

High Energy Density Science with FELs, Intense Short Pulse Tunable X-ray Sources

R. W. Lee^a, S. J. Moon^a, H.K. Chung^a, R. C. Cauble^a, S. Glenzer^a, O. L. Landen^a, S. J. Rose^b, H. A. Scott^a, G. Gregori^c, D. Riley^d

^aL-399 Lawrence Livermore National Laboratory, Livermore, CA

^bDepartment of Physics, Clarendon Laboratory, Oxford University, Oxford, UK

^cCentral Laser Facility, Rutherford Appleton Laboratory, Chilton, UK

^dPhysics Department, Queen's University Belfast, Belfast, Northern Ireland

ABSTRACT

Short pulse (< 100 fs) tunable X-ray and VUV laser sources, based on the free electron laser (FEL) concept, will be a watershed for high energy density research in several areas. These new 4th generation light sources will have extremely high fields and short wavelength (~ 0.1 nm) with peak spectral brightness—photons/(s/mrad²/mm²/0.1% bandwidth)— 10^{10} greater than 3rd generation light sources. We briefly discuss several applications: the creation of warm dense matter (WDM), probing of near solid density plasmas, and laser-plasma spectroscopy of ions in plasmas. The study of dense plasmas has been severely hampered by the fact that laser-based probes that can directly access the matter in this regime have been unavailable and these new 4th generation sources will remove these restrictions. Finally, we present the plans for a user-oriented set of facilities that will incorporate high-energy, intense short-pulse, and x-ray lasers at the first x-ray FEL, the LCLS to be opened at SLAC in 2009.

Keywords: High Energy Density Science, Free-Electron lasers, Fourth Generation Light Sources

1. INTRODUCTION

Since the late 1960's plasma-based research has moved toward higher density regimes. The advent of laser-produced plasmas and laser-based plasma diagnostics has fueled interest in the formation of plasmas at densities nearing solid density. There are two separate areas where the next generation sources can play a critical role in moving these fields substantially forward. The first is in the area of warm dense matter research, where FLASH, a VUV-FEL at DESY, the Linac Coherent Light Source (LCLS), a XFEL in construction at SLAC, and a proposed XFEL at DESY, will provide major improvements over the current state of the field. The second is in the area of plasma spectroscopic techniques where the role of the 4th generation light sources, which are in this regard essentially x-ray lasers, will provide substantive improvements. For more information on these facilities see the website <http://www-ssrl.slac.stanford.edu/LCLS/> for information on the LCLS facility, the website <http://www-hasylab.desy.de/facility/fel/> for information on the XFEL facility and the FLASH VUV-FEL a soft x-ray FEL facility.

For the 4th generation sources we note that whether we are interested in creating warm dense matter, performing Thomson scattering, or probing a plasma the LCLS/XFEL capability provides a major advance on any capability that exists with 3rd generation sources. The key to the advance is the tunable, narrow band x-ray source with very short pulse duration. Since the individual bunch photon intensity is the essential quantity for all the plasma-based research, using the peak spectral brightness best summarizes the comparison of the LCLS/XFEL to current synchrotron sources. Indeed, one finds a 10 orders of magnitude enhancement that will make the LCLS/XFEL a most promising source for plasma-based research. The utility of the high repetition rate of other sources, *e.g.*, APS or ESRF, are not useful here since we require a single photon pulse to either heat, scatter, or probe matter that is transient. Indeed, to create solid matter that is at temperatures greater than 1 eV temperature, which can be studied before expansion, on an x-ray light source will require the capability of x-ray FELs like LCLS/XFEL. This, then, will provide for WDM samples that have gradients that are small when compared to the size of the warmed volume. Further, the incorporation of a high-energy laser source at the future XFEL facility provide, *e.g.*, shock heating. In this manner the x-ray FELs will make significant advances into the study of the warm dense matter regime.

Second, the spectroscopic probing of high energy density plasmas requires a short pulse high-energy source. For the source to be a useful spectroscopic probe requires spectrally tunability and a number of photons per mode that is on the order of unity. Due to the high peak brightness the 4th generation sources this capability will be possible in the x-ray regime for the first time.

Third, and finally, to measure the Thomson signal in a dense plasma or the warm dense matter regime requires a Thomson probe, here the 4th generation light sources, with temporal duration that is short compared to the evolution of the system but long enough to probe the plasma collective modes. The nominal 200 fs pulse duration will be able to probe the electron feature, arising from the collective behavior of the electrons.

1.1 Warm Dense Matter

With a short duration pulse containing a substantial number of high-energy photons or ions one can generate solid matter at temperatures of ≤ 10 eV, *i.e.*, warm dense matter. The interest in the warm dense matter regime arises because in dense plasmas the atoms and/or ions will start to behave in a manner that is intrinsically coupled to the plasma. That is, the plasma starts to exhibit long- and short-range order due to the correlating effects of the atoms/ions. This intriguing regime where the plasma can no longer be considered a thermal bath and the atoms are no longer well described by their isolated atom behavior provides a tremendous challenge to researchers. In the limit of dense cool plasmas one obviously arrives at the threshold of condensed matter. Here the problem has changed from a perturbative approach to ground-state methods where complete renormalization of the atom/ion and its environment is essential.

From the perspective of plasma studies the defining quantity is the coupling parameter Γ , *i.e.*, the ratio of the interatomic potential energy to the thermal energy given by the equation:

$$\Gamma = \frac{Z^2 e^2}{r_0 k T} \quad \text{with} \quad r_0 = \left(\frac{3Z}{4\pi n_e} \right)^{1/3} \quad (1)$$

where Z is the ion charge and r_0 is the interparticle spacing given in terms of the electron density n_e .

The regions of interest span the density-temperature phase space going from modestly coupled ($\Gamma \leq 1$) to strongly coupled ($\Gamma > 1$), while bridging the transition regimes between solid to liquid to plasma.

