The science of dynamic compression at the mesoscale and the Matter-Radiation Interactions in Extremes (MaRIE) project

Cris W Barnes, David J Funk, Mary P Hockaday, John L Sarrao and Michael F Stevens
Los Alamos National Laboratory, Los Alamos, NM 87545
E-mail: cbarnes@lanl.gov

Abstract. A scientific transition is underway from traditional observation and validation of materials properties to a new paradigm where scientists and engineers design and create materials with tailored properties for specified functionality. Of particular interest are the regimes of materials’ response to thermo-mechanical extremes including materials deforming under imposed strain rates above the quasi-static range (i.e. $> 10^{-3}$ s$^{-1}$), material subjected to imposed shocks, but also material response to static, high-pressures. There is a need for the study of materials at the “mesoscale,” the scale at which sub-granular physical processes and inter-granular organization couple to determine microstructure, crucially impacting constitutive response at the engineering macroscale. For these reasons Los Alamos is proposing the MaRIE facility as a National User Facility to meet this need. In particular, three key science challenges will be identified: Link material microstructure to macroscopic behavior under dynamic deformation conditions; Make the transition from observation and validation to prediction and control of dynamic processes; and Develop the next generation of diagnostics, dynamic drivers, and predictive models to enable the necessary, transformative research.

1. Introduction
Materials research is on the brink of a new era from observation of performance to control of properties. The confluence of unprecedented experimental capabilities (e.g. 4th generation light sources, controlled synthesis and characterization, etc.) and simulation advances are providing remarkable insights at length and time scales previously inaccessible. New capabilities will be needed to realize this vision: In situ, dynamic measurements providing simultaneous scattering and imaging; well-controlled and characterized materials providing advanced synthesis and characterization; extreme environments of dynamic loading and irradiation; all coupled with predictive modeling and simulation to achieve materials design and discovery.

The science requirements set by these challenges have been identified, and functional requirements for experimental facilities defined to meet them. Key will be the study of materials at the “mesoscale,” the scale at which sub-granular physical processes and inter-granular organization couple to determine microstructure, crucially impacting constitutive response at the engineering macroscale. Present or currently being constructed facilities can meet some but not all of these functional requirements, leaving a capability gap to meet the needs of the science. The Matter Radiation in Extremes (MaRIE) facility has been proposed that, building on
LANSCE success, would be a key first step towards this vision. This paper presents background on the requirements and how they are met in the MaRIE 1.0 proposal.

2. The key science questions in materials dynamics

Workshops of the science community have defined very important challenges for material science, especially in extremes at the mesoscale using light sources [1–4]. At one workshop [5], five challenges were identified that capture the scientific needs that are required to achieve full understanding and that ultimately support the ultimate goal of moving from “observation to control.” These five challenges are as follows:

- Acquire time and spatially resolved in-situ measurements at all length scales (i.e., atomistic, micro-, meso-).
- Discover new physics and chemistry in extreme environments.
- Incorporate material complexity into multi-scale simulations to achieve predictive capability.
- Unify static and dynamic compression understanding across relevant length and timescales.
- Leverage scientific knowledge derived from theory and experiment to the design and control of real materials (i.e., chemistry, microstructure, defects, etc.)

Grand challenge priority research directions were identified including:

- Creating and exploiting the chemistry of a new periodic table through extreme Pressure-Temperature (P-T)
- Predicting, characterizing and controlling the performance of matter between solids and plasmas
- Determining the transport of energy, momentum and mass under extreme density and temperature

These challenges and priority research directions for materials dynamics are related to broader grand issues or key science questions for all of materials science research. While variants exist of the questions, they can be described as:

- How does one link micro-to-macro under dynamic loading? (the Multiscale Problem, related to BESAC Mesoscale challenge [6])
- How do we enable going from observation to control of dynamic processes? (the Co-Design Problem, related to the Materials Genome Initiative [7])
- What are the transformative new diagnostics, drivers, and predictive models needing development? (Transformative Technology)

The MaRIE project is intended to provide the experimental facility of the next decade and future that can provide the answers to these questions. The MaRIE 1.0 facility proposal [8] was officially developed in response to a call for future New Flagship Experimental Science Technology and Engineering Facility Concepts by the National Nuclear Security Administration. MaRIE 1.0 provides for the subset of capabilities and environments envisioned for a larger project [9].

