Final Technical Report

Title	**Experimental Study of the Fundamental Properties of Warm Dense Mixtures**
Lead PI name, telephone number, email	**James Hawreliak** **(509) 335-1901** **James.Hawreliak@wsu.edu**
Address/City/State/Zip	**Institute for Shock Physics** **Washington State University** **1455 E. College Avenue** **Pullman, WA 99164-2816**
Performing Organization	**Washington State University** **PO Box 643140** **Pullman, WA 99164-3140**
Funding Office Contract Number	**DOE/Office of Science Program Office (FES)** **DE-SC-0016360**
Reporting Product Number	**DOE-WSU-0016360**

Abstract

The aim of the proposed research was to provide a multi-scale study of the properties of warm dense hydrocarbons, by studying the thermodynamic properties through the equation of state and the microscopic properties by x-ray scattering. Understanding the fundamental properties of warm dense mixtures is an intellectual challenge due to the complexity of the system. Unlike liquids or gases, where constituent particle interactions occur through collisions or bonding between valence electrons, in these strongly coupled systems the atoms will be partially ionized and compressed so tightly together that interactions between the inner core electrons can play a role in the systems chemistry. Advances in computational capabilities and development of new theoretical models have been used to predict the properties of mixtures but there is currently no experimental data of the fundamental interaction between the particles in mixtures to test these predictions against. The proposed research was to experimentally investigate the interaction of the different species in hydrocarbons by measuring the compressibility of substances with different carbon and hydrogen ratios and the complexity of the microscopic interactions through elastic and inelastic x-ray scattering.

To study the bulk properties of warm dense hydrocarbons we established the Warm Dense Matter Research Laboratory (WDMRL) in the Institute for Shock Physics at Washington State University. The goal was for experiments in the WDMRL to determine shock loading conditions of interest in the hydrocarbon mixtures. We planned on using Hugoniot EOS measurements and a range of carbon and hydrogen concentrations to determine conditions when the EOS of the mixture varied significantly from that of the classical mixing model. Even though shock transit measurements through aluminum foils suggested pressures upto 400GPa, the experiments in the WDMRL were unsuccessful in getting usable shockwave compression data above 100GPa in polystyrene which was below the pressure of interest for hydrocarbon mixtures (>200GPa).

To complete the project, we tested a technique using x-ray phase contrast imaging to map the location of tracer layers in a test sample of polycarbonate to record the material motion in dynamically compressed samples. A technique that will be useful for future warm dense matter experiments. These experiments used <200nm gold layers in polycarbonate samples to measure the material velocity and shock speed using x-ray phase contrast imaging. These results were compared to continuum surface measurements performed at ISP and show that the tracer layer technique can measure hydrodynamic properties accurately in dynamically compressed materials

Table Of Contents

Final Technical Report 1
Abstract 2
Table Of Contents 3
Introduction 4
Establishing the Warm Dense Matter Research Laboratory 5
  Experimental Capability 5
Work Force Development 7
  Benjamin Hammel 7
  Kory Mercer 8
Tracer Layer Experiments 9
Conclusion 11
References 12

Introduction

The goal of the project was to provide insight into chemical bonding in the extreme thermo-physical conditions inside giant planets[1] and inertial confinement fusion experiments[2] where traditional scientific approaches for describing the properties of matter breaks down[3]. This dense plasma state or warm dense matter (WDM) is too hot to be condensed matter and too dense to be a classical high temperature plasma. The study of mixtures and bonding in these conditions provides both intellectual excitement and challenges. The excitement comes from pushing the boundaries of our understanding of the phenomena by which atoms interact in extreme conditions. The intellectual challenge of warm dense mixtures lies in the complexity of the system. Unlike liquids and gases, where constituent particle interactions typically occur through collisions or bonding between valence electrons, in warm dense mixtures the atoms will be partially ionized and compressed so tightly together that interactions between the inner core electrons can play a role in the systems chemistry[4]. While advances in computational capabilities and the development of new theories have been used to predict the properties of mixtures in extreme conditions experimental data is still required to build an understanding and test the predictions of these models[5]. This project planned to study the properties of warm dense hydrocarbons, shock and multiple shock compressed to high-pressure and high-temperature states.

The scientific importance of warm dense matter continues to grow as observations of planets outside our solar system challenge current models and ICF science plots a course for the future. Experimental data that illuminates the fundamental properties of warm dense mixtures is critical in building accurate models of materials in extreme conditions. The "Report of the 2009 Workshop on Basic Research Needs for High-Energy-Density Laboratory Physics" [6] in this regime to support ICF science and astrophysical modeling. Hence, the need for accurate WDM data has never been greater.