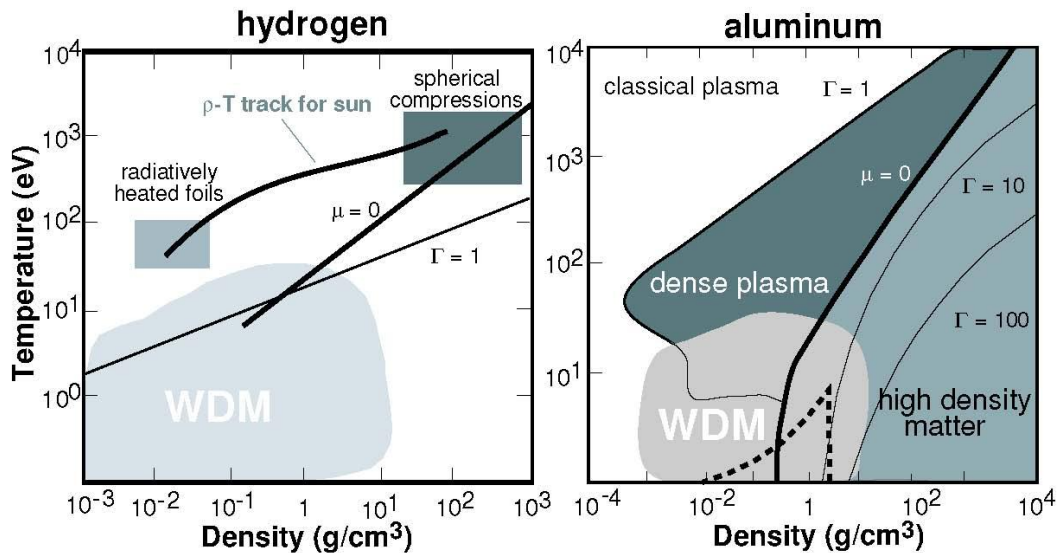


Fig. 1. The temperature-density phase diagrams that show hydrogen on the left and aluminum on the right. The relevant regimes are noted, as are the various values of the strong coupling parameter Γ . The regions greatest uncertainty are roughly noted by the black outlined areas. Also indicated is the region where degeneracy will become important: it is the region to the right of the line where the chemical potential $\mu = 0$. The hydrogen data is taken from a compilation of data from the NRL Plasma Formulary¹ while the aluminum data is derived from the QEOS formalism².

In Fig. 1 above we show the region of the temperature-density plane where warm dense matter studies are important. Here we show the temperature (T) in eV versus the density (ρ) in g/cm^3 both for hydrogen, a low Z element, and aluminum, a moderate Z element. The region where the theoretical uncertainties are largest are those where the standard theoretical approaches fail and experiments are exceedingly difficult. The difficulty arises theoretically from the fact that this is a regime where there are no obvious expansion parameters, as the usual perturbation expansions in small parameters used in plasma phase theories are no longer valid. Further, there becomes an increased importance on density-dependent effects, *e.g.*, pressure ionization, as the surroundings starts to impinge on the internal structure of the ion or atom. Experimentally the study of warm dense matter is difficult, as the isolation of samples in this regime is complicated. Indeed, although the plasma evolution of every ρ - T path that starts from the solid phase goes through this regime and plays an important role in its evolution, trying to isolate warm dense matter remains a major challenge.

It has been exceedingly difficult to perform experiments in the warm dense matter regime, which is, simply, why we know so little about it. As a first step, one must create a well-characterized warm dense matter state; the second is to gain information on the state through experiments. The first step has been the problem: warm dense matter is not a limiting case of matter, *e.g.*, high- or low-temperature. When created in a laboratory environment, it does not tend to remain in a specified thermodynamic state for very long, making characterization difficult. The only other imaginable method to produce the kind of warm dense matter of interest with in a laser-based experiment may be to use sub-30-fs laser pulses on sub-100-Å-thick foils and perform thermodynamic measurements on a few-fs timescale over extremely small spatial dimensions. To be able to do this on comparatively macroscopic samples with 4th generation sources will be a boon.

1.2 Plasma Spectroscopic Studies

There is great interest in the higher temperature dense plasma regime. Here the problem arises from the production of high temperature plasmas at electron densities in excess of 10^{22} cm^{-3} . In any experiment where a high intensity, *e.g.*, $I \geq 10^{12} \text{ W/cm}^2$, laser irradiates a solid target there will be a region of the solid that is hot and near solid densities. Lasers with wavelengths $> 0.25 \mu\text{m}$ do not directly heat the solid as they can not propagate beyond the critical electron density, $n_{\text{cr}} \sim 10^{29} \text{ cm}^{-3}/\lambda^2(\text{\AA})$ (see Rosenbluth & Sagdeev³ for general references to the concepts of laser-produced plasmas and plasma physics); however, heat flow from the surface efficiently generates the hot dense medium. The spectroscopic information derived from these plasmas provides, on the one hand, diagnostic information about the plasma itself, while on the other hand we can investigate, using spectroscopy, our understanding of the mechanism at play in the creation of the plasma and the interaction of the atoms/ions with the plasma in which it is embedded. The LCLS/XFEL will provide two related and intriguing possibilities. First, there is the possibility to perform diagnostics on plasmas at solid density.^{4,5} Second, we can explore laser pump-probe techniques for high density plasmas that have been used in low densities plasmas to measure line shapes, observe radiation redistribution, and determine the kinetics processes. For an example of the radiation pumping see Back⁶ and Koch⁷ where it can be seen that to date in higher density plasmas only the total emitted fluorescence has been studied in photopumping experiments of ion emitters. While for discussions of the kinetics processes see Foord⁸ and Bar-Shalom⁹.

The mechanisms involved in the formation of a plasma and the details of the kinetics processes taking place can be illuminated by using a laser as a pump to selectively populate levels and thus redistribute radiation. In a particularly intriguing possibility one will be able to study the formation of laboratory x-ray lasers that currently depend on kinetics processes.¹⁰ Thus, one could disentangle the plasma production from the inversion-forming processes that lead to the x-ray lasing. It is clear that numerous aspects of plasma spectroscopy have been severely constrained by a lack of data. The 4th generation sources will provide a substantial improvement in the development of our understanding of intrinsic line shape formation, level shifts, radiation transfer, and detailed kinetics processes.

