Generation and probing of warm dense matter (Al) created by laser-accelerated proton beams

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Abstract. A study of isochoric heating of Al foil by laser-accelerated proton beam is presented. The proton source that induces the heating has been characterized in details. The heated sample conditions at different times during heating were inferred by coupling a time- and space- resolved interferometry diagnostic with a 1-dimensional hydrodynamic code (that includes proton stopping) that uses the previously characterized proton source as input. In this way, we have minimized the number of free parameters in the modeling of the heating. We have shown that heating was isochoric and almost uniform along the target thickness with a maximum temperature of ~ 18 eV. Finally, a X-ray Near Edge Absorption Spectroscopy (XANES) diagnostic has been used to study the structural modification of the warm dense aluminum sample. We have observed a progressive loss, within ~ 10 ps, of lattice ordering within solid density Al samples as the temperature rises from 300 K to > 10⁷ K.

1. Introduction
The possibility of producing, in a controlled way, matter at solid density (1-10 g/cm³) while maintaining it at high temperature (1-100 eV) has been for a long time a desired goal. At the boundary between condensed matter and plasma physics, the study of matter in the warm dense matter (WDM) [1] regime involves, nowadays, many laboratories worldwide. Indeed, the matter in this state is found a large array of physical environments: in astrophysical objects, in the transition from solid density matter to the plasma state found in numerous plasma generators and during the implosion of an inertial confinement fusion capsule. Accurate modeling of inertial plasmas requires physical data, equation of states and transport coefficients in this WDM regime where classical theories such as plasma kinetic theory and condensed matter theory fail as WDM is defined by ion-ion correlation parameter and Fermi degeneracy parameter both of the order of the unity. The lack of data in the WDM regime motivates the need for the design of experiments able to produce with high fidelity and very good characterization the warm dense matter sample in copious quantity.

Different methods and approaches have been used to produce matter in this regime: laser-driven shock heating [2], x-ray heating [3, 4], direct heating with subpicosecond laser pulses [5]. These methods, though relatively fast (10⁻⁶-10¹¹ s) either produce heating that is longer than the typical hydrodynamic expansion time scale (10⁻¹² – 10⁻¹⁷ s) or result in highly nonuniform heating through the sample.

Recent progress in laser matter interaction makes it now possible to produce high proton currents with protons energy spanning spectrum from few hundreds of electronvolt up to several tens of MeV [6]. These proton beams present a promising source for volumetric heating. What makes them good
candidate for this is their (protons’) longer penetration depth in matter in comparison to x-rays or laser light together with the fact that they deposit most of their energy at the end of the range in the material (so called Bragg peak). Also, being produced in a very short time (few ps), protons can heat the sample before it expands. The isochoric heating by laser-accelerated proton beam been demonstrated by P. Patel [7].

In this paper we present the results of a study of isochoric heating of Al foil by laser-accelerated proton beams. By coupling the experimental measurements of heated target expansion and self-consistent simulations of the same experiment, we have inferred the heated target conditions at different times during heating. In order to achieve this, we have characterized experimentally the proton source that induces the heating with the emphasis on the low energy part of the proton spectrum (< 10% of the proton maximum energy, i.e. < 2 MeV in our case) [8] which plays a dominant part in the heating process (for target thickness of the order of µm) and which was not studied in detail in previous experiments conducted in similar conditions [9]. The experimental results have been compared with the results of 1-D hydrodynamic simulations of aluminium foil expansion that use the measured proton beam characteristics as an input for the energy deposition. In this way, we have eliminated free parameters in the modeling of the heating.

Once the induced heating was characterized, giving us an insight into the target conditions, we have studied the structural modification of the heated sample. We exploit the technique of ultrafast X-ray Absorption Near-Edge Spectroscopy (XANES) [10] which is related to the interpretation of the modulations observed above a photoabsorption edge. The XANES absorption spectrum is very sensitive to the local atomic arrangement and can provide direct insight into the structural properties of matter. X-ray photoabsorption was previously used in slow (ns) shock-compression experiments [11, 12] and calculations [13] but the main focus was on the position and the width of the K-edge as the density and temperature increased. However the full structure of the spectrum was not exploited to get structural information about the phase transition.