The interiors of giant gas planets are complex warm dense mixtures. The understanding of these mixtures is key to building accurate models of these planets and building the knowledge of particle interactions in extreme conditions. Demonstrating these techniques as a means of studying the fundamental properties of mixtures will open up research to a broad range of mixtures like hydrogen/oxygen mixtures where the bonding is predicted to be ionic in nature, and more complicated mixtures like carbon/hydrogen/oxygen mixtures. Since the information is of a fundamental nature the results will be used to validate models that can be used to predict transport properties.

Our objectives were to establish the Warm Dense Matter Research Laboratory in the Institute for Shock Physics. The WDMRL at ISP provides access to an important physical regime and bridges the gap between gas gun experiments and the extremes generated by the facilities at the national laboratories. Having this capability at an academic institution gives WSU the responsibility for great scientific research, the duty to educate and grow the next generation of scientists, and the privilege to foster intellectual liberty to explore novel ideas that plant the seeds for future research. The education and exploration requires rapid access to smaller scale facilities, like the

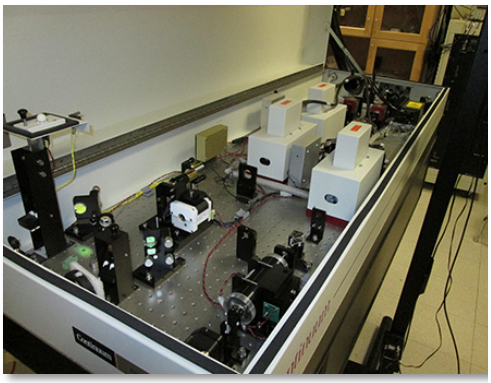
laser and line-VISAR system at ISP, where researchers can perform simple experiments and determine the value of pursuing. It may also be preliminary experiments are required to determine what capabilities are needed. In this project we successful established a rapid pump-probe capability.

Establishing the Warm Dense Matter Research Laboratory

We developed the Warm Dense Matter Research Laboratory in the Institute for Shock Physics at Washington State University. It is a unique table-top scale facility were extreme states generated through shock compression can be explored in an academic environment. Under this project we have been combining the pump laser with the line-VISAR diagnostic. The equipment for the line-VISAR (Velocity Interferometer System for Any Reflector) was funded from a grant from the Murdock Charitable Trust. The pump laser is a 12J, 1064nm Nd:YAG system was already present in ISP. A fast Pockel's cell is being used to pulse shape the beam to generate a 10nsec semi-flattop laser pulse from the Gaussian profile that comes from the oscillator. The shaped pulse is amplified through two larger amplifiers. The pulse can also be stretched to 60nsec gaussian and used to launch fliers upto 6km/sec. The line-VISAR uses a 532nm laser and fast streak camera to measure velocity in one spatial dimension as a function of time. The line-VISAR system has demonstrated 5 micron spatial resolution and 50psec temporal resolution. We have used shock transit measurements through 50 micron thick aluminum foils to infer a shock pressure of 400GPa.

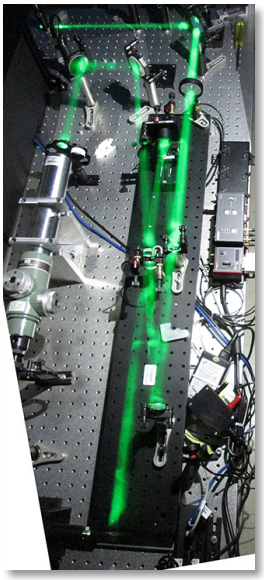
Experimental Capability

The line-VISAR system is a custom-built instrument used to provide time dependent velocity information along one spatial dimension. It is comprised of three main sub-systems: a laser source, streak camera detector, and interferometer. To obtain the desired time resolution and interference characteristics a bright single mode pulsed laser with temporal pulse shaping capability is required. The interferometer provides the interference from which the velocity can be interpreted. This needs to be aligned and stable to a fraction of the wavelength of the laser (532nm) to ensure optimal velocity resolution. The streak camera detector requires picosecond timing to provide the temporal resolution. The line-VISAR uses a Continuum Agilite laser provides a bright, pulsed, single mode laser source of 30mJ per 200ns pulse at 532nm wavelength run at 10Hz operation.