1.3 Thomson Scattering

Thomson scattering provides an *in situ* measurement of the temperature, density, charge state, and collective behavior of the plasma. Indeed, the Thomson scattering diagnostic is directly related to the dynamic structure factor, $S(k, \omega)$, of the plasma and thus provides insight into the theoretical predictions from different theories. It is fair to say that in recent years each effort at diagnosing a higher density plasma, *i.e.*, higher than 10^{20} cm^{-3} , using Thomson scattering has led to new and important discoveries.¹¹ These experiments have, of course, been few since the constraints on the experiments are substantial. Here we believe that the 4th generation sources will provide a major advance in diagnosing dense plasmas. This is clearly a complement to the concept of creating warm dense matter, as Thomson scattering can provide a diagnostic of the warm dense matter conditions. However, the preconditions for the interpretation of the scattering data is that there is a valid theoretical model for the $S(k, \omega)$ in the high density regime, and this in itself will be a

challenge. The tunable nature of the x-ray source, the high energy, bandwidth, the short pulse duration and, importantly, the very high peak photon flux make this source the only one that can address the Thomson scattering of transient plasmas.

2. OVERVIEW OF THE HEDS EXPERIMENTAL STATION AT LCLS

There is now a plan which has been submitted to the LCLS Science Advisory Committee, and approved, that set out a proposal for an experimental station at the Linac Coherent Light Source (LCLS). This will create a capability to perform High Energy Density Science (HEDS) covering a vast range in the temperature-density phase space. While HED experiments, in which the response of solid material to heating to various degrees of ionization, are performed on existing facilities, the capabilities provided by the x-ray XFEL will remove the temperature and density gradients that presently limit the volume of material that can be studied. Thus, HEDS, which has been slowly developing on x-ray light sources, will burgeon with the advent of the LCLS. For example, experiments on current light sources, where heating and/or perturbing matter is achieved, the light source is the probe and not the pump. We will use the unique x-ray laser-like qualities of the LCLS – short pulse duration, high bunch photon numbers, 120 Hz repetition rates, and photon energy tunability – to make a full range of HEDS experiments possible. As there are no alternatives to this unique x-ray source the research proposed here is dependent on access to the LCLS.

Table 1: List of experiments incorporated in the High Energy Density Science experimental station proposal

Experiment	Brief Description
Warm Dense Matter Creation	Use XFEL to uniformly warm solid density samples
Equation of State Measurements	Heat and probe a solid with an XFEL to provide a diagnostic of material properties
Absorption Spectroscopy	Heat a solid with an optical laser or XFEL and use the XFEL to probe
Shock Phenomena	Create shocks with a high-energy lasers and probe with the XFEL
Surface Studies	Probe ablation/damage process to study structural changes and disintegration processes
XFEL / Gas Interaction	Create exotic, long-lived highly perturbed electron distribution functions in dense plasmas
XFEL / Solid Interaction	Use XFEL directly to create extreme states of matter at high temperature and density
Plasma Spectroscopy	Use XFEL as a pump to excite bound-state populations and study radiation redistribution
Diagnostic Development	Develop the XFEL for Thomson scattering, interferometry, and radiographic imaging

In broad outline, it is assumed that the experimental station will be placed in the far hall contingent on the possibility for focusing the LCLS to ~10 micron spot sizes. This is also a consideration for the implementation of interferometers, and monochromators needed for the experiments discussed below. The experimental capability includes two experimental areas, one is an independently placed laser hall, see Fig. 2, and the other is in an experimental hutch in the far experimental hall, see Fig. 3. The laser hall would have two optical lasers systems, one would be a short pulse system, ~1 J 50 fs, and the other would be a high-energy laser, >100J, 1 ns. The combination of the LCLS with these optical lasers will open a new frontier in HED research.

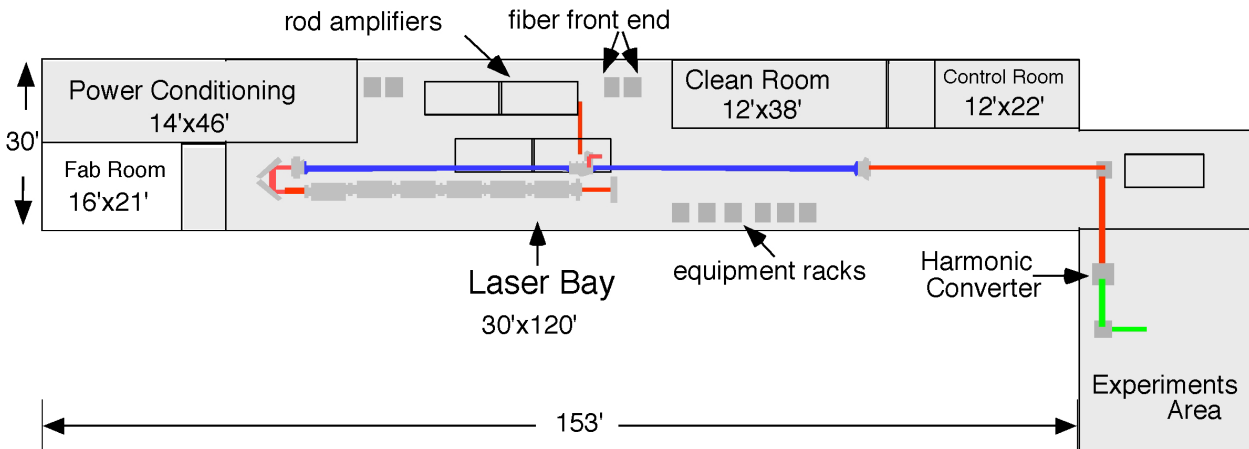


Figure 2) The laser hall with an 8 kJ long pulse laser (shown) and a TW short pulse laser. The laser beams will be transported to the end station shown in Fig. 2. There will also be a stand-alone experiments area in the laser hall that will have a capability to match that of the end station.

