the elastic to plastic structural changes in lithium fluoride serve as two excellent examples of the important role X-ray diffraction has in advancing our understanding of material changes under dynamic loading.

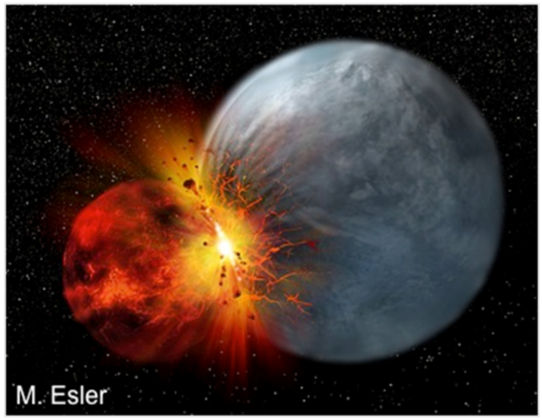
The Dynamic Compression Sector (DCS) at Argonne National Laboratory's Advanced Photon Source will provide the dynamic materials community with a dedicated facility for performing research on phase transitions and structural changes using X-ray diagnostics, and open up these research areas to a broader scientific community. The opportunities for greater involvement within the academic community are a particularly important benefit. The data that will emerge from these explorations will undoubtedly provide many difficult challenges for the theoretical and modeling communities, an essential aspect of continued advances in those areas.

We have highlighted three priority research directions for DCS: structural changes during loading, kinetics and dynamics of phase transitions, non-crystalline diffraction. In the case of structural changes during loading, a key word is *during.* Time resolved diffraction measurements open a window on the crystal distortions, translations, and dislocation densities that emerge during the loading process. These insights will provide a wealth of data to inform better models of material strength under dynamic loading and extreme conditions. Research on kinetics and dynamics of phase transitions will provide much needed information on the role of loading rates in phase transitions. Our current state of knowledge here remains quite thin and yet the potential impact on our ability to model these important aspects of phase transitions is great. At sufficiently high dynamic loading stresses, all materials are believed to reach a liquid state. However, little is known about the true structure of these high-pressure liquids. Most of our understanding comes from molecular dynamics simulations that will benefit greatly from experimental validation. In recent years, the subject of liquid-liquid phase transitions has emerged as an important topic for understanding a number of important materials. Diffraction in non-crystalline materials presents the additional challenge of obtaining a sufficient scattering strength at a given angle. Recent work in this area is very exciting and hints of a rich and vibrant future.

Implications for Planetary Science

Shock compression studies have important applications to a wide range of processes in Earth and planetary science ranging from planetary formation to shock metamorphism to synthesis of high-pressure phases otherwise only found in the inaccessible deep interior. Impact events play a major role in planetary accretion, the early chemical evolution of planets, planetary surface features, formation and evolution of atmospheres, and the energy budget of planets. The transient shock loading of rocks and minerals during impact events produces unique deformation features and phases that are diagnostic of impact events and have been studied intensively in samples from terrestrial craters and meteorites. DCS will provide a new tool to understand how the physical processes during impact events produce these new phases and unique shock-induced features.

Quartz, SiO_2, is one of the most abundant minerals of Earth's crust, and is the archetype of planetary silicates. Shock compression of quartz produces a broad spectrum of complex phenomena including phase transitions, strength changes, amorphization, and inelastic deformation. *In situ* studies using X-ray techniques at DCS will provide a unique, new window to observe these deformation processes as they are on-going. In particular, X-ray studies at DCS will address whether the high-pressure phase formed in SiO_2 under dynamic loading is the stable polymorph stishovite, a metastable crystalline phase, or an amorphous phase. It will enable us to answer questions about the mechanism of high-pressure phase formation, and unravel the detailed history of samples from compression through release to recovery. Thus, DCS will provide a revolutionary new tool to answer fundamental questions about quartz and a host of other important geological and planetary materials.



Planetary impact phenomena are major forces that shape the accretion and early evolution of planets. A late-stage giant impact as shown here can influence a broad range of phenomena including satellite formation, orbital evolution, magma oceans and interior stratification, and devolatilization and atmosphere formation.

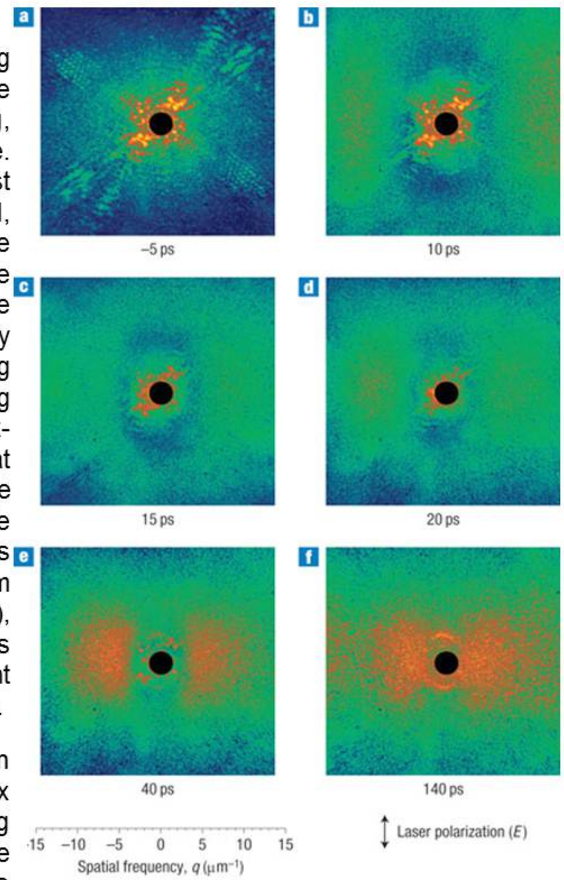
Shock metamorphic features are diagnostic of impact events such as those producing the ~1-km diameter Meteor Crater in Arizona. DCS will enable major advances in understanding the dynamic processes governing the formation of such features throughout the impact process. Shock wave studies also provide unique insights into the nature of planetary interiors through the synthesis of high-pressure phase and constraining thermodynamic and elastic properties.

Sidebar 2. Implications for Planetary Science

Coherent Imaging of Structural Changes and Phase Transformations

APS' short high-energy x-ray pulses can be used to image both the structural and phase *dynamics* of matter out of equilibrium. High-intensity x-ray synchrotron radiation allows us to record multiple high signal-to-noise-ratio snapshots, which strung together provide information about the evolution of the material. Since synchrotron radiation is monochromatic and tunable, it also enables element-specific imaging. Imaging artifacts are kept to a minimum since synchrotron radiation is highly paraxial.

Of particular interest are *coherent* x-ray imaging techniques that significantly improve the resolution over absorption-contrast imaging, which is typically several µm and more. Absorption-contrast imaging is the most common x-ray imaging technique that is used, for example, in hospitals. The contrast in the shadowgraph images is due to differences in the attenuation of x rays in the sample. The resolution can be enhanced somewhat by combining small spot sizes with scanning methods. On the other hand, diffractive imaging techniques require at least partially coherent x-ray radiation, which can be produced at synchrotrons by minimizing the radiation source size and maximizing the distance of the sample from the source. Diffractive imaging allows probing different length scales, ranging from macroscopic phase-contrast imaging (sub-µm), over mesoscopic speckle correlation techniques (10 to 100 nm), to microscopic coherent diffractive imaging, possibly to atomic resolution.



Example of using x-ray diffractive imaging to explore the nanoscale optical-laser-induced ablation dynamics in patterned iridium/Si_3N_4 bilayers. Shown are single-shot diffraction patterns at different times after laser excitation. The gradual degradation of the nanofabricated sample is visible by the loss of high spatial frequency information in the diffraction patterns [A. Barty et al., Nat. Phot. 2, 415 (2008)].

Phase contrast imaging is based on differences in the real part of the refractive index that create phase shifts in the x rays traversing the sample. In propagation-based phase contrast imaging, the interference pattern is recorded at a distance from the sample and contains coherence-enhanced image contrast. Phase-retrieval algorithms can be used to obtain an image with improved contrast. Typically sub-micrometer resolution can be achieved with this technique, which makes it particularly useful for shock fronts, phase domains, including bubbles, and inclusions.

Coherent diffractive imaging allows high-resolution imaging of material structure down to atomic resolution by computationally inverting large-angle diffraction patterns, thereby overcoming the limitations of x-ray lenses. In the past, coherent diffractive imaging has been used to image both the unit cell in crystal structures and small, non-periodic samples. More recently, keyhole techniques have been proposed to perform region-of-interest coherent diffractive imaging. For crystalline materials, a combination of resonant Bragg and diffuse scattering can be used to obtain information about strain distributions and defects.

Sidebar 3. Coherent Imaging of Structural Changes and Phase Transformations

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DEFORMATION PROCESSES AND FRACTURE DYNAMICS IN MATERIALS

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Introduction

Materials are central to many emerging technologies intended to meet national needs. Future technologies will place increasing demands on performance in a range of extremes: stress, strain, temperature, pressure, chemical reactivity, photon or radiation flux, and electric or magnetic fields. The condensed phase (for most materials) defines a pressure and temperature range of interest which may be approximately fixed at less than a TPa and less than 10,000 K respectively (Figure 8) [1]. Compression induces changes in bonding properties at the atomic scale. Indeed, thermo-mechanical loading allows new forms of matter to be produced. These compressions (reducing interatomic spacings by up to a factor of two and increasing densities by over an order of magnitude) result in changes in the electronic structure that begin to shift notions of chemical interaction and atomic bonding. For example electrons surrounding nuclei or ions become localized between nuclei rendering concepts of strength difficult at pressures under extreme conditions above a few Mbar. Under dynamic loading (shock compression or ramp compression), these extreme conditions may be exploited to explore the delicate balance between mechanical ($P\Delta V$) and thermal ($T\Delta S$) energies by examining how this dichotomy governs physical and chemical phenomena in the condensed state. The strength of materials is intimately connected with the second of these terms ($T\Delta S$). Additionally, shortened loading periods provide a filter to select governing mechanisms according to operating kinetics. The system may then attain a final metastable state that lies beyond thermodynamic constraints [2].

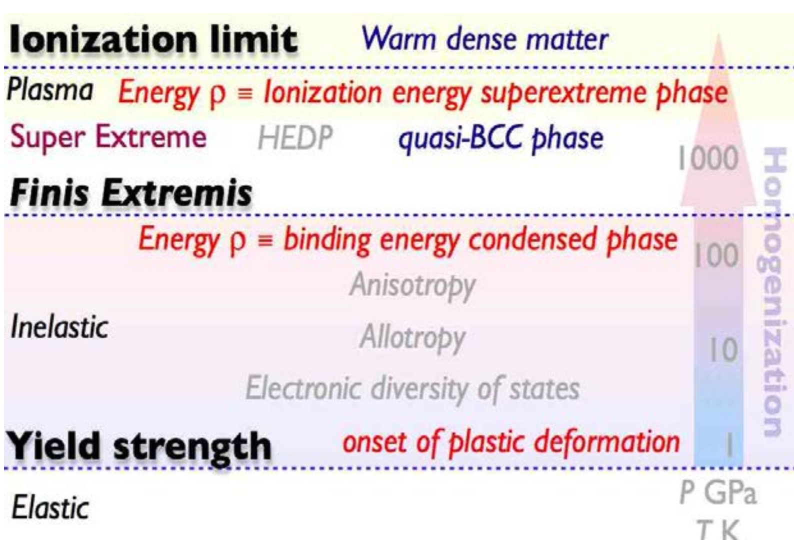


Figure 8. Schematic of matter with increasing pressure.

If the fundamental basis for these transitions can be understood, exciting opportunities to use such extreme thermo-mechanical conditions to design and manufacture new classes of materials will open up. Such advances may allow materials engineered to display their theoretical strengths. Empirical discovery techniques can achieve incremental advances. However understanding operational mechanisms allows defeat of nature's traditional linkages such as high strength with poor ductility and low toughness. Thus, critical questions remain to be answered. What are the most important length scales and defect distributions that control deformation and fracture? What are the ultimate strength and temperature performance limits for structural materials? Finally, can dynamic processes be harnessed to capture and maintain theoretical limits for some operating period if not permanently? The only means of truly attacking and conquering these ideals is to understand the fundamental processes operating as bonds break or rearrange to yield new structures. DCS gives a tangible future for determination of real-time properties at the atomic scale as materials deform under these states.

The characterization of condensed matter under extremes of loading is confined within a region of common physics not by the pressure applied but by the strength of the structure it adopts. This is bounded by the elastic limit at the lower boundary whilst at the upper, the electronic states become changed so that valence electrons no longer determine bonding and delocalization of core electronic states occurs. As pressure increases at all scales, the response homogenizes and, beyond the upper limit for valence electron bonds, high energy density physics describes a homogeneous state [3]. Within the bound below this threshold, the panoply of subject interests range from the atomic physics studies excited by laser impulses, to integrated systems of behavior found in planetary science. Within this suite, the observed behavior is defined by the pressure applied and characterized by the strength observed. While the former is a thermodynamic variable, defined at scales beyond the unit cell, the latter is a function of the volume element sampled by the experiment or phenomenon of interest. The interpretation of measurement in the laboratory thus depends on this sampled volume (a function of the defect population contained with the material under load), the length of the impulse applied, and the resolution limit of the detector.

Multiple Scales

A complete understanding of material response to an arbitrary loading condition is further complicated by the fact that the majority of engineering alloys are complex multi-phase systems at the mesoscale, where a variety of effects, such as number and type of phases, morphology, distribution and grain size all influence the material response. Thus, advances must be made first with pure metals, representative glasses and pedigreed polymers to describe effects when microstructure is controlled [4]. Such research has been carried out statically for many years. However, intermediate loading rate and particularly the dynamic and shock-loading impact regimes have seen little significant progress. Further, materials have been treated as homogeneous (*i.e.* structureless) continua, which has provided a qualitatively reasonable means of representing the compressive behavior of solid materials. This has been adapted naturally into solid mechanics descriptions over the past decades. This route is to some extent self-consistent and further, the success of such constitutive laws is aided by the fact that in compression, high isotropic pressure loading represents a small strain process due to the strong interatomic potential resisting motion. On the other hand, in shear and tension, large strain effects can nucleate at defects in the material structure. Further, defect assemblies evolve through a sequence of stages in their lifetimes, from creation as point defects, through relaxation by migration and interaction with others, to assembly into large clusters. These agglomerations ultimately degrade materials' performance at the macroscopic level and finally lead to failure. Each of these stages can be tracked by the transfer of energy among the electronic, atomic, vibrational, and structural systems of the microstructure, and by the time and length scales required for each transfer. For all of these reasons, the next generation of measurements at DCS must track development of the defect populations in order to capture the relevant physics.

In research today, the availability of relatively inexpensive but powerful cluster machines and the continued development of high-performance computers (at the petaflop or exaflop/s scale) make it possible to model ever larger (and complex) systems [5]. However these are generally of little help in addressing the timescale over which simulations can be performed. At the quantum level, state-of-the-art calculations are typically limited to a few picoseconds, even for quite simple systems; for classical potential atomistic molecular dynamics simulations, typical times rarely exceed nanoseconds. Given the need to address the long time stability of materials under extreme conditions, there is a clear need for the development of new theoretical techniques that can be linked to diffraction measurements at DCS. Further, the gaps in experiments between the meso- and atomic scales require new classes of measurement described below to inform the nature of the modeling and simulation strategy. Unless these gaps are closed, models cannot hope to attack problems that relate to larger scale issues such as the ductile–brittle transition, fracture toughness properties, adiabatic shear banding, material responses to ballistic impacts and shock or ramp compression experiments. Finally, after an initial compressive stress excursion has ended, further release mechanisms follow conditioning the microstructure for later reloading. Amongst such mechanisms are dynamic recrystallization and spallation or fragmentation.

Thus, it will be necessary to make measurements at small length scales to image the development of assemblages of defects or structures responding dynamically. One must then embrace the related inevitable consequence that the measurement will therefore have to be made at faster rates than are presently typical. Thus the challenge for diagnostics in the present situation is to image at greater resolution and in 3-D, but also much faster, than has been required in previous development in the field, in order that the theoretical descriptions are more statistically descriptive and become physically based. With greater resolution, and smaller length scales modeled, there is a need to look at interactions of defect assemblies in three dimensions. Further, once descriptions have been constructed, one needs to map details of the response at these scales up to the continuum scale. The devices and techniques to apply to DCS to accomplish the former, and the frameworks for constructing theory to deliver the latter represent the challenges for the next decade. The outcome will be fully physically-based dynamic material descriptions. These, with increased computer power, offer the promise of virtual design for structures capable of operating in the extreme environments from high-pressure energy generation to structures in space.

Material Response

Continuum observations drive the application of a material to its operational environment, but to understand materials' deformation and to construct a physical description of its response requires information from the subscale processes that occur. In the case of metals, the available data indicate that aspects of processes occurring at the atomic scale (such as phase transformation thresholds) can be modeled accurately with advanced quantum mechanical techniques whereas the kinetics cannot. Processes leading to plastic deformation are however dependent upon defect populations, their activation and subsequent transport and interaction, and these processes occur over times that map to length scales at the micro- and mesoscales. An evolving microstructure and a material with a continuous strength history must be captured by analytical kinetic descriptions at the macroscale. Amorphous metals offer high hardness and superelastic response combined with free volume that impacts a diverse array of applications from coatings, to vibrational damping, strength reinforcements, and hydrogen storage. Existing descriptions derived using continuum tests for quasistatic loading, function well for the regime that they were fit to. However, these models cannot describe transient loading, or be coupled to electromagnetic loading. The next generation of constitutive models must explicitly account for the operating physics informed by fundamental measurements at DCS if they are to be useful in this class of problems.

The inhomogeneity of nature results in materials that deform accessing key physics tied to their crystalline or amorphous structures and the inherent defects within their volumes. It is the appreciation of statistical physical relationships within these populations and their effects upon response that will drive materials to be developed for use under extreme conditions in the future. To achieve physical understanding will require detailed mechanisms describing the evolution of defect populations in order to define response. Sensing the development of this population and determining a means of introducing its effect into continuum models will occupy a major fraction of DCS related research over the next decade in collaboration with the workers developing material physics models across different length scales. Multiscale modeling and simulation must encompass the analytical descriptions of material response, to allow physically-based prediction of the performance of designed microstructures or composites on the one hand, but also the prediction of constitutive and thermodynamic response.

The role of DCS will be to extend traditional crystallography and imaging techniques beyond their present state to quantify and categorize the microstructure and defects and their distribution in disordered materials in real time as the dynamic compression occurs. A challenge in single event dynamic compression experiments is obtaining maximum information in each shot; unlike traditional crystallography and imaging experiments, the X-ray source, the sample and the detector must be stationary during the experiment. Thus, obtaining maximum information in each experiment necessitates using area detectors. The elements of plasticity can then be followed and quantified using Debye, Laue or Bragg diffraction patterns. This will allow determination of phase as well as dislocation distributions, lattice parameters and even texture in the materials under investigation. Further, the determination of correlation lengths in amorphous solids and liquids will be possible. Small angle X-ray scattering will provide insight into void formation and growth. At the engineering scale, 2D imaging will provide insight into macroscopic inhomogeneities and tomography will allow the imaging of deformation dynamically and in 3D with application of new analysis and cameras. The following two sections describe, in more detail, some of the approaches that can be used at DCS to obtain the microscopic information necessary for the development of physically based models for the response of materials to dynamic compression and dynamic tension, respectively.

PRD: Examining Microscopic Response to Dynamic Compression

Among the most compelling applications of the Dynamic Compression Sector is the study of the relaxation of materials forced deeply into metastable states by high-rate compression. Related phenomena include strength and plasticity, twinning, and phase transitions in shock- and ramp-loaded materials. Short-pulse, high-energy X-ray diffraction and imaging permit *in-situ* observations in materials as they are being deformed, revealing mechanistic details and potentially answering long-standing fundamental questions about these processes.

Immediately after rapid uniaxial compression, a material is typically in an unsustainable non-equilibrium state. The deviatoric stress may be many times what the material's strength could support on more conventional time scales, while the pressure may be considerably beyond the range at which the existing crystal structure is stable. A material in such a state will quickly relax through many competing mechanisms. These include dislocation-mediated plasticity, twinning, and phase transformations. A better understanding of these mechanisms is essential for both pure and applied studies of matter under extreme conditions. Essential questions to be answered include:

<u>Materials science:</u> What is the material's ability to withstand irreversible deformation (i.e. its strength) at different time scales? How does the microstructure evolve, especially on passing through a phase transition, and how does this affect the strength? How different are the mechanisms of deformation and the magnitude of sustainable deviatoric stresses on time scales of ps, ns, and µs? How well can we deduce macro-scale properties from micro-scale processes and fluctuations?

<u>Condensed matter physics:</u> How are phase transitions modified when deviatoric stress is an important thermodynamic variable? As a material relaxes from a nonequilibrium state, the stage is set for unstable growth of small perturbations and the development of emergent structure from homogeneous initial conditions. What are the boundaries of stability, what are the competing mechanisms, what kinds of structures can form, and what determines the length scales, time scales, and scaling laws?

Most rapid compression experiments designed to study these phenomena are limited to very indirect information about the internal mechanisms. Measurements are most often limited to time-resolved information about continuum variables and post-mortem information about the recovered microstructure. From these one must deduce the thermodynamic paths and the kinetics of various relaxation mechanisms. While it is possible to make such deductions [6-9], often we must face the fact that our data interpretation problem is badly underconstrained.