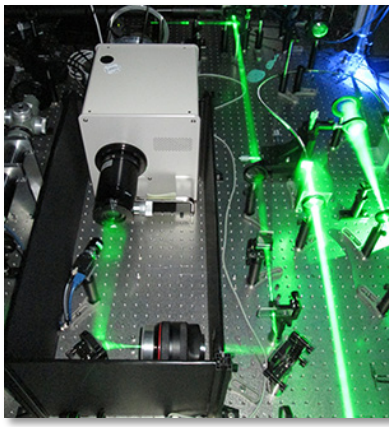


The Continuum Agilite laser setup for use in the Warm Dense Matter Research Laboratory

The interference is generated by a custom-fabricated Mach-Zehnder interferometer with a thick glass etalon in a single arm of the interferometer. The etalon provides a fixed path length difference between the two arms, while maintaining the imaging needed for spatial resolution. The interferometer needs to be a robust diagnostic that is stable and can be rapidly aligned to the needed accuracy. The interferometer uses beam splitters from BMV Optical Technologies to meet the demanding specifications required. The path length difference is established by large glass etalons ordered through Precision Optical. The mechanical components were purchased through Thorlabs and assembled in the WDMRL. The interferometer has been aligned to better than a wavelength of light (532nm) and has demonstrated stability that has exceeded expectations. Due to the compact design, the mirror mounts that hold the beam splitter are not true gimbal mounts and care must be taken when determining the zero-path length difference reference



Mach-Zehnder interferometer in the line-VISAR system



Hamamatsu streak camera incorporated into the line-VISAR system

The detector needs to have both high spatial and temporal resolution. The Hamamatsu C7700-01 is a newly developed high dynamic range streak camera that can handle a large number of photoelectrons. This feature enables single-shot measurements of ultrafast phenomena with a dynamic range as high as 10,000:1 along with a temporal resolution of 5ps. The Hamamatsu has advantages over other streak cameras because it provides an intuitive graphical user interface and easily adjustable probe window time of 500ps to 1ms. The Hamamatsu C7700-01 has demonstrated picosecond resolution and the large dynamic range necessary.

The fabrication and implementation of the line-VISAR system was part of this project and required engineering expertise from Institute for Shock Physics personnel. As described above, the line-VISAR system required the integration of a pulsed laser system, complex optical components, and fast streak camera detector. The biggest challenge in fabrication and implementation was obtaining picosecond time synchronization. The diagnostic system needed to be synchronized across a wide range of timescales for a single event experiment. The system needs to maintain a 10Hz repetition rate firing the flash lamps in the laser to ensure stability of the probe laser, millisecond timing for the flash lamps to pump the laser medium, and picosecond synchronization of the probe pulse to the streak camera detector. While challenging at times,

diagnosing and correcting a range of issues related to system timing developed a very robust line-VISAR system for experiments in the WDMRL.

Figure 1 is representative data from the line-VISAR system in the WDMRL for both the direct ablation step targets and laser launched flyers. In figure 1a) since the free surface velocity measurement is not clear we can use shock transit times to get an estimate of stress. The laser launch flyer in figure 1 b) reaches a velocity of 5.2 km/sec

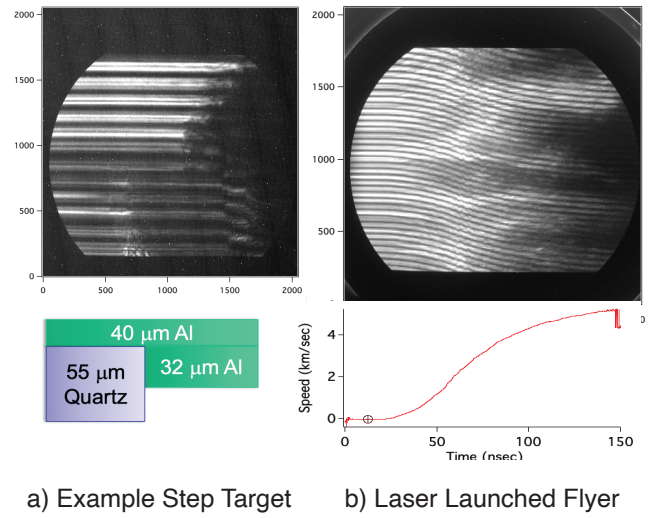


Figure 1: Representative line-VISAR data from the Warm Dense Matter Research Laboratory.