The uniqueness of the facility has attracted a number of researchers to a capability that is by definition in its formative stages. We present the case for research into the two broad areas of which the HED regime is composed, *i.e.*, the Warm Dense Matter (WDM) region and the Hot Dense Matter (HDM) region. They correspond to ideal and highly correlated plasmas, respectively. The case for the uniqueness of the LCLS can be made for each separately. 1) The HDM regime requires the use of x-rays to probe the dense medium, sub-picosecond source to perform the measurements on the timescales of importance to the hot dense systems, and a focusable source to probe the spatial scales of importance. 2) The WDM regime can be created directly by the LCLS or other sources (e.g., laser driven shocks) and can be probed by the LCLS with the requirements of short time duration, intense bursts of x-rays.

Below we will schematically indicate the areas of research, representing them as three areas of research covering a wide range of topics from condensed matter physics to plasma science. The experiments will be grouped into three categories, see Table 1: WDM regime (WDM creation, Equation of State studies, Absorption spectroscopy, Shock Phenomena, Surface studies); HDM regime (gas/XFEL interactions, Focused XFEL/Solid matter interactions, Plasma Spectroscopy); and, Diagnostic Development (Thomson scattering, Interferometry, radiographic imaging).

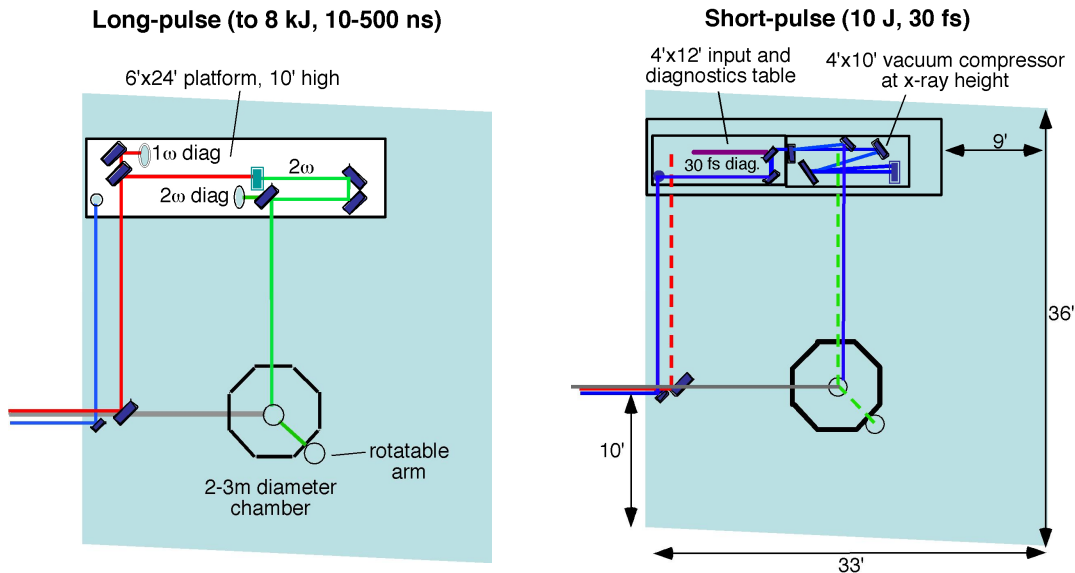


Figure 3) The end station in the LCLS Far Experimental Hall showing the transported long pulse laser in red and frequency doubled to green, and the short pulse TW laser in read, along with the XFEL beam that come straight into the chamber.

3. OVERVIEW OF HIGH ENERGY DENSITY SCIENCE EXPERIMENTS

As an illustration of the type of experiment that could be performed with the XFEL we provide the following simple example. First, we assume that we have an 200 fs pulse of 10^{12} photons with 8 keV focused to a $10 \mu\text{m}$ by $10 \mu\text{m}$ spot. The sample irradiated will be a normal density aluminum slab of $100 \mu\text{m}$ extent chosen to use only 66% of the beam energy in photoabsorption. We calculate the state of the aluminum in two ways: First, we use a simple model to deposit the absorbed energy, estimated by using the photoabsorption of Al to 8 keV photons, as a balance between total kinetic energy of the system and the ionization created. In this way we can estimate temperature, T_e and ionization of the sample by equating energy E , deposited in volume V to the internal and kinetic energy of the sample. The energy per unit volume goes into electron kinetic energy and ionization and thus is given by the formula

$$E/V = 3/2 n_e T_e + \sum n_i IP_i, \quad (2)$$

where IP_i is the ionization potential of ion stage i and n_i is the number in that ion stage. For Al there are three electrons in the conduction band, so that when we heat these electrons without ionization, which will occur, for example, at 1 eV, we assume that the deposited energy, with sufficient rapidity, goes into these conduction band electrons.

This model indicates that we obtain a state of Al that is at 10 eV and has an electron density of $2 \times 10^{22} \text{ cm}^{-3}$ giving a mean charge state of $\langle Z \rangle \sim 0.3$. To verify that this simple model is correct we performed a hydrodynamic simulation¹² using the specifications of the sample and the XFEL beam and found agreement. In fact the assumption that the heating

is uniform was verified and the expansion after heating was found to be isentropic. The track of this simple example is seen in Fig. 1 on the aluminum phase diagram as the dashed line. The hydrodynamic simulation further indicated that the pressure reached 4 Mbar and the velocity of release would be $\sim 1.6 \times 10^6$ cm/s. Indicating that movement of the surface would be 160 Å which is much less than a fraction of a percent of the aluminum bulk.

3.1 Experiments on LCLS in the WDM regime

3.1.1 WDM creation using the LCLS at 1 Å can volumetrically heat a sample via photo-absorption. Calculations indicate that $10 \mu\text{m} \times 10 \mu\text{m} \times 100 \mu\text{m}$ of Al can be heated to 10 eV in this manner. Here one would strive to determine the evolution of the system from strongly non-thermal non-equilibrium to an equilibrium state while determining the spatial uniformity.

3.1.2 Equation of State measurements (EOS) of matter at high pressure (exceeding >1 Mbar) are fundamental to numerous applications in astrophysics, geophysics, high-pressure science, plasma physics, laboratory laser experiments, inertial confinement fusion, and related fields. Experimentally accessing these material states offers opportunities to study phase transitions in solids near the critical point, strong coupling effects in dense plasmas, and transport properties at high densities and temperatures. However, even in the most studied materials one finds that in the WDM regime there are significant differences between models. Since the EOS relates temperature, volume, and pressure an EOS measurement requires determining two of the three state variables independently. The experiments envisioned would build on WDM creation techniques and LCLS probing to provide measures of the pressure-density-temperature relationship defining the EOS.