Experimentally, we observe the smoothing of the XANES structure of solid sample (aluminum) as it is isochorically heated from room temperature to several eVs. The observations are correlated to Quantum Molecular Dynamic (QMD) calculations as well as a simpler liquid description [14, 15, 16, 17], and good agreement, qualitative as well as quantitative, is found. Such comparison allows conclusively to relate the observed XANES spectral modifications to structural disordering of the heated material. We can thus, for the first time put an upper limit of ~10 ps on the loss of short range ordering during this solid to plasma transition.

2. Experimental set-up
The experiment was performed using the LULI 100 TW laser facility working in the chirped pulse amplification mode at a wavelength $\lambda_0=1.057$ µm. The experimental set-up is shown in figure 1(a). The main laser pulse had a duration of 320 fs and irradiated 10 µm thick Au flat foil (‘proton source’ target) at normal incidence with a peak intensity of $3 \times 10^{19}$ W/cm². This created a pulsed (few ps), highly directional, laminar proton beam containing about $10^{13}$ protons and having multi-MeV energies up to 20 MeV. This beam was incident on flat Al foils (‘heated’ target) of different thickness (ranging from 0.5 to 15 µm) which were placed 200 µm away from the proton source target. Protons coming from the ‘proton source’ target deposited energy within the ‘heated’ target. The energy deposition varies with incident proton energy and with the heated target thickness producing isochoric heating.

In order to characterize the proton source we used a magnetic spectrometer equipped with an image plate [18] (IP), which was absolutely calibrated as a proton detector [19], in its dispersion plane in order to obtain the continuous energy spectrum of the proton beam and stacks of radiochromic films [20] (RCF) to obtain the transverse characteristics of the beam [see figure 1(b)]. The IP was placed in the magnetic spectrometer in a way that enabled the measurement of the low energy part of the proton spectrum (down to $E_{\text{min}} \sim 0.13$ MeV), which plays a dominant role in the heating, as well of as the high energy one. Examples of measured proton spectra are shown in figure 2(a). Note that there is a shot to shot variation of the proton spectrum which can influence the heating. From the RCFs the data
regarding the proton beam’s divergence angle were obtained [figure 2(b)]. This represented the first characterization of the low-energy part of a high-energy laser-accelerated proton source [8].

![Experimental Set-up Diagram]

**Figure 1.** (a) Schematic of the experimental set-up, (b) Set-up for the proton source characterization.

![Proton Spectra Diagram]

**Figure 2.** (a) Measured proton spectra from three different shots obtained by irradiating a 10 µm thick Au flat foil with a main laser beam; (b) Proton beam divergence half-angle versus proton energy. Dots represent the measured data that were fitted with a function of a type (energy = x):

\[
\text{Halfangle}(x) = p_0 + p_1 x + p_2 x^2 + p_3 x^3 + p_4 x^4.
\]

The main diagnostic used to characterize the induced heating by the proton beam was time- and space- resolved interferometry [21] (TASRI) of a chirped probe beam (1.057 µm) of 50 ps duration, reflecting from the rear (nonirradiated) surface of the ‘heated’ target [8]. It allows us to measure the expansion velocity of the heated target critical density interface. For the targets that are heated, since the probe is reflected from the expanding rear surface, there is an induced phase shift compared to the case when the beam is reflected from an unperturbed surface. This shift is related to the induced heating in the target. Two other diagnostics were implemented in the experiment: time- and space-resolved self-emission measurement of the heated target in the visible range (450-600 nm) [22], and X-ray absorption spectroscopy.

3. Accurate description of warm dense Al

To assess the warm dense matter condition of the heated aluminum sample, we have adopted the methodology that is illustrated in figure 3: from the TASRI diagnostic, we retrieve information about the phase-shift of the probe beam reflecting from the rear surface of the heated target, while from the self-emission diagnostic, we retrieve the information about the emissivity that has been collected.
Then, we perform simulations using as input the proton spectra measured during the same experiment. At the output, we obtain heated target conditions as well as rear surface expansion velocity that can be compared to the measured ones. Also, using the calculated temperature, density and ionization degree we can calculate the emissivity that should be produced by this target. This computed emissivity is then compared with the measured emissivity.

Figure 3. Schematic of the methodology adopted to retrieve the heated target condition using both a simulation code and diagnostics.