DCS offers the opportunity to perform exactly the same kinds of experiments but with the crucial added element of short-pulse high-energy X-ray diffraction. This adds the ability to determine strains, phase fractions, and twin fractions from inside the material during the actual deformation. Such data can provide the crucial missing information to transform a tentative, inexact interpretation into a detailed, precise, sophisticated story of what the material did, when it did it, and what the consequences were. In the following pages, we discuss some of the expected measurement capabilities at DCS. This is followed by several examples of current research efforts and how these efforts would be tremendously enhanced by the additional information provided by DCS.

For example, consider the problem of microscale heterogeneity in a compression wave front, arising from physical instabilities and pre-existing microstructure. Velocity interferometry has been an invaluable tool in examining the continuum response of shock compressed solids and has even been used to demonstrate that the shock front is not homogeneous [10]. However, velocity interferometry is limited to measuring variations in local surface velocity and does not provide detailed information regarding microstructural heterogeneities. In contrast to velocity interferometry, use of high-energy X-rays at the DCS will provide direct insight into the lattice level response of shocked solids. The periodic short duration (~100 ps) X-ray pulses at the APS will provide time evolution of the lattice response throughout the loading/unloading cycle. Whereas velocity interferometry is a Lagrangian measurement examining the response of a material point, X-ray diffraction allows one to "look into" a material. Varying the X-ray energy allows the depth of the probe volume to be varied. Additionally, the X-ray beam spot size can be varied from millimeter to micron dimensions. X-ray diffraction measurements on macroscopic probe volumes will provide the average lattice response with the broadening corresponding to the distribution in microstructure. X-ray diffraction measurements on small probe volumes in well-characterized samples will provide the local lattice response (elastic strains, grain rotations, and subgrain formation) that can be compared with mesoscale simulations.

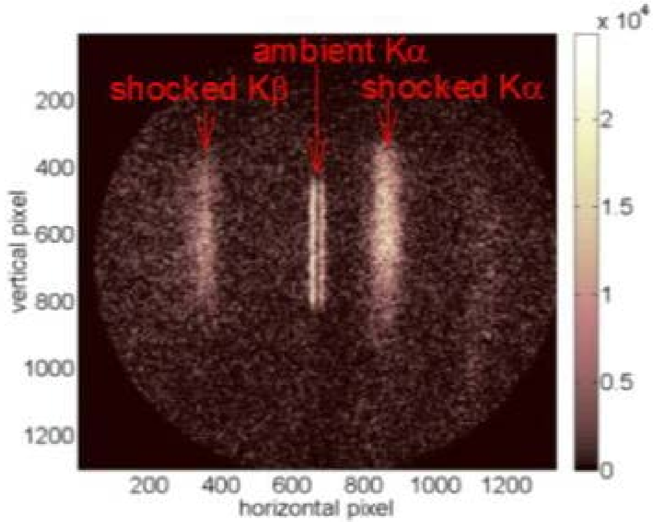


Figure 9. High-resolution XRD from ambient and shocked Si (100). The 400 peak shift provides the elastic strain along the shock loading direction and the peak broadening is due to microstructural heterogeneities. Similar high-resolution Laue measurements of multiple hkl points at the DCS will provide more complete information. From Ref. [12]

Shifts of diffraction peaks provide elastic strains which can be related to stress and strength through elastic constants (Figure 9) [11]. Laue diffraction measurements on single crystals can provide the 3-dimensional deviatoric elastic strain tensor. The volumetric strain can be recovered by employing either a complementary monochromatic diffraction measurement (see Figure 9) or velocity interferometry to obtain the complete strain tensor. Velocimetry analysis also provides an estimate of the normal stress [8, 9], thus providing either a redundant check on the data or—in the case where the pressure-dependent elastic constants are unknown—a direct measurement of both the strength and the equation of state. Such a measurement resolves a long-standing and very important problem: distinguishing normal stress from mean stress, and thus measuring the strength, even in the face of extreme rate dependence and complex nonsteady wave propagation. Multiple Laue diffraction measurements from small volumes can also provide the partitioning of elastic deviatoric strains amongst individual subgrains analogously to past work on static samples [13].

The appearance of new diffraction peaks during shock compression is related to phase transformation [14-17] or twin formation (Figure 10). Powder diffraction methods on polycrystalline materials can be used to identify new phases. Laue diffraction on single crystals can be used to identify both phase and twin variant formation. Relative intensities of the peaks can provide the relative volume fractions between parent and specific product variants in both cases.

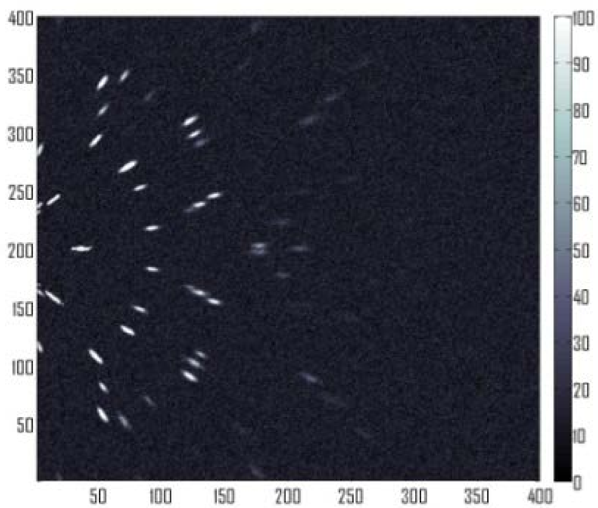


Figure 10. Simulated Laue diffraction pattern from Fe single crystal shock-compressed along [100]. Simulation corresponds to a snapshot while the Fe exists in a mixed bcc-hcp phase. Multiple snapshots as the transformation occurs can provide direct information on the phase change kinetics and transformation pathway.

Broadening of diffraction peaks (see Figure 9) can be used to understand microstructural heterogeneities caused by shock compression. Forward calculations of diffraction pattern broadening can be compared with measured diffraction patterns to determine the best microstructural description of the shocked solid within a given model microstructural parameter space [18]. Because of uniqueness issues, detailed microstructural examination of shock recovered samples will be useful in restricting the microstructural parameter space to include in the analysis of the diffraction broadening in the shocked state. Additionally, anisotropic intensity in powder patterns can provide insight into shock-induced texture.

Several recent studies of strength, twinning, and phase transformation in shock-compressed solids can be improved upon by using capabilities that will be available at the DCS. The strength of shocked Al(100) single crystals was recently determined from X-ray diffraction measurements of the longitudinal elastic strain in the shock compressed state [11]. Strength determination required additional knowledge of density and longitudinal stress which were determined from continuum methods. Similar measurements on single crystals at the DCS using Laue measurements – both in transmission and back reflection - will provide the 3-dimensional elastic deviatoric strain tensor which will significantly reduce the uncertainty in the strength determination. Additionally, the previous strength measurements [11] were obtained from single 50 ns duration X-ray diffraction measurements. Future measurements at the DCS incorporating multiple temporally spaced diffraction measurements will demonstrate whether shear stress remains constant in the peak state or whether the shear stress relaxes indicating a time-dependent strength loss.

Twinning and phase transformations have a profound effect on functional strength, as both effectively dissipate deviatoric stresses. Moreover, the driving forces and subsequent variant selection for twinning and phase transformations depend strongly on the relative orientation of the deviatoric stress and the crystal lattice. Recent measurements of twinning in single-crystal tantalum (Figure 11) reveal a complex dependence of twin fraction on temperature, strain rate, and crystal orientation. While models have been developed to help explain these results, it is extremely difficult to validate the models in detail because of one crucial missing piece of information: We do not know when the twinning happened. Twinning relaxes deviatoric stress in much the same way as dislocation motion, and there is no "smoking gun" in velocimetry traces to tell when twinning is occurring. Twin fraction measurements were all done on post-mortem recovered materials, many days after the shock experiment. There is no way of knowing how many of the twin grains formed early in the compression, near the point of peak strain rate, during the post-shock plateau, or long afterward (perhaps even as a byproduct of the capture process, despite all efforts to minimize such effects). Just a few measurements of the twin volume fraction at well-chosen points in time would resolve the most challenging issues in such experiments and allow very direct validation of twin-mediated strength models.

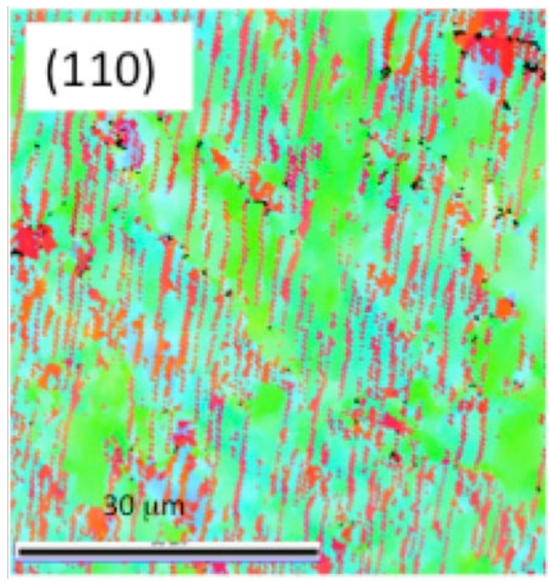


Figure 11. Electron backscatter diffraction image of a shocked (110) single crystal of tantalum showing twins in red.

Orientation relations between low- and high-pressure phases of KCl [14, 15] and Fe [16, 17] have been determined using real time X-ray diffraction on shocked single crystals. For KCl, the orientation relation between the low and high pressure phases was found to depend on the loading direction; interestingly, loading along [111] produced an unidentified Bragg peak in the data collected in the shocked state. Laue diffraction measurements at the DCS will elucidate the origin of this unidentified peak and will also provide the complete orientation relationship(s) between the parent and product phases. In the case of martensitic transformations, these data provide direct information on the underlying crystallographic mechanisms, among which there may be several choices, as well as dependence of the driving force on deviatoric stress. Laue measurements can be designed to provide the 3-d deviatoric strain tensor, which includes critical information about transverse strains and hence strength. For the case of Fe, significant kinetic effects in the $\alpha-\varepsilon$ phase transformation have been observed both using static experiments and shock compression experiments [19]. The time evolution of the phase transformation can be observed at the lattice level by measuring the relative intensities of the Laue spots of the low and high pressure phases.

Another recent advance in the study of loading rate-dependent strength under shock compression comes in the field of ultrafast velocimetry, coupled with highly repeatable short-pulse laser drives [20]. This method produces velocity-time traces similar to those produced by VISAR but with dramatically higher time resolution, extending down to a few picoseconds. Ultrafast velocimetry is emerging as a probe of fundamental mechanisms of dissipation, dispersion, and nonlinearity and how they compete and cooperate to govern complex wave evolution. Elastic wave amplitudes up to ~10 GPa have been observed in aluminum [20], a material which at more ordinary time scales starts to deform at considerably lower stresses. At strain rates of 10^{10} s^{-1} or more, many of the conventional assumptions about the mechanisms of wave propagation in materials will break down, and we can expect thermal transport and even nonlinear acoustic phonon dispersion to play ever-increasing roles. Just as in conventional shock-wave strength experiments, the ultrafast experiments would directly benefit from a time-resolved measurement of deviatoric strain. The laser-driven shocks also enjoy the benefit of very large throughput, such that with an automated system potentially hundreds of shots could be performed in a few days of experiment time. Such experiments therefore complement the more conventional gas-gun-driven nanosecond to microsecond scale experiments.

The thread that links these various efforts is this: Conventional velocimetry analysis, coupled with a robust understanding of thermodynamics and (where possible) post-mortem analysis of recovered material, provide powerful tools for extracting information about strength and phase transformations from complex compression wave experiments (Figure 12). However, we often find that our data interpretation is hampered by the lack of direct, time-resolved information about the processes happening inside the materials. DCS can provide exactly this information, and many distinct experimental campaigns are poised to take advantage of this capability as soon as it is available.

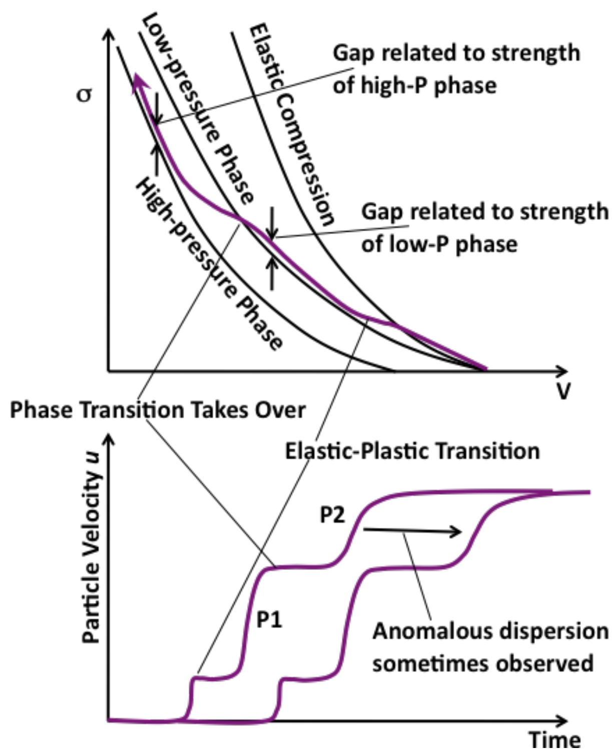


Figure 12. Time-resolved high-energy X-ray diffraction can resolve key ambiguities in the interpretation of conventional shock-wave strength and phase transformation studies. Conventional analysis demands that we assume specific wave features correspond to specific material processes, and that the stress-volume curves for various asymptotic "pure" states are precisely known. Just a few measurements of phase fractions and deviatoric strains at key points in time will resolve the most vexing interpretation issues in such experiments. Such shots could be fielded on DCS quite quickly; they are essentially repeats of existing experiments with the addition of one crucial diagnostic. The experiment could potentially even measure the strength of otherwise inaccessible high-pressure phases as the microstructure evolves during and after the compression wave.

PRD: Examining Dynamic Tensile Damage and Spall

The integrity of structural elements subjected to dynamic loading is often limited by their resistance to failure caused by dynamic tension following the initial dynamic compression. Thus, it is important to understand the detailed mechanisms through which dynamic tensile damage initiates and how the damage coalesces as it proceeds to macroscopic material failure. The large deformation, damage and failure process for many polycrystalline metallic materials is inherently ductile in nature. In general, this means that the material will choose four specific physics mechanisms for accommodation of imposed deformation field or resistance to loading – solid-solid phase transformation, large deformation plasticity, shear localization or adiabatic shear banding, and cavitation. This discussion at present restricts itself to the materials and physics in this class of behavior. At present, materials models to represent this damage process contain the elements of pore initiation or nucleation, pore growth, pore coalescence, and ultimate failure [21-27]. The nucleation process is believed to depend heavily on microstructural based heterogeneities and the spatial distribution of defects. These include grain boundaries, impurity inclusions, intersection of twin planes, dislocation sub-cells. Additionally, the compressive loading modifies the initial microstructure as described in the previous section. Figure 13 gives one such example, void nucleation associated with grain boundaries in high purity copper.

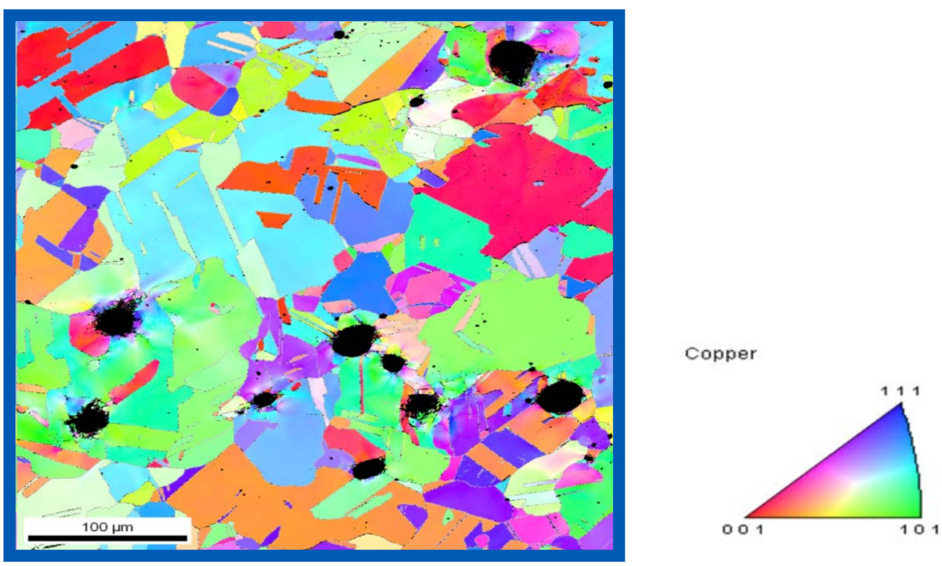


Figure 13. Electron back scattered diffraction (EBSD) generated inverse pole figure map of incipiently spalled copper showing damage nucleating at grain boundaries rather than within grains. Colors of grains pertain to crystallographic orientations and regions in black represent voids. This observation raises questions about deterministic features of grain boundary failure during shock and how shock loading affects grain boundary regions in ways that promote void formation.

The statistical spatial distribution of inherent (grain boundaries, inclusions, initial dislocations) or deformation induced heterogeneities (twinning, dislocation sub-cell) is believed to act in combination with the spatial and temporal intensity of loading to determine which of the weakest defect sites will initiate a pore. The size of grains and the amount of stored work in the form of alterations to the microstructure including changes in crystallographic texture have also been shown to influence the way in which this damage process evolves within the material. A nucleated field of pores will then grow in size until they become large enough such that the deformation field surrounding individual pores begins to overlap with neighboring pores – at which time the process of damage coalescence begins. The coalescence phase is when the established pore field begins to join and when localization or adiabatic shear banding facilitates this process. Of course, ultimate failure will occur when the coalescence process brings about a percolated region of damage. For dynamic and shock loading conditions, these events occur within very small length and time spans and so velocities and accelerations are very large. Therefore, in addition to spatial effects, local inertial effects must also be accounted for [27-29]. This process is extremely complex, statistical and inherently loading path/rate dependent which our present day models do not adequately represent in complex loading histories. Figure 14 gives one such example for shock loaded Ta showing our current damage modeling capability.

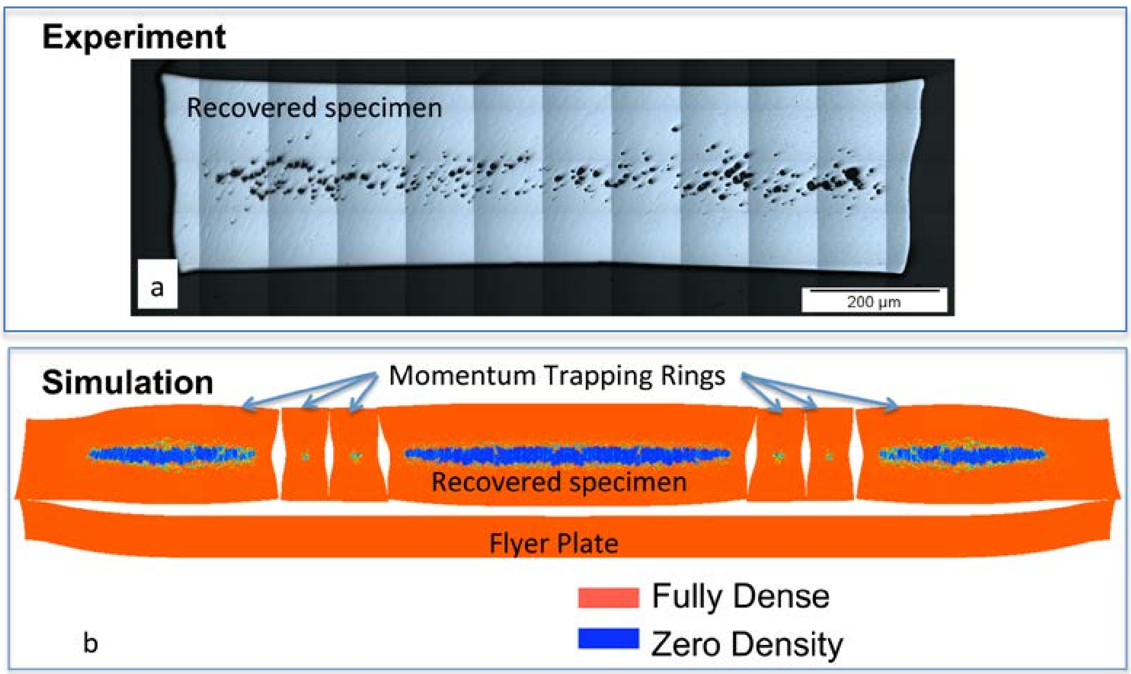


Figure 14. (a) Cross section of a shock loaded Ta specimen displaying incipient damage in the form of voids and (b) finite element simulation of this experiment. Comparison of the recovered specimen in experiment with that in simulation reveals that current models do not accurately capture the process of damage evolution**.**

Although we presently are able to perform detailed characterization of the initial and final microstructural states (see Figure 14) of the material for dynamic loading conditions, we are unable to experimentally probe the complex series of events introduced above which link the initial to the final state of the material. Our only prompt diagnostic at present is free surface velocity as a function of time (see Figure 15). Although valuable, we require new *in-situ* data in order for us to decouple the complex series of events which give us the spatially integrated result of free surface velocity. We need new tools to understand the three-dimensional link between the defect structure/microstructure and detailed loading characteristics and also new models and theoretical approaches to address this substantial problem. It is this process of physical events which our models are tasked to represent and require new experimental insight. This understanding will then be used to motivate new physically based models for micromechanical and continuum representation of these events. This understanding can also be used to design new materials and/or tailored processing of old materials for specific applications which are based upon functional requirements and known physics.

