White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	***24 July 2015***

Indicate the primary area this white paper addresses by placing "P" in right column.
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	"P", "S"
• Plasma Atomic physics and the interface with chemistry and biology	P
• Turbulence and transport	
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	"X"
Oral	
Poster	
Either Oral or Poster	X
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Title:	Exploring non-equilibrium material behavior under rapid compression with high throughput at LCLS
Corresponding Author:	Michael R. Armstrong
• Institution:	Lawrence Livermore National Laboratory
• email:	Armstrong30@llnl.gov
Co-Authors:	Jonathan C. Crowhurst, Joseph M. Zaug, Sorin Bastea, Nir Goldman, Alexander Goncharov (CIW)

(Limit text to 3-pages including this form. Font Times Roman size 11.
1 page of references and 1 page of figures may also be included. *Submit in PDF format.*)

- ***Describe the research frontier* and importance of the scientific challenge.**

The examination of transient states in materials under rapid compression is a scientific opportunity comparable to the examination of transition states established decades ago in physical chemistry ("femtochemistry"). Particularly when coupled with the powerful x-ray diagnostics available at MEC/LCLS (which are ideal for studying order in extended material), high throughput examination of transient material states driven by rapid dynamic compression is transformational. In contrast to electron-nuclear equilibration addressed in typical photoexcitation experiments, the examination of transition states subsequent to rapid compression of materials addresses the flow of energy from initially mostly potential (compressive) degrees of freedom to kinetic degrees of freedom such as thermal kinetic energy or chemical bond breaking. This broad topic covers a vast range of tantalizing unexplored phenomena, including *in situ* chemical kinetics (ultrafast "beaker" chemistry), correlations in mobile materials (such as liquids and plasmas) under metastable phase conditions, and mechanochemistry, while also enabling a revolution in throughput for the study of EOS of rapidly equilibrating materials such as hydrogen[1-5].

Here we broadly propose to study transient states in materials upon rapid compression over time scales less than 1 ns. Although dynamic compression experiments have historically been used to determine equilibrium equations of state (requiring relatively long time scale experiments), the exploration of transition states is particularly well suited to time and length scales accessible to LCLS. Beyond the compelling scientific potential of this proposal, short time scale experiments (which are well suited to study transition states in materials) enable several transformational practical advantages.

- ***Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.***

As we show below, relatively very low energy can be used for short time scale (< 1ns) compression experiments, enabling high repetition rates at average powers available from even commercial laser systems. Ultimately (as we also show), this enables substantially higher x-ray scattering throughput than available to longer time scale experiments.

This is a great practical advantage of short time scale experiments. Multiple shot, high rate data acquisition is a necessary element of several broad scientific topics, including the characterization of protein structure and the kinetics of material transformations in physical chemistry and material science. Compared to single shot experiments, high data rate experiments can explore larger phase spaces of material behavior (over a wider range of control samples), can discern subtle changes in behavior (via high signal to noise obtained with averaging), explicitly obtain statistical information relevant to error analysis, inhomogeneous variation of sample environments and stochastic material behavior, and enable the observation of unlikely events such as, for example, the homogeneous nucleation of melting in a superheated metastable state.

In particular, for ultrafast (< 1 ns) compression the laser drive energy required to obtain a given final state (at equilibrium or otherwise) varies as the third power of the laser drive pulse duration, which is typically comparable to the minimum compression time required to reach the state of interest. So, if a compression time of 1 ns is required to obtain a given final equilibrium state, 1000x more energy than necessary would be required to compress to the same state over 10 ns. As such, short time scale experiments are well suited to high repetition rate and low energy.

Traditionally, laser driven dynamic compression experiments have been designed to approach 1D compression (to more easily interpret data) by employing near-planar compression profiles. Analytically, the deviation of an actual experiment from planar compression is quantified by[6,7]:

$$\left|\frac{\delta v}{v}\right| < \left|\frac{\delta r}{r}\right| \propto \frac{1}{A^2}$$

where $\delta v/v$ is the deviation of the compression wave speed (for a curved wave front) from the wave speed in the 1D case, $\delta r/r$ is the fractional change in wave front curvature over the duration of the experiment, and A is aspect ratio (typically the sample width/thickness). Critically, the deviation of the wave speed from 1D is only a function of the aspect ratio of the experiment, and is independent of the time scale of the experiment. Thus, if the compression time scale is sufficient to obtain the final state of interest, *the deviation of the material state from what would be obtained in an ideal 1D experiment will be no worse than that obtained in a longer time scale experiment with the same aspect ratio*. Assuming similar final states are achieved, experiments at short time scales have the same deviation from planarity and exhibit similar systematic errors due to geometry as longer time scale experiments.

Further, the x-ray scattering throughput of high repetition rate compression experiments may be substantially higher than longer pulse experiments, primary due to the third power scaling of energy with drive duration shown above. The scattered intensity per shot increases at most linearly with sample thickness (i.e. drive pulse duration), but the energy per shot increases as the third power of the sample thickness (at constant aspect ratio as described above). So, the scattered x-ray intensity per unit laser drive energy varies as the inverse square of the scale of the experiment. Summarizing, for a drive laser with a given average power at variable repetition rate, *a 10x shorter experiment will have a 100x higher throughput* – this is a strong incentive to keep experiments as short as possible with a high repetition rate.

Generally, high repetition rate also uses less sample area per unit signal, generates less volumetric debris per unit signal, and, compared to high energy lasers with comparable average power, high repetition rate lasers (with commensurate smaller pulse energy) typically obtain better shot-to-shot repeatability. Also, high repetition rate lasers typically can achieve higher average power overall. Pulse shaping technology used for ultrafast lasers (a variation of which is used for sub-ns dynamic compression experiments[1]) is mature, and laser systems which already achieve transformational specifications for high repetition rate dynamic compression experiments are commercially available.

Small scale experiments may still be subject to issues like shot to shot pointing jitter (due to small drive spatial profiles), but this is typically accounted for in other contexts by characterizing the actual drive on a shot-to-shot basis, where a given diffraction pattern is directly correlated to the conditions of the sample at the probe position ("binning").

- ***Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit.***

The potential to study hydrogen under very dense, high temperature conditions is a strong driver of this proposal, since compressed hydrogen and its isotopes are fundamental to fusion energy generally. Further, from a practical standpoint, hydrogen is an ideal candidate for study on very fast time scales since it intrinsically equilibrates on a very rapid time scale.

More generally, analogous to transition states found in physical chemistry, materials under rapid compression exhibit complex behavior, often incorporating phenomena which do not occur in any other context, such as extreme elasticity, very rapid solid-solid phase transitions and chemistry, and, potentially, exotic hypothesized phenomena such as mechanochemistry[1-5]. Although it is difficult to predict how such a broad range of topics might impact society, the study of extremely rapid "ballistic" chemistry driven by the direct application of anisotropic stress has great potential to introduce new methods to synthesize materials, increase product yields, and enable detailed engineering of chemical synthesis.

References (Maximum 1 page)

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