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<td>• Plasma self-organization</td>
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<td>• Statistical mechanics of plasmas</td>
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Indicate type of presentation desired at Town Hall Meeting.

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Title: Resolving the structure of warm dense materials

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• Describe the research frontier and importance of the scientific challenge.

When materials are compressed with high power laser driven shock waves, they will undergo phase transitions into a dense charged-particle system that is dominated by strong correlations and quantum effects. This complex state, known as warm dense matter (WDM), exists in planetary interiors\(^1\)-\(^4\), high power laser laboratory experiments, and during the initial compression phase of inertial confinement fusion implosions\(^5,6\). The theoretical description of matter at these conditions is very challenging as standard solid-state theories and plasma expansion techniques do not apply. The experimental investigation of these states are equally challenging because these plasmas are extremely dense, and require active probing by high-power penetrating x-rays or particle beams. Additionally, such measurements become progressively difficult due to the transient nature of this state of matter. Many dynamic techniques such as Velocity Interferometer System for Any Reflector (VISAR) and Streaked Optical Pyrometry (SOP) used to study this regime are limited to the material surface boundary\(^7,8\), and are not directly probing the structure and microphysics of the system.

The growing interest in understanding and predicting the properties of warm dense matter necessitates the development of accurate \textit{in situ} temperature and density probes throughout the entire material-shock evolution. In many laboratory, laser-based shock-wave experiments, a variety of plasma regimes are created, and of particular interest are the ones where the ions are strongly coupled, and the electron subsystem is degenerate. Whereas degeneracy in weakly coupled systems can be solved, strongly coupled systems represent a greater challenge for a complete theoretical description. The plasma-to-solid phase
transition \cite{9,10} in these degenerate systems plays a critical role in the understanding of the equation of state (EOS) of materials under extreme conditions \cite{11}. Understanding how materials react to pressure is a critically important scientific challenge in high energy density (HED) physics, as the thermodynamics and the hydrodynamics of WDM cannot be accurately predicted without knowledge of the EOS.

Simultaneous high-resolution angularly and spectrally resolved x-ray Thomson scattering has proven to be an accurate method to measure the electron density, temperature, and pressure of highly compressed dense plasmas \cite{12,13}. This technique probes the bulk properties of matter deep inside dense materials and is not limited by refraction and reflection at the surface boundary. Previous x-ray scattering experiments from WDM have focused on measuring and studying the inelastic scattering by free electrons in the plasma and in some cases, bound-free or resonant inelastic scattering transitions \cite{14-16}. The characteristics of an ultra bright, ultrafast, collimated, and narrowband coherent x-ray source have allowed for the development of a novel experimental platform from which accurate measurements of the total elastic ion feature from x-ray scattering can be measured \cite{17}. This measurement is directly related to the ion-ion static structure factor and also contains information about the electron density from both the bound electrons and the screening electron cloud around the nucleus \cite{18}. Such novel experiments of the elastic scattering peak promise to be a sensitive and informative test of the ionic structure of the plasma.

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

Experiments performed at the Linac Coherent Light Source (LCLS) Matter under Extreme Conditions (MEC) end-station have demonstrated novel in situ measurements of the electron temperature, pressure and density by simultaneous high-resolution angularly and spectrally resolved x-ray scattering from shock-compressed aluminum (Fig. 1) in the warm dense regime \cite{13}. The experiments use an ultra-bright seeded narrowband x-ray beam to spectrally resolve plasmons for density and temperature measurements \cite{19}. Simultaneously using the unique high x-ray brightness properties of LCLS, to measure the ion-ion static structure factor, offers the capability to independently verify the measured properties from first principles while additionally yielding information about the structure and precise quantum mechanical state of the plasma.

The high peak brightness x-rays at the LCLS are required to resolve ionic interactions at atomic (Ångstrom) scale lengths and to determine their physical properties. Moreover the high photons per pulse make it possible to obtain data in a single shot during the short lived state of highly compressed phenomenon. Measurements on aluminum have characterized the compressed lattice and resolved the transition to WDM (Fig. 1), demonstrating that short-range repulsion between ions must be accounted for to obtain accurate structure factor and EOS data. The angularly resolved elastic scattering amplitude from WDM has recently been measured with higher precision and accuracy than has been possible before the commissioning of the MEC end-station at the LCLS. However the capabilities at MEC and the LCLS need to be improved to push the physics into highly unknown states of matter where large discrepancies in theory have yet to be tested by experiments.