An example of how MaRIE 1.0 can address the key science questions of material dynamics can be seen by considering a theoretical and computational approach to linking science at multiple scales by adaptive physics refinement. Figure 1 illustrates an approach to refining the type of model used in different regions of a shock traversing a material. From continuum hydrodynamics models at the largest scale, through refinement to poly-crystal, single-crystal, molecular dynamics and even atomic density functional theory approach, the different models can be used to reduce predictive uncertainty where the simulation requires it. Experimental observation at the mesoscale will couple multi-scale theory and multi-probe experiment on next-generation computing architectures. Coupled with flexible and responsive synthesis and
characterization capability, one can take theoretical predictions and experimental results and iterate on the materials themselves to provide a co-design capability. This leads to not just observation but designed and controlled functionality of the materials under dynamic extremes. To achieve this success, a new and transformative experimental facility will be required.

### 3. The challenge of dynamic mesoscale imaging

The challenge of mesoscale imaging is to observe the dynamic evolution of polycrystalline materials at the granular and sub-granular level. Coherently scattered hard X-ray photons provide the primary diagnostic tool. “One must balance the photons you need (coherently scattered), versus the photons you have (flux), versus the photons you can withstand (heating)” [10]. Peak brilliance can provide faster time resolution; the resulting energy deposition can modify significantly or destroy the sample and not allow observation of dynamic change.

Doing dynamic Mesoscale imaging appears possible with the use of very hard (> 40- to 70-keV) photons [11]. Subplots (a) and (b) in figure 2 provides examples of such analysis, showing how for mesoscale samples (multiple grains across and thus hundred’s of microns thick) such hard x rays are optimum for transmitting and, because the elastic cross section does not drop as fast with energy as the total cross section, even better for creating a larger fraction of coherently scattered signal. And because the total cross section and absorption does drop so rapidly with energy, as well as larger spot sizes reducing the intensity of the incident beam on the sample, even the peak brilliant pulses from an x-ray free electron laser (XFEL) can possibly heat a sample in only a small way, allowing multiple pulses.

Large spot sizes and high energy photons imply that achieving conditions of far-field, Fraunhofer diffraction require unrealistic sample to detector distances. The Fresnel number, $\text{Fr} = \frac{d^2}{\lambda L}$, where $d$ is the diameter or FWHM of the radial beam profile, $\lambda$ is the photon wavelength, and $L$ is the sample-to-detector distance, needs to be much less than 1 to be in the far-field. Coherent diffractive imaging is possible in the $\text{Fr} \geq 1$ regime as well [11], and would be used at MaRIE 1.0.

---

**Figure 1.** Variable-resolution models are synergistic with multi-probe, in-situ, transient measurements.
4. The MaRIE 1.0 Facility proposal

A rich set of dynamic materials science is currently underway at present light sources such as LCLS and APS [12, 13]. However such facilities have a capability gap for large-spot-size mesoscale imaging with multiple-pulse time resolution. The MaRIE 1.0 proposal fills that capability gap.