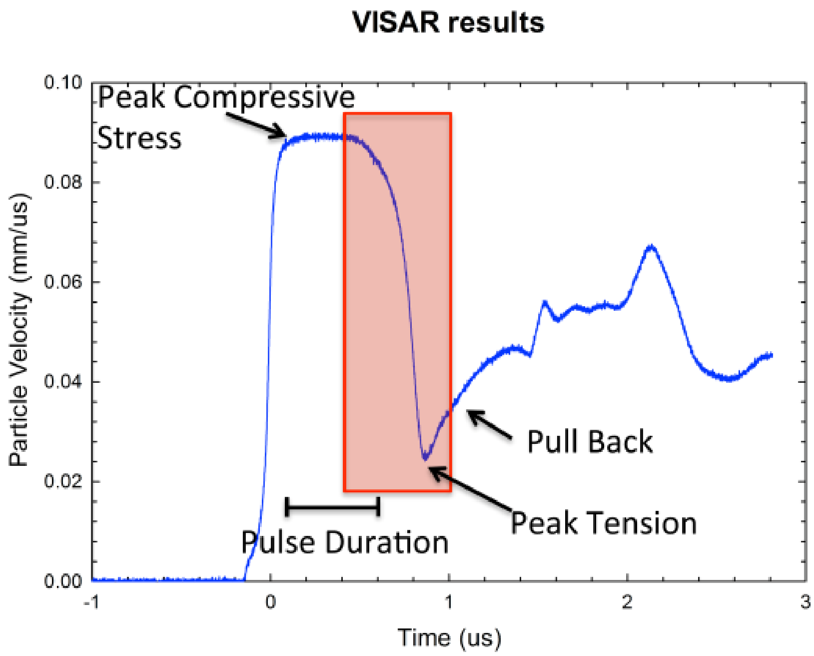
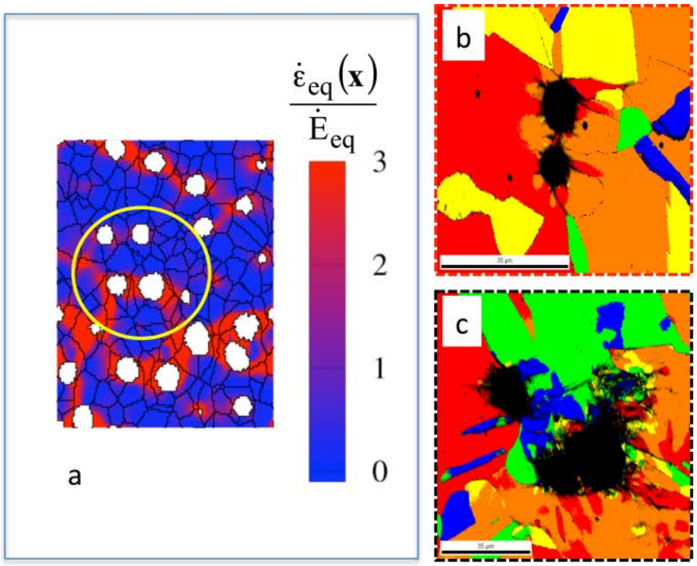


Figure 15. Velocimetry record for incipiently spalled copper. Time frame within the continuum measurement for which information about the microstructure is desirable has been highlighted by the red box. These data will be utilized to link microstructure with damage generated under peak tensile conditions.

Deformation of metals under shock loading conditions is not limited to but is thought to be dominated by slip. This type of massive slip is thought to contribute to material failure in a number of distinct ways. For example, this slip leads to alteration of the microstructure through significant reorientation of grains. This can make grains relatively hard against further deformation and prevent coalescence of voids nucleated on either side of the grain. This has been predicted and experimentally supported through post mortem characterization, as shown in Figure 16.



Min	Max	Total Fraction	Partition Fraction
2.26886	2.54993	0.082	0.085
2.54993	2.83101	0.157	0.163
2.83101	3.11208	0.104	0.108
3.11208	3.39316	0.249	0.259
3.39316	3.67423	0.371	0.386

Figure 16. (a) Fast Fourier Transformation simulation of void coalescence in a polycrystalline matrix. Circled region of the simulation indicates that voids with a propensity to coalesce are linked by grains in a soft orientation with respect to slip and those that do not coalesce are linked by grains with a hard orientation with respect to slip. EBSD generated Taylor factor maps (b and c) confirm this simulation.

Slip can also lead to non-uniformity in the way in which grains become misoriented. This is thought to lead to the stress concentrations required to nucleate voids. While we observe this in post mortem specimens, it is difficult to isolate features of shock-induced deformation due to the compressive and unloading cycles of the shock experiment. Ideally, we would like to characterize and understand the microstructural development that occurs just before and during unloading in a dynamic experiment. This region is shown schematically in Figure 15.

Future capabilities for *in-situ* damage characterization should include the ability to make time resolved misorientation measurements within individual grains of the polycrystal. This capability should include the ability to examine void formation and coalescence and concurrent velocimetry measurement for connection to previous experiments as well as future correlation of microstructure with continuum response. In addition, it is critical to use diffraction techniques to measure texture evolution in large grained specimens to examine how misorientation evolves as a function of pulse shape, pulse duration, crystal orientation, and granular neighborhood. This should be coupled with small angle X-ray scattering, 2D imaging, and ideally tomography to observe void formation. This capability will likely require a bright, high energy, short pulse, multi-frame diffraction capability and controlled synchronization of diagnostic and firing capability. This will in turn require high data throughput with multiple framing and rapid firing. Software developments will also be required to process this new class of data for physical feature extraction in automated ways.

Experimental drive platforms should include the ability to shape the profile of the wave and include a range of drive conditions. An example of the types of drive conditions such as square wave, triangular wave, Taylor wave, and ramp wave loading is provided in Figure 17.

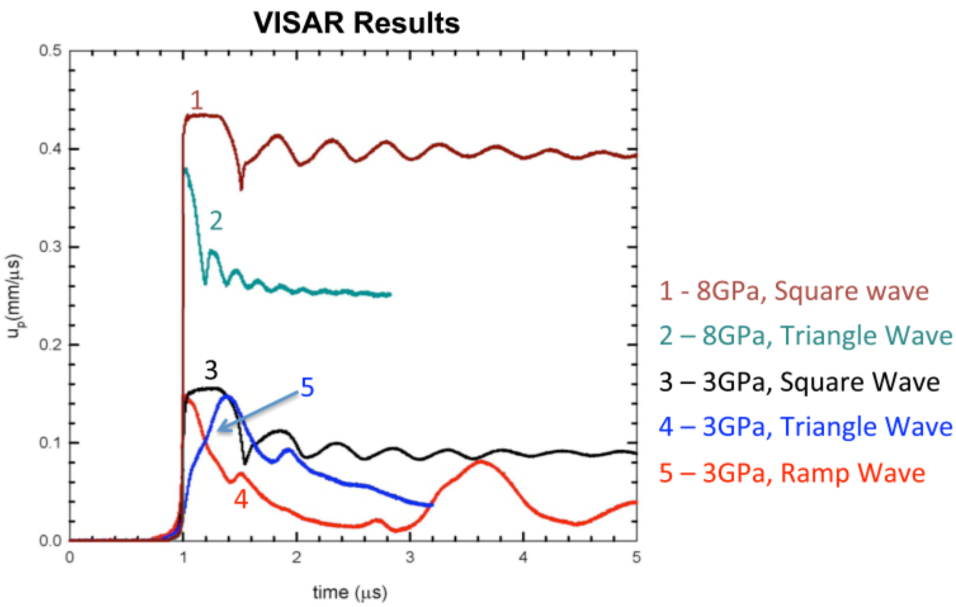


Figure 17. Velocimetry results displaying an example of the range of wave shaping experiments that would be desirable for DCS.

The drive system should also handle large specimens, on the order of mm's. Additionally, to couple with metallographic and tomographic capabilities, the drive system should include the ability to soft capture the sample post loading. Finally, concurrent development of analytical tools must be part of the DCS facility to link measurements with features of interest. Examples of such correlations could include: bulk texture to misorientation of individual grains and linking these measurements with information about dislocations and twin densities.

Linking the heterogeneous nature of polycrystalline metallic material deformation behavior to damage and failure processes in those materials remains a tremendous challenge. Not only are the materials aggregate composites in composition but phase transformations, twinning, dislocation sub-cell structures develop with deformation and contribute to potential nucleation sites for damage. This experiment is designed around the need to observe these physical events inside the material while being loaded dynamically. The length scale of observation needs to be sufficiently small to enable the geometric linking of several different simultaneous physical events within the material. Therefore, we desire to track the propagation and growth of dislocation and polycrystal aggregate heterogeneity and link these features to the nucleation, growth, and coalescence of a damage field during dynamic loading. Implicit in the aggressive nature of the experiments proposed is that the information derived from such experiments are maximized by parallel theory and model development.

Potential Impacts

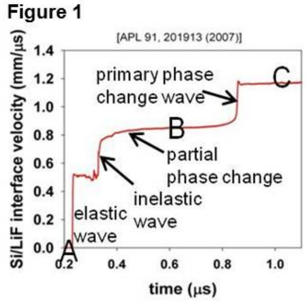
Materials are central to every national need (i e., security, energy, environment, transportation, energy security) and future technologies will place increasing demands on performance in a range of extremes: stress, strain, temperature, pressure, chemical reactivity, photon or radiation flux, and electric or magnetic fields. To lower fuel consumption in transportation, future vehicles will demand lighter-weight components with increased strength and damage tolerance. Next-generation nuclear fission reactors require materials capable of withstanding higher temperatures and higher radiation flux in extremely corrosive environments and for longer service lifetimes without failure. To enhance national defense, energy security, and counter security threats, defense agencies require the means to field protection for the populace against terrorist attack and to protect critical facilities and buildings against human or atmospheric extremes. Finally, to exploit new deep sea or space environments requires technologies constructed from materials capable of withstanding the range of operational conditions found in these locations. The only means of achieving these ambitious goals is to evolve a suite of tools and a knowledge base that is built upon the physical mechanisms that operate both in the formation and processing of the materials at their inception, but also in the means by which the microstructure responds to loads delivered in the environment in which the material must operate. At present such mathematical descriptions are derived from a limited suite of tests in a small part of loading phase space and fitted to the responses observed.

To address the primary deficit in materials' understanding requires the means to find the operating physical mechanisms that occur in the microstructure. If

High Pressure Polymorphs of Silicon

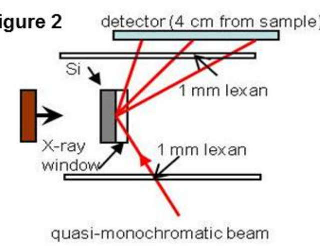
Silicon is a high strength brittle material that has several high pressure polymorphs. For quasistatic loading of silicon up to 16 GPa, four stable phases have been found: ambient diamond phase, b-Sn, Imma, and simple hexagonal.

Figure 1



Stress waves propagated through impulsively loaded silicon are complex due to the high strength and phase transformations. (Figure 1) Although the velocimetry data are consistent with the peak state being fully transformed to a high pressure phase, they cannot elucidate which of the several high pressure phases are present throughout the loading process.

Figure 2



A plate impact powder diffraction experiment, shown schematically in Figure 2, at the DCS could identify the volume fractions of each phase as the loading wave passes through the silicon.

Figure 3

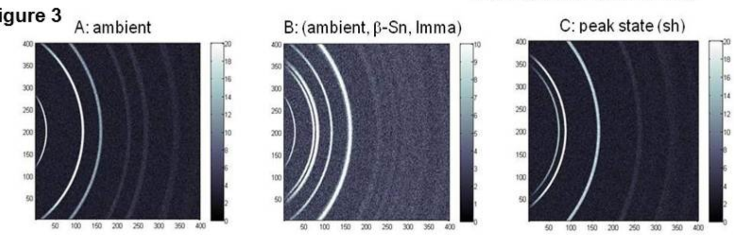


Figure 3 shows simulations for what the powder diffraction patterns might look like in A) the ambient state B) the ramp wave region and C) the peak state.

Sidebar 4. High Pressure Polymorphs of Silicon these fundamental processes can be understood, exciting opportunities to use such extreme thermo-mechanical conditions to design and manufacture new classes of materials will open up. Such advances may allow extremes such as the theoretical strength to be achieved through confinement effects on dislocations. Thus, a shift in perception and boundary conditions is necessary to bridge the gap between materials today and those theoretically achievable. Further, these processes are different across material classes. Metals respond differently than polymers and they in turn behave differently than brittle solids.

When such knowledge has been assembled, one will be able to apply the designed materials or use existing stock outside of the existing design envelopes accepted in modern engineering. This will allow extension of the abilities of materials both in service life for existing components but also to access new operating conditions for use in the field. The future will see faster, better structures capable of lasting longer times in safety to address the challenges of the modern world.

Conclusion

The current understanding of dynamically compressed solids is derived largely from real-time continuum measurements (velocity interferometry, stress gauges, etc…) and detailed microstructural examination of shock recovered samples. DCS will provide the opportunity to obtain new real-time information regarding the time evolution of microstructural heterogeneities: defects need to be identified; time evolution of defect densities and defect spatial arrangements need to monitored during the dynamic compression event; the time evolution of phase fractions needs to be monitored for materials that undergo phase transformation during dynamic compression; void initiation, growth, and coalescence need to be monitored in real-time as dynamic tension leads to failure. Such information will lead to more realistic physically-based models for the response of dynamically compressed materials.

Due to the short measurement durations and the large microstructural changes taking place on nanosecond timescales during dynamic compression, the real-time X-ray measurements on dynamically compressed condensed matter at the DCS will always be limited in comparison to what can be accomplished on static samples. Some measurement signatures at the DCS, such as the appearance of new diffraction peaks, can be correlated with high confidence to specific microstructural changes such as new phases or twin formation. Other measurement features in dynamic compression experiments, such as broadening of diffraction peaks due to microstructural heterogeneities, are more subtle; multiple microstructures may give rise to similar broadening of a given peak. For this reason, optimal use of the experimental output generated by DCS will require both knowledge regarding microstructure gained from shock recovery experiments and strong collaboration with theory and computation. Current computational techniques for simulating shock compression such as classical molecular dynamics are able to provide details on atomic motions, defect formation and phase transformation, but length scales are currently limited to a few microns and time scales are limited to a few nanoseconds. Large disconnects in either time or length scales currently exist between experimental and computational approaches. Future theory and computations must be extended to longer length and time scales such that they are able to simulate the experimental output generated at DCS. To help bridge the gap between experiments at DCS and current computational capabilities, initial experiments should be performed on ideal, well-characterized systems such as single crystals, bicrystals, or designer samples seeded with defects where voids will preferentially nucleate.

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DYNAMICS OF CHEMICAL REACTIONS

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Introduction

Chemical reactions are sensitive to a range of environmental factors, including heat, light, and pressure. One of the more common, but least understood, of these environmental factors is dynamic compression. Dynamic compression occurs during astrophysical impacts, at defect sites during mechanical failure, during high explosive detonations, and in the interiors of planets to name a few situations. Dynamic compression events often include combinations of high pressure, temperature, and large mechanical deformations; dynamic tension is not to be excluded as it often arises during such events. Chemistry under dynamic compression is a severe test of our ability to understand the fundamental quantum mechanical processes controlling changes of bonding in condensed matter. As the number of known exoplanets rapidly grows, and as the search for life within the solar system deepens by the use of chemical probes, so grows the scientific need to understand how chemistry is modified by compression. Dynamic compression events have been demonstrated to produce bulk quantities of novel nanomaterials with industrial interest; detonation synthesis of nanodiamond is perhaps the most successful to date [1]. Recently, controlled compression has been used as a way of synthesizing ultra-low density carbon with a local diamond structure. The ultra-low density carbon has properties that could be useful in quantum computing [2].

Dynamic compression is of direct relevance to the mission of the National Nuclear Security Administration (NNSA). Detonation is a self-propagating shock wave, or dynamic compression, and the resulting work drives metals during implosion. The NNSA must be able to certify the precise behavior of high explosives across a wide range of environments, and for periods in excess of 40 years. High explosive sensitivity depends on complicated mechano-chemistry that is not well understood, and detonations can achieve conditions similar to many planetary interiors (up to 40 GPa and 4000 K). The development of adequate understanding of chemical kinetics at such extreme conditions such as detonation is still in its infancy. Many of the difficulties involving explosives also apply to polymeric materials, which are often used in stockpile applications. Furthermore, polymers are used widely in dynamic experiments, notably in the capsules used in National Ignition Facility (NIF) fusion experiments.

Chemistry during dynamic compression also cross-cuts Department of Defense and other national security missions. For example, energetic materials with enhanced structural properties are currently of great interest. Improved reactive armor materials could save lives during armed conflicts. Energetic materials based on nano-components could dramatically improve the safety and mechanical properties of energetics.

Progress in understanding chemistry during dynamic compression, however, has been impeded by numerous experimental difficulties. Dynamic compression typically generates condensed phases with a disordered nanostructure. Such materials are often optically opaque, especially when chemical reactions occur. This makes optical probes of chemistry during dynamic compression challenging. In addition, the complicated chemical environment induced by dynamic compression dramatically reduces the information content of optical spectra. Although chemical simulation methods have improved significantly over the past several decades, the ns-μs timescales typical in many dynamic compression simulations are too challenging for condensed matter simulation methods such as ab-initio molecular dynamics. The importance of mechano-chemistry in dynamic compression couples the spatial scales of chemical events at about 0.1 nm, to material defects in the 1 – 100 μm range.

The X-ray diagnostics offered by the Dynamic Compression Sector at the Advanced Photon Source promise to generate a revolutionary increase in our understanding of chemical processes during dynamic compression. Bragg X-ray scattering could be used to probe the creation and destruction of crystalline phases in chemically reacting systems. Time-resolved small-angle X-ray scattering could be used to monitor the kinetics of nanoparticle formation or consumption during detonation. For instance, the precipitation of graphite and diamond-like nanocarbon has long been recognized as essential to understanding the product Equation Of State (EOS) generated from the explosive material triamino-trinitro-benzene (TATB; see Sidebar: Watching Carbon Cluster Formation and Void Collapse in Detonating Energetic Materials). Finally, dynamic X-ray spectroscopies could offer revolutionary information on chemical bond changes induced by dynamic compression. This suite of new experimental techniques promises to deliver unprecedented insight into fascinating chemical processes occurring at extreme conditions.

PRD: Mechanics Leading to Chemical Change

The chemistry panel identified two broad priority research directions for the Dynamic Compression Sector. The first of these is the study of Mechanics Leading to Chemical Change. This research direction is inspired by the scientific challenge of understanding the nature of configurational changes in solids, such as phase transformations, mechanical deformation, and creation of defects such as crystal twins, stacking faults, and slip/shear bands. There is currently little detailed knowledge of these processes, due to a lack of time-resolved experimental methods applicable to dynamic compression. These configurational changes are known to influence the transition states and the resulting chemistry during dynamic compression. The mechanisms by which configurational changes lead to chemistry, however, are not well understood.

This scientific challenge could be overcome by performing X-ray imaging and diffraction experiments at the appropriate length and time scales. This would allow evolution of defects and transition states to be directly observed for the first time. X-ray scattering experiments could allow monitoring of sites for nucleation of void collapse and hot-spot formation, voids and nano-sized phases, as well as their growth and consumption. Finally, we anticipate the simultaneous measurement of mechanical and chemical changes combining e.g., interferometry, diffraction and energy dispersive X-ray edge spectroscopies or X-ray emission spectroscopies.

As an example, single crystals of the energetic materials pentaerythritol tetranitrate (PETN) and cyclotrimethylene trinitramine (RDX) exhibit shock sensitivity (sensitivity here refers to the initiation of exothermic chemical reactions) that depend on the orientation of the crystal relative to the shock wave [3]. In the case of PETN, greater steric hindrance or resistance to "slip," has been identified as a principal mechanism. However, extension of these concepts to RDX has not proven as simple [3]. Thus, developing an ability to monitor mechanical deformation in real time, enabling the identification of the active mechanism(s), will lead to a better understanding of mechano-chemistry and ultimately, the development of a predictive capability for energetic material initiation.

Conformational changes also play a large role in photochemical pathways. Energy and charge transfer processes occur on time scales from femtoseconds to hundreds of picoseconds and thus require ultrafast spectroscopy techniques to explore physical and electronic structure fluctuations. Examining the photo-reduction/photo-oxidation conformational dynamics with these techniques would help identify intermediate charge transfer states offering insight into photosynthesis mechanisms.

Observation of charge transfer fluctuations in the photosynthetic Mn_4Ca cluster, a key reaction center complex is Photosystem II, will become possible with new dynamic characterization techniques. X-ray Raman spectroscopy would be ideal for these advanced investigations. Specifically, X-ray Raman scattering reflects the features due to the charge-transfer transitions from metal to ligand states. Elucidating ultrafast transitions from filled valence band states to empty conduction band states would provide quantitative data for phenomenological models. A thorough understanding of energy-transfer reactions is necessary for engineering new and advanced photosynthetic structures.

