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X-ray free-electron laser studies of dense plasmas

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The high peak brightness of X-ray free-electron lasers (FELs), coupled with X-ray optics enabling the focusing of pulses down to sub-micron spot sizes, provides an attractive route to generating high energy-density systems on femtosecond time scales, via the isochoric heating of solid samples. Once created, the fundamental properties of these plasmas can be studied with unprecedented accuracy and control, providing essential experimental data needed to test and benchmark commonly used theoretical models and assumptions in the study of matter in extreme conditions, as well as to develop new predictive capabilities. Current advances in isochoric heating and spectroscopic plasma studies on X-ray FELs are reviewed and future research directions and opportunities discussed.

1. Introduction

The study of high energy-density physics (HEDP) is of great importance for both fundamental and applied physics. Matter is loosely defined to be in this regime if it has an energy density exceeding about 10^{11} J m^{-3}, which corresponds to temperatures above a few eV at a typical density of a solid, or pressures above one Mbar. As illustrated in figure 1, such matter is widespread in the universe in a variety of forms, ranging from the interiors of giant planets, such as Saturn and Jupiter, to the constituent matter of all the different types of stars. Importantly, understanding the properties and the dynamics of matter in these extreme conditions, and its interaction with radiation, is essential for inertial confinement fusion investigations.

Within the realm of HEDP, a further practical distinction is often made between warm-dense matter (WDM), describing partially degenerate, dense systems at temperatures around the Fermi energy of a typical metal (5–10 eV), and hot-dense matter (HDM), where the temperatures are sufficiently high that there is no degeneracy in the free-electron distribution. What these systems have in common is a high density of charged particles, so that their Coulomb energies are non-negligible compared with their kinetic energies. This significantly complicates the theoretical description of such systems, as approaches based on kinetic theory become inadequate, many-body effects become important, and systems exhibit strong correlations between their constituent particles. In this sense, theoretical work on HEDP often lies on the boundary between condensed matter physics and classical plasma physics, borrowing techniques from both, but generalizable to neither. Further, the complexity of these systems, combined

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with the vast scale of interactions and processes that need to be accounted for, makes the development of simplified, approximate models necessary; their validation with experimental data is thus all the more essential.

To date, creating a homogeneous system, which is both very dense and has a high energy density, has proved to be a remarkable experimental challenge. While such states of matter are produced any time plasmas are created from solid samples, they are invariably highly transient, and also exhibit steep, juxtaposed gradients in density and temperature. As such, creating and isolating well-defined WDM and HDM states is an important experimental goal in its own right, in addition to the clear interest in the development of dedicated diagnostic techniques aimed at the study of their properties and dynamics. Importantly, the experimental difficulties in studying these systems impact theoretical developments, as little quantitative data is available to validate and guide calculations and modelling.

In this context, X-ray FELs are now starting to prove their potential as a tool to both create and study HED systems. On the one hand, X-rays, characterized by photon energies much larger than the plasma frequency of a typical dense system (10–20 eV), are capable of penetrating deep into samples, depositing energy and probing them volumetrically. On the other, the high peak brightness afforded by FELs, combined with sub-micron focusing X-ray optics, now enables the deposition of millijoules of energy into volumes of a few cubic microns, within about 100 fs, sufficient to generate high energy-density systems on inertial confinement time scales. In what follows I will discuss some of the first results of experiments where X-ray isochoric heating was used to create extreme states of matter, and the advances that this has led to in terms of our understanding of the properties of dense plasmas.

2. Isochoric heating on X-ray FELs

Using intense X-rays to deposit energy in free-standing foils is an attractive route to creating solid-density plasmas, both because of its simplicity in terms of experimental setup, and because the energy deposition of X-rays is volumetric, i.e. not limited only to a surface layer. Provided the thickness of the target foil is comparable to the absorption depth of the X-rays used, this deposition can also be made fairly homogeneous. For these reasons, this route towards the controlled generation of HED matter was seen as very promising and was proposed and extensively investigated theoretically over the past decade, forming a key component in the drive for HEDP to be performed on X-ray FEL facilities (Lee *et al.* 2002, 2003; Ng *et al.* 2005; Tschentscher & Toleikis 2005; Fisher *et al.* 2006; Rose 2009). With the commissioning and beginning of user operation of XUV and X-ray FELs worldwide, these techniques have now become experimentally feasible, and, as will be discussed in the following, are starting to fulfil their promise. Of course, this method is not limited to foils. For example, isochoric heating of cryogenic hydrogen droplets to WDM conditions has recently been demonstrated on the FLASH XUV FEL (Zastrau *et al.* 2014).

2.1. *Saturable absorption*

While the absorption of X-rays is volumetric in dense matter, in the linear absorption regime the X-ray intensity still decays exponentially within a thin foil following Beer's law. For this reason, targets thicker than about one absorption length are generally not used to create homogeneous HED systems, and even at a single absorption length, gradients of over a factor of two may form between the front and back surfaces

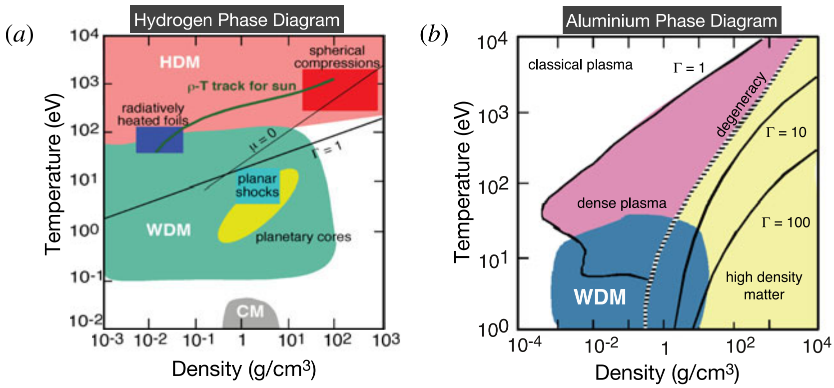

FIGURE 1. Phase diagrams of high energy-density systems. Temperature–density phase diagrams for hydrogen and aluminium plasmas, denoting the regions of warm- and hot-dense matter. Various values of the plasma coupling parameter $\Gamma = Z^2e^2/(r_0k_BT)$, denoting the ratio of the interatomic potential and thermal energies, are shown. Here $r_0 = (3Z/(4\pi n_e))^{1/3}$ is the inter-particle spacing, Z is the ion charge and n_e the electron density. Strong-coupling conditions are for $\Gamma > 1$. Also shown are the set of conditions for which $\mu = 0$. The chemical potential, μ, is a decreasing function of temperature, and zero denotes the lower bound of the region of electron degeneracy. Figures reproduced from Lee *et al.* (2003).

of samples. However, the X-ray intensities that can now be achieved at FELs are sufficiently high for non-linear absorption effects, such as saturable absorption, to take place. Besides being of interest to the study of fundamental atomic physics and plasma dynamics, saturable absorption is an important process to consider in the generation of HED systems because it can reduce the longitudinal gradients in energy density within samples. In fact, it is becoming increasingly clear that at the highest FEL intensities, which are also those often used to create HED samples, photo-excitation, photo-ionization and electron collisional ionization can compete with atomic recombination rates, such as fluorescence and Auger decay, saturating absorption channels. This effect is generally welcomed, as the dense plasmas created tend to be more homogeneous than if the X-ray deposition process were linear. The reason for this is that as a particular absorption channel starts saturating at the surface of the sample, more energy can be transmitted through and deposited more deeply, reducing or eliminating the overall exponentially decreasing energy deposition.