MaRIE 1.0 facility definition derives from “First Experiments and Campaigns” functional requirements and identified performance gaps. LANSCE provides significant site credit and accelerator operations expertise for MaRIE 1.0, including the ability to do both open and classified national security experiments on materials of interest (Pu, HE, etc.). Leveraging LANSCE’s existing 1-MW, 0.8-GeV proton accelerator, MaRIE 1.0 will provide: the world’s first very hard (42-keV) XFEL; a new Multi-Probe Diagnostic Hall (MPDH), coupling hard, coherent, brilliant x-ray photons with 12-GeV electron and 0.8-GeV proton radiographic tools in dynamic extremes; and a unique Making, Measuring, and Modeling Materials (M4) Facility for materials synthesis and characterization with high-performance computational co-design focused on the mesoscale. As figure 3(a) shows, MaRIE 1.0 will be unique amongst present and planned light sources in both its higher photon energy for an FEL and also its ability to provide a rapid micro pulse train of pulses on time scales to see sound speed motion across single grains. MaRIE 1.0, conceptually illustrated in figure 3(b), will be a national user facility, building on LANSCE infrastructure and leveraging community engagement.

5. Summary

The grand challenges of dynamic compression will be at the Mesoscale. Dynamic imaging at this scale requires large spot size and hard X rays for penetration of the multigranular samples. This leads to needing Fresnel-regime coherent imaging (which has been demonstrated). Further, probing polycrystalline metals non-destructively with femtosecond X-ray pulses is achievable with hard X rays, providing the opportunity to study materials dynamics at the mesoscale. MaRIE 1.0 will be the future facility meeting the requirements for this grand challenge.

Acknowledgments

The technical contributions of John Barber, Richard Sandberg, and Richard Sheffield are acknowledged with appreciation. The MaRIE 1.0 project would not exist without the strong support of the MaRIE Core team at Los Alamos.

Figure 2. (a) The fraction of incident photons coherently and elastically scattered once in a sample of a given thickness that then transmit through the sample. (b) The temperature rise in a sample normalized by the area of the incident beam when sufficient photons to generate a diffractive image are incident in a single pulse on a sample.
Figure 3. (a) Comparison of key facility functional requirements in multi-pulse repetition rate and photon energy to alternative facilities present or planned. (b) A conceptual drawing of the proposed MaRIE 1.0 facility.

[1] Wadsworth J, Crabtree G W and Hemley R J (eds) 2007 Basic Research Needs for Materials under Extreme Environments Office of Basic Energy Sciences
[2] Eberhardt W and Himpsel F (eds) 2008 Next Generation Photon Sources for Grand Challenges in Science and Energy Basic Energy Sciences Advisory Committee
[3] 2009 Decadal challenges for predicting and controlling materials performance in extremes Los Alamos National Laboratory Tech. Rep. LA-UR 10-02959
[4] Sarrao J, Mailhiot C, Gupta Y, Mills D and Kao C C (eds) 2012 New Research Opportunities in Dynamic Compression Science (Washington State University)
[5] Funk D, Gray R, Germann T and Martineau R (eds) 2009 A Summary Report on the 21st Century Needs and Challenges of Compression Science Los Alamos National Laboratory Tech. Rep. LA-UR 09-07771
[6] Crabtree G and Sarrao J 2012 From quanta to the continuum: Opportunities for mesoscale science A Report from the Basic Energy Sciences Advisory Committee
[7] Office of Science and Technology Policy 2011 Materials Genome Initiative Tech., Rep. Executive Office of the President
[8] Sarrao J et al. 2012 MaRIE 1.0: A flagship facility for predicting and controlling materials in dynamic extremes Los Alamos National Laboratory Tech. Rep. LA-UR 12-00500
[9] Experimental Physical Sciences VISTAS 2010 MaRIE: Matter-Radiation Interactions in Extremes Los Alamos National Laboratory Tech. Rep. LA-LP-10-059
[10] Stephenson B 2009 private communication
[11] Barber J L, Barnes C W, Sandberg R L and Sheffield R 2013 Diffractive Imaging at Large Fresnel number: The Challenge of Dynamic Mesoscale Imaging with Hard X rays Submitted to Phys. Rev. B
[12] Jensen B, et al. 2013 Dynamic experiment using IMPULSE at the Advanced Photon Source Submitted to these proceedings
[13] Ramos K, et al. 2013 In situ investigation of the dynamic response of energetic materials using IMPULSE at the Advanced Photon Source Submitted to these proceedings