The DCS would meet a significant capability gap in performing the envisioned experiments. Although some experiments will be accessible with current diagnostics, others will require more advanced diagnostic development. Examples

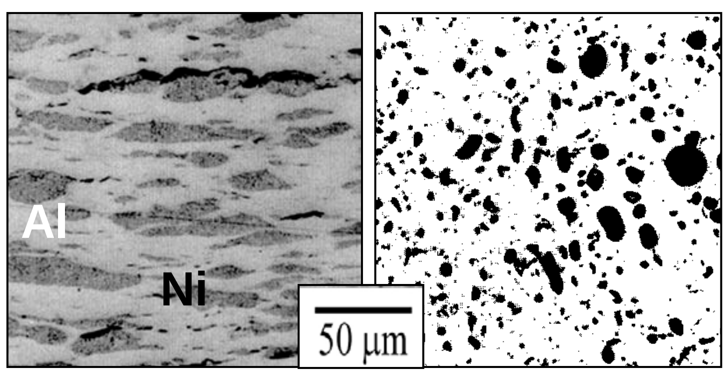

Figure 18. Complex configurations are generated when Ni and Al undergo deformation during dynamic compression experiment as shown in the figure on the left for a recovered shock-compressed sample. However, once reaction occurs, the resulting microstructure, shown on right, reveals no signature of the reaction process nor the transition states. DCS will enable the first time-resolved *in-situ* characterization of microstructure evolution coupled to chemistry.

are energy-dispersive detectors, large area detectors, pink beam optics, and X-ray Raman and polarization capabilities. Precise synchronization of the DCS beamline with dynamic compression events is a worthwhile challenge. A detonation containment chamber will be required to study energetic materials and high-energy impacts.

We anticipate that DCS would enable time-resolved imaging of configurational changes at the 100 nm to 1 μm scale. We could observe the evolution of the first stages of chemical changes caused by the influence of configurational effects, leading to transition states and final equilibrium states. These results would be vital in understanding stress-induced chemistry in solids. The measurements would validate and challenge current first principles modeling approaches to chemistry under dynamic compression. The diagnostics developed for DCS would also be relevant to other physical sciences, for instance understanding chemical processes in biological systems.

PRD: Chemical Reaction Mechanisms in Extreme Conditions

Dynamic compression experiments offer the ability to generate the widest possible extremes in pressure, temperature, density, strain, and strain-rate. In this priority research direction, the scientific challenge is to develop an understanding of the influence of extreme conditions on chemical bonds, electronic structure, and nuclear motion. Studies have shown that extreme environments induce changes in mechanism that are difficult to anticipate or predict. Important examples include the initiation and detonation of energetic materials, synthetic routes for industry, and charge transfer for energy harvesting.

Possible research directions include the measurement of chemical changes using energy dispersive edge spectroscopies or emission. We anticipate that a combined experimental/computational approach will be most fruitful in lending definitive interpretations of experimental results. The picosecond time resolution afforded by the APS could make it possible to capture transition states of chemically reacting materials under dynamic compression for the first time.

The hard X-rays present at the APS offer an excellent opportunity to examine chemical processes in extreme conditions due to their large penetration depth and due to the difference in energy between the APS X-rays and the optical emissions that occur in abundance in typical shock induced chemical reactions. A noted deficiency in current detector technology is high efficiency detectors that are both spatially resolved and energy-dispersive. Further development of these detectors and of the related tomographic spectroscopy methods will surely advance our ability to measure dynamic reactions with elemental and chemical contrast.

An excellent demonstration of chemically selective direct tomography has recently been published by Huotari, *et al.* [4]. They were able to reconstruct 3D structures using either diffraction or X-ray Raman scattering. While Huotari's technique used scanning of the sample to build up the 3D reconstruction, it could easily be simplified to provide data only along a single line through a sample. Capitalizing on the dynamic nature of a shocked sample, one could strategically position the probed axis to also provide data about the chemistry as a function of time after the passage of the shock front. In this way, a single experiment could provide information about the chemical time evolution.

An interesting chemistry problem to consider is the response of aromatic compounds to dynamic loading when compared with non-aromatic "relative" compounds. These changes are often observed through slope changes in the Hugoniot (locus of points generated by a single shock in a material starting from ambient condition). For example, benzene shows a slope change in the Hugoniot starting at a pressure near 13.3 GPa, yet the non-aromatic cousin, cyclohexane, shows a smooth Hugoniot up to 40 GPa. One difference between benzene and cyclohexane is the existence of π-bonding in benzene and the lack of it in cyclohexane. The existence of a slope change also occurs in the Hugoniot of the insensitive explosive PBX 9502, which is primarily TATB – an aromatic molecule. Thus, the use of techniques that can distinguish between molecular orbital symmetries and bonding would be useful for understanding the chemistry occurring in shock compressed aromatic species.

Industrial Applications

The DCS is applicable to a wide range of dynamic processes, including but not limited to compression. The full spectrum of DCS applications will garner significant industrial interest. The enabling unique capability of DCS, particularly in materials synthesis characterization, is a fast, precisely timed camera of deeply penetrating light. The committee envisions exciting industrial innovations from DCS studies of nanomaterials systems, super-hard materials, and energy-related applications.

Many industrial applications would utilize a full spectrum of compression drivers, and materials failure and toughening studies are perhaps most obvious among them. Phase-transformation toughening, cavitation, abrasion, penetration, and fracture are examples of dynamic mechanisms—both good and bad—that have not been adequately "caught in the act" yet may lend themselves to immediate elucidation with DCS capabilities. Super-hard and super-tough materials are of great interest to geophysicists, prospecting geologists, and petroleum engineers pressed by ever increasing technological demands for deep drilling and mining. Although armor has universal application in hostile environments, such as down-hole instrumentation, automotive safety, and personnel safety, rational design awaits fast diagnostics under dynamic impulses.

The committee foresees outstanding opportunities for industrial participation in materials synthesis not always utilizing a compression drive. As a general strategy, highly dynamic formation yields ever-greater heterogeneity at ever-finer scales, which appeals to the advantages of nanomaterials. The fast camera at DCS should provide a new, high definition view of rapidly quenched alloys and glasses, thermal sprayed coatings, and flame-synthesized materials unusually far from equilibrium. In other important synthetic processes, a dynamic drive is crucial, such as explosive forming and joining of disparate materials.

Sidebar 5. Industrial Applications. Contributed by Alan Hurd, Santa Fe Institute and Los Alamos National Laboratory

One technique that would provide these data is polarized inelastic X-ray spectroscopy, which has been used to measure the density of σ– and π-states by Nagasawa et al., in their work on graphite [5]. Measurement of the spectra of the σ and π electrons will help elucidate how pressure influences bonding in these systems. Coupling these measurements with direct X-ray absorption spectroscopies (EXAFS, XANES) will help to answer fundamental questions regarding the details and mechanisms of chemical change initiated through dynamic compression.

While dynamic compression techniques would be expected to dominate at the DCS APS beamline, static and quasi-static methods, which are complementary in the range of deformation rates and time-scales examined, can use essentially the same X-ray diagnostic methods and will greatly benefit from technical developments at DCS APS. The experimental platforms using quasistatic methods will include dynamic diamond anvil cells (dDACs) and pulsed laser heated DACs with the associated pressure/temperature metrologies. The use of quasistatic compression methods is justified by the ability to reach time scales and pressure-temperature conditions that are unattainable with conventional dynamic-compression techniques.

The DCS will close a gap in present capabilities by allowing for a suite of X-ray spectroscopies and simultaneous optical spectroscopies. We anticipate that energy dispersive detector development will be required. Multi-frame fast detectors and pink beam optics will enable adequate signal to noise in combination with unprecedented time resolution (see synchrotron techniques below).

The DCS will significantly advance our understanding of chemical reaction mechanisms in extreme conditions. We expect that under increasing pressure and temperature, chemical reaction rates will dramatically accelerate. For instance, DFT models predict an H_2O molecule lifetime for shock compression at 40 GPa of less than 50 fs [6]. While DCS is not expected to resolve chemistry at these timescales, the DCS will challenge DFT first-principles simulations by providing the first dynamical images of chemical reactions in extreme environments. We anticipate that a wide range of novel experiments could be performed on the DCS. We consider some promising examples below:

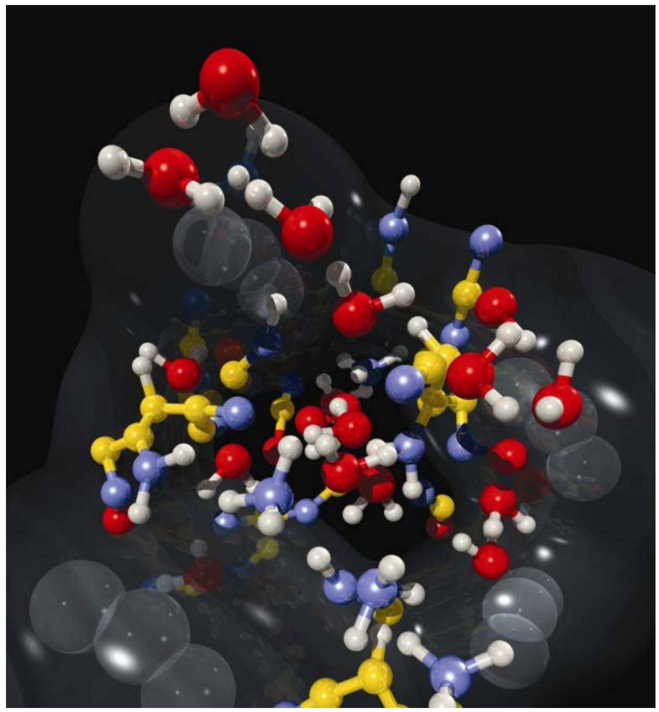

Figure 19. Chemical reactions during dynamic compression are often characterized by complex mixtures of short-lived species. In this figure a simulation of shocked nitromethane is shown.

1. Synchrotron techniques for studying the dynamics of chemical reactions

A suite of synchrotron techniques, complimentary to the enabling capabilities at free-electron laser facilities, can be developed and used for studying configurational changes in dynamically compressed materials at DCS. To take advantage of the APS source, the DCS should focus on the effective use of the wide energy available with synchrotron radiation and the high repetition rate (~MHz) of the X-ray pulses. Furthermore, to enable these techniques, all beam line optics components need to be designed to deliver the maximum possible flux.

2. *White/pink beam X-ray spectroscopy*[1]

X-ray spectroscopy allows the measurement of chemical bonds, the associated electronic structure, and charge-transfer states, etc. From the brilliant source at the APS, it is feasible to use white/pink beam together with energy dispersive detectors for single bunch X-ray near edge absorption spectroscopy (XANES) and X-ray emission spectroscopy (XES) measurements. These two techniques should be considered as primary spectroscopy techniques that together with polarization capability at DCS enable unprecedented measurements of dynamic chemical changes to be made. In addition, efforts for developing time resolved X-ray inelastic scattering (IXS) technique (sometimes incorrectly called X-ray Raman) should be pursued. However, due to the relative weak cross-section in IXS, new developments in energy dispersive detecting efficiency and multiple detector configurations will be required to enable IXS probes in real time and on a regular basis.

3. *X-ray imaging*

Density contrast, phase contrast, and Laue imaging provide detailed information critical for understanding configurational changes associated with dynamic compression experiments, including mechanical deformation, crystal twins, stacking faults, slip bands, and phase transitions. X-ray imaging will be a primary technique at DCS. Developing X-ray imaging will require answering key questions such as whether a longer beamline and/or ancillary optics are needed for a larger field of view, considering the typical sample size (1-10 mm) to be utilized in dynamic experiments. Typical spatial resolution X-ray imaging the full width of such objects is 1-5 microns. In order to have enhanced spatial resolution for fine structures (e.g., clusters, voids, grain boundaries), the transmission X-ray microscope (TXM) technique may be employed, which can have high spatial resolution (down to 10 nm) at cost of field of view (as small as tens of microns).

4. *Small-Angle X-ray Scattering/Wide-Angle X-ray Scattering (SAXS/WAXS)*

SAXS and WAXS experiments may be performed simultaneously by adopting multiple detectors for single bunch time resolved measurements. The capability will allow for obtaining integrated dynamic information on phase transition, detonation, void evolution, particle growth, etc. This is essential to connect phenomena occurring on ~100 nm scales to phenomena on molecular or atomic scales that will be interrogated with diffraction. Such a setup could either incorporate area detectors and a high-speed chopper to isolate single pulses [8], or a high-speed silicon annular detector [9]. Both technologies have been developed and implemented at the APS.

5. *White beam diffraction*

White beam in Laue and/or Bragg geometry is well suited in dynamic experiments, as it provides the full scattering profile for the sample being interrogated, to maximize the

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[1] A white beam is usually referred to entire energy spectrum, 4-150 keV; while a pink beam has a typical bandwidth of 0.5-1 keV.

information available on a per shot basis. In fact, single bunch white beam diffraction measurement has been well developed in protein crystallography. The technique provides detail information on microscopic properties including crystal chemistry, lattice disorder, crystal deformation, twins, stacking faults, mosaicity etc., that can be used to study the mechanics that lead to chemical change.

6. Synchronization and Conformational or Chemistry Movies

Gas gun experiments on the DCS will require “movie mode” data acquisition analogous to oscilloscope traces and streak cameras, where rapid acquisition of a window of single shot data is synchronized to the compression event via an electronic signal from, for instance, a strain gauge. The jitter on this timing signal must be less than the desired time resolution. This scheme requires a continuous X-ray pulse train from the synchrotron to periodically illuminate the sample, synchronized to a fast framing camera to obtain data from each X-ray pulse, or a well-defined number of pulses per image. The time resolution of such a scheme is ultimately limited by the repetition rate of the X-ray pulses, but in practice will be more limited by the frame rate of the acquisition camera, which depends on the acquisition rate of CCD electronics and the decay rate of the scintillation phosphor, where faster decay rates are typically traded off against signal to noise in the detected image. State of the art cameras can achieve MHz framing rates, with faster rates achievable by multiplexing either cameras or regions of the image plane with a single camera. This is a research direction that must be pursued to enable “movie-making” with DCS

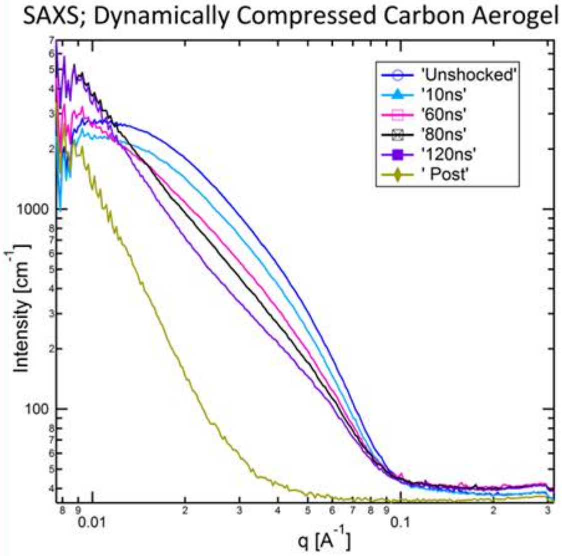
Generally, experiments that require extremely high time resolution (i.e. as high as the duration of an X-ray pulse, less than 100 ps) would benefit from a pulsed laser compression drive to accommodate low-jitter synchronization of the probe to the drive. Such a drive has been used at X-ray sources for decades and can be well synchronized to the probe X-ray pulse. At very high (sub-μs) time resolution, framing cameras must be multiplexed to obtain “movie mode” data acquisition. Alternatively, for sufficiently repeatable laser drive and sample conditions, distinct pump-probe experiments at many delays may be superposed to build up an interleaved aggregate time history of the material dynamics, as is commonly used in time-resolved spectroscopy. Note, though, that such experiments are intrinsically averaged and may not be appropriate for situations where the detected signal depends on parameters that are difficult to control, such as the specific grain distribution, or the distribution of dislocations. Typically, such parameters would only be relevant for imaging experiments – diffraction experiments are already averaged over large (with respect to the unit cell) volumes.

Watching Carbon Cluster Formation and Void Collapse in Detonating Energetic Materials

Major challenges in detonation science and associated modeling can be resolved with proposed capabilities at the DCS. First, explosives that have solid carbon condensates in the detonation byproducts cannot be modeled without including a time dependent behavior such as diffusion-controlled carbon particulate growth or other kinetic behavior. The time scale in which the carbon condensation is formed after the detonation front passes is required for developing an understanding both the physics of this phenomenon, and for an evaluation of the amount of energy released during exothermic growth of carbon clusters. These dynamics will be explored on the nanosecond to microsecond time scale. Second, elucidating the dynamics of void collapse will provide insight regarding hot spot formation during HE detonation. Both initiation sensitivity, as well as propagation of detonation in various explosives, depends on voids that models predict are in the ~100 nm to ~1 um size regime. The endstation will ultimately be able to interrogate void collapse at the detonation front with better than 100 ps temporal resolution.

Void collapse and the condensed carbon nanoparticles produced during the detonation of high explosives are not easily measured: the detonation front is optically opaque and therefore not amenable to optical interrogation. Early Russian studies using dynamic small-angle x-ray scattering measured the condensed carbon nanoparticles; however these results have been limited to low signal-to-noise and microsecond temporal fidelity[1]. The Advanced Photon Source currently has over an order of magnitude shorter pulse-length (<100 ps) and typical APS undulators, especially in polychromatic pink-beam mode, will generate orders of magnitude more photons per pulse. In addition to diffraction, small-angle x-ray scattering is an ideal technique to study these two phenomena. As an example, void/structure collapse in carbon foams on the requisite length scales has recently been performed at the APS with dramatic, high-fidelity results (see Figure)[2]. This proof-of-principle experiment at the APS points to the future success and potential of DCS@APS.

SAXS; Dynamically Compressed Carbon Aerogel



Time-resolved SAXS using ~100 ps single pulses at the APS.

The temporal resolution is ultimately limited by the APS pulse width, and in the near-term, detector temporal capabilities. Carbon soot kinetics is reaching an opportune time for interrogation: fast SAXS detectors, such as the silicon annular detector[3] are now able to measure at intervals as fast or faster than 3.67 microsecond intervals between hybrid-mode single bunches. The kinetics is thought to occur on the microsecond

Continued on next page…

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Watching Carbon Cluster Formation and Void Collapse in Detonating Energetic Materials
(Continued)

timescale; thus multiple data points could be obtained from a single detonation event. As detector technology improves, we may eventually be able to directly utilize all bunches in, e.g. a 24 bunch mode with ~34 ps pulses separated by 153 ns. To achieve comprehensive temporal data, synchronized beam choppers and multiple acquisitions/shots with interleaved data can give resolution approaching the bunch temporal resolution of the particular mode used by APS. Interleaving multiple shots will be required to interrogate void collapse and hot-spot formation. These events occur on much shorter, ns timescales.

The capabilities immediately required are an endstation incorporating a detonation tank capable of containing full detonations that are started through either gas-gun shock initiation, or detonator and booster-based initiation. For these tasks, the beamline requires a fast small-angle scattering capability with the ability to be tuned to cover scattering angles between ~10^{-3} and ~1 $Å^{-1}$.

Sidebar 6. Watching Carbon Cluster Formation and Void Collapse in Detonating Energetic Materials. Contributed by Trevor Willey, Lawrence Livermore National Laboratory

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[7] A white beam is usually referred to entire energy spectrum, 4-150 keV; while a pink beam has a typical bandwidth of 0.5-1 keV.

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TIME RESOLVED DYNAMIC PROCESSES IN MATERIALS BEYOND THOSE GENERATED BY COMPRESSION

Panel Chairs

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Introduction

The primary mission of the Dynamic Compression Sector (DCS) is to enable time-resolved, *in-situ* experiments to achieve a fundamental understanding of condensed matter phenomena under dynamic loading. Experiments in these regimes requiring the most extreme conditions of pressure and/or loading rate will be performed using shock compression facilities in the downstream hutch of DCS. Many of the proposed dynamic compression experiments are complex and will require considerable setup time, resulting in low repetition rates (*e.g.,* ~10 per day for gas gun experiments). This means that a large fraction of the beam time available to the sector could profitably be used for other experiments in an upstream hutch in the intervals between dynamic compression experiments.

This panel was tasked with identifying the most scientifically beneficial and effective ways to use this beam time. The overarching conclusion from the panel regarding the use of the upstream hutch is that the research needs to be complementary to the primary mission of DCS, which is to understand the behavior of materials under dynamic loading. We identified three approaches to optimize the use of the available beam time:

1. Detailed three dimensional microstructural characterization of materials to be examined in single event experiments;
2. Dynamic experiments under conditions complementary to the primary dynamic compression experiments in the downstream hutch or;
3. Technique and diagnostic development including the development of new techniques such as stereoscopic imaging

Besides scientific complementarity, it was important that these experiments take the best possible advantage of the expected properties of the source, tailored as it will be for the needs of the dynamic compression experiments with ultra-fast high-resolution detectors. Finally, we also sought to identify areas where the upstream hutch activities could contribute to the mission of the Advanced Photon Source as a whole.

Complementing the Primary DCS Mission

In this section, we comment briefly on the three approaches (listed above) by which the upstream hutch operations can complement the primary dynamic compression mission of DCS.

Full Characterization of Dynamic Compression Specimens

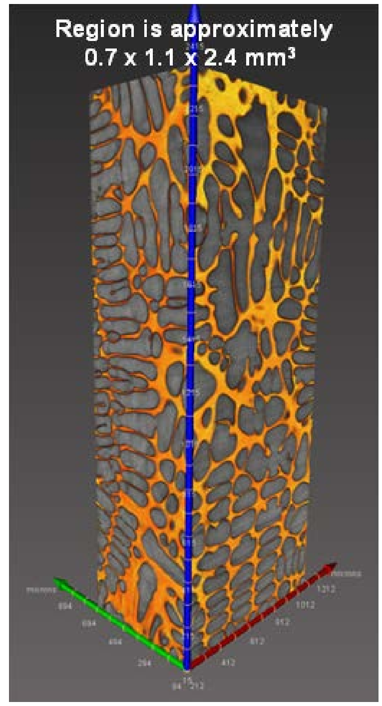


Figure 20. X-ray tomograph of an Al-Cu alloy. The interdendtitic eutectic regions are colored orange, and the Al-rich dendrites appear gray in color. *(Courtesy of B.M. Patterson, A.J. Clarke, and J.C. Cooley, LANL)*

One area in which the dynamic compression community has frequently fallen short is in adequate characterization of materials that are examined. Sample pedigrees have not always been fully documented, which makes comparison of results between different dynamic drivers and from different institutions challenging at best. Given that the number of shots of the primary compression drivers at DCS will be limited, it is essential that every shot count. For instance, it is within the power of DCS management to require a full suite of chemical and microstructural characterization capabilities such that every sample that is tested at DCS has a known and fully documented pedigree. Critical aspects of the microstructure, including bulk composition, grain size, and the presence of second phase precipitates or inclusions, should be quantified. Doing so would ensure that the results obtained from the dynamic compression experiments are as reliable and reproducible as possible.