The first experimental proof of saturable absorption at FELs was observed in very thin Al samples at the XUV FEL FLASH in Hamburg (Nagler *et al.* 2009). Performed at a photon energy of 92 eV, the intense 15 fs pulse saturated the L-shell absorption channel, leading to a large increase in the transmitted signal, and an energy deposition gradient between the front and rear of the target foil that was deduced to be linear rather than exponential. Spectroscopy was used to deduce heating to a temperature of a few eV during the FEL pulse (Vinko *et al.* 2010), and at later times the sample was estimated to have thermalized to around 20 eV, sufficient to create a warm-dense Al plasma (Medvedev *et al.* 2011). Saturable absorption has since been observed in a range of systems in the XUV (Yoneda *et al.* 2009; Inubushi *et al.* 2010; Di Cicco *et al.* 2014).

Given that core hole lifetimes decrease with increasing binding energy, one might expect it to become increasingly difficult to observe saturable absorption when

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interacting with deeply bound states, such as those pumped by harder X-rays in mid- and higher-Z elements. In fact, this is not the case. As described by Rackstraw *et al.* (2015), at X-ray wavelengths these high-intensity experiments also deposit a large amount of energy in the samples, heating them to significant temperatures. If this temperature is sufficient to thermally ionize bound states, the charge state distribution of the system will naturally evolve over the duration of the pulse and will never return to the initial state, regardless of core hole lifetimes. Additionally, recombination processes filling deep core holes predominantly occur from other, shallower bound states, leading to the generation of new excited or further ionized ion configurations. If the higher charge states generated through these various processes have lower absorption cross-sections at a given X-ray wavelength, as is generally the case, the total absorption in the sample will tend to decrease with increasing X-ray fluence. It follows that rather than an exception, saturable absorption is a characteristic phenomenon of high-intensity FEL interactions with matter. Experimentally it has now been observed in multiple systems and over a range of different conditions, both at the Linac Coherent Light Source (LCLS) X-ray FEL in California (Hoener *et al.* 2010; Young *et al.* 2010; Rackstraw *et al.* 2015) and on the Japanese X-ray FEL SACLA (Yoneda *et al.* 2014).

2.2. *Generating warm-dense matter*

The idea that intense X-rays from an FEL source could produce HED states in well-defined conditions via isochoric heating of thin foils was initially proposed over a decade ago (Lee *et al.* 2002, 2003), and was recently put to the test in an experiment at the LCLS X-ray FEL by Lévy *et al.* (2015). Here, a 0.5 μm thick silver foil was irradiated and heated by X-rays at a photon energy of 8.9 keV. The target was designed to be much thinner than an absorption length at this wavelength (∼6 μm), so as to minimize energy deposition gradients between the front and the back of the foil. The X-ray pulse was approximately 60 fs long and was focused to a spot size yielding an intensity of about 5×10^{15} W cm^{-2}. The energy absorption mechanism of the X-ray pulse in the target was predominantly Ag L-shell photoionization, although some M- and N-shell photoionization also took place. These processes occurred during the ultra-short FEL pulse, creating core-ionized and excited electron configurations, which quickly equilibrated via X-ray fluorescence, and Auger and collisional processes, leading to a heating of the electron sub-system. The ions, in turn, remained cold during the X-ray irradiation, and were subsequently heated by the hot electrons to the equilibrium temperature on the time scales of electron–ion coupling, on the order of 10 ps (Cho *et al.* 2011; White *et al.* 2014). Because of the difference in equilibration time scales for electrons and ions, the X-ray heating mechanism was largely decoupled from any ion density gradient in the sample.

The peak temperatures reached in the experiment were deduced via hydrodynamic simulations to be of 10–15 eV while the sample remained at solid density. At later times, because of the high internal pressure, the sample expanded into the surrounding space and cooled, with its evolution dictated by its equation of state. This expansion was the main observable in the experiment, measured via time-and-space resolved interferometry, simultaneously on both the front and back surfaces of the WDM sample. The expansion was observed to be similar on both sides of the foil, broadly within the margin of experimental error and in agreement with hydrodynamic calculations. This is the first experimental confirmation of isochoric heating by direct examination of the sample expansion on the relevant, picosecond time scales, and has also quantified the temperature uniformity in the longitudinal direction to be within about 10 % (Lévy *et al.* 2015).

2.3. *Generating hot-dense matter*

The first demonstration of generating HDM with an X-ray FEL from thin foils was also recently performed on the LCLS, by irradiating a 1 μm thick Al foil (∼1 absorption length) at X-ray intensities up to 10^{17} W cm^{-2} (Vinko *et al.* 2012). In the experiment, the X-ray wavelength of the FEL was tuned around the various K-edges of increasingly ionized Al ions, in the range 1460–1800 eV (Cho *et al.* 2012; Vinko *et al.* 2012).

Provided the X-ray photon energy of the FEL is above the ionization threshold of the ions in the system, the dominant absorption mechanism in the sample is K-shell photoionization. Below threshold, only L-shell and free-free absorption can take place, with significantly smaller cross-sections. The various absorption channels transfer energy from the X-ray pulse to the target, heating it and ionizing it further, creating a range of ionic configurations with inner-shell vacancies. These unstable ionic configurations recombine in two ways: radiatively, where an electron fills the core hole and a photon is emitted, or non-radiatively, via Auger processes, which further ionize the ion by emitting electrons. In Al, Auger processes dominate, accounting for over 96 % of the recombination events (Bambynek *et al.* 1972). As the Auger electrons have very high collisional cross-sections, very few escape the sample, and most transfer their energy to the remaining electrons and ions in the system via collisional processes on ultra-short time scales, so that the energy deposited by the X-ray beam is efficiently retained in the newly generated plasma. Peak temperatures approaching 200 eV and electron densities exceeding 5×10^{23} cm^{-3} were reported, deduced from detailed atomic kinetics calculations, and consistent with the observed spectral emission (Vinko *et al.* 2012).