3.1.3 Absorption Spectroscopy using the LCLS can serve as a probe of material dynamics of WDM or the interatomic dynamics of shocked materials. By closely examining the structure of the spectrum in the vicinity of an absorption edge, information about local electronic and atomic structure can be extracted. The absorption spectrum very close to an edge yields information on the local bonding properties, ionization and chemical environment of a particular element. Further above the edge, small oscillatory modulations due to interference from photoelectron scattering gives information on the short range nuclear order near the absorbing atom. Forays into the realm of time-resolved spectroscopy have recently approached the ultrafast time-scale.^{13,14} Although alternate short time-duration sources and fast detectors suited for x-ray absorption are under development, scientific progress in this area will be greatly advanced by high peak brightness of the LCLS.

3.1.4 High Pressure Studies using the LCLS in tandem with a high-energy laser will allow one to measure EOS in a complementary manner to the direct x-ray heating techniques. The shock or isentropic compression technique can sample the Hugoniot, the locus of points in pressure-density-temperature phase space accessible by single shocks, which for these experiments will be in excess of 1 Mbar. Here we will augment the usual measurements by providing faster time resolutions and smaller spatial scale probing than will be possible elsewhere. The possibility to probe the microscopic state of the shock front, i.e., measuring the local temperature, composition and ionization state will provide critical new capability to the understanding of matter in these extreme conditions. The connection between the strong shocks described here and the warm dense matter regime is very strong. As a example, the highest compression that can be achieved by single shock in solid H_2 is approximately 4-fold, which gives a density of 0.3 g/cc. However, the pressure (and temperature) can be varied by many orders of magnitude by changing the energy of the driver. Thus, the shock is essentially heating the sample isochorically with a large degree of tunability in the pressure. Further, higher densities can be accessed with multiple shocks.

3.1.5 Surface Studies using either a short pulse optical laser or the LCLS directly can create novel state of matter that can be probed by the short pulse x-rays of the LCLS. The interaction of intense x-rays with matter differs from conventional laser-matter interaction as one will be able for the first time, to excite directly deep lying electron states in the atomic time scale. The high photon density is expected to lead to new kinds of matter excitations such as collective motion of inner atom shells or nonlinear processes due to interactions of highly excited states. Further, the role of non-thermal phenomena such as Coulomb explosions, photo-induced bond breaking and ionization excited by multi-high-energy-photon absorption can be studied for the first time. Since the goal here is to study the processes modifying the irradiated surfaces, time dependent reflectivity measurements, time and wavelength dependent fluorescence measurements, energy and mass spectra of the ejecta, and photoelectron spectroscopy will be employed.

3.2 Experiments on LCLS in the HDM regime

3.2.1 XFEL-gas interactions is built on two unique properties of the intense x-ray source: the ability to photoionize gas and create an electron distribution function (EDF) with sharp energy distribution defined by ionization stages, and the small value of the ponderomotive potential compared to optical lasers at the same intensity. The latter allows creation of the plasma with high degree of spatial uniformity, free of parametric instabilities and sources of super-energetic particles. Thus, for the first time one can engineer hot uniform plasmas with prescribed EDFs over a wide range of conditions. In the proposed experiment, the gas will be both irradiated and probed (via Thomson scattering) by the same LCLS beam. Thomson scattering (TS) measurements are used to determine plasma parameters and details of the EDF. They will provide a proof-of-principle for x-ray TS and will enable further developments and applications of this powerful diagnostic. The anisotropy of EDF in photoionized gases gives rise to unique plasma physics processes, such as electrostatic two-stream instability, Weibel instability, magnetic field generation, and terahertz radiation emission. Measurements of the magnetic field and of the emitted radiation will provide additional detailed diagnostics of these plasmas.

3.2.2 XFEL-solid interactions have a strong precedent in the long history of the use of high-power laser beams to generate high energy density matter in the laboratory. An outstanding application is inertial confinement fusion. In this context, it is of interest to use short-wavelength laser light to couple efficiently to the solid. The plasma generated by the LCLS will be unique as one can expect primary photo-electrons to have energies ~ 10 KeV, while simple estimates using Spitzer formulas indicate that free electrons thermalize within 10 fs, while excited bound states live longer, and ions equilibrate only after 1 ps. However, more detailed studies indicate that the electron distributions stay non-thermal for substantially longer, which will be monitored via the emission spectra observing the effects on the line shapes, line shifts and ionization potential depression. Optical probing of x-ray heated non-conducting material via reflection and transmission will be used to characterize the time dependent evolution of the electron relaxation.

3.2.3 Plasma spectroscopy is concerned with the measurements of the kinetic rates or the populations. However, a major impediment has been the inability to probe *in situ* HDM plasmas. Further, the population kinetics of highly stripped ions that occur in HDM plasmas is difficult due to the large number of states that must be considered in a model and the detail to which one must incorporate these states. The situation is made more difficult due to the fact that these plasmas tend to have rapid time evolution and large spatial gradients. The initial goal for the LCLS experiments will be to create a plasma using a high-energy laser and then with the LCLS selectively pump a single line transition. Variations on the idea of pumping individual transitions in high energy density plasma include the selective pumping of the wings of a line transition to observe redistribution within the line profile and pumping of selected transitions to attempt to understand the inversion mechanisms for the production of x-ray lasers.