Some microstructural characterization can clearly be done ahead of time at the home institution, or via collaboration. Other techniques will require synchrotron radiation and thus will need to be performed at the APS.

One possibility would be to establish a suite of characterization tools in the upstream hutch of DCS for non-destructive 3D reconstruction of sample microstructure (Figure 20). Possibilities in this regard are discussed in more detail below. It is important to note, however, that the X-ray beam characteristics that would be optimal for 3D characterization (e.g. high coherence, small spot size, high energy, and good energy resolution) may not be entirely compatible with the primary DCS need for a high flux, moderate energy (15-35 keV) beam. This is not to say that good data could not be collected in the front hutch with the beam characteristics proposed for DCS. But if front-hutch microstructural characterization is to be a priority, we urge that there be some flexibility in the beam properties to optimize its characterization capabilities.

Although the upstream hutch can provide much-needed quantitative data, we recognize that it is not feasible to perform all aspects of microstructural characterization there. This capability gap can be accommodated through experiments conducted in other sectors at the APS. For this to be possible, some organizational and administrative changes may be necessary at the APS. For example, it is probably not reasonable to expect experimenters to make multiple beam time applications, and potentially multiple trips to the APS, to enable this type of characterization. We therefore recommend that the beam time application and allocation process among multiple beam lines be streamlined as much as possible.

Experiments under a Wider Range of Conditions

The dynamic compression experiments in the downstream hutch will use compression drivers designed to provide the highest possible strain rates or shock pressures. There are, however, many dynamic experiments that one could contemplate that would not reach these extremes yet be very valuable to the DCS mission. Possibilities for these other drivers include:

- Dynamic loading at lower strain rates (*e.g.*, dynamic diamond anvil cell, Kolsky-Hopkinson bar, fatigue, creep)
- Different loading conditions (*e.g.*, tension, torsion, Charpy impact, or multiaxial)
- Rapid heating (e.g., pulsed laser)
- Magnetic fields
- Electric fields

One appealing aspect of these experiments is that they could be tailored to use the same characteristics of the DCS X-ray beam, especially its high flux, that make it well suited for the *in-situ* dynamic compression experiments. Additional discussion of the possibilities in this area is described below, in our **Priority Research Directions**.

Technique and Diagnostic Development

The third area of complementarity we identified is using the upstream hutch beam time for development of techniques that could ultimately be applied in the downstream hutch, or elsewhere at the APS. Taking advantage of the ultra-fast detectors, the upstream hutch could be used to develop faster, three-dimensional, CT techniques optimized for the detectors. This capability is essential for the deformation and failure and the phase transition dynamics priority research directions described below. A second example is the development and implementation of stereoscopic imaging. Going beyond the static imaging techniques described above, the source characteristics of the APS make it well suited for real time, *in-situ* studies of phenomena with characteristic length scales on the order of microns, and time scales on the order of microseconds or shorter. In particular, the high repetition rate of the APS (271 kHz, vs. ~100 Hz for LCLS) allows for continuous imaging of dynamic processes with sub-microsecond intervals between frames (depending on the electron bunch structure and the contrast provided by the sample). It is not surprising; therefore, that high-speed imaging has become a major focus for several sectors at the APS.

One option, which has been discussed at the APS but not implemented, is the possibility of using dual canted undulators on a single beamline to provide two high-brightness beams incident on the sample at different angles. In principle, if each of those beams is used to image the specimen separately, the results can be combined into stereoscopic images of the specimen. Although this approach would not yield the kind of complete 3D structural information available from X-ray tomography, having any depth information at all would be a tremendous advance over the 2D projections typical of high-speed imaging. Such a capability would be, to our knowledge, unique in the world.

As an example of the kind of investigation this capability would enable, consider crack propagation through a heterogeneous microstructure, such as a composite material. With stereoscopic imaging, one could watch the crack tip advance through the material and observe its interaction with the microstructure. Detailed observations could be made, for instance of fiber bridging behind the crack tip. These kinds of observations have been made of fatigue crack propagation in a static tomography mode [1], but never for dynamic crack propagation.

We note that the kind of dual canted undulator arrangement necessary for stereoscopic imaging may not be compatible with the source characteristics desirable for the primary compression experiments in the rear hutch. Nevertheless, we believe that the research directions that would be enabled by a stereoscopic high-speed imaging capability are so compelling that the possibility should at least be explored.

Opportunity for Accelerated Discovery

Full materials characterization (T. Hufnagel)

As discussed above, the high opportunity cost associated with dynamic compression experiments in the downstream hutch makes it essential that those specimens be fully characterized prior to those experiments. Clearly, a wide range of characterization techniques is possible, and different techniques will be appropriate for different kinds of specimens. Here, we focus on hard X-ray techniques that could, in principle, be implemented in the upstream hutch at DCS for static measurements of structure.

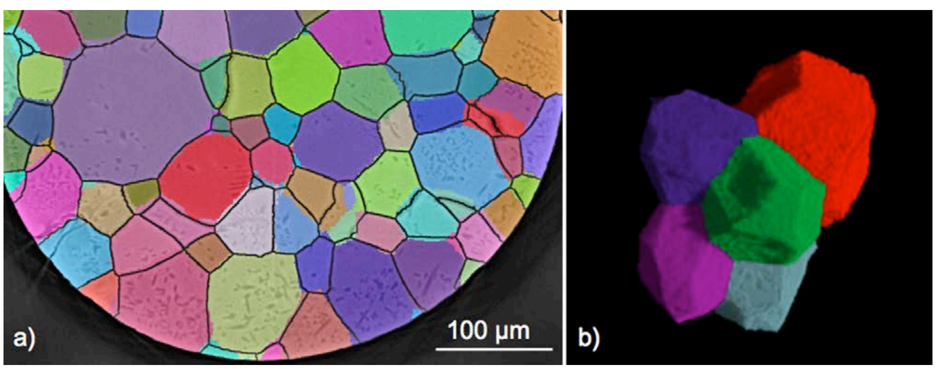


Figure 21. (a) Overlay of grain reconstruction in a titanium alloy from 3D X-ray microscopy with grain boundaries determined from phase contrast tomography (black lines) (b) 3D rendition of a small grain cluster. *Figure from [Ludwig 2009]*

X-ray imaging techniques have many advantages, among the most important of which is that they are non-destructive, raising the possibility that a sample could be extensively characterized by imaging prior to a dynamic compression experiments. Depending on the particular imaging mode, many aspects of structure can be addressed, including density, chemical composition, crystallography and strain, and the presence of interfaces [3]. In particular, the coherency of radiation from the APS makes it possible to obtain image contrast based on the phase shift of the transmitted beam, rather than differential absorption. *Phase-contrast imaging* is exquisitely sensitive to the presence of interfaces, for example, which might reveal the presence of defects, which could lead to premature failure of specimens under dynamic compression. Similarly, one could imagine full 3D (tomographic) characterization of heterogeneous materials prior to dynamic loading.

A related technique is *3D X-ray diffraction microscopy*, which, because it is based on diffraction, is capable of probing the crystallographic orientation of individual grains in a polycrystalline structure, along with their shapes (Figure 22) [4]. The information is similar to that obtained by serial sectioning techniques using electron backscatter diffraction (EBSD), but with the significant advantage that the X-ray technique is non-destructive. This raises the tantalizing prospect that one could run a multi-scale simulation of the mechanical response of a specimen to a dynamic loading, using the actual microstructure of the specimen to be examined as input to the model. This, combined with the downstream hutch capability of DCS to track the evolution of the structure of a specimen *in-situ* during a dynamic compression experiment, would allow for unprecedented fidelity in comparing the results of multi-scale simulations to real-world behavior of materials.

Priority Research Directions

The discussion above notwithstanding, we believe that the most beneficial use of the upstream hutch at DCS would be to explore dynamic behavior of materials under conditions other than those provided by the primary compression driver in the rear hutches. In this section, we describe several promising lines of investigation, which differ somewhat in their scientific objectives, but also in their choice of drivers. Because it will not be possible to implement all drivers that might be desired, judicious choices will have to be made as to which lines of investigation have the highest priority.

Figure 22. ParaDIS simulation of dislocation interactions. (*Figure courtesy of A. Schwartz)*

PRD: Deformation and Fracture of Materials (A. Schwartz, T. Hufnagel)

Throughout the evolution of materials science, the fundamental tenet of materials—the structure–properties-processing–performance relationship—has remained the same. At the heart of this tenet is that the microstructure, which for crystalline materials consists of overall and local composition, crystal structure of the matrix and secondary phases, morphology of the grains, and the interfaces between grains or between the matrix and precipitates dictate the properties and thus performance of the material. Composition and processing are used to control the microstructural parameters, which in turn control properties and performance, for example deformation and failure performance of materials.

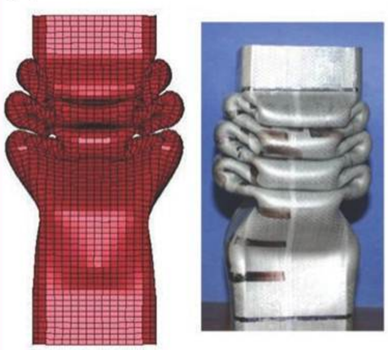
Over time, these ideas have naturally evolved into the Integrated Computational Materials Engineering (ICME) paradigm, the essence of which is to "develop materials models that quantitatively describe processing-structure-property relationships for use by the engineering community" [5]. Although the ICME paradigm includes quantitative predictive models of materials properties, it goes further by incorporating modeling of materials processing as well as the validation of material performance over its lifetime of service.

In order to achieve these goals, coupling experiments and computational modeling is essential and the experiments must be on the time and length scales of simulations. Increasing computational power enables simulations of longer real times and larger computational cells. While high-performance computing continues to extend the boundaries of time and length scales available for simulation, significantly extending the time scale is problematic. Experiments must make up the gap, necessitating better spatial and temporal resolution. With small probe areas, variable energy, and fast, high-resolution detectors, researchers conducting deformation and failure investigations will probe the microstructure at unprecedented levels.

The rate of development of robust materials models is often proportional to the rate of experimental capabilities as well as the power of high performance computers. Materials simulations unthinkable just 10 years ago are now quite routine on today's high performance computers. Molecular dynamics simulations of 10s of billions of atoms are now commonplace. Specialized codes, like the dislocation dynamics code ParaDiS, can model the generation and interaction of multiple dislocations, implicitly involving interactions among many thousands (Figure 23). To provide input for material model development, the deformation and failure community needs high-resolution data focused on specific materials responses.

Automotive Materials

Metals used in automotive structures can encounter moderate-to-high strain rates both during forming operations and during crashes. Advanced multi-scale models of materials performance require detailed microstructural information both as input and for validation. For example, crashes can subject metals to global strain rates on the order of 50 s^{-1} with local strain rates exceeding 500 s^{-1}. These rates are considerably lower than those proposed for the rear hutch at DCS, but could profitably be explored in the upstream hutch. This strain rate regime has been largely unexplored, but there are a host of important problems to study. For example, aluminum has been reported to transition from strain-rate-softening to strain-rate-hardening at moderate strain rates [1].



Comparison of finite element model (left) and experimental testing (right) of axial crushing of hydroformed aluminum alloy tubes. Figure from [2].

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2. Williams, B.W., Simha, C. H. M., Abedrabbo, N., Mayer, R. and Worswick, M.J. Effect of anisotropy, kinematic hardening, and strain-rate sensitivity on the predicted axial crush response of hydroformed aluminium alloy tubes, *International Journal of Impact Engineering* **37**, 652-661 (2010).

Sidebar 7. Automotive Materials

Materials models require quantitative data at all length and time scales, starting with the earliest stages of plastic deformation and extending all the way through failure.

Fracture of materials is a complex, multi-scale process that is dependent on all aspects of the microstructure, including, bulk and local composition, impurities, morphology and distribution of second phase precipitates and inclusions, grain size, morphology, and grain boundary character distribution, texture, residual stress, and many other factors. It also depends on the externally imposed loading, including loading rate and loading path. Developing a complete predictive capability for deformation and failure of all materials will require concurrent advances in material model development, high performance computing, and high resolution experiments like those possible at DCS. Dynamic experiments that probe the structure (stress-induced phase transformations), strain (at precipitates, grain boundaries, and crack tips), crack velocity, shear localization, and void growth rates will be enabled by the upstream hutch at DCS.

- *Single crystals:* Onset and evolution of shear localization, formation and evolution of subgrains, subgrain rotation, void nucleation and growth, and void linkage and failure.

- *Polycrystals:* Evolution of texture by grain rotation, evolution of stress state, stress state around second phase particles and/or inclusions, stress-assisted or strain-induced phase transformation (*e.g.* TRIP steels), void nucleation and growth, and void linkage and failure.

- *Brittle materials*: Microplasticity, crack initiation and growth, and fracture.

- *Granular materials:* Particle flow, compaction, particle rotation, fusing, and fracture of particles.

Each of these experiments will provide unprecedented levels of detail beyond any experiments conducted to date and, if properly executed, should provide for direct implementation in multi-scale computational models of deformation and fracture processes, especially phase field models.

PRD: Phase Transition Dynamics (A. Clarke, W. Evans)

The ability to predict and control microstructure evolution to create materials by design requires *in-situ* monitoring during processing, since post-mortem examination does not generally reveal how critical processing parameters define the final microstructure. Tremendous opportunity will exist at DCS to create microstructures by design with application-tailored properties, using directed synthesis and processing. DCS will also provide opportunities to explore the fundamentals of phase transformations and kinetics, including nucleation, growth, and microstructural evolution.

A generalized pressure-time-transformation diagram (Figure 23) illustrates the influence of pressure and time (compression rate) on the formation of thermodynamically stable, metastable, and amorphous phases. In addition to the examination of phase transformations at pressure, unprecedented observations of phase transformations at non-ambient temperatures will be possible at DCS.

As an example of the microstructural characterization that is possible *in-situ* during microstructural evolution, Figure 24 shows synchrotron X-ray radiography images collected during continuous heating and cooling of an Al-Cu alloy. DCS will permit direct observations and control of dynamic phenomena during melting and solidification as well as solid-state phase transformations. It will also be possible to examine the influence of other drivers (*e.g.*, electric or magnetic fields) or extreme environments.

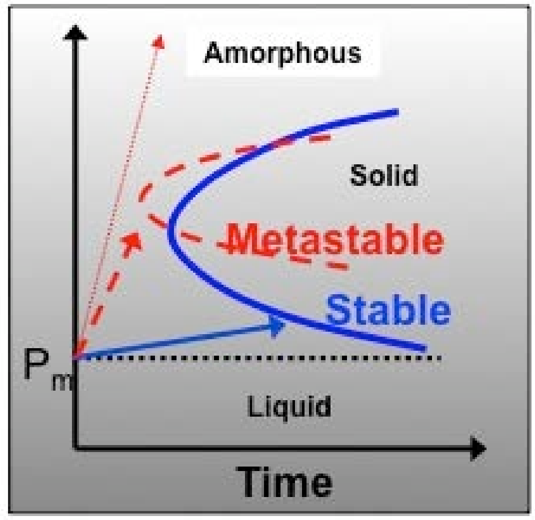


Figure 23. Generalized pressure-time-transformation diagram. The slope of the trajectory in pressure-time space denotes the compression rate. Slow compression rate results in a thermodynamically stable phase, whereas rapid compression rate results in metastable or amorphouse phases. *(Courtesy of W.J. Evans, LLNL)*

To achieve the creation of microstructures by design through directed synthesis and processing and to further our understanding of phase transformation kinetics, several important research directions are envisioned. These directions include *in situ* monitoring of microstructure evolution by diffraction, imaging and tomography, and spectroscopy, with the appropriate temporal and spatial resolution. The simultaneous characterization of phase transition dynamics using a suite of complementary probes is also of interest. New experimental findings from investigations of phase transition dynamics at DCS will also enable multiscale models of phase transformations from the atomic to the microstructural scale, allowing for predictive capability over the length scale relevant for phase transformations and microstructure evolution.

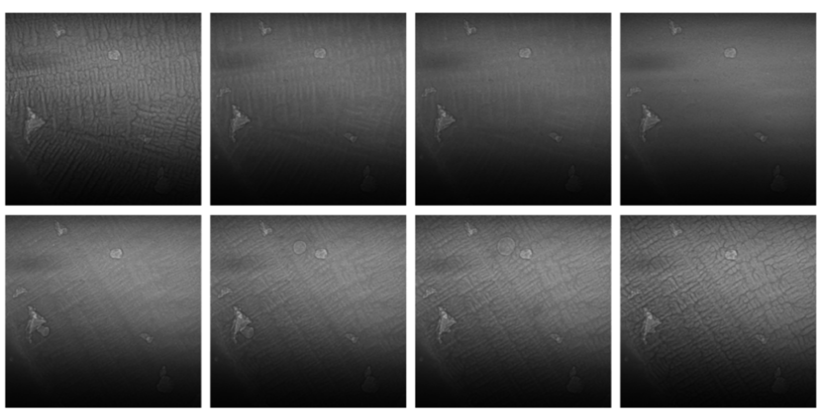


Figure 24. Images obtained during continuous heating (upper images) and slow continuous cooling (lower images) of a 100 micron thick Al-Cu sample using synchrotron X-ray radiography at APS beam line 32-ID-C. The field of view is approximately 1.4 x 1.4 mm^2. *(Courtesy of A.J. Clarke, J.C. Cooley, B.M. Patterson et al., LANL; W.-K. Lee, K. Fezzaa et al., APS)*

To facilitate the study of melting and solidification (liquid-solid phase transformations) and solid-state phase transformations and kinetics using DCS, the following experimental capabilities are desirable:

- High spatial resolution (less than 100 nm) and high flux (to enable temporal resolution better than 100 ns) for imaging, diffraction, and spectroscopy
- High-speed, high-efficiency, high-resolution detectors
- The ability to process large data sets
- Moderate-to-high X-ray energies for the examination of bulk materials.

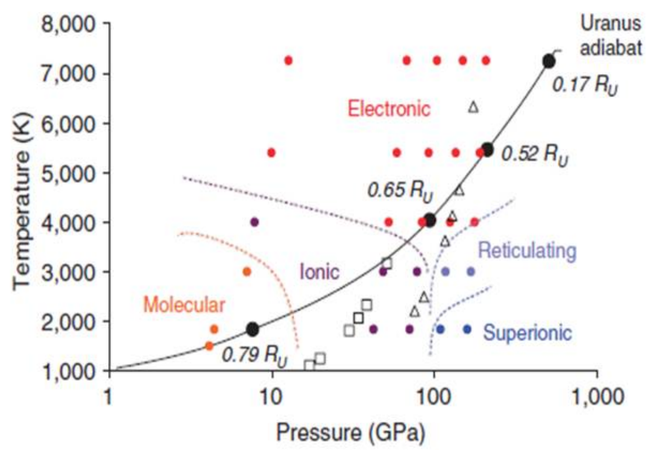
We note that these requirements are largely compatible with the beam characteristics planned for DCS to enable the time-resolved compression experiments in the rear hutch.

PRD: Chemical Reaction Dynamics (A. Goncharov)

Chemical reaction dynamics and kinetics are an example of a fascinating new frontier for high-pressure science. Under pressure, most chemical bonds change to favor polymeric states [6]. Moreover, many examples of charge transfer, ionization, molecular disproportionation and electronic structure changes have been documented [6, 7], but these phenomena remain incompletely understood. Other challenges for studies of materials under extreme environments include microscopic reaction mechanisms, formation of molecular

Planetary Chemistry

Planetary ices (H_2O, NH_3, CH_4), earth volatiles (CO_2, CH_4), and their mixtures (*e.g.*, CH_4-H_2O) are predicted to form polymeric phases or transform to superionic states at moderate compressions (>0.5 Mbar); at further compression/heating they are expected to ionize (>5000 K) [1-3]. The existing data, however, are largely inconclusive, and the kinetics and dynamics of chemical transformations are unknown, particularly for mixed ices. This information is important for understanding the behavior of these complex mixtures as a function of depth in Earth and planets, especially super Earths and super Neptunes outside of our solar system. DCS provides a unique opportunity to study these kinds of systems.



P–T space diagram of synthetic Uranus. The filled circles are from the FPMD calculations. Opened squares are the measured P and T after the initial shock. Opened triangles are the estimated P and T reached in the reverberating shock. The colors represent different phases of the fluid. The black line is the planetary adiabat of Uranus [4].

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Sidebar 8. Planetary Chemistry

alloys, and reactivity at interfaces. An improved understanding of these phenomena would benefit a range of disciplines, including chemistry, physics, materials science, and earth and planetary sciences.

The capabilities of DCS will allow probing material states using a range of X-ray techniques, enabling technically demanding research under conditions of dynamic extremes, which were previously unavailable due to the lack of a suitable dedicated facility. For example, time-resolved diffraction and spectroscopy measurements can be made on statically pre-compressed or pre-heated specimens, subjected to periodic mechanical loading in a dynamic diamond anvil cell [8] or by laser drives [9,10]. *In-situ* diffraction measurements can reveal changes in crystal structure, while spectroscopy (EXAFS and XANES) provides information about electronic structure. Moreover, *in situ* X-ray measurements can be complemented by simultaneous optical spectroscopy diagnostics including, spectro-radiometry, optical and vibrational (*e.g.* CARS) spectroscopies.