The plasma was diagnosed via X-ray spectroscopy of the emission generated from the radiative K-shell recombination process. Despite the relatively small X-ray fluorescence yield, the K_α emission (photons generated by the 2p–1s transition) for the FEL-driven sample was observed to be sufficiently strong to record spectra pumped by a single, sub-100 fs, X-ray pulse. This emission is driven by the FEL pulse, since the intense X-ray beam is the only significant generator of K-shell core holes, making the technique very different from normal X-ray (self-) emission spectroscopy (XES). In fact, this diagnostic has several similarities to X-ray absorption spectroscopy (XAS) measurements, since the narrow bandwidth of the X-ray pulse (∼0.4 %) will only be absorbed if a 1s electron can be either excited to some higher-lying bound state, or ionized. However, unlike XAS, which for a single photon energy yields only a transmitted intensity, here the absorption process gives rise to inner-shell radiative recombination in the plasma, providing an entire emission spectrum. The integrated photon yield of this spectrum is, of course, directly related to the absorbed energy fraction via the radiation transport function. This measurement is therefore unique, and by scanning the FEL X-ray photon energies across the various ion energy levels, the authors reported a measurement of the electronic structure of all the ions present in the dense plasma, as shown in figure 2. Therefore, this is an extremely information-rich experimental technique for dense plasma studies, and shows great potential for future investigations.

3. Ionization potential depression

The ionization potential depression (IPD) of ions in a plasma due to their interaction with the environment is a fundamental component of dense plasma studies, as the ionization thresholds are needed to specify all ionic binding energies within the

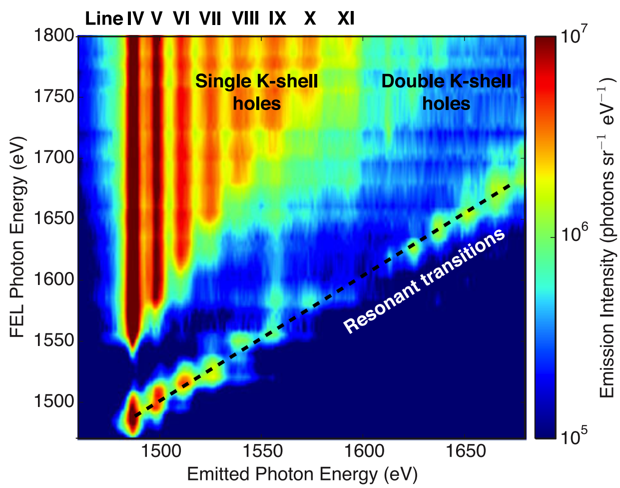

FIGURE 2. X-ray-driven emission spectroscopy. By pumping a hot-dense plasma with intense, narrow-bandwidth X-rays, the electronic structure of ions immersed in a dense plasma can be investigated on ultra-short time scales, via X-ray-driven K_α emission spectroscopy. The ions will only emit K_α photons if the X-ray pulse from the FEL can ionize or excite a 1s electron and create a K-shell hole. Therefore, emission will be observed for each ion only if the X-ray photon can drive a transition from the 1s level to some other available free state, allowing the states to be experimentally mapped in energy, as shown here for an Al plasma (Vinko *et al.* 2012). Bound–bound transitions can be observed as islands in the emission spectrum; here these correspond to the resonantly driven 1s–2p transitions (Cho *et al.* 2012). Photoionization corresponds to driving the 1s electrons into some state in the continuum. Such transitions will yield broadly flat emission intensities over an extended range of FEL wavelengths, observed experimentally at higher FEL photon energies. Data in figure taken from Cho *et al.* (2012) and Vinko *et al.* (2012).

system. This is essential for determining virtually every property of partially ionized plasmas, from the ionization balance and absorption cross-sections, to the plasma equation of state, the temperature and the emission spectrum. While it underlines so many plasma properties, extracting accurate IPDs from experimental observations in hot-dense plasmas prior to the advent of X-ray FELs had proved to be extremely challenging, in part because of difficulties in creating well-defined WDM and HDM conditions, and in part because of a lack of experimental techniques capable of tracking ionization thresholds with sufficient accuracy. The IPD is most significant in conditions of both high density and high ionization (temperature), so it is clearly crucial for the correct description of HDM systems, but it is also essential for WDM, even though the degree of ionization is much lower and the absolute IPD energies are small (Fletcher *et al.* 2013, 2014).

The ability to measure the electronic structure of ions in hot plasmas at a known ion density (and structure) has proved to be crucial for the investigation of the physics of continuum lowering in these extreme conditions. As can be seen from figure 2, alongside bound–bound resonant features, the experimental data from the LCLS campaign show clear emission thresholds as a function of FEL photon energy, with near-constant emission intensities above them. At least for the first several charge states, Ciricosta *et al.* (2012) identified these thresholds as the K-edges of Al ions

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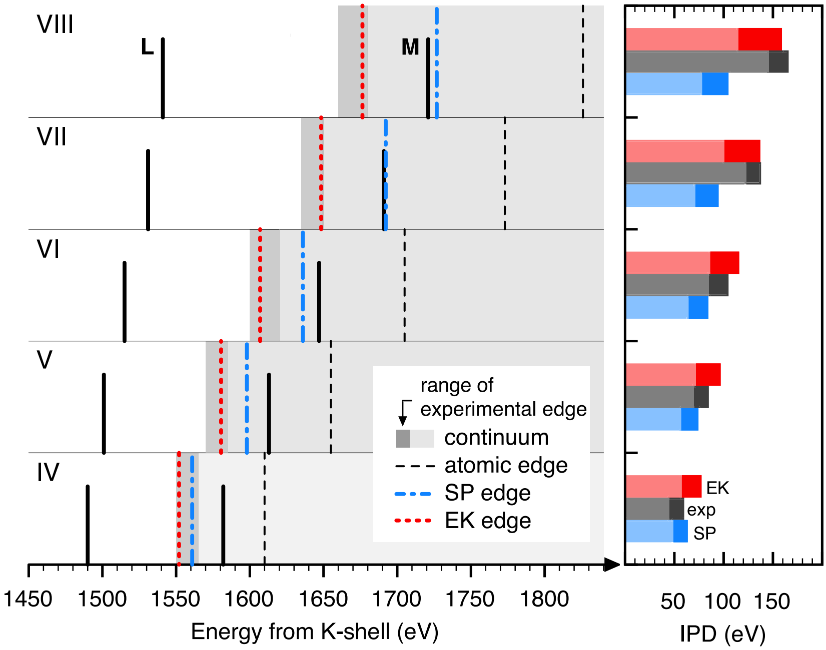

FIGURE 3. Experimental measurement of the ionization potential depression. Experimentally measured K-edges for Al charge states 3–7 (dark grey, width indicates the full extent of the K-edge) in a hot-dense plasma, compared with the K-edges for isolated Al ions (no plasma, dashed lines), and two IPD models: the Stewart–Pyatt model (blue) and a modified version of the Ecker–Kröll model (red). The energies of atomically bound L- and M-shell states with respect to the K-shell are also shown; the M-shell is seen to be pressure-ionized (binding energy is less than the IPD) for all charge states within the EK model, and up to charge states 6 or 7 in the SP model. Figure reproduced from Ciricosta *et al.* (2012).

embedded in the solid-density plasma, and could therefore extract the experimental IPDs in these conditions, for the first time.