The proposed LCLS-II x-ray capabilities will greatly enhance the opportunity for discovery in this regime \cite{20}. Using the higher photon energies available with LCLS-II, higher order correlations in the ionic structure can be probed thereby allowing even more accurate measurements of the material structure. These explorations of WDM will greatly benefit from LCLS-II, with higher x-ray energies allowing the study of denser material and higher-Z elements, and with multiple x-ray pulses facilitating investigations of ps-scale shock wave dynamics. Additionally, upgrading current lasers capabilities from the current 15 J/pulse to 100 J/pulse will allow access to greater density regimes where phenomenon such as shell effects in degenerate matter (Fig. 2), exotic material phase transitions, and pressure ionization can be directly investigated.
• Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit.

Accurate knowledge of the ionization balance, the thermodynamic properties, and the EOS of dense plasmas are of fundamental importance towards precisely modeling WDM and materials in the HED physics regime. Such information is relevant for understanding matter at pressures above 1 Mbar and temperatures beyond 1 eV. The ability to directly measure the underlying structure and properties of matter in this regime can have a significant impact to the scientific community and help solve some of the most contested and important science questions in the high-pressure community.

Hydrogen

Hydrogen, the simplest element in the universe, has a surprisingly complex phase diagram. Owing to fundamental questions about the structure and core stability of gas giants in our solar system \(^{[1-3]}\), hydrogen’s high-pressure properties have been the subject of intense study over the past two decades. Of critical importance is the EOS of hydrogen and deuterium. A precise determination will affect our understanding of the formation and evolution of the solar system and will further affect our ability to achieve inertial confinement fusion in the laboratory. The need for reliable EOS at degenerate electron densities and temperatures of a few eV’s becomes critical. Numerous experiments (Fig. 3) \(^{[21]}\) using laser drivers and z-pinchs have provided EOS data that show significant uncertainties for pressures above 50 GPa \(^{[2,3,21,22]}\). What these studies have in common is the use of VISAR to measure shock and particle velocities to infer pressure and compression. Accurate structure factor information from angularly resolved scattering could resolve these discrepancies.

Atomic shell structure of degenerate matter

For plasmas close to solid density, the simplest and most widely used Thomas-Fermi (TF) model of the electrons shell structure becomes reliable only for \(T_e > 50\ eV\). However, at Mbar shock pressures when a degenerate material is partially ionized the statistical TF model is no longer applicable as it treats the electron states as a continuum \(^{[23]}\). The EOS, for materials in this regime, depends on the precise quantum mechanical state (i.e. the electronic shell structure), and becomes difficult to model accurately. In such extreme environments small fluctuations in the structure of neighboring ions will drastically influence ionization mechanisms and electron localization of the subsystem resulting in predicted material pressure conditions from the EOS that can be different by two orders of magnitude (Fig 2.). The error bars from many experimental measurements in this regime are either too large to provide the necessary insight into the existence of shell effects, or accurate measurements are incapable of recording data at high enough material densities to evaluate the discrepancies (Fig 2.). Structure factor measurements are a direct test of the ionic structure predicted by many models of warm dense systems. The high accuracy of such a measurement allows one to directly evaluate the precise quantum mechanical state. It is a very promising prospect for studies of WDM that data on the elastic feature can be obtained with enough accuracy to clearly distinguish between these models at material densities approaching 9 g/cm\(^3\).
References

Figure 1 - Wavenumber-resolved scattering data $W(k)$. Bottom: Bragg peaks from Debye–Scherrer rings at $t = 0$ ns. Second from bottom: data from compressed solid aluminium show a shift of the (111) Bragg peak by $3^\circ$ and compression of $1.21 \rho_0$. Middle: the appearance of a broad ion–ion correlation peak is observed, together with a Bragg peak shifted further to larger angles. Second from top: at higher compression, the aluminium melts, Bragg peaks disappear, and the ion–ion correlation peak shifts to $50^\circ$. Top: the angle of the correlation peak increases further to $56^\circ$ when higher densities are reached after coalescence. The data (black curves) show excellent agreement with DFT-MD simulations (top three panels). The data can also be described by a model that accounts for both Yukawa screening and short-range repulsion (SRR) (red, blue and green solid curves), but a Yukawa-screened potential alone is not sufficient (dashed curves). [13]

Figure 2 - Theoretical EOS models, plotted for high pressure and compression, where the various models exhibit differences in predicted conditions due to k–shell ionization behavior in highly dense aluminum. A comparison of the pressure data obtained from experiments along with data from the ion–ion structure factor for compressed dense aluminium during shock coalescence compressed along an isentrope [13].

Figure 3 - Phase diagram of hydrogen and deuterium showing the equation of state versus compression along the shock Hugoniot [21]. Shock data obtained from various platforms on cryogenic D$_2$ and H$_2$ compared to various EOS models.