A wide range of problems is ripe for investigation. For example, data on the structure and electronic properties of planetary ices are, for the most part, inconclusive. Similarly, formation of metal hydrides under pressure is common [11] yet the kinetics of this industrially and scientifically important process are largely unexplored. As a final example, microscopic details of the reactivity of energetic materials are mostly unknown, because direct measurements under appropriate extreme conditions have not been performed. *In-situ* time-resolved diffraction and spectroscopy measurements on nanosecond time scales would provide vital information about the reaction routes and intermediate products. This kind of information is vital for development of predictive models of the chemical processes in energetic materials under detonation conditions and, ultimately, for creation of energetic materials with desired properties.

Conclusion

The relatively low repetition rate of experiments using the primary compression drivers in the downstream hutches of DCS creates opportunities to use the beam in complementary experiments in the upstream hutch. Three broad areas of complementarity are envisioned: (i) detailed three-dimensional microstructural characterization of materials to be tested in the single event experiments, (ii) dynamic experiments under conditions complementary to the primary shot dynamic compression experiments in the downstream hutch and (iii) technique and diagnostic development including the development of new techniques such as stereoscopic imaging. Of these, the most compelling are the use of dynamic drivers not available in the downstream hutch, including dynamic loading under different strain rates or strain states, or entirely different drivers such as laser heating or magnetic fields. Our priority research directions, which emphasize these different drivers, include deformation and fracture of materials, phase transition dynamics, and chemical reaction dynamics. Each of these areas is complementary to the primary science mission of DCS.

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INSTRUMENTATION CHALLENGES

Panel Chairs
Dean Haeffner, *Argonne National Laboratory*
Marcel Demarteau, *Argonne National Laboratory*

Introduction

While a wide range of experiments are likely to occur, and indeed hoped for at DCS, the following prototypical descriptor sufficiently captures most experiments: a loading device will impart a dynamic stress to a sample, and the material response will be probed in real time through imaging and diffractive measurements.

Thus, the primary challenges for the dynamic compression sector are: (1) beam optics, (2) gating and synchronization, (3) detector technologies, and (4) loading platforms. What will vary among the experiments may include the amplitude and duration of loading, the desired spatial resolution, and the required temporal detection characteristics. Also important will be (5) complementary diagnostics, both coupled to the experiments and to the efficient use of other beam and characterization resources to support desired pre- and post-characterization of targets. The final challenge is (6) offline support needs unique to DCS and specific to the precision assembly and modification of experimental targets. These are the six priority research directions identified for DCS instrumentation.

This section will briefly describe the capability gaps, potential impact and priority research directions for these six high priority challenges.

Capability Gaps

Beam Optics

The basic beam characteristics are defined by the specifications of the APS, which are routinely used at many sectors across the facility. For example, the choices of 324 bunch mode, 24 bunch mode, and hybrid mode define both pulse spacing and flux and bandwidth characteristics. The select use of monochromators, focusing and detection optics can follow existing technologies to modify the pulses to match the experiment's needs. While ultimately the photon numbers and optics might present a limitation at some time in the future, detection technologies cannot currently make ideal use of even these existing characteristics, so the primary challenge here is defining how to make investments for the initial operation of DCS. While it may vary by hutch, at the simplest level, it should enable operation in all of the storage ring modes, have capability for pink and monochromatic beams, and have in place a set of lenses for focusing to some desired range of beam sizes. As the facility definition progresses, this specific definition should be straightforward and can follow successful approaches at other sectors.

Gating and Synchronization

While time-resolved experiments have been ongoing at synchrotrons for many years, and remain an area of intense interest across many disciplines, the typical experiment at DCS will require different, and perhaps more difficult, synchronization challenges. These stem primarily from the specific operation of the loading devices themselves.

However, we can draw from and apply approaches that have been successfully developed elsewhere. For laser-driven experiments, for example, the synchronization of the laser to the X-ray bunch is now done routinely at synchrotrons worldwide. At the APS, master-oscillator-power-amplifier (MOPA) lasers such as Ti:Sapphire lasers are phase-locked to a subharmonic of the radiofrequency and the amplifier output triggered with a subharmonic of the revolution clock. The laser and X-rays can be synchronized to a few picosecond rms. Mechanical and electronic shutters can be synchronized with the laser to isolate a single synchrotron bunch in the 24-bunch mode of the APS. Sector 14 (BioCARS) already employs 3 inline shutters for pump-probe experiments and this technique is directly transferrable to planned laser-loading experiments at DCS (Figure 25).

However, such a technique will not work well for timing with a gas-gun driven loading event, where the firing operation and the desired loading state in the sample are separated by milliseconds with unacceptable jitter. Approaches to solve this problem include the low-jitter gun operations previously demonstrated at the Los Alamos Proton Radiography facility, and the synchronization of a gas gun with inline slow and fast shutters at Sector 32 (Figure 26). An approach similar to the latter using shutters and a gated ICCD X-ray detector has been demonstrated to isolate the desired X-ray pulse with certainty when using monochromatic X-rays. However, further work is needed to correlate the impact event with faster shutters which provide a beam for less than 100 μs in order to avoid significant sample heating when using a focused pink or white beam.

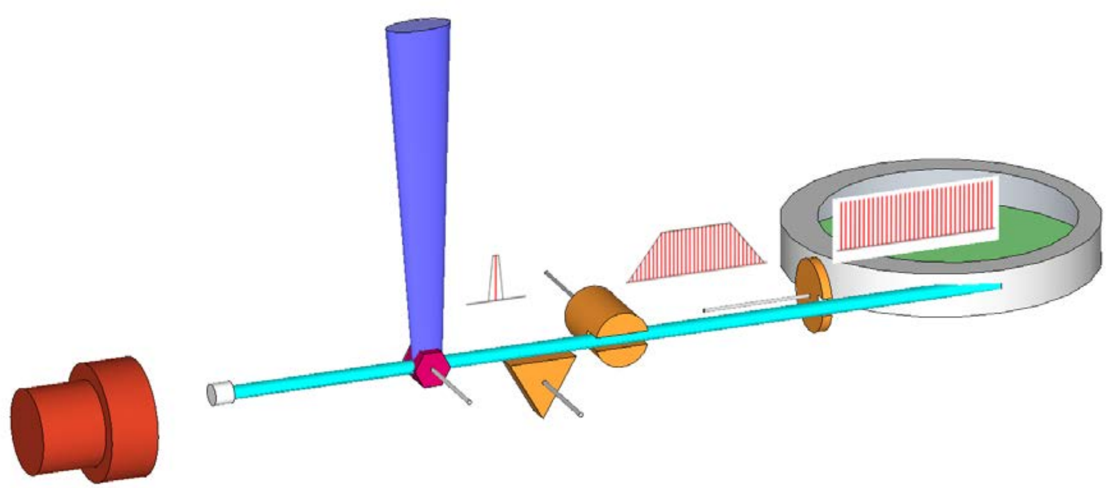

Figure 25. The inline shutter system at Sector 14 should be directly applicable to laser loading experiments at DCS.

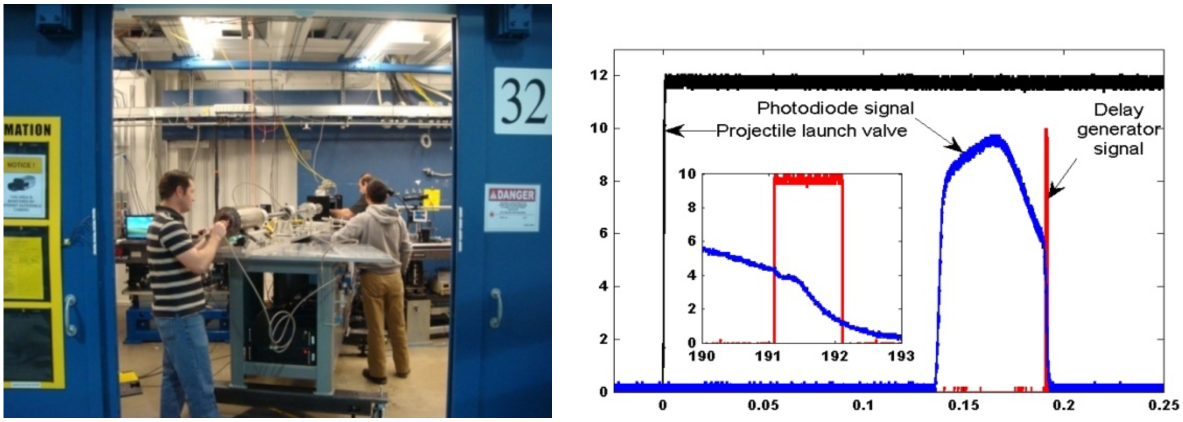

Figure 26. Gas gun research at Sector 32 has demonstrated synchronization of the loading event with 1-2 bunches in 24-bunch mode, but this will not be directly useful for multi-frame instrumentation.

Detector Technologies

The most challenging aspect of preparation for DCS is the limitations of current detector technologies. A broad suite of detector technologies over a wide range of X-ray energies will be required to fully exploit the information present in the brief, intense DCS events. The scientific challenge is the development of detectors to record shock movies, that is, *in-situ* multi-frame imaging and diffraction. Ideally, DCS should be capable of delivering a shock movie using X-rays of energies from ~10 – 35 keV with the time between frames matched to possible fill patterns at the APS, that is 153.4ns, 11.4ns or 2.8 ns. It is highly unlikely that detector developments for other light sources will universally meet the demands of the DCS community.

Loading Platforms

The choice of loading devices or drivers will largely be driven by the desired experiments, although cost and space are important considerations (see Table 2). Single and two stage gas guns, laser drive, and ramp loading are the presumed candidates, and indeed their use has been demonstrated in many government and academic laboratories and should be straightforward. However, the development of a compact 2-stage gun that will practically fit in a hutch enclosure will require detailed engineering considerations. Development of this particular capability is important, and is currently a focus at Sandia National Laboratories and other institutions.

Loading Device	Shock or Ramp	Pulse Duration or Risetime	Jitter	Synchronization / Gating	How Desirable
Gas gun (powder, gas, 2-stage)	Shock (>150 ns)	Up to 2μs	ms	Unsynchronized / Shutters and gated detectors	Most
Laser (direct drive)	Ramp	5-50 ns	1-10 ns	Synchrotron RF triggers laser / Choppers, shutters	Most
Laser driven flyer	Shock	5-50 ns	10-50 ns	Synchrotron RF triggers laser / Choppers, shutters	Most
HE driven flyer	Shock		ms	Unsynchronized ?/ Shutters and gated detectors?	
HE direct	Ramp		10-100 ns?	Synchrotron RF triggers HE / Choppers, shutters	
Pulsed Power Ramp Generator	Ramp	100-500 ns	10-100 ns	Synchrotron RF triggers PPRG / Choppers, shutters	

Table 2. Possible loading devices and their characteristics

Schematics of some possible impact and laser drive experiments are shown in Figures 27 and 28. The plate impact configurations shown in Figure 27a-d are for diffraction experiments, which have a low divergence incident beam focused to a spot size between 10-500 μm diameter. For plate impact or laser shock radiography experiments, the configuration shown in Figure 27e or Figure 27b could be used. Due to the nature of laser shock measurements, a forward scattered geometry can also be used, as shown in Figure 28a.

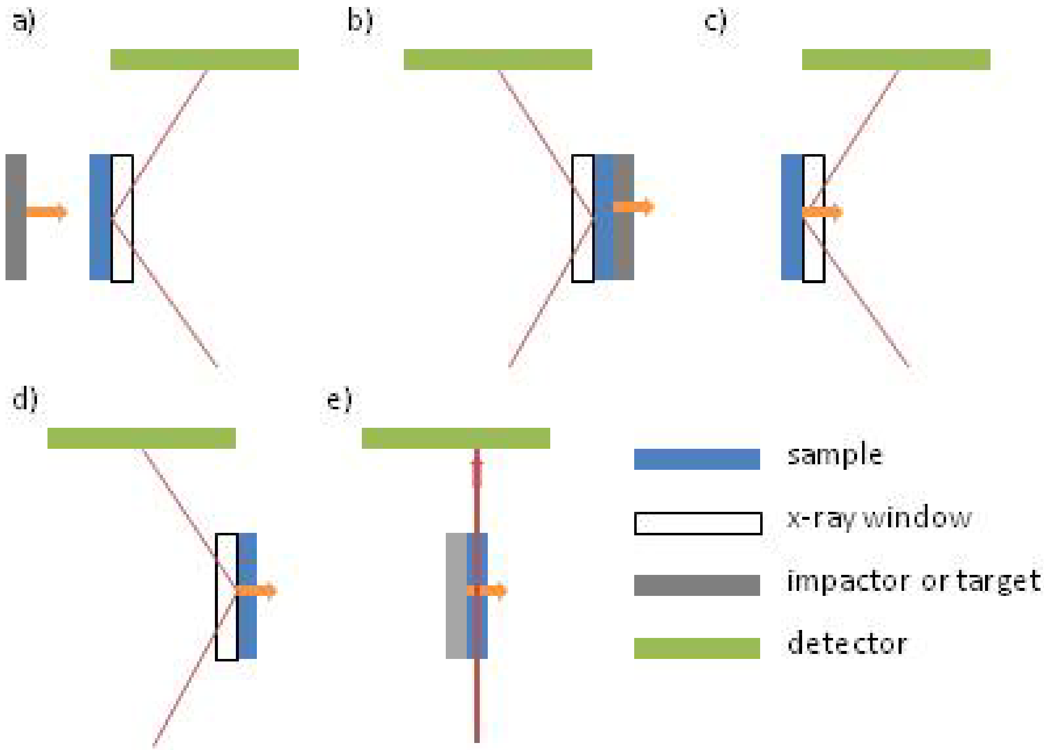

Figure 27. Possible configurations for plate impact experiments. (a)-(d) are for diffraction or scattering experiments and (e) is for radiography.

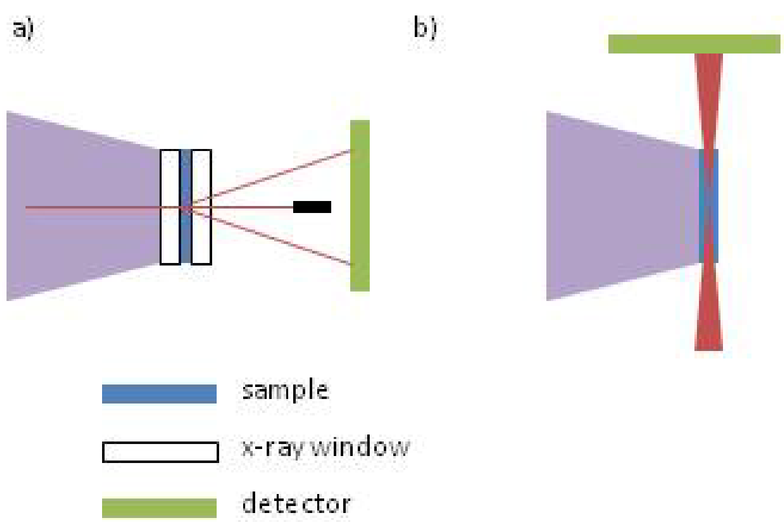

Figure 28. Possible configurations for direct drive laser drive experiments. (a) is for diffraction or scattering experiments and (b) is for radiography.

Complementary Diagnostics

To complement the rich new data that will be provided by the new technologies of DCS, and to connect with a vast body of previous research, it will be desirable to also have some traditional diagnostics available. Traditional velocimetry, such as VISAR, PDV, and perhaps line-VISAR, should be available and fielded routinely. Other diagnostics specific to laser-shock (e.g. Spectroscopy) might be desirable as a routinely available diagnostic. The hutch designs should also be able to accommodate "in-house" and user diagnostic development programs through user-provided "routine-to-them" diagnostics and beamline-scientists collaborative efforts, such as the development of temperature, pressure, chemical probes. These developments will be specific, and will necessarily be user and beamline scientist defined, but the initial DCS definition should include a basic set of complementary diagnostics.

Furthermore, there are other possibilities for pre- and post-characterization of targets that will certainly benefit from increased interactions with other sectors at the APS. For example, tomographic and topographic characterization is possible at other sectors and has been applied in both high pressure and pre- and post-shock experiment samples already. It is conceivable that dynamic implementations of these techniques could be developed, and needs to be studied. Furthermore, if these techniques are possible at neighboring sectors it may not be immediately necessary to achieve dynamic data in the near term, and their application to dynamic techniques might be better explored at their existing implementations. Thus, during development and operations, DCS can benefit from the maximal use of existing beam resources at the APS and elsewhere.

Offline Support

Finally, the support elements required to maintain an operational dynamic compression facility are considered separately, as these will share some of the aspects of adjacent lab facilities typically at other APS sectors, but will have some significant differences. Target design (uniformity for users showing up expecting to do a number of shots), real-time onsite assembly and measurement, modifications (specific to shock experiments – microns over centimeters), operational staff to keep guns and diagnostic equipment working, standard projectiles and target holders all need to be supported.

Priority Research Directions

Beam Optics

Choosing the correct beamline components will be critical in obtaining the flux and bandwidth necessary for the envisioned DCS experiments. The challenge here is to identify the most common or critical experiments to be conducted at DCS and then compile the components that will meet those needs.

Gating and Synchronization

Nanosecond (ns) synchronization with arbitrary time delays between the arrival of a fast projectile and the gating of an image intensified CCD detector that images the visible fluorescence from an X-ray sensitive scintillator or phosphor has been demonstrated at the APS by WSU and LANL. Relatively long time gating of the X-ray bunches was accomplished using commercially available slow millisecond mechanical X-ray shutters which were opened simultaneously with the firing of the projectile. The afterglow of the scintillator coupled to the CCD detector was important in isolating XRD or PCI images in these measurements, and will be an important issue for future sequential imaging studies. This is an area where high-Z, but fast scintillators are important.

To help quantify this issue, the bunch spacing in 24-bunch mode at the APS is 153 ns. A scintillator with a 10 ns fluorescence half-life will be contaminated by $(1/2)^{15}$ = 30 ppm of the background intensity. This residual scattering from the previous X-ray bunch is a source of ghost images or spurious diffraction patterns. Typical scintillators used at synchrotrons have decay times in the range of 25-100 ns, and faster scintillators, in particular for phase contrast imaging, will be needed at DCS.

At the commissioning of DCS, single-pulse time-resolved data can be expected. However, with the current technology there will be serious limitations for multi-frame diagnostics. Separate from the development of diagnostic technologies, immediate research on approaches to synchronize loading platforms and diagnostics to match the available mode structures of the APS is imperative. Work should begin on matching synchronization to utilize each individual pulse of the 24 bunch mode, and matching the 324 bunch mode will be a challenge for the future.

Detector Technologies

An intense emphasis should be placed on the development and fielding of trigger-able, gate-able X-ray detectors suitable for shock experiments. There currently is a tremendous technology gap, which severely limits the full potential of the DCS experiments. Since a first wave of DCS experiments needs to be ready by mid-2014, the time available does not allow development of new technologies and a short-term and long-term strategy is advisable. First generation experiments will have to employ and further develop existing technologies, including phosphor-based imagers, which may have a capability of about 10 frames and direct (Si) detectors for XFELs, capable of capturing 10s to 100s of frames at ~5 MHz. For the long term, new technologies will have to be developed with a frame-rate capability matching the APS bunch structure.

Although fast full-frame detectors are the ultimate goal, if the timing has a precision at the ns-level, a one-frame movie with micron-level resolution would already be a great step forward. Typical shock wave velocities are between a few mm to 10 mm per microsecond, so it is relatively easy to determine the time window and temporal resolution needed for a given sample size to get a good one-frame image with micrometer resolution and minimal blur.

The various detector technology approaches can be separated into direct and indirect technologies. Promising technologies, including some that have been demonstrated in limited ways, are the analog pixel array detector (PAD), intensified CCDs (iCCD), and scintillator camera approaches. Furthermore, the use of fiber optic tapers and geometric multiplexing schemes has been demonstrated, but suffer significant limitations. The development of faster scintillators and triggering/gating schemes for these technologies is required and evaluation and fielding of these technologies for shock-specific experiments should be an ongoing task prior to DCS commissioning, as detection represents the primary limitation to experiments at DCS. This need highlights an imperative for the development and subsequent operation of the sector: successful commissioning and operation will rely on efficient use of and collaboration with all available beam resources and the much broader interest in time-resolved science at synchrotrons – not just DCS sector specific.

An initial attempt at summarizing the detector requirements for both scattering and radiography experiments is given in Table 3. The primary physics drivers will refine these requirements. In general, these requirements are extremely challenging and will require a significant investment in the development of these detectors to exploit the full potential of the DCS science. For reference, EuXFEL has invested about $50M to develop detectors capable of meeting the low end of the requirements for the scattering experiment listed above.

Parameter	Scattering	Radiography
Frame Rate	6.5 , 88, 357 MHz	
Number of Frames	Record 1 μs (~2^3, 2^{6-7}, 2^{8-9} frames)	
Energy range(s)	10 - 22 keV	>25 (30) keV
Number of Pixels	~500 x 500	
Pixel size	100 μm	10 μm
Max. Photons/Pixel/Frame	~10^4	~10^5
Corresponding charge	4 - 10 pC	130 pC
Single Photon Detection?	Yes	Yes
Dynamic Range	17 bits	19 bits
Spatial Resolution?	No	Yes
Energy Resolving?	No	No

Table 3. Detector requirements for scattering and radiography experiments.

Loading Platforms

A high priority is to develop an understanding of the needs of the various science experiments in order to establish necessary loading devices. Once these initial loading needs are established, particular platforms, i.e. guns, lasers, etc., should be designed to meet those needs within the confines of the DCS Sector.