The IPD measurements at the LCLS proved to be particularly interesting, not only because they were the first measurement of this kind but also because the results disagreed by a considerable margin with the predictions of the most commonly used models for dense plasmas, such as the Zimmerman & More (1980) ion sphere model, or the commonly used model of Stewart & Pyatt (1966) (SP). Instead, a modified version of a model put forward by Ecker & Kröll (1963) (EK) (where the leading constant in the expression was replaced by unity: see Ciricosta *et al.* 2012, Preston *et al.* 2013), was found to reproduce both the experimental edges, and the full range of emission spectra shown in figure 2. A comparison between the experimental edges and the prediction of the models is shown in figure 3, reproduced from Ciricosta *et al.* (2012). The experimental edges, indicated in dark grey in the figure, correspond to the observed emission thresholds, which are much lower than those predicted by the SP model for all charge states, indicating that larger IPDs are needed to explain the experimental results.

Recently, experiments at the Orion laser at AWE in the United Kingdom have also attempted measurements from which the IPD could be extracted, on small Al dots

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buried in plastic or diamond (Hoarty *et al.* 2013*a*,*b*). These dots were compressed to several times solid density and heated to temperatures up to 700 eV by both short- and long-pulse optical laser beams. The plasma conditions produced by this method at the Orion laser were more weakly coupled compared with those generated at the LCLS via X-ray isochoric heating, but the result is worth mentioning in this context because the IPDs deduced from this experiment were seen to lie about halfway between the predictions of the EK and SP models, agreeing with neither within the quoted experimental error.

In light of the failure of the popular SP model to reproduce the measured results in the conditions of either experiment, and the somewhat enigmatic success of the EK model in reproducing the LCLS data, a renewed interest in the topic from the theoretical point of view has emerged, highlighting the need for a much better treatment of density effects in strongly coupled plasmas. Crowley (2014) has argued that at least two different IPDs must be used in considering a plasma, a thermodynamic IPD to describe equilibrium phenomena (such as the equation of state), and a non-equilibrium, spectroscopic IPD to describe processes such as photoionization, and, as a consequence, the experimental results from the LCLS described above. This approach yields an improvement over the predictions of the Stewart–Pyatt model when applied to the conditions of both the LCLS and the Orion experiments. Son *et al.* (2014) have developed a two-step Hartree–Fock–Slater model, which they claim also yields a better agreement with both the Orion and LCLS results. Vinko, Ciricosta & Wark (2014) have conducted a finite-temperature density functional theory (DFT) study of the electronic structure of ions in a dense plasma with a view to investigating the effect of continuum lowering. In this work, the electronic structure and the K-edges for the various charge states were seen to agree very well with the LCLS experiments; however, the authors were unable to reach the conditions of the Orion experiments due to the difficulty in using DFT methods at high temperatures. The results from these models are shown alongside the LCLS measurements in figure 4. Most recently, Calisti, Ferri & Talin (2015) have presented an IPD model based on classical molecular dynamics. While they have yet to perform calculations in the conditions of the experiments described here, their initial results indicate IPD values in between those of SP and EK.

The disparity of IPD calculations points to several important deficiencies in the way dense plasmas are currently modelled. In particular, a clear cut-off in the energy of the bound state partition function is, in general, too crude an approximation to model the depression of the ionization potential due to high density. Worse still, it entirely ignores the fundamental mechanism by which the continuum forms in dense systems at energies which would otherwise correspond to bound states in isolated ions: the overlapping of valence wave functions giving rise to a non-zero probability of finding an electron at some later time far from the ion from which it originated. This is, in the zero-temperature limit, simply the tight-binding theory of band formation, a continuous process, which complicates the definition of a free or bound electron, and of ionization in a dense plasma in general. Dharma-wardana & Perrot (1992) were the first to discuss some of these effects, and their importance in dense plasmas, via the neutral-pseudoatom model with an ion correlation sphere. Today, modern spherically symmetric, average-atom models have the ability to address many of the complications arising from density effects in dense, partially ionized plasmas, but several difficulties remain in the systematic treatment of the transition between fully bound and free states, and in the determination of a meaningful mean plasma ionization (Murillo *et al.* 2013). Recently, Vinko *et al.* (2014) performed fully three-dimensional DFT

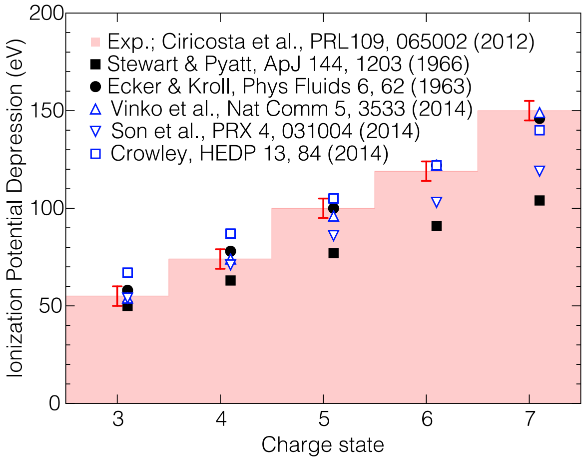

FIGURE 4. Comparison of IPD models. The IPD models of Stewart & Pyatt (1966), Ecker & Kröll (1963), Crowley (2014), Son *et al.* (2014) and Vinko *et al.* (2014) are compared with the experimental results by Ciricosta *et al.* (2012) obtained on the LCLS. Good agreement between all the models and the data within the experimental uncertainty can only really be found for the ground state (Al 3^{+}) even for the relatively simple Al plasma, exemplifying the current difficulties in the theoretical treatment of density effects in strongly coupled plasmas.

calculations of the electronic structure in a partially ionized dense plasma, accounting for a realistic ion and charge distribution, and treating bound and free valence electrons equally. As shown in figure 5, the calculations show good agreement with the experimental measurements from the LCLS campaign, and the authors claim to match all measured edges to within the experimental error. However, the calculations remain limited to relatively small, simple systems, and the computational cost of extending them to a wider range of plasma conditions remains prohibitive.

While improvements in modelling the effect of high density on atomic states are certainly required, it seems unlikely that the current challenges will be solved by theoretical work alone, and further experimental effort, both on X-ray FELs and other facilities, will be crucial in guiding computational and theoretical investigations.

4. Outlook

Further experimental results on the ionization potential depression in Si, Mg, and their compounds, have been reported recently by Ciricosta (2014) and are expected to be published soon. While the interpretation of the experimental results is still being debated (e.g. see Iglesias & Sterne 2013; Iglesias 2014), as are the methods used to model these effects in codes dedicated to simulating dense plasmas, there now exists for the first time a clear set of comprehensive experimental data from a well-defined hot-dense plasma, with which codes and models can be compared and benchmarked. In this context, further experimental data using X-ray FELs, comprising a more extended set of materials and temperature–density conditions, will undoubtedly help further our predictive and modelling capabilities for this key plasma property.