3.2.4 Laser Pump Probe Techniques

The 4th generation sources will be employed in plasma-based experiments to address the foundation of plasma creation in transition to hot dense matter providing a truly unique method to probe the spectroscopy of the hot dense matter. The probing of dense plasmas, whether these be warm or hot, will move to a new level of sophistication with the use of x-ray Thomson scattering. While, the active probing of the a hot dense plasma will be advanced by extending the methods of laser fluorescence spectroscopy that are employed in low density plasmas with visible lasers to high density using these x-ray laser sources. Several papers indicate that the field of laser induced fluorescence is currently limited to neutral or near-neutral species.^{15,16,17}

Since the creation of high-density laser produced plasmas there have been virtually no quantitative *in situ* measurements of the kinetic rates or the populations. This is a major impediment to progress, as population kinetics of highly stripped ions is a complex problem. The complexity derives from the large number of states that must be considered in a model and the detail to which one must incorporate these states. The situation is made more difficult yet due to the fact that these plasmas tend to have rapid time evolution and large spatial gradients.³

Indeed, much of the effort to improve the situation has been focused on target design and advanced diagnostic development; however, the difficulties in determining the level populations or the kinetic rates remain. Therefore the interest, which comes from all areas involved with dense plasma studies and its underlying theoretical problems, *e.g.*, laboratory x-ray laser generation, laser plasma production, astrophysics, and inertial fusion, has never been met with substantial improvements in experiments.

The import of the 4th generation sources for high energy density plasma experiments is that one can use these x-ray sources to pump individual transitions in a plasma creating enhanced population in the excited states that can be easily

monitored. The idea has been used in lower density plasmas with visible lasers and can, with the 4th generation sources, be employed to advance the study of high density plasmas. As examples, one can look at the work of Burgess¹⁸ and Razdobarin¹⁹.

Variations on the idea of pumping individual transition in high energy density plasma include the selective pumping of the wings of a line transition to observe redistribution within the line profile and pumping of selected transitions to attempt to understand the inversion mechanisms for the production of laboratory x-ray lasers. In all of these applications the tests of the theoretical developments in the areas of atomic processes, kinetics model creation, line shape formation, and x-ray gain studies would be the first of their kind as there are currently no available probes.

There are several constraints on the x-ray source for it to be useful as a laser probe of the high energy density regime. First, the probe must be tunable and this is easily satisfied. Second, the line width of the pump must be such that it can pump entire line profiles and also be capable – for studies of redistribution within line profiles – of pumping parts of the line profile. Again, these conditions will be readily met. Finally, we need to have a pump that can move enough population from one state to another so that the population changes can be monitored. This last requirement can be verified by looking at the radiative pumping rate, R_{LU} , due to the source compared to the spontaneous emission rate, A_{UL} , of the transition being pumped. This is proportional to the number of photons per mode and is given by Elton²⁰

$$\frac{R_{LU}}{A_{UL}} = 6.67 \times 10^{-22} \frac{g_U}{g_L} \lambda_L^5 I_o^{laser} \left(\frac{W}{cm^2} \right) \frac{[,] }{\delta_\lambda \Delta_\lambda} \quad (3)$$

where the g 's are the statistical weights of the upper and lower states, λ_L and I_o are the source wavelength and intensity. The δ_λ and Δ_λ are the bandwidths of the x-ray source and the line shape of the transition being pumped, respectively, while $[,]$ represents the minimum of the two. Two important insights emerge when evaluating Eq. 3. First, if we conservatively assume $I_o \sim 10^{14}$ and $[,]/\delta_\lambda \Delta_\lambda \sim 0.001$ we find that the ratio is approximately 1 for λ_L of 10 Å. This number is at least 10^3 larger than can be obtained by using a plasma source to pump a transition. Second, the ratio does not increase with decreasing source wavelength, indicating that large numbers of photons per mode will not be available as we move toward shorter wavelengths. This is due to the fact that the spontaneous rate has a strong inverse dependence on wavelength. Of course, matching, or at least controlling the source bandwidth can have salutary effects as indicated by Eq. 3.

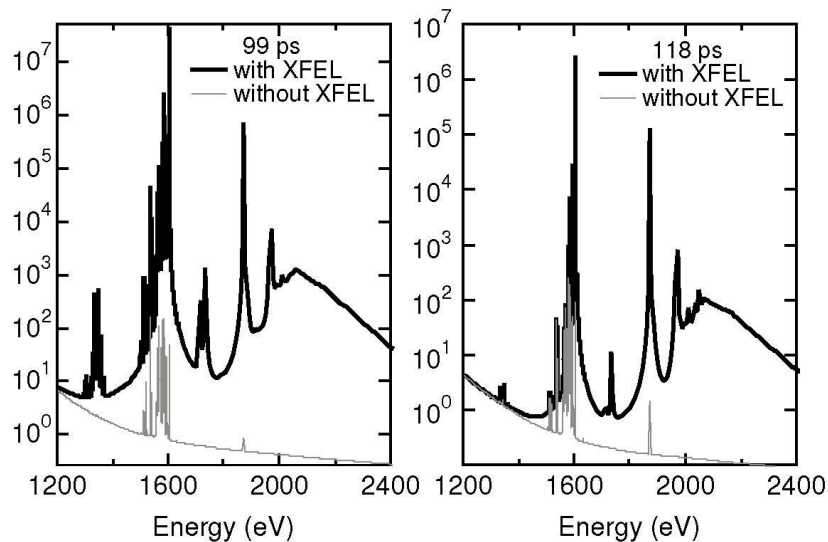


Fig. 4. The logarithm of the emissivity versus spectral energy for two times in the evolution of an exploding aluminum foil. The emission from the plasma with no x-ray pump is the thin line while the thick line spectra indicate the emission when the XFEL pumps the He-like $n=1$ to $n=3$ transition. The He-like emission, from, *e.g.*, $n=2, 3$ and 4 at 1598 eV, 1868 eV, and 1963 eV, respectively, increases substantially while the H-like $n=2$ emission at 1724 eV arises with the pump.