The ability to handle and fire small amounts of HE is crucial for NNSA science. This is necessary, not only for HE loading of materials, but also as an opportunity to make significant strides in HE science: detonation and deflagration properties, which include Equation of State and burn model development that relate directly to NNSA program interests.

Because most measurements at DCS will be destructive and require more setup time than typical experiments at the APS, the throughput of experiments should be maximized without compromising safety, scope, or the quality of experiments. As such, the design of the loading platforms must encompass the necessary components to enable rapid experimental turn around. This will likely entail the need for multiple guns and/or gun components that can be rapidly interchanged. The expertise of the APS in automating processes should be utilized in this regard.

The choice of bore size(s) for the guns should also be carefully considered. A bore size of 0.5 inch may suffice for most measurements and has been previously fielded at the APS by WSU and LANL. Larger or smaller bores should also be considered as dictated by the desired experiments.

Lastly, defining ancillary equipment and instruments, maintenance/backup equipment, and proven operating procedures will be important. The support infrastructure and space needs must be well defined, with an emphasis placed on reproducible and rapid turnaround of measurements.

Complementary Diagnostics

Top priority in the area of complementary diagnostics is the evaluation and development of the full complement of diagnostics techniques required by the community. Traditional diagnostics such as VISAR, PDV, and perhaps line-VISAR, should be available. Visible high-speed imaging, shock wave sensing, both electrical pin and optical fiber based, pressure transducers, and others should also be part of the diagnostics suite.

There are other possibilities for pre- and post-characterization of targets that will certainly benefit from the increased possibility for interactions at the APS. It needs to be studied if dynamic implementations of these techniques could be developed. Furthermore, if these techniques are possible at neighboring sectors it may not be immediately necessary to achieve dynamic data in the near term, and their application to dynamic techniques might be better explored at their existing implementations. This needs to be evaluated.

Offline Support

Finally, the support elements required to maintain an operational dynamic compression facility are considered separately, as these will share some of the aspects of adjacent lab facilities typically at APS sectors, but will have some significant differences. Target design (uniformity for users showing up expecting to do a number of shots), real-time on site assembly and measurement, modifications (specific to shock experiments – microns over centimeters), operational staff to keep guns and diagnostic equipment working, standard projectiles and target holders all need to be supported. An evaluation of the needs of the various science groups is required.

The heat load on the sample from a focused pink beam is a concern. It may be that as long as the power density on the sample is comparable with the power density of an unfocussed white beam, a metallic sample can survive for a few ms exposure. This should be investigated for each of the target samples under consideration. It would be helpful for DCS to develop computing tools for experimenters to estimate the time-resolved heating of the sample depending on the X-ray flux, sample composition, and boundary conditions.

Data storage and offline reconstruction tools are another critical area that needs to be addressed.

Conclusion

Scientific achievements at DCS will depend heavily on the available instrumentation. Of primary note, existing detector technologies at light sources by far do not meet the challenges for the science at DCS. The current dedicated detector developments for light sources will still fall short in their capabilities to fully exploit this science sector. A short-term and long-term strategy is envisioned to develop a full set of desired and minimum specifications for each area of dynamic compression science. A significant investment in the development of these detectors is required to meet the stringent requirements of the DCS community.

GOVERNANCE MODELS AND USER EXPERIENCE CONSIDERATIONS

Panel Chairs

Roger Falcone, *Lawrence Berkeley National Laboratory*
Alan Hurd, *Los Alamos National Laboratory*

Introduction: Guiding Principles

The panel on governance models and user experience set several overarching principles from which other considerations flow. These principles derive from examples of successful Office of Science (SC) user facilities, including the APS:

- Maximize the quality of science emerging from DCS
- Develop the unique attributes of DCS
- Provide effective leadership in the field
- Consistency with operating principles of DOE/SC user facilities

In a sense the last item is the easiest to address as there are examples of highly successful beamlines at the APS itself. The attributes of a successful user facility are detailed in the following sections. Some of the key attributes are open access for a broad range of experiments, a transparent scientific and administrative peer review process for proposals, support for users at the beamline, periodic reviews, and the development of a DCS user community.

Developing a user community is essential to maximize the scientific payoff from DCS and for this first-of-a-kind user facility to grow into a stable and unique resource. DCS is an instrument at the crossroads of disparate research fields with no extant user community: dynamic vs. static compression/deformation; single-shot vs. continuous measurements; nanosecond vs. millisecond resolution. DCS constitutes a unique opportunity for dynamic compression science. To ensure success, the appropriate community must be assembled, aided, and nurtured. It is imperative that DCS leadership, sponsors, and partners actively cultivate the user community by being open, scientifically broad, attentive to user needs, and energetic in growing the user base by directly engaging diverse technical areas and by outreach. To achieve this goal, DCS Leadership and Partners must work closely with sponsors (NNSA and SC/BES) and all stakeholders.

DCS should operate as a Partnership Sector as defined within the standard APS model:

- Establish shared time allocation between the Partner and General Users;

- Use the standard model for APS Partnerships in which all proposals for experiments go through Proposal Review Panels;

- Make consistent beam time allocations. The panel felt that the standard APS practice meets this intent, with 25% time for top-ranked General Access Users decided by the APS and 75% time allocations by the Partner.

Several formal agreements have been made, are being made, and should be made:

- between Washington State University's Institute for Shock Physics and NNSA to design and develop the DCS program concept;
- between DOE NNSA and SC/BES to define roles and responsibilities regarding construction, commissioning, stewardship, and operations;
- among APS, SC/BES and NNSA to construct the beamline (from undulator to hutch);
- between WSU/ISP and APS to develop and construct DCS with NNSA funding;
- among WSU/ISP and APS to operate DCS with NNSA funding according to a clear and well-defined management plan.

The DCS facility will benefit from broad engagement with users. The User Community should have early and continued input on choices being made regarding the construction of the beamline, instrumentation for core capabilities, photon parameters, pulse chopper, optics, gas-gun drivers, laser drivers, non-single event experiments, detectors, data collection, and the like. Moreover, users should be involved in governance activities such as the review processes, advisory committees, and assessment reviews. DCS leadership should consult the user community about appropriate staffing for the DCS beamline. It is expected that the level of resources available will be influenced by community demand. Broadening the community through significant and proactive outreach will be required, and strong interaction between DCS and other APS Sectors will be extremely beneficial.

WSU and the Institute for Shock Physics are in a unique position in enabling science as the originator and partner-operator of the DCS beamline at the APS. As such, WSU is strongly encouraged to ensure continual engagement with both NNSA and SC/BES agencies, NNSA and SC/BES Labs, and the broader academic and other scientific and industrial communities needed to ensure success of DCS. The operation of DCS will require WSU attention to leadership, organization, and long-term planning. Above all, careful attention should be paid to the roles of users within and outside the Partnership. *Because of the cooperative stewardship role of NNSA in DCS, the long-term viability of DCS will benefit from the NNSA Laboratories playing a special role in DCS operation.*

Policy and Governance

Access modes

Following user facility practices within DOE SC/BES and around the world, three access modes are advised: General Access, Partner Access, and Proprietary Access. Variants of these such as "fast access" and "remote access" may be added as needs arise. An important variant "classified access" for national security research of a sensitive nature may be required, depending on user demand, though not currently planned.

For each access mode, the DCS facility should provide for user agreements that define roles and responsibilities, including financial obligations of the facility, and its personnel, and users. Such matters as intellectual property are defined in DOE model agreements, and the DCS facility agreements should be consistent with these models. Authorship and acknowledgment arrangements should be clear and consistent with the APS practice. While international users have no special restrictions to beam time following the BES access model, special cases may arise involving programmatic sensitive research. When applicable, preference to government-to-government treaties and agreements should be considered in formulating facility policies. Example international partners include NNSA's stockpile stewardship partners, AWE in the UK and CEA in France, who may want to use DCS.

Preliminary DCS Expectations and Parameters

- DCS@APS will be a NNSA-funded user facility for the dynamic compression science community established by a WSU/APS partnership following APS guidelines with approval from NNSA.
- DCS will be operated and managed by the WSU/APS partnership on behalf of the NNSA sponsor.
- All aspects of DCS policies require NNSA approval.
- NNSA funds have been provided to construct DCS.
- An MOU between WSU and APS will be finalized, with NNSA approval, defining the governance model and relevant committees.
- Commissioning experiments begin in late 2014.
- All science at DCS will be open, unclassified science using the existing APS access model (75% Partner Access and 25% General Access).
- WSU and APS will finalize a joint plan for DCS Operations and associated costs to be submitted to NNSA for approval and funding.
- WSU represents the Dynamic Compression User Community.
- DCS@APS will have its own user group coordinated with the broader APS user group.
- All APS beam modes are compatible with timing experiments expected at DCS. Present modes are singlet, 24-, and 324-bunch at 0.27, 6.5, and 88 MHz, respectively running 15%, 65%, and 15% of the time.

Sidebar 9. Preliminary DCS Expectations and Parameters

Membership

Partner membership, to be clearly stipulated by written agreement, defines institutions that enjoy Partner Access in exchange for certain obligations such as, but not limited to, operating the beam line. The panel cautions DCS against limiting partnership to the founding institution(s) for all time; a robust partnership may evolve with diverse institutional partners (including industry and national labs) joining WSU. The steady-state partnership will emerge as the user base matures, and the panel anticipates diverse user constituencies interested in single-shot vs. continuous experiments; different time scales, and perhaps domains of interest such as geoscience vs. materials science.

Partner obligations may include team formation and organization, on-site experiment observations, outreach and education among other aspects. As currently planned, the partner-operator will provide engineers to operate experimental facilities, beam-line scientists, and research scientists. Following the Cooperative Stewardship model, the facility should supply support for safety, compliance, and security. The relationship between DCS and the academic community must be carefully nurtured because major aspects of DCS mission will naturally emphasize national lab users.

Leadership

In consultation with the APS management, the DCS leadership must effectively integrate all components of the program. Proactive governance captures the sense of supportive yet directed administration through hands-on management and effective communication. The DCS leadership must address the hiring and retention of top-quality staff as early as possible. Flexibility and willingness to sacrifice partner beam-time for general access for the good of the facility may be necessary at times.

The Director of DCS must practice and exemplify the guiding principles as well as a proactive governance style. She/he should be open to staff and users alike, should network well, and nurture innovation. A clear organizational chart will empower the Director, defining a line of authority and resource flow. Owing to the multiple sponsorships of DCS, the governing agreements should address the relationship of the DCS Director to APS management, NNSA program officers, BES authorities, and partner institutions. The roles and responsibilities of the key operational staff should be spelled out when ambiguous or complex interpretations could arise. For example, what is the relationship of the DCS Director to the APS Management, the WSU Administration, and the NNSA Program Office? All three stakeholders listed should be involved in reaching a consensus decision.

Advisory Bodies

As noted in the Guiding Principles, the formation of advisory and planning committees is fundamental to any governance model consistent with SC/BES facility practices. The charter for each body must, at a minimum, define the roles, responsibilities, membership type, selection procedure, terms of appointment, and frequency of meetings. Purview domains should dovetail but not overlap. The DCS leadership should form executive committees addressing research needs, capabilities planning, and user group needs although chartering the last is in the hands of the users themselves.

The most fundamental of these is a strategic advisory board that involves, in the course of time, representatives of all stakeholders and potential stakeholders. Here "stakeholders" includes but is not limited to users, customers, and sponsors.

As a first-of-a-kind facility, DCS brings up unfamiliar engineering and operational challenges. A concerted effort in beamline design will best serve future users. The panel felt that DCS might need a group of high-level experts to guide planning and to collaborate with early users for the most efficient and impactful studies. This "hands-on" group may be transient—established primarily to help plan and conduct a series of pilot studies and to explore alternative approaches—yet panelists felt that an enduring presence would be prudent. While the feasibility experiments in 2007 were a good trial run, they were not optimized; following experiences at HPCAT and NSLS-2 with such a hands-on advisory group, superior designs invariably resulted.

Stewardship models

A concerted effort in governance will pay forward in improved efficiency and success rate of DCS science as well as enhance cooperation with related programs at the APS such as HPCAT, GSE-CARS, Bio-CARS, and others (Figure 29). Implementing a robust user program will have unique opportunities, rewards, and challenges inherent in multi-mission facilities.

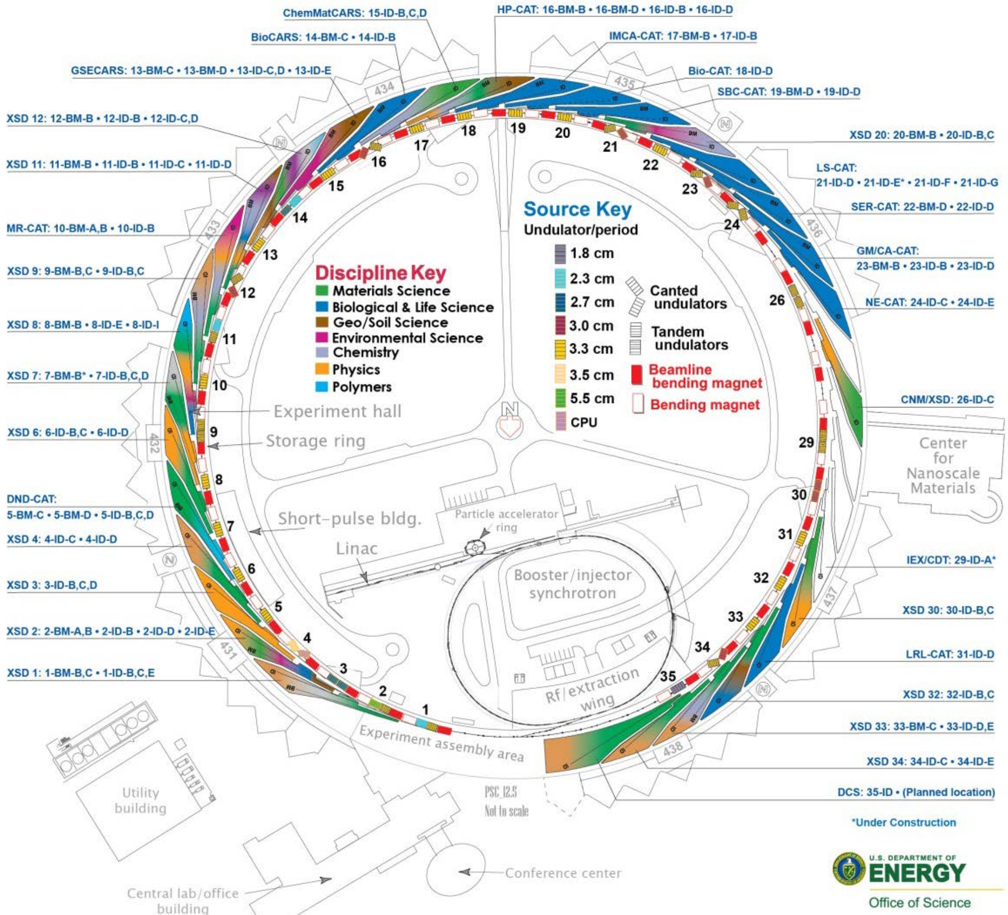

Figure 29. DCS will be operated by Washington State University and the Advanced Photon Source. (*Figure courtesy of the APS/ANL*)

As a baseline, best practices from operations of the APS facility should inform the stewardship principles and organizational structures. Many of these lessons are codified in *Cooperative Stewardship* (National Academy Press, 2003) specifically to address multidisciplinary user facilities for research with more than one sponsor to work with the natural tension that arises under differing program goals, types of experiments, agency cultures and user community cultures. Panelists recommend learning from several examples involving NNSA partnerships such as Z, Omega, NIF, Jupiter, Lujan Center at LANSCE, and HPCAT. Many of these programs have in common that they produce excellent science (but not at the same rate) while managing the tensions between program and science. Success depends on a combination of scientific breadth, diversity of support, and leadership. "The rest is detail" according to one panelist.

As an example of lessons from these facilities, DCS could establish a council for Stockpile Stewardship Program (SSP) work similar to that of NIF, or a National Security Program Advisory Review Committee as similarly implemented at Lujan Center. As epitomized by various APS Sectors, the academic community at these partnership facilities contributes strongly to fundamental science underpinning NNSA interests. For well-chosen research areas, the physics is the same for academia and NNSA.

Diversity of support carries the advantage of a larger footprint in funding advocacy. Assuming a welcoming governance structure, DCS will benefit from this diversity. Specifically, DCS will serve NNSA indirectly through national security programs at Argonne National Laboratory and possibly through the NNSA Laboratories buy-in in LDRD and overhead funding. DCS should benefit directly and indirectly from NNSA funding through grants to the Stockpile Stewardship Academic Alliance partners, and partnerships with NNSA Laboratories may further underwrite the DCS program in the fullness of time. Finally, DCS should be viewed as the nexus of other funding through industrial and academic users.

The governance system must adjust to the life phases of DCS. During design and construction, startup, commissioning, and steady-state operations, priorities will necessarily shift. During all phases, it is necessary to develop a set of metrics appropriate for assessing the performance, overall experimental availability, and effectiveness of operation of the facility, and an advisory body will be instrumental in managing expectations at each phase. For example, good estimations of construction budget and annual operations budget are essential in the early phases whereas satisfaction of the user community should be considered an important metric in steady state. Flexibility is the key, and the panel recommends that DCS define a change-control process for making changes to facility governance in order to adjust to life phases and shifts in the political environment. We recommend developing a review schedule with concurrence requirements for the Governance Plan, as well as how to communicate and distribute revisions.

Self-assessment is a necessary management function tied to governance. Agreement on metrics is essential. The panel anticipates that facility utilization (user numbers, time resource allocations, number of shots and experiments in both dynamic compression and non-single event categories) and scientific productivity (papers, patents, invited talks, awards) will be important. Educational impact metrics (students, theses, postdocs, outreach, etc.) are essential. So too will programmatic impact on DOE/NNSA and energy programs such as in mission-directed reports. However, these are largely steady-state metrics, and it is critical to have clear metrics (cost, schedule) for construction, startup, and commissioning; compliance measures are a fact of facility life. A schedule of annual performance self-assessments will keep the facility well prepared for external assessments by sponsors.

Embedding DCS in the APS offers major scientific opportunities and the quality of multi-mission governance essential to realize this vision. The challenge for management will be to uphold this vision for NNSA and SC/BES as well as other stakeholders WSU, APS, ANL, and NNSA Laboratories.

Review Processes

DCS must provide for an effective infrastructure, both technical and administrative, to support users, from proposal solicitation through planning and execution of experiments to dissemination of scientific results. A "user program group" should develop and publish a timeline associated with the process for facility scheduling, beginning with calls for proposals, through review, selection, and notification for scheduling of experiments. Then, clearly communicate the processes to users for proposal solicitation, technical review using well-defined evaluation criteria by a committee of peers, ensuring no conflicts of interest. Proposal selection, notification, and scheduling are standard. Importantly, DCS must provide for grievance and appeals procedures.

Data Handling and Availability

Good governance embodies credible and mutually accepted review processes for both the programmatic and fundamental science communities. As mentioned earlier, DCS may someday require a review process for sensitive or even classified experiments. Similarly, a data policy for programmatic (NNSA) experiments will need to be in place; otherwise, APS data policies may suffice.

Export Control and Other Security Issues

Leaving aside the possibility of classified experiments, APS and ANL policies for handling sensitive data, appropriately modified for DCS as needed, should be established. Considerations include export control, ITAR, and proprietary issues.

Operations

With the right governance model, DCS should become a mecca for dynamic experiments for academic research and NNSA missions in stockpile stewardship and nonproliferation. Setting the operational capacity expectations starting with commissioning is a task of first priority. *Commissioning, producing high-impact early science, and achieving a rapid and orderly transition to a general user program will set the course for many years to come.*

Capacity

Utilization split between dynamic and non-single event experiments was discussed by the panel. How can DCS bridge dynamic compression and non-single event experiments while making best use of beams and detectors? Dynamic compression experiments require longer set-up time but only a very short time for the actual experiment. Meanwhile, DCS can be operated with non-single event experiments without any effect on the main dynamic mode. That said, dynamic compression science is the “raison d'être” for having DCS. Once operations commence at this first-of-a-kind user facility, optimal utilization of different experimental stations will become clearer.

The presence of dynamic compression drivers at DCS is a significant operational difference compared to other APS stations requiring enhanced attention to operation, maintenance, safety, and upgrades. In addition to a lower duty cycle for dynamic experiments than is possible for non-single event experiments, the panel anticipates, conversely, that drivers may sometimes need to be available on their own (i.e. without APS beam) as exemplified by the Z-Beamlet laser at SNL. Therefore, DCS management should consider the challenge of establishing a strong scientific and programmatic synergy among dynamic compression, non-single event, and photon source experiments. This task may require coordination with NNSA funding flow to build a concerted effort.

Access to Sample Fabrication

Laboratories supporting user preparation of samples are needed along with the normal APS-style user offices. Sample fabrication for dynamic experiments will be more complicated and take more intervention on the part of DCS personnel than typical APS experiments. Although, at other APS Sectors, users bring their samples to the beamline already prepared, on-site sample preparation capabilities are essential along with knowledgeable personnel to aid users. Facilities at DCS should permit significantly more sample fabrication than normally envisioned at other APS Sectors. This is also addressed in comments on the user experience, as well as in the comment below.

Staff and Resources

Dedicated staff and resources are needed to support DCS design and (sometimes) simulate experiments. A clear understanding of staffing levels is essential between the DCS management and the user community. In addition to an understanding of the staffing levels because of dynamic compression drivers, it will be important to consider levels of each type of staff required. Ideally separate scientific and engineering staff would be charged with development and user support, though often beamline staff at other Sectors carry both responsibilities. The management structure and reporting responsibilities must be considered carefully given the complexity of DCS and, particularly, the dynamic compression drivers.