Few experimental investigations dedicated to generating high-Z hot-dense plasmas have been conducted so far on X-ray FELs, and initial results indicate that much higher intensities may be needed to drive non-linear processes similar to those

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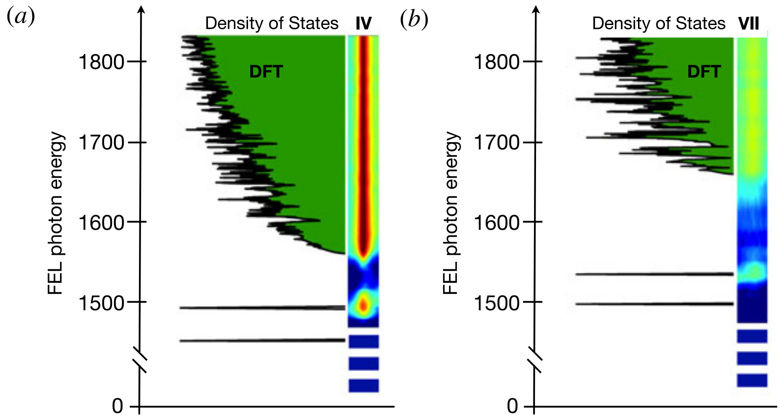

FIGURE 5. Electronic structure of ions in a dense plasma. Density functional theory calculations of core-excited Al configurations in plasmas at finite temperatures (Vinko *et al.* 2014) can provide a theoretical basis for investigation of the electronic structure of ions embedded in a dense, strongly coupled plasma with minimal assumptions. The calculated density of states (left) providing the structure and position of the continuum is seen to agree well with charge-state-resolved measurements (right) recently performed at the LCLS (Vinko *et al.* 2012), shown here for lines IV in (*a*) and line VII in (*b*), corresponding to charge states 3^+ and 6^+, respectively. The sharp lines at lower energies in the density of states correspond to the 2s and 2p orbitals in ion charge states 4^+ and 7^+, respectively.

observed on lower-*Z* elements such as Al (Yoneda *et al.* 2014). Such high-*Z* systems, while very important from a physical point of view, may also be harder to make and study using the spectroscopic techniques presented above, for several reasons. Firstly, atoms with higher atomic number contain many more electrons, and the core states are more strongly bound, so that more energy is required for significant ionization. There are also many more weakly bound electrons, for which very different charge states can produce similar and overlapping emission spectra. Further, inner shell (K or L) spectroscopic techniques require much higher photon energies to generate core holes in high-*Z* elements, a process which generates higher-energy Auger and photoemission electrons that are in turn less likely to thermalize within the small plasma volume. Isochoric heating via core-state photo-absorption with increasingly harder X-rays is also less efficient because of higher energy losses due to radiative recombination (the probability of radiative recombination between two energy levels grows with their energy separation). Some of these issues may be mitigated by using higher drive X-ray intensities. The peak intensities generated so far on X-ray FELs are in the region of 10^{20} W cm^{-2}, achieved by focusing to spot sizes a few tens of nanometres across (Mimura *et al.* 2014). Although these intensities are very promising for a range of (non-linear) studies, at some point electron losses during the collisional thermalization process, due to hot electrons escaping the small plasma, may become important, placing a limit on their usefulness for creating homogeneous HDM states.

It is important to note that isochoric heating experiments performed with a single X-ray FEL pulse are limited to the study of ion densities of materials in their ground state. Future work involving X-ray pump–probe configurations, or coupling X-ray FELs with intense optical lasers, will be needed to investigate plasma systems at different densities and at temperatures beyond those currently reachable on FEL facilities by X-ray heating alone. These are perhaps the most promising future

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developments, and should significantly advance our capabilities for studying the evolution and dynamics of dense plasma systems.

5. Conclusions

The isochoric heating of foils to warm- and hot-dense matter conditions with FEL radiation is now an established technique capable of creating extreme and exotic states of matter in well-defined conditions of temperature and density on inertial confinement time scales. Warm-dense matter states in Ag plasmas at solid density and temperatures of 10–15 eV have now been demonstrated with a thermal gradient of as little as 10 % (Lévy *et al.* 2015), and homogeneous hot-dense plasmas at temperatures approaching 200 eV have been observed in micron-sized Al samples (Vinko *et al.* 2012). These apparently very different conditions have been created by the same isochoric heating process, the only variation being the wavelength and focusing of the FEL beam, illustrating how a vast range of parameter space can now be accessed to investigate specific questions regarding the behaviour of dense plasmas in strong-coupling conditions. Encouragingly, the high X-ray intensities required to generate the highest energy-density systems invariably drive non-linear processes in the samples under investigation. Such processes, as was shown in the case of saturable absorption, can be very beneficial to/for the homogeneity of the sample in isochoric heating, but a detailed understanding of the interaction dynamics of the intense X-ray pulse with the sample is required to exploit this capability in full.

The first investigations of plasma properties of mid-Z elements have been performed on the LCLS using novel spectroscopic techniques, and have yielded the first measurements of continuum lowering, resolved by charge state, in a 100–200 eV solid-density Al plasma (Ciricosta *et al.* 2012). These measurements were made possible by the bright, narrow-bandwidth X-ray pulse, which allowed for the determination of the K-edges in a range of Al charge states to within a few eV, providing by far the most stringent constraint to date on IPD models in HED systems. Perhaps unsurprisingly, the measurements have revealed the extent to which many currently used models fail to describe ionization energies in a dense plasma appropriately – a critical property for any system – with repercussions for its equation of state (Fletcher *et al.* 2014), transition rates and collisional ionization rates (Vinko *et al.* 2015). It should be noted that on the theoretical side, the increased effort in modelling continuum lowering has not, so far, led to a unique picture of the ionization dynamics in the challenging WDM and HDM regimes, with a plethora of available models yielding significantly different predictions (see figure 4). In this context, a strong need is emerging for modelling capabilities which can bridge between the accurate but expensive multi-centred Kohn–Sham DFT calculations, and the much more efficient self-consistent average-atom models, combining some level of accuracy and completeness of the former with the speed and scalability of the latter.

Current and future experimental platforms at X-ray FELs, in conjunction with computational and theoretical advances, present a truly unprecedented opportunity to further our understanding of matter in extreme conditions at the microscopic level. By providing insight into its elementary interactions, structure and dynamics on the relevant spatial and temporal scales, these investigations afford the potential to address outstanding questions in a range of fields from fundamental plasma physics and astrophysics to inertial confinement fusion research.

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REFERENCES

BAMBYNEK, W., CRASEMANN, B., FINK, R. W., FREUND, H. U., MARK, H., SWIFT, C. D., PRICE, R. E. & RAO, P. V. 1972 X-ray fluorescence yields, Auger, and Coster–Kronig transition probabilities. *Rev. Mod. Phys.* **44** (4), 716–813.