Work Force Development

Benjamin Hammel

Dr. Benjamin Hammel was hired at WSU as a Postdoctoral Research Associate to investigate warm dense mixtures using the line-VISAR system in the WDMRL. Dr. Hammel received his Ph.D. from the Department of Physics at the University of Nevada, Reno before coming to WSU in Oct 2016. Dr. Hammel's primary focus was to measure the shock-compressed properties of polystyrene (CH) at high pressures and temperatures. Dr. Hammel has learned the safe operations of a class 4 laser systems used for both the pump and the probe of the shock compressed matter. For the Warm Dense Matter Research Laboratory this involved the challenging aspect of synchronizing the two lasers systems with the fast streak camera diagnostic. Dr. Hammel developed skills in interferometry and a better understanding of how to measure velocity from the interference signal. He developed computational routines required to analyze the data. He also worked on 2D hydrodynamic modeling of the experiments. Dr. Hammel was partially supported on this project.

Dr. Benjamin Hammel, a postdoc hired to perform warm dense matter experiments with line-VISAR system

Kory Mercer

Korey Mercer was a fourth year undergraduate student in the Department of Physics and Astronomy at Washington State University. He built a soft x-ray pinhole camera that can be used to image the ablation plasma during the shock experiments. The goal of the project was two fold: 1) to have an imaging diagnostic that could give spatial resolution of 10microns of the ablation plasma and 2) a spectral diagnostic that could be used to determine the emission from the plasma. The spatial images would be used to determine the spot size and correlate laser intensity to shock pressure. The spectral information would be used to measure the photon spectrum and assuming the plasma and emitted radiation are in thermal equilibrium determine the temperature of the plasma.

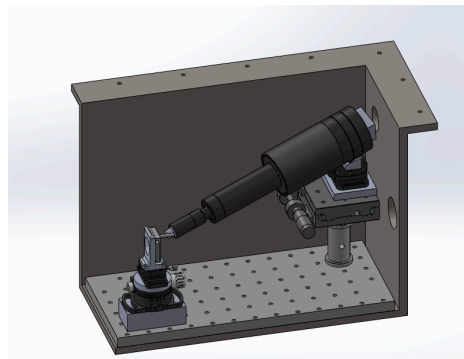


Figure 2: Soft X-ray Pinhole camera design for WDMRL.

The pinhole camera can do both, the first by classically imaging the emission, the second by reducing the photon flux to the single hit regime and then the CCD signal will correlate to the incident photon energy. Korey was responsible for determining the pinhole camera characteristics to achieve both objectives. His design is shown in Figure 2. This included: determining the sample, pinhole and CCD distances to achieve the desired magnification; the pinhole size to get the desired resolution; filtering to get the desired photon flux assuming a 1keV blackbody spectrum from the plasma. He also was responsible for the engineering aspects of fitting the camera into the target chamber.

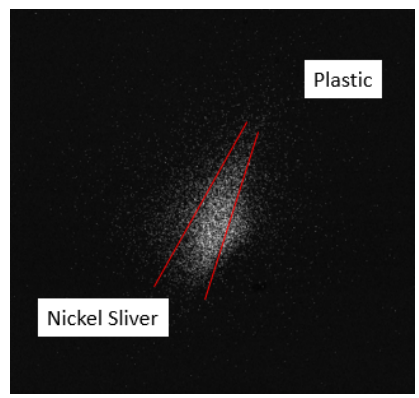


Figure 3: X-ray Pinhole Camera image

Korey's pinhole camera was successful on both fronts. First the imaging capabilities could image engineered targets with specific features , figure 3. This was critical in the development of the Warm Dense Matter Research Laboratory as the pinhole camera was able to image the astigmatic laser focal spot that was not observed in other diagnostics. This required significant changes to the system. With changes in filtering the camera was able to resolve line emission from a nickel plasma that could be used to calibrate the energy spectrum measured from the plasma, figure 4. This was a great introduction to High Energy Density Physics research to Korey.

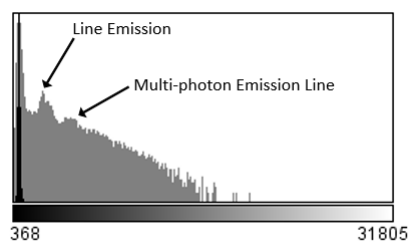


Figure 4: Spectrum in single hit mode showing Ni K-shell peak,

Korey Mercer is currently a staff scientist at the Dynamic Compression Sector.