The possibilities provided by plasma spectroscopic probing are illustrated with the simulation of an aluminum layer tamped on both sides with a thin layer of CH plastic irradiated by x-rays.²¹ An undiluted radiation field emitted from the

rear side of a 1000 Å Au target impinges on the Al foil, heating it uniformly. The Au foil is irradiated by a single 1 ns, temporally square-shaped pulse of 0.52 μm light at an intensity of 1.6×10^{14} W/cm².^{22,23,24}

The emission spectrum will be the observable and it is shown in Fig. 4 at two times in the evolution of the plasma, one near the initiation of the x-ray pulse and the other at 18 ps later. The most notable feature is that there is a substantial increase in the hydrogenic transitions. For example, the Lyman α line at ~ 1724 eV which is unobservable in the He-like background emission without the LCLS/XFEL pump, rises well above the background with the x-ray pump. Further the structure of the He-like resonance series starting at ~1598 eV and ending at the bound-free continuum near 2086 eV is substantially changed by the pumping. Indeed the Li-like satellite transitions seen on the low energy side of the He-like $1s^2-1s2l$ transitions are substantially enhanced. The major effect of the pump, although it is tuned to a particular transition, is to cause photoionization due to the pump strength. The ionization of the Li-like stage and the pumping of population from the He-like ground state up to the H-like ion stage cause a slow recombination decay back towards the He-like ground state.

It is clear from the emission spectra shown in Fig. 4 that one could use a fraction of these x-ray sources as pumps to generate observable signals. The detailed information that can be obtained from these measurements would provide unique constraints on the complex processes necessary to construct a complete kinetics model for the highly charged ions. Indeed, we chose to use as an example the K-shell spectra as it is easily interpretable; however, the generation of L-shell and M-shell models is also of importance and raise the level of complexity substantially. Thus, one can understand the need for experiments that can provide basic information on the processes necessary to build kinetics models.

3.3 Diagnostic Development for High Energy Density Science

3.3.1 Thomson scattering, which will be a primary focus for HEDS end station, provides an *in situ* measurement of the temperature, density, velocity, charge state, and collective behavior of a plasma. Indeed, the Thomson scattering diagnostic is directly related to the dynamic structure factor, $S(k, \omega)$, of the plasma and thus provides insight into the theoretical predictions of the constituent particle velocity distribution functions predicted by different theories. In recent years each effort at diagnosing a higher density plasma, *i.e.*, higher than 10^{20} cm⁻³, using Thomson scattering has led to new and important discoveries. These experiments have, of course, been few since the constraints on the experiments are substantial. Here we believe that the next generation sources will provide a major advance in diagnosing dense plasmas, particularly in the WDM regime. The advantage of Thomson scattering using the LCLS as a probe versus a conventional optical laser is three fold: First, the short wavelength increases the critical density to which the probe beam can reach. Secondly, the susceptibility of the probe beam to refraction effects is reduced making the probe focus location more reliable, particularly in the steep density gradients close to the target surface in a laser produced plasma from a solid target. Lastly, the tunable photon energy from LCLS makes it possible to select a wavelength so $S(k, \omega)$ is optimized for the desired diagnoses. However, the preconditions for the interpretation of the scattering data is that there is a valid theoretical model for the $S(k, \omega)$ in the high density regime, and this in itself will be a challenge. The tunable nature of the x-ray source, the high energy, bandwidth, the short pulse duration and, importantly, the very high peak photon flux makes the LCLS the only source that can address the Thomson scattering of high-density transient plasmas.

One can extend the power of spectrally resolved Thomson scattering to the x-ray regime for direct measurements of the ionization state, density, temperature and the microscopic behavior of strongly coupled plasmas and warm dense solids.⁴ This would be the first direct measurement of microscopic parameters of solid density matter, which could be used to properly interpret measurements of material properties such as thermal and electrical conductivity, equation of state (EOS) and opacity found in astrophysical environments as well as in virtually all plasma production devices. We note that in standard current plasma physics use the concept of Thomson scattering is use to explain the spectrum derived when a probe scatters from the free electron in a plasma. Actually, ignoring photoabsorption, there are three possible scattering processes that occur when photons interact with a plasma: 1) There is true Thomson scattering, a coherent process, in which in the photons interact with the tightly bound electrons – indeed as the electrons are coupled to the nucleus there is essential no Compton shift; 2) there is the Compton shifted scattering from those weakly bound electrons that have ionization energies less than the Compton shift – this is an incoherent process; 3) there is the Compton shifted scattering from the unbound, *i.e.*, free, plasma electrons – this is an incoherent process. In the standard plasma literature the latter scattering process has come to be called the “Thomson” scattering. As we speak of plasma type studies we will retain this nomenclature here, but note the various components schematically.

Thomson scattering is characterized by the scattering parameter α , proportional to the ratio of the laser probe scale-length, λ_L , to the screening length, λ_S , and the scattering angle Θ :

$$\alpha = \lambda_L / 4\pi\lambda_S \sin(\Theta/2) = 1 / [2k_L\lambda_S \sin(\Theta/2)] \quad (4)$$

For $\alpha < 1$, spectrally-resolved incoherent Thomson scattering provides information on the velocity v , hence temperature, and the directed flow of free electrons from the Doppler shifts experienced by scattered probe photons. For $\alpha > 1$, the collective scattering regime, the scattering is sensitive to temporal correlations between electron motion separated by more than a Debye length, hence the scattering is dominated by ion-acoustic and electron plasma wave resonances, the latter set by the Bohm-Gross dispersion relation.³ The frequency shift of the resonance is dependent on density through the plasma frequency while the width of the resonances yields information on the wave damping rates. In the intermediate regime, *i.e.*, near $\alpha = 1$, the form of the high frequency electron plasma component depends strongly on both the electron temperature and density, providing a robust internal measurement of these basic plasma parameters, confirmed by spectroscopy. Clearly Eq. 4 will break down as the concept of Debye shielding becomes invalid as the coupling becomes large, thus necessitating the use of the screening model; however, the definition of the boundary between the weakly and strongly coupled regimes can be examined by using this formula.

A highly schematic of the expected generic scattered spectrum features is shown in Fig. 5. Coherent scattering from tightly bound electrons (Z_{tb} per atom) will provide an unshifted peak at the probe wavelength whose intensity is proportional to Z_{tb}^2 . Incoherent Compton scattering from weakly bound electrons (Z_{wb} per atom) should provide a second peak downshifted in energy by the order of $h\nu/mc^2$, with an intensity proportional to Z_{wb} . Thomson scattering from free electrons (Z_f per atom) should provide a dispersed spectrum centered on the Compton peak, with a spectrally integrated intensity varying as Z_f . The form of the spectrum will in general depend on the free electron density, n_e , free electron and/or Fermi temperature T_{Fermi} and electron-ion collisionality ν_{ei} . Hence, by spectrally resolving the scattered x-rays we would gain access, for the first time, to an unparalleled source of information on warm dense matter.