CALISTI, A., FERRI, S. & TALIN, B. 2015 Ionization potential depression in hot dense plasmas through a pure classical model. *Contrib. Plasma Phys.* **55** (5), 360–365.

CHO, B., ENGELHORN, K., CORREA, A., OGITSU, T., WEBER, C., LEE, H., FENG, J., NI, P., PING, Y., NELSON, A., PRENDERGAST, D., LEE, R., FALCONE, R. & HEIMANN, P. 2011 Electronic structure of warm dense copper studied by ultrafast x-ray absorption spectroscopy. *Phys. Rev. Lett.* **106**, 167601.

CHO, B. I., ENGELHORN, K., VINKO, S. M., CHUNG, H.-K., CIRICOSTA, O., RACKSTRAW, D. S., FALCONE, R. W., BROWN, C. R. D., BURIAN, T., CHALUPSKÝ, J., GRAVES, C., HÁJKOVÁ, V., HIGGINBOTHAM, A., JUHA, L., KRZYWINSKI, J., LEE, H. J., MESSERSMIDT, M., MURPHY, C., PING, Y., ROHRINGER, N., SCHERZ, A., SCHLOTTER, W., TOLEIKIS, S., TURNER, J. J., VYSIN, L., WANG, T., WU, B., ZASTRAU, U., ZHU, D., LEE, R. W., NAGLER, B., WARK, J. S. & HEIMANN, P. A. 2012 Resonant $K\alpha$ spectroscopy of solid-density aluminum plasmas. *Phys. Rev. Lett.* **109** (24), 245003.

CIRICOSTA, O. 2014 Radiative properties of hot dense matter 2014. *Density Independent Continuum Lowering in Hot Dense Matter*, In https://sites.google.com/site/rphdm14/schedule-abstracts.

CIRICOSTA, O., VINKO, S. M., CHUNG, H.-K., CHO, B.-I., BROWN, C. R. D., BURIAN, T., CHALUPSKÝ, J., ENGELHORN, K., FALCONE, R. W., GRAVES, C., HÁJKOVÁ, V., HIGGINBOTHAM, A., JUHA, L., KRZYWINSKI, J., LEE, H. J., MESSERSCHMIDT, M., MURPHY, C. D., PING, Y., RACKSTRAW, D. S., SCHERZ, A., SCHLOTTER, W., TOLEIKIS, S., TURNER, J. J., VYSIN, L., WANG, T., WU, B., ZASTRAU, U., ZHU, D., LEE, R. W., HEIMANN, P., NAGLER, B. & WARK, J. S. 2012 Direct measurements of the ionization potential depression in a dense plasma. *Phys. Rev. Lett.* **109**, 065002.

CROWLEY, B. J. B. 2014 Continuum lowering: a new perspective. *High Energ. Dens. Phys.* **13**, 84–102.

DHARMA-WARDANA, M. W. C. & PERROT, F. 1992 Level shifts, continuum lowering, and the mobility edge in dense plasmas. *Phys. Rev.* A **45** (8), 5883–5896.

DI CICCO, A., HATADA, K., GIANGRISOSTOMI, E., GUNNELLA, R., BENCIVENGA, F., PRINCIPI, E., MASCIOVECCHIO, C. & FILIPPONI, A. 2014 Interplay of electron heating and saturable absorption in ultrafast extreme ultraviolet transmission of condensed matter. *Phys. Rev.* B **90**, 220303.

ECKER, G. & KRÖLL, W. 1963 Lowering of the ionization energy for a plasma in thermodynamic equilibrium. *Phys. Fluids* **6** (1), 62–69.

FISHER, D. V., HENIS, Z., ELIEZER, S. & MEYER-TER-VEHN, J. 2006 Core holes, charge disorder, and transition from metallic to plasma properties in ultrashort pulse irradiation of metals. *Laser Part. Beams* **24**, 81–94.

FLETCHER, L. B., KRITCHER, A., PAK, A., MA, T., DÖPPNER, T., FORTMANN, C., DIVOL, L., LANDEN, O. L., VORBERGER, J., CHAPMAN, D. A., GERICKE, D. O., FALCONE, R. W. & GLENZER, S. H. 2013 X-ray Thomson scattering measurements of temperature and density from multi-shocked CH capsules. *Phys. Plasmas* **20** (5), 056316.

FLETCHER, L. B., KRITCHER, A. L., PAK, A., MA, T., DÖPPNER, T., FORTMANN, C., DIVOL, L., JONES, O. S., LANDEN, O. L., SCOTT, H. A., VORBERGER, J., CHAPMAN, D. A., GERICKE, D. O., MATTERN, B. A., SEIDLER, G. T., GREGORI, G., FALCONE, R. W. & GLENZER, S. H. 2014 Observations of continuum depression in warm dense matter with x-ray Thomson scattering. *Phys. Rev. Lett.* **112**, 145004.

HOARTY, D. J., ALLAN, P., JAMES, S. F., BROWN, C. R. D., HOBBS, L. M. R., HILL, M. P., HARRIS, J. W. O., MORTON, J., BROOKES, M. G., SHEPHERD, R., DUNN, J., CHEN, H., VON MARLEY, E., BEIERSDORFER, P., CHUNG, H. K., LEE, R. W., BROWN, G. & EMIG, J. 2013*a* The first data from the Orion laser; measurements of the spectrum of hot, dense aluminium. *High Energ. Dens. Phys.* **9** (4), 661–671.

HOARTY, D. J., ALLAN, P., JAMES, S. F., BROWN, C. R. D., HOBBS, L. M. R., HILL, M. P., HARRIS, J. W. O., MORTON, J., BROOKES, M. G., SHEPHERD, R., DUNN, J., CHEN, H., VON MARLEY, E., BEIERSDORFER, P., CHUNG, H. K., LEE, R. W., BROWN, G. & EMIG, J. 2013*b* Observations of the effect of ionization-potential depression in hot dense plasma. *Phys. Rev. Lett.* **110** (26), 265003.

HOENER, M., FANG, L., KORNILOV, O., GESSNER, O., PRATT, S., GÜHR, M., KANTER, E., BLAGA, C., BOSTEDT, C., BOZEK, J., BUCKSBAUM, P., BUTH, C., CHEN, M., COFFEE, R., CRYAN, J., DIMAURO, L., GLOWNIA, M., HOSLER, E., KUKK, E., LEONE, S., MCFARLAND, B., MESSERSCHMIDT, M., MURPHY, B., PETROVIC, V., ROLLES, D. & BERRAH, N. 2010 Ultraintense x-ray induced ionization, dissociation, and frustrated absorption in molecular nitrogen. *Phys. Rev. Lett.* **104**, 253002.

IGLESIAS, C. A. 2014 A plea for a reexamination of ionization potential depression measurements. *High Energ. Dens. Phys.* **12**, 5–11.

IGLESIAS, C. A. & STERNE, P. A. 2013 Fluctuations and the ionization potential in dense plasmas. *High Energ. Dens. Phys.* **9** (1), 103–107.