Tracer Layer Experiments

Experiments were performed at the Dynamic Compression Sector (DCS) at the Advance Photon source in Argonne National Laboratory[7]. The experiments used a two stage light gas gun to launch polycarbonate and copper impactors up to 5km/sec to impact into our samples. The DCS is a unique user facility dedicated to dynamic compression.

With the advent of bright single pulse x-ray sources coupled to high energy density drivers it is now becoming possible to probe the properties of warm dense systems generated by dynamic compression in-situ. The performed work investigated using tracers layers in a sample to study the material flow. X-ray Phase Contrast Imaging (PCI) offers the ability to non-destructively interrogate the processes ongoing inside a material during dynamic compression[8]. The challenge is that these images are sensitive to density and opacity variations, not material movement. So to visualize the motion of material requires regions with varying opacity. In this case the tracer layers we use are <200nm gold layers vapor deposited onto 0.5 mm thick polycarbonate sheets stacked to form a 1.5mm thick sample which is bounded to a 1.5mm buffer and 1.5mm window, all of the sample polycarbonate, figure 5.

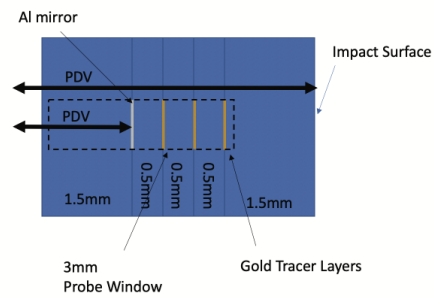


Figure 5 : Schematic diagram of samples used in tracer layer experiments

An example of a static image (taken before the experiment) and a dynamic image are shown in figure 6. The detector uses four framing cameras which can record upto two frames each. This gives a total of eight frames which can be captured at 153nsec intervals, the storage ring frequency. The static image a) shows the gold tracer layer and the aluminum mirror which separates the sample and window as sharp features. The sample and gun are turned at an angle until the x-rays probe along the interface. The probe window is large enough that the free surface is observable. The dynamic image shows the displacement of the tracer layers and the visible shock front propagating through the sample. The displacement of the tracer layers is measured with respect to the impact surface.

Figure 7 is a plot of the relative transmission of the x-rays through the sample. The features associated with the gold tracer layers and the shock front can easily be identified. The near constant opacity suggests the density is uniform after the shock. In regions where there is a rapid variation in density (or more specifically index of refraction) there are characteristic phase

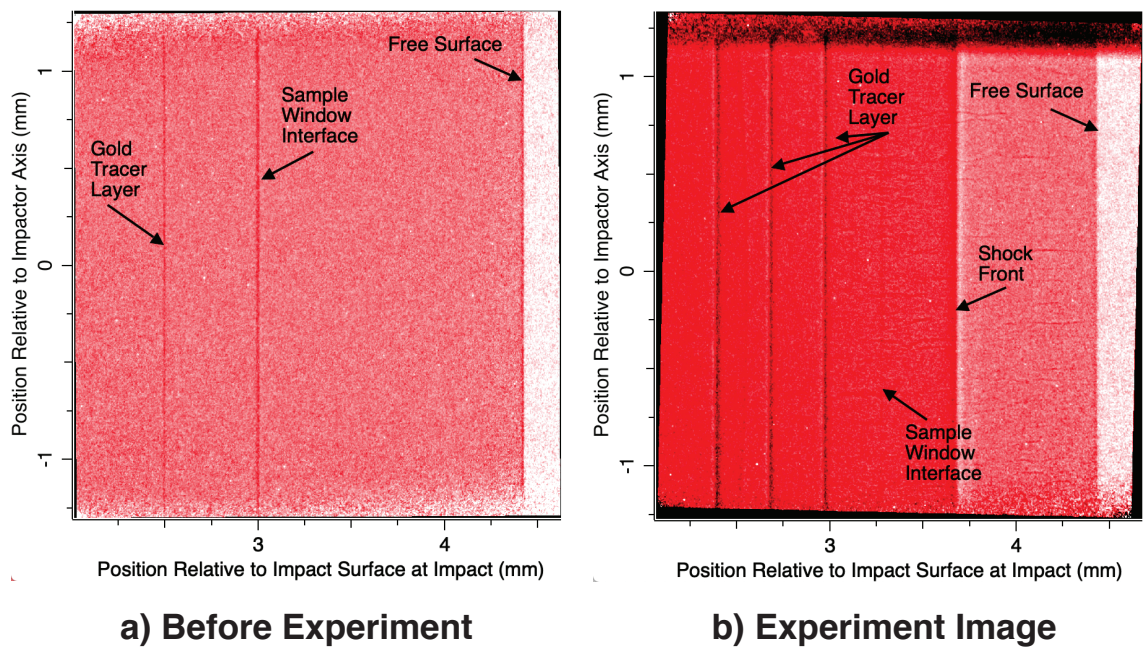


Figure 6: a) a static image of the sample before experiment, b) a dynamic image during shock compression.

contrast features. Particularly noticeable are the features at the shock front where the transmission is high before the shock front and then is much lower after. Because the x-rays propagate after passing through the sample the refraction of the x-rays due to the density variation causes the features which enhanced sharp density changes.