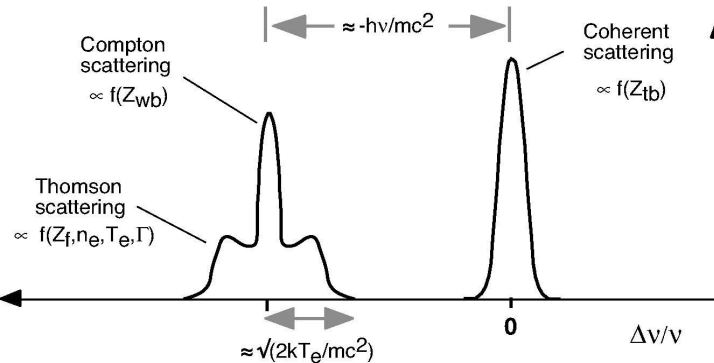


Fig. 5. Schematic of spectrally-resolved x-ray scattering spectrum expected, with information provided by each feature noted. The definition of weakly bound and tightly bound electrons depend on their binding energy relative to the Compton energy shift. Those with binding energies (ionization potentials) less than the Compton shift are categorized weakly bound.

For example, we should be able to infer Z_f , Z_{tb} , and Z_{wb} from the relative importance of coherent, incoherent and free electron scattering contributions. This would allow us to discriminate between different ionization balance models used to define the EOS of these plasmas.²⁵ We should be able to infer the free electron temperature and density (and hence ionization state since the ion density is effectively hydrodynamically frozen) from the shape of the Thomson scattered spectrum for $\alpha \approx 1$, hence shedding further light on the equilibrium states of warm dense matter. We should be able to infer the plasma collisionality from the shape of the free electron spectral peak for $\alpha > 1$, hence allowing us to test the validity of various strongly-coupled statistical physics models.

3.3.2 Interferometry has been demonstrated with long wavelength plasma x-ray lasers employing a wide range of techniques from Mach-Zehnder, Michelson, Lloyd's mirrors, and Fresnel bi-mirrors. The other major difference in using interferometers is the intrinsic requirement on beam coherence, either temporal or spatial. Michelson or Mach-Zehnder instruments do not require any coherence. However, for Lloyd's mirror or Fresnel bi-mirror the useful field of view is limited by the transverse coherence length. The full transverse coherence of the LCLS opens the possibility to use these two last techniques that are technically simpler. Further, these latter two interferometers work in grazing incidence

enabling one to use the same interferometer over a wide wavelength range. The short wavelength and short pulse length of the LCLS will enable interferometric studies of denser plasmas at higher time resolution than currently possible with plasma x-ray lasers.

3.3.3 *Radiography* will be developed as a complement to interferometry, which cannot give a fully two-dimensional image of the plasma. Indeed, for interferometry the spatial resolution is quite high along the fringes but is about 10 times smaller in the perpendicular direction. This might make the interferometry inappropriate for probing small-scale structures that would arise from hydrodynamic or laser induced parametric instabilities. X-ray laser radiography has been used for measurement of hydrodynamic instabilities growing in thin foils. With the short pulse LCLS this type of experiments will be easily reproduced and extended to high time resolution and denser and longer plasmas than has been previously possible.

4. DISCUSSION

It is interesting to make comparison of the next generation sources discussed above to other methods that can provide additional information in the areas discussed. First, we note that the plasma spectroscopic studies are not possible with the current sources. The use of lasers sparked a revolution in spectroscopy when it first appeared over three decades ago, but this left those interested in dense plasmas to wait for an x-ray laser that could probe these plasmas. As there seems no logical way around the problem that hot dense systems are short-lived, require x-ray probes, and the techniques of laser based spectroscopy will have to wait for x-ray lasers we feel that these experiment will be performed by the x-ray FELs of the future. Of course, we must remain aware that there have been major advances in the generation of laboratory x-ray lasers, e.g., see the work of Fajardo²⁶, which may efficiently reach the sub-100 Å regime in the future.

On the other hand, the generation of warm dense matter could potentially be performed with numerous sources. Thus, we can imagine that particle beams, high explosives, short pulse lasers, and high-energy lasers could be employed to create the warmed state. To start we point out the visible lasers have very short penetration depths as their critical density is well below solid densities so that only ultra-thin foils can be heated with a degree of uniformity. Experiments of this type will be important for technique development; however, due to the constraints on accuracy required to measure an equation-of-state, x-ray sources and particle beams will be necessary. The other possible method to generate x-rays, not discussed above is to use a large-scale high-energy laser to generate x-rays that in turn warm a sample. This will, of course, be pursued and indeed there are ongoing efforts to use the LLE Omega Laser at University of Rochester to develop these experiment.²⁷ We note that the eventual goal will be to study the WDM regime and develop x-ray Thomson scattering for use on the National Ignition Facility (NIF) currently being constructed at Lawrence Livermore National Laboratory. Given the nature of the NIF, a MJ laser with a high cost of operation, it would appear that the x-ray Thomson scattering will be extremely important, while the only WDM probing that will occur will be limited to those cases relevant to the generation of higher energy densities.

Further, there are several other possible facilities that can generate finite temperature dense samples where WDM experiments could be performed. These are gas guns, laser heated diamond anvil cells, and high explosives. Currently we are not aware of any experiments where either of the first two capabilities has created matter that is in the WDM regime, as this would require in the low-temperature solid-density limit obtaining temperatures in excess of the Fermi energy. There is one effort, of which we are aware, that uses high explosives to obtain temperatures in excess of 1 eV using aerogel foams. These experiment use two-sided planar converging shocks with a view to eventually seeding metals into the foams to gain access to the WDM regime.²⁸

Thus, we see that the next generation of light sources provides one of the better options for quantitative warm dense matter experiments, while the study and probing of the hot dense matter regime will be uniquely matched to these intense short pulse x-ray lasers.

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