INUBUSHI, Y., YONEDA, H., HIGASHIYA, A., ISHIKAWA, T., KIMURA, H., KUMAGAI, T., MORIMOTO, S., NAGASONO, M., OHASHI, H., SATO, F., TANAKA, T., TOGASHI, T., TONO, K., YABASHI, M., YAMAGUCHI, Y. & KODAMA, R. 2010 Measurement of saturable absorption by intense vacuum ultraviolet free electron laser using fluorescent material. *Rev. Sci. Instrum.* **81**, 036101.

LEE, R. W., BALDIS, H. A., CAUBLE, R. C., LANDEN, O. L., WARK, J. S., NG, A., ROSE, S. J., LEWIS, C., RILEY, D., GAUTHIER, J.-C. & AUDEBERT, P. 2002 Plasma-based studies with intense X-ray and particle beam sources. *Laser Part. Beams* **20**, 527–536.

LEE, R. W., MOON, S. J., CHUNG, H.-K., ROZMUS, W., BALDIS, H. A., GREGORI, G., CAUBLE, R. C., LANDEN, O. L., WARK, J. S., NG, A., ROSE, S. J., LEWIS, C. L., RILEY, D., GAUTHIER, J.-C. & AUDEBERT, P. 2003 Finite temperature dense matter studies on next-generation light sources. *J. Opt. Soc. Am.* B **20** (4), 770–778.

LÉVY, A., AUDEBERT, P., SHEPHERD, R., DUNN, J., CAMMARATA, M., CIRICOSTA, O., DENEUVILLE, F., DORCHIES, F., FAJARDO, M., FOURMENT, C., FRITZ, D., FUCHS, J., GAUDIN, J., GAUTHIER, M., GRAF, A., LEE, H. J., LEMKE, H., NAGLER, B., PARK, J., PEYRUSSE, O., STEEL, A. B., VINKO, S. M., WARK, J. S., WILLIAMS, G. O., ZHU, D. & LEE, R. W. 2015 The creation of large-volume, gradient-free warm dense matter with an x-ray free-electron laser. *Phys. Plasmas* **22** (3), 030703.

MEDVEDEV, N., ZASTRAU, U., FÖRSTER, E., GERICKE, D. & RETHFELD, B. 2011 Short-time electron dynamics in aluminum excited by femtosecond extreme ultraviolet radiation. *Phys. Rev. Lett.* **107** (16), 165003.

MIMURA, H., YUMOTO, H., MATSUYAMA, S., KOYAMA, T., TONO, K., INUBUSHI, Y., TOGASHI, T., SATO, T., KIM, J., FUKUI, R., SANO, Y., YABASHI, M., OHASHI, H., ISHIKAWA, T. & YAMAUCHI, K. 2014 Generation of 10^{20} W cm^{-2} hard X-ray laser pulses with two-stage reflective focusing system. *Nat. Commun.* **5**, 3539.

MURILLO, M. S., WEISHEIT, J., HANSEN, S. B. & DHARMA-WARDANA, M. W. C. 2013 Partial ionization in dense plasmas: comparisons among average-atom density functional models. *Phys. Rev.* E **87** (6), 063113.

NAGLER, B., ZASTRAU, U., FÄUSTLIN, R. R., VINKO, S. M., WHITCHER, T., NELSON, A. J., SOBIERAJSKI, R., KRZYWINSKI, J., CHALUPSKY, J., ABREU, E., BAJT, S., BORNATH, T., BURIAN, T., CHAPMAN, H., CIHELKA, J., DÖPPNER, T., DÜSTERER, S., DZELZAINIS, T., FAJARDO, M., FÖRSTER, E., FORTMANN, C., GALTIER, E., GLENZER, S. H., GÖDE, S., GREGORI, G., HAJKOVA, V., HEIMANN, P., JUHA, L., JUREK, M., KHATTAK, F. Y., KHORSAND, A. R., KLINGER, D., KOZLOVA, M., LAARMANN, T., LEE, H. J., LEE, R. W.,

https://doi.org/10.1017/S0022377815000902 Published online by Cambridge University Press

MEIWES-BROER, K.-H., MERCERE, P., MURPHY, W. J., PRZYSTAWIK, A., REDMER, R., REINHOLZ, H., RILEY, D., RÖPKE, G., ROSMEJ, F., SAKSL, K., SCHOTT, R., THIELE, R., TIGGESBÄUMKER, J., TOLEIKIS, S., TSCHENTSCHER, T., USCHMANN, I., VOLLMER, H. J. & WARK, J. S. 2009 Turning solid aluminium transparent by intense soft X-ray photoionization. *Nat. Phys.* **5** (9), 693–696.

NG, A., AO, T., PERROT, F., DHARMA-WARDANA, M. W. C. & FOORD, M. E. 2005 Idealized slab plasma approach for the study of warm dense matter. *Laser Part. Beams* **23**, 527–537.

PRESTON, T. R, VINKO, S. M, CIRICOSTA, O., CHUNG, H.-K., LEE, R. W. & WARK, J. S. 2013 The effects of ionization potential depression on the spectra emitted by hot dense aluminium plasmas. *High Energ. Dens. Phys.* **9**, 258–263.

RACKSTRAW, D. S., CIRICOSTA, O., VINKO, S. M., BARBREL, B., BURIAN, T., CHALUPSKÝ, J., CHO, B. I., CHUNG, H.-K., DAKOVSKI, G. L., ENGELHORN, K., HÁJKOVÁ, V., HEIMANN, P., HOLMES, M., JUHA, L., KRZYWINSKI, J., LEE, R. W., TOLEIKIS, S., TURNER, J. J., ZASTRAU, U. & WARK, J. S. 2015 Saturable absorption of an x-ray free-electron-laser heated solid-density aluminum plasma. *Phys. Rev. Lett.* **114**, 015003.

ROSE, S. J. 2009 The effect of a radiation field on excitation and ionisation in non-LTE high energy density plasmas. *High Energ Dens. Phys.* **5** (1–2), 23–26.

SON, S.-K., THIELE, R., JUREK, Z., ZIAJA, B. & SANTRA, R. 2014 Quantum-mechanical calculation of ionization-potential lowering in dense plasmas. *Phys. Rev.* X **4**, 031004.

STEWART, J. C. & PYATT, K. D. JR. 1966 Lowering of ionization potentials in plasmas. *Astrophys. J.* **144**, 1203–1211.

TSCHENTSCHER, T. & TOLEIKIS, S. 2005 Investigations of ultrafast phenomena in high-energy density physics using X-ray FEL radiation. *Eur. Phys. J.* D **36**, 193–197.