Importantly, we wanted to compare the results obtained using X- PCI to more classical techniques for the Hugoniot properties of the polycarbonate. To this end we preformed classical symmetric impact experiments on the power and 2-stage light gas gun at the institute for shock physics. In these experiments the shock speed was measured by the transit time of the shockwave through the sample and the material velocity was measured using half the impactor velocity in a symmetric impact experiment. We plot the measured shock speed versus material speed for both this experiments in figure 8. The points on the graph show excellent agreement between the DCS and ISP data (and the LANL shock material compendium). In the Figure the marker sizes are larger than the error bars.

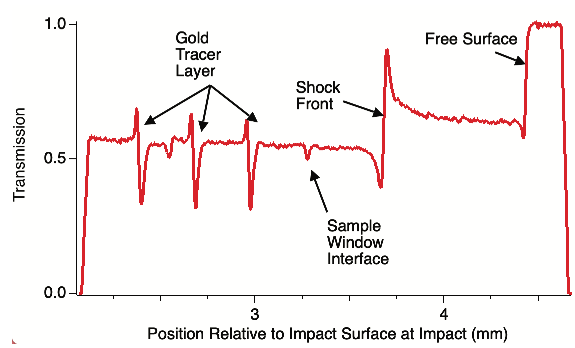


Figure 7: Relative transmission through the sample.

Conclusion

This project had some challenges achieving its desired goals but was successful in establishing a unique pump probe capability using a laser driven shock and line-VISAR diagnostic in the Warm Dense Matter Research Laboratory at Washington State University. It was also successful demonstrated that tracer layers can provide reliable information about material flow inside materials using PCI. The advantage of the PCI over conventional imaging is that sharp edges can be enhanced by the phase contrast.

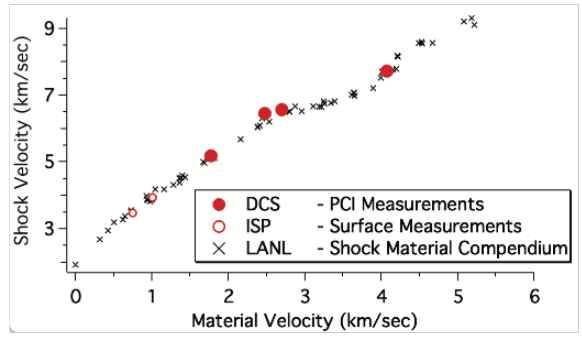


Figure 8: Shock speed versus material velocity from ISP and DCS experiments.

References

1. Chau, R., S. Hamel, and W.J. Nellis, *Chemical processes in the deep interior of Uranus.* Nat Commun, 2011. **2**: p. 203.
2. Haan, S.W., et al., *Point design targets, specifications, and requirements for the 2010 ignition campaign on the National Ignition Facility.* Physics of Plasmas, 2011. **18**(5): p. 051001.
3. Murillo, M.S., *Strongly coupled plasma physics and high energy-density matter.* Physics of Plasmas, 2004. **11**(5): p. 2964.
4. Militzer, B. and H.F. Wilson, *New Phases of Water Ice Predicted at Megabar Pressures.* Physical Review Letters, 2010. **105**(19).
5. Redmer, R. and G. Röpke, *Progress in the Theory of Dense Strongly Coupled Plasmas.* Contributions to Plasma Physics, 2010. **50**(10): p. 970-985.
6. Dehmer, J., R. Staffin, and E. Smith, *Report of the Interagency Task Force on High Energy Density Physics*, in *National Science and Technology Council Committee on Science Interagency Working Group on the Phyiscs of the Universe*. 2007.
7. https://dcs-aps.wsu.edu.
8. Hawreliak, J.A., et al., *Dynamic Behavior of Engineered Lattice Materials.* Sci Rep, 2016. **6**: p. 28094.