User Experience

Training and Access

The user conduct policy, operations, data handling, training, and safety envelope should be consistent with the APS policies. As noted above, the user access policy and governance should follow established BES norms with fair, open access, and transparent processes that make best use of the facility. Time and experience have shown that these factors maximize scientific impact under the enrichment of regular reviews. Users expect clear communication of the call for proposals, fair peer review using broad technical review committee, and short time lag between proposal and experiment consistent with these measures.

User Support

The panel identified crystallography and spectroscopy as fundamental adjuncts to DCS to optimally understand and interpret dynamic data. Coordinated or joint proposals to other beam lines should be encouraged.

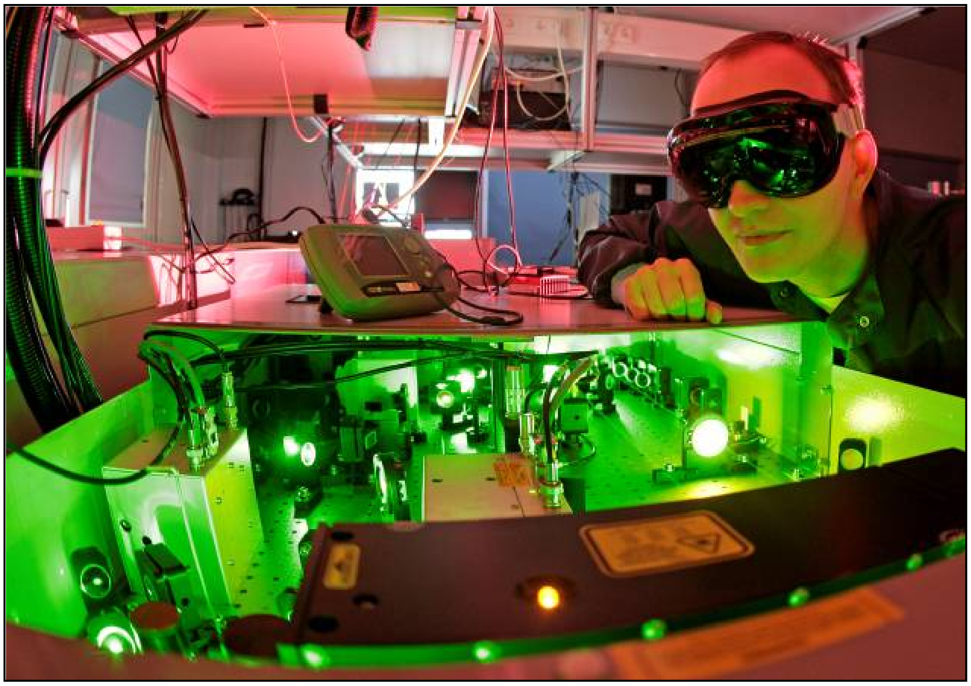

Figure 30. User support before, during, and after experiments at DCS will be critical to the user experience. *(Courtesy of Helmholtz Institute)*

A user base will develop organically if the appropriate infrastructure—experimental and policy—is put in place (Figure 30). DCS has the potential to attract diverse users and to enable a broad set of important science that takes advantage of DCS's unique capabilities (Figure 31). The anticipated user community will focus on the scientific problems related to the compression and deformation of materials, ideally addressed by accessing both dynamic compression and static compression sectors at the APS. Knowledgeable users will develop the best approach and intuition for utilizing the enormous capabilities of dynamic and static compression sectors at the APS in their specific scientific interests. A concerted effort between DCS and other Sectors, with clearly defined scientific objectives, will benefit all users.

Access to Theoretical Support

Many users will benefit by access to simulations, design, and theoretical support in association with their DCS activities. ANL theory staff may find collaborations stimulating and useful to their own ends. Within the budget scope of DCS, a win-win arrangement could include non-APS staff through external program funding.

Data Handling and Computations

A strong motivation for advanced computational modeling is that fact that DCS is unique. Provision for user software with new data reductions and interpretation, at high data rates and in large quantities, will be a cost-effective measure within the project.

Users' Group

An important step to developing an effective user community is to establish a user group that involves the full spectrum of participants (academia, national labs, and private sector). DCS management should foster the formation of a user executive committee while maintaining a comfortable distance in order to maintain user group independence. The user group is important for integrating NNSA programmatic and fundamental science users, for direct input to the DCS Director, and for independent advocacy in federal circles.

The founding documents of the user group should clarify steward, WSU, APS, and partner roles. For example: Does APS, WSU, or the user-group support the user group meeting? Should the user group be incorporated?

Outreach and Education

An energizing activity for DCS staff and users alike is a robust program of outreach and education for developing and cultivating relevant user communities. Components may include workshops, summer schools, student programs, and printed materials. The panel recognized interest within the DCS community for high compression science collaboration at Euro-XFEL and other non-US-based facilities.

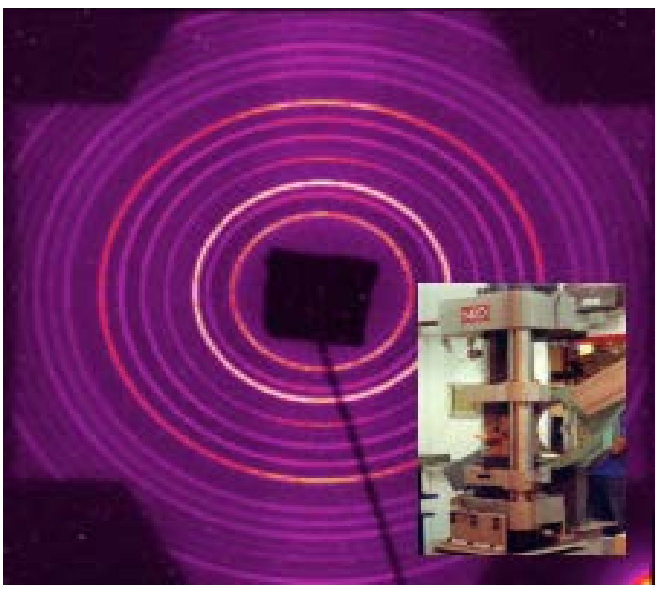

Figure 31. High pressure X-ray diffraction from alumina performed on anvil press at NSLS. (*Courtesy Yusheng Zhao, University of Nevada at Las Vegas)*

The scientific footprint of DCS, determined to zeroth order by the science payoff as a function of driver energy and rep-rate, can be anticipated to be broad enough to encourage out-of-the-box thinking within the user community. Examples of exciting outreach science are magnetic phenomena in compressed matter and meteorite impacts in which time-resolved X-ray diffraction data are novel and incisive. The characteristics of such disciplines are that they will be attractive to NSF and other funding agencies and will attract students to the DCS program. NNSA may have special interest in the science and general interest in recruiting talent to NNSA mission-related topics. DCS should explore collaborations and partnerships with other facilities in which NNSA plays a stewardship role (see e.g., Table 4).

Ideally, one FTE would be devoted to education, training and outreach in order to establish a good program fully coordinated with partners and users.

Scientific Breadth

Over time, DCS will help develop a cadre of experimenters and theorists who specialize in dynamic compression science. The panel recognizes that a broad scientific footprint is needed for DCS including academia, national labs, and industry. Reaching the vision of a large user constituency delivering excellent publications in broad areas will require extensive efforts to enlarge the community. Thoughtful governance will set the tone for encouraging the full breadth at DCS.

Facility Partnerships involving NNSA			
Facility	**Discipline**	**Steward**	**Partners**
HPCAT	Static, high P-T	NNSA	SC-APS, Carnegie
Z		NNSA	SNL
Omega Laser	HED Science	NNSA	Rochester
NIF	HED Science	NNSA	LLNL
Lujan Center	Neutron scattering	SC-BES	NNSA-LANSCE

Table 4. User facilities NNSA partnerships have governance models similar to that envisioned for DCS@APS.

CONCLUSION

The grand challenge of predicting and controlling materials in extreme environments is important not only to the Department of Energy's National Nuclear Security Administration but also to a broad array of sponsors and communities. Recent progress in this area has been fueled by advances in measurement capabilities that enable *in-situ* studies while a material is being subjected to extremes, especially thermo-mechanical extremes (e.g., high strain, high pressure, high temperature). These studies provide tests of theories with resolution and physics fidelity that are also being advanced by recent gains in modeling and computations. An important overarching goal of time- and space-resolved investigations of dynamically compressed condensed matter is to perform experiments on the time and length scales of numerical simulations. **This is the frontier of dynamic compression science**. In particular, recent community-based workshops have concluded that *in-situ, time-resolved measurements at microscopic length scales constitute the overarching science need for achieving a fundamental understanding of the mechanisms governing time-dependent condensed matter phenomena (structural transformations, inelastic deformation and fracture, and chemical reactions) under dynamic loading*.

The Dynamic Compression Sector (DCS), currently being built at the Advanced Photon Source (APS), will constitute a key advance in this area. DCS@APS will focus on time-resolved X-ray diffraction and imaging measurements in materials subjected to dynamic compression. The X-ray energies (hard X-rays) and the time structure (ns-separated pulses) of the APS are well suited to examine time-dependent changes in materials subject to a broad range of peak stresses (~ 5 GPa to above 100 GPa) and time durations (tens to several hundred ns). DCS will emphasize dynamic compression science of solid phase materials and will complement user facilities that emphasize static high pressure studies of materials or warm dense matter/plasma physics regimes.

A user workshop of approximately 110 international leaders spanning the frontiers of dynamic compression science was convened at the Advanced Photon Source on January 19-20, 2012. The objective of the workshop was to explore "Basic Research Directions for Dynamic Compression Science" and to identify the broad spectrum of scientific challenges and opportunities afforded by the integration of dynamic compression platforms and advanced X-ray capabilities. Topics of discussions included structural changes and phase transformations in condensed matter, deformation processes and fracture dynamics in materials, dynamics of chemical reactions, and time-resolved dynamic processes in materials beyond those generated by compression. From their deliberations, workshop participants identified a set of priority research directions (summarized below) that were both timely and important for advancing the field.

Panel	Priority Research Directions (PRDs)
Structural Changes and Phase Transformations in Condensed Matter	Structural Changes During Loading
	Kinetics and Dynamics of Phase Transitions
	Non-Crystalline Diffraction for DCS
Deformation Processes and Fracture Dynamics in Materials	Examining Microscopic Response to Dynamic Compression
	Examining Dynamic Tensile Damage and Spall
Dynamics of Chemical Reactions	Mechanics Leading to Chemical Change
	Chemical Reaction Mechanisms In Extreme Conditions
Time Resolved Dynamic Processes in Materials Beyond Those Generated by Compression	Deformation and Fracture of Materials
	Phase Transition Dynamics
	Chemical Reaction Dynamics

Table 5. Dynamic Compression Science Priority Research Directions

In addition to exploring these frontiers of dynamic compression science, the workshop identified instrumentation opportunities, including advanced diagnostics and X-ray optics, and governance considerations for the DCS facility that would maximize the impact of this first-of-a-kind capability on dynamic compression science.

In the end, workshop attendees enthusiastically concluded that DCS would enable a broad suite of frontier dynamic compression science opportunities and would further fuel the development of additional techniques and capabilities for advancing this science. Finally, workshop attendees recognized that DCS should be the first, not the last, of a next generation of advanced measurement capabilities that coupled e.g., advanced X-ray sources to relevant extreme environments with measurement resolution designed to validate and stretch state-of-the-art modeling capabilities to advance our predictive understanding of materials in extreme environments.

Appendix A: Workshop Agenda

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Dynamic Compression Sector (DCS) User Workshop Agenda
January 19 - 20, 2012
Advanced Photon Source, Building 401

Thursday, January 19, 2012

7:00	Poster Set Up (7:00 - 8:00) *Location: Lower Level Gallery*
7:15	Registration and Continental Breakfast *Location: Registration - First Floor Atrium; Continental Breakfast*
8:15–12:30	Plenary Session Chaired by: John Sarrao (LANL) and Christian Mailhiot (LLNL) *Location: 402 Auditorium* DCS: NNSA Perspective Chris Deeney (NNSA) APS, the Upgrade and the DCS Brian Stephenson (APS/ANL) DCS: Overview and Scientific Opportunities Yogi Gupta (WSU) *10:00 – 10:15 Morning Break* New Opportunities in Compression Science Rus Hemley (CIW) Next Generation Light Sources for Studying Materials at Extremes Roger Falcone (UC – Berkeley and LBNL) Shock Compression of Condensed Systems: Role of Emerging Computing Platforms Priya Vashishta (USC)
12:30 – 1:45	Lunch in the Lower Level Gallery (*Please ensure your poster is set up.)*

1:45 – 5:00 Parallel Technical Breakout Sessions

- Structural Changes and Phase Transformations in Condensed Matter; Chaired by: Gilbert Collins (LLNL) and Mike Desjarlais (SNL)
- Deformation Processes and Fracture Dynamics in Materials; Chaired by: Neil Bourne (AWE) and Mukul Kumar (LLNL)
- Dynamics of Chemical Reactions; Chaired by: Larry Fried (LLNL) and Dave Funk (LANL)
- Time Resolved Dynamic Processes in Materials Beyond Those Generated by Compression; Chaired by: Todd Hufnagel (Johns Hopkins Univ.) and Adam Schwartz (LLNL)
- Instrumentation (Including Diagnostics and X-ray Optics); Chaired by: Dean Haeffner (APS/ANL) and Marcel Demarteau (ANL)

3:00 – 3:15 *Afternoon Break*
Location: Inside Auditorium

5:30 – 7:30 Poster Session; Chaired by: Denny Mills
And Reception
(Heavy Hors d'oeuvres and Refreshments)
Location: Lower Level Gallery

Friday, January 20

7:30 Continental Breakfast
Location: Inside Auditorium

8:15 – 9:00 Phase Contrast Imaging, a New Frontier for Dynamic EOS Examination
Jon Eggert and Damien Hicks (LLNL)
Location: 402 Auditorium

9:00 – 10:15 Initial Report-Out by Breakout Groups (15 mins each)
Location: 402 Auditorium

10:15 – 10:30 *Morning Break*
Location: Inside Auditorium

10:30 – 12:15 Parallel Technical Breakout Sessions

- Structural Changes and Phase Transformations in Condensed Matter; Chaired by: Gilbert Collins (LLNL) and Mike Desjarlais (SNL)
- Deformation Processes and Fracture Dynamics in Materials; Chaired by: Neil Bourne (AWE) and Mukul Kumar (LLNL)
- Dynamics of Chemical Reactions; Chaired by: Larry Fried (LLNL) and Dave Funk (LANL)

- Time Resolved Dynamic Processes in Materials Beyond Those Generated by Compression; Chaired by: Todd Hufnagel (Johns Hopkins Univ.) and Adam Schwartz (LLNL)
- Instrumentation (Including Diagnostics and X-ray Optics); Chaired by: Dean Haeffner (APS/ANL) and Marcel Demarteau (ANL)
- Governance Models and User Experience Considerations for the DCS Facility; Chaired by: Roger Falcone (UC Berkeley and LBNL) and Alan Hurd (LANL)

12:15 – 1:30	Hosted Lunch *Location: Lower Level Gallery*
1:30 – 2:30	Summary Comments *Location: 402 Auditorium*
2:30 – 3:00	DCS Beamline: Photon Requirements and Challenges Kevin D'Amico *Location: 402 Auditorium*
3:00 – 3:15	*Afternoon Break* *Location: Inside Auditorium*
3:15 – 5:00	Next Steps *Location: 402 Auditorium*
5:00	Adjourn

Appendix B: Workshop Participants

Appendix B: Workshop Participants

Last Name	First Name	Organization
Akin	Minta	Lawrence Livermore National Laboratory
Alp	Esen	Argonne National Laboratory
Armstrong	Michael	Lawrence Livermore National Laboratory
Barnes	Cris	Los Alamos National Laboratory
Bartlett	Roger	National Security Technologies
Bayramian	Andy	Lawrence Livermore National Laboratory
Belak	James	Lawrence Livermore National Laboratory
Belof	Jonathan	Lawrence Livermore National Laboratory
Bernier	Joel	Lawrence Livermore National Laboratory
Bi	Wenli	Argonne National Laboratory
Bolme	Cindy	Los Alamos National Laboratory
Bourne	Neil	AWE
Bronkhorst	Curt	Los Alamos National Laboratory
Brown	Ronald	Naval Postgraduate School
Campbell	Andrew	University of Chicago
Capatina	Dana	Argonne National Laboratory
Cauble	Robert	Lawrence Livermore National Laboratory
Cerreta	Ellen	Los Alamos National Laboratory
Clarke	Amy	Los Alamos National Laboratory
Collins	Rip	Lawrence Livermore National Laboratory
Cowan	Thomas	Helmholtz-Zentrum Dresden-Rossendorf
Crandall	David	Department of Energy
D'Amico	Kevin	Argonne National Laboratory
Demarteau	Marcel	Argonne National Laboratory
Denes	Peter	Lawrence Berkeley National Laboratory
Desjarlais	Michael	Sandia National Laboratories
Distel	James	Los Alamos National Laboratory
Duffy	Thomas	Princeton University
Dufresne	Eric	Argonne National Laboratory
Eggert	Jon	Lawrence Livermore National Laboratory
Evans	William	Lawrence Livermore National Laboratory
Falcone	Roger	UC Berkeley and LBNL
Fezzaa	Kamel	Argonne National Laboratory
Flicker	Dawn	Sandia National Laboratories
Foiles	Stephen	Sandia National Laboratories
Fried	Larry	Lawrence Livermore National Laboratory
Funk	Dave	Los Alamos National Laboratory
Goldstein	William	Lawrence Livermore National Laboratory

Last Name	First Name	Organization
Graber	Timothy	University of Chicago
Gramsch	Stephen	Carnegie Institution of Washington
Goncharov	Alexander	Carnegie Institution of Washington
Gupta	Yogi	Washington State University
Haeffner	Dean	Argonne National Laboratory
Hau-Riege	Stefan	Lawrence Livermore National Laboratory
Hemley	Russell	Carnegie Institution of Washington
Hooks	Daniel	Los Alamos National Laboratory
Hu	Michael	Argonne National Laboratory
Hufnagel	Todd	Johns Hopkins University
Hurd	Alan	Santa Fe Institute
Iverson	Adam	National Security Technologies
Jacobsen	Chris	Argonne National Laboratory
Jensen	Brian	Los Alamos National Laboratory
Keane	Christopher	Lawrence Livermore National Laboratory
Kong	Lingping	Harbin Institute of Technology
Konopkova	Zuzana	Deutsches Elektronen-Syncrotron
Kumar	Mukul	Lawrence Livermore National Laboratory
Lavina	Barbara	University of Nevada Las Vegas
LeChien	Keith	DOE/NNSA
Lee	Hae Ja	SLAC National Accelerator Laboratory
Li	Liangliang	Harbin Institute of Technology
Liu	Haozhe	Harbin Institute of Technology
Luo	Sheng-Nian	Los Alamos National Laboratory
Mailhiot	Christian	Lawrence Livermore National Laboratory
Manghnani	Murli	University of Hawaii
Mao	Agnes	Carnegie Institution of Washington
Mao	David	Carnegie Institution of Washington
Meredith	Chris	Army Research Laboratory
Mills	Dennis	Argonne National Laboratory
Moy	Ken	National Security Technologies
Nelson	Art	Lawrence Livermore National Laboratory
Occelli	Florent	CEA
Patterson	Reed	Lawrence Livermore National Laboratory
Paz-Pasternak	Moshe	Tel Aviv University
Peralta	Pedro	Arizona State University
Pivovaroff	Michael	Lawrence Livermore National Laboratory
Prakapenka	Vitali	University of Chicago
Reed	Bryan	Lawrence Livermore National Laboratory
Rodriguez	George	Los Alamos National Laboratory

Last Name	First Name	Organization
Root	Seth	Sandia National Laboratories
Ruff	Jacob	Argonne National Laboratory
Sandy	Alec	Argonne National Laboratory
Sarrao	John	Los Alamos National Laboratory
Sattelberger	Al	Argonne National Laboratory
Schwartz	Adam	Lawrence Livermore National Laboratory
Schwegler	Eric	Lawrence Livermore National Laboratory
Seagle	Christopher	Sandia National Laboratories
Shen	Guoyin	Carnegie Institution of Washington
Smith	Jesse	Carnegie Institution of Washington
Stephenson	Brian	Argonne National Laboratory
Stevens	Lewis	Cooperative Resources International
Sun	Chengjun	Argonne National Laboratory
Tewari	Surya	University of Hyderabad
Thadhani	Naresh	Georgia Institute of Technology
Tschauner	Oliver	University of Nevada Las Vegas
Turneaure	Stefan	Washington State University
van Buuren	Tony	Lawrence Livermore National Laboratory
Vashishta	Priya	University of Southern California
Vernon	Stephen	Lawrence Livermore National Laboratory
Walko	Donald	Argonne National Laboratory
Wallin	Brad	Lawrence Livermore National Laboratory
Wang	Jin	Argonne National Laboratory
Wang	Lin	Carnegie Institution of Washington
Wang	Luhong	Harbin Institute of Technology
Weber	Franz	National Security Technologies
Willey	Trevor	Lawrence Livermore National Laboratory
Yang	Wenge	Carnegie Institution of Washington
Young	Linda	Argonne National Laboratory
Zahir	Islam	Argonne National Laboratory
Zellner	Michael	Army Research Laboratory
Zhao	Yusheng	University of Nevada Las Vegas
Zhuravlev	Kirill	University of Chicago
Zimmerman	Kurt	Washington State University

Washington State University

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