VINKO, S. M., CIRICOSTA, O., CHO, B. I., ENGELHORN, K., CHUNG, H.-K., BROWN, C. R. D., BURIAN, T., CHALUPSKÝ, J., FALCONE, R. W., GRAVES, C., HÁJKOVÁ, V., HIGGINBOTHAM, A., JUHA, L., KRZYWINSKI, J., LEE, H. J., MESSERSCHMIDT, M., MURPHY, C. D., PING, Y., SCHERZ, A., SCHLOTTER, W., TOLEIKIS, S., TURNER, J. J., VYSIN, L., WANG, T., WU, B., ZASTRAU, U., ZHU, D., LEE, R. W., HEIMANN, P. A., NAGLER, B. & WARK, J. S. 2012 Creation and diagnosis of a solid-density plasma with an X-ray free-electron laser. *Nature* **482** (7383), 59–62.

VINKO, S. M., CIRICOSTA, O., PRESTON, T. R., RACKSTRAW, D. S., BROWN, C. R. D., BURIAN, T., CHALUPSKÝ, J., CHO, B. I., CHUNG, H. K., ENGELHORN, K., FALCONE, R. W., FIOKOVININI, R., HÁJKOVÁ, V., HEIMANN, P. A., JUHA, L., LEE, H. J., LEE, R. W., MESSERSCHMIDT, M., NAGLER, B., SCHLOTTER, W., TURNER, J. J., VYSIN, L., ZASTRAU, U. & WARK, J. S. 2015 Investigation of femtosecond collisional ionization rates in a solid-density aluminium plasma. *Nat. Commun.* **6**, 6397.

VINKO, S. M., CIRICOSTA, O. & WARK, J. S. 2014 Density functional theory calculations of continuum lowering in strongly coupled plasmas. *Nat. Commun.* **5**, 3533.

VINKO, S. M., ZASTRAU, U., MAZEVET, S., ANDREASSON, J., BAJT, S., BURIAN, T., CHALUPSKY, J., CHAPMAN, H. N., CIHELKA, J., DORIA, D., DÖPPNER, T., DÜSTERER, S., DZELZAINIS, T., FÄUSTLIN, R. R., FORTMANN, C., FÖRSTER, E., GALTIER, E., GLENZER, S. H., GÖDE, S., GREGORI, G., HAJDU, J., HAJKOVA, V., HEIMANN, P. A., IRSIG, R., JUHA, L., JUREK, M., KRZYWINSKI, J., LAARMANN, T., LEE, H. J., LEE, R. W., LI, B., MEIWES-BROER, K.-H., MITHEN, J. P., NAGLER, B., NELSON, A. J., PRZYSTAWIK, A., REDMER, R., RILEY, D., ROSMEJ, F., SOBIERAJSKI, R., TAVELLA, F., THIELE, R., TIGGESBÄUMKER, J., TOLEIKIS, S., TSCHENTSCHER, T., VYSIN, L., WHITCHER, T. J., WHITE, S. & WARK, J. S. 2010 Electronic structure of an XUV photogenerated solid-density aluminum plasma. *Phys. Rev. Lett.* **104**, 225001.

WHITE, T. G., MABEY, P., GERICKE, D. O., HARTLEY, N. J., DOYLE, H. W., MCGONEGLE, D., RACKSTRAW, D. S., HIGGINBOTHAM, A. & GREGORI, G. 2014 Electron–phonon equilibration in laser-heated gold films. *Phys. Rev.* B **90**, 014305.

YONEDA, H., INUBUSHI, Y., SATO, F., MORIMOTO, S., KUMAGAYA, T., NAGASONO, M., HIGASHIYA, A., YABASHI, M., ISHIKAWA, T., OHASHI, H., KIMURA, H., TOGASHI, T. & KODAMA, R. 2009 Observation of saturable absorption of Sn metal film with intense EUV laser pulse. In *Proc. SPIE 7501, International Conference on Ultrafast and Nonlinear Optics 2009*, 75010H.

YONEDA, H., INUBUSHI, Y., YABASHI, M., KATAYAMA, T., ISHIKAWA, T., OHASHI, H., YUMOTO, H., YAMAUCHI, K., MIMURA, H. & KITAMURA, H. 2014 Saturable absorption of intense hard X-rays in iron. *Nat. Commun.* **5**, 5080.

YOUNG, L., KANTER, E. P., KRÄSSIG, B., LI, Y., MARCH, A. M., PRATT, S. T., SANTRA, R., SOUTHWORTH, S. H., ROHRINGER, N., DIMAURO, L. F., DOUMY, G., ROEDIG, C. A., BERRAH, N., FANG, L., HOENER, M., BUCKSBAUM, P. H., CRYAN, J. P., GHIMIRE, S., GLOWNIA, J. M., REIS, D. A., BOZEK, J. D., BOSTEDT, C., MESSERSCHMIDT, M. & MESSERSCHMIDT, M. 2010 Femtosecond electronic response of atoms to ultra-intense X-rays. *Nature* **466** (7302), 56–61.

ZASTRAU, U., SPERLING, P., HARMAND, M., BECKER, A., BORNATH, T., BREDOW, R., DZIARZHYTSKI, S., FENNEL, T., FLETCHER, L. B., FÖRSTER, E., GÖDE, S., GREGORI, G., HILBERT, V., HOCHHAUS, D., HOLST, B., LAARMANN, T., LEE, H. J., MA, T., MITHEN, J. P., MITZNER, R., MURPHY, C. D., NAKATSUTSUMI, M., NEUMAYER, P., PRZYSTAWIK, A., ROLING, S., SCHULZ, M., SIEMER, B., SKRUSZEWICZ, S., TIGGESBÄUMKER, J., TOLEIKIS, S., TSCHENTSCHER, T., WHITE, T., WÖSTMANN, M., ZACHARIAS, H., DÖPPNER, T., GLENZER, S. H. & REDMER, R. 2014 Resolving ultrafast heating of dense cryogenic hydrogen. *Phys. Rev. Lett.* **112**, 105002.

ZIMMERMAN, G. B. & MORE, R. M. 1980 Pressure ionization in laser-fusion target simulation. *J. Quant. Spectrosc. Radiat. Transfer* **23**, 517–522.

Culham Prize Winner 2012

Sam Vinko received his Master's degree from the University of Rome Tor Vergata in 2007, and his doctorate from the University of Oxford in 2011 working with Professor J. Wark, investigating the interaction of intense XUV light with solid-density matter on the FLASH free-electron laser (FEL) in Hamburg. As part of a large international team he worked on producing record intensities in the XUV, and showed how these could be used to saturate atomic processes in metals on femtosecond time scales, leading to the creation of homogeneous dense plasmas. He was awarded the Culham thesis prize for this work in 2012. As a postdoctoral researcher at the University of Oxford he then worked extensively on the Linac Coherent Light Source FEL in Stanford, showing how intense X-rays could be used to create and study plasmas at temperatures and densities similar to those found halfway towards the centre of the Sun, with exquisite precision and control. Dr Vinko was awarded a Royal Society University Research Fellowship in 2014 and now leads a small research group at the University of Oxford focused on investigating matter in extreme conditions using X-ray FEL sources. He shares the 2015 John Dawson Award for Excellence in Plasma Physics Research, awarded by the American Physical Society for outstanding achievement in plasma physics.

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