We have performed simulations of the heating and expansion of a thin aluminium foil irradiated by a laminar high-energy proton source using a one-dimensional hydrodynamics code ESTHER [23]. This code solves, according to a Lagrangian scheme, the fluid equations for the conservation of mass, momentum and energy. The target material is described either by the Bushman-Lomonosov-Fortov (BLF) multiphase equation of state (EOS) [24] or the SESAME EOS [25]. We sample the experimentally measured proton spectra [see figure 2(a)] and use it as an input parameter in the simulation. For the proton stopping power, the values given by the SRIM database are used [26]. The simulation assumes electron-ion equilibrium, i.e. $T_e = T_i = T$. A laser probe diagnostic is implemented in the code in order to model the TASRI diagnostics. Thus, we obtain, as in the experiment, the phase after reflection off the heated target surface. For this post-processing of the hydrodynamic data the interaction between the laser field and the target is given by the solution of the Helmholtz wave equation. Details on the simulations parameters can be found in [8].

Figure 4 shows the result of the simulation of heating of a 10 µm thick Al target. Density normalized to the solid density and temperature profiles are presented for different simulation times (time ‘0’ corresponds to the moment when first, most energetic protons leave the proton source target). “Warm dense matter” conditions, i.e., degeneracy parameter equal to one half and ion-ion correlation parameter of the order of two, are obtained after 25 ps [8]. It can be seen that the bulk of the target stays at solid density while the density decreases at the edges due to expansion induced by heating. Also, in the early stage, one can notice a small shock in the density profile facing the incident proton beam that is due to energy deposition in a thin layer of the target by electrons co-moving with proton beam [27]. Note that these simulations are performed self-consistently using, where possible, measured parameters as inputs.

In order to assess the validity of these simulations, we have compared the measured probe beam phase shift with the simulated one and found a good agreement between the two [8]. This is illustrated in figure 5(a) for 10 µm thick Al target where several experimental probe beam phases obtained from different shots are plotted. The fluctuation of these phases is likely resulting from shot-to-shot fluctuations of the incident proton beam irradiating the target [see figure 2(a)]. Indeed, simulations that have been performed using as input a range of measured proton spectra, which were measured on separate shots, give the shaded areas in figure 5(a) that indicate the resulting range of probe beam phases derived from the simulations. We observe that the simulated phase spread is of the same order as the experimental phase spread. There are two shaded areas corresponding to the two equations of state that can be used in the code (the BLF EOS - area with vertical lines and the SESAME table - area with diagonal lines).
Figure 4. Density (solid line) normalized to the solid density and temperature (dotted line) profiles (normalized to the maximum value 15 eV in the target center) as a function of target thickness plotted for different simulation times: (a) 10 ps; (b) 50 ps. The time ‘0’ corresponds to the moment when first protons leave the ‘proton source’ target.

Similarly, the measured values of emissivity in time can be compared to the emissivity calculated by using the simulated target conditions at different times by the code ESTHER and the equation of radiative transfer in an expanding plasma [8]. This is illustrated in figure 5(b) for a 10 µm thick Al target. A difference between the measured and calculated emissivity can be attributed to the fact that the target was modeled as a black body radiating at a certain temperature where in reality the target is partially reflective and not perfectly absorbing.

Figure 5. Comparison of the measured and computed (with two different EOS describing the target material) probe beam phase shift (a) and emissivity (b) from the rear surface of the heated 10 µm thick Al target (b). Shaded area with vertical lines corresponds to BLF EOS, area with diagonal lines corresponds to SESAME EOS.

This work is an advance on previous studies where the proton energy deposition was not experimentally measured but, rather, fit to the heating diagnostics measurements. While in [28] the proton spectrum was scaled to reproduce the measured temperature and then used as an input to hydrodynamic simulation, here the measured proton spectrum was directly used as an input for the simulation and then the output compared with the measurements. Similarly, in [7], it was shown that in the proton beam there is enough energy to induce isochoric heating of a material, while the temperature was deduced by comparing the measured rear surface emission with the simulated one assuming the material to be heated to some given initial temperature.
4. Time-resolved study of structural properties of cold to warm dense matter transition

Once the proton source and the induced heating were characterized, giving us an insight into the target conditions, we studied the local atomic structure of warm dense aluminium sample using X-ray Absorption Near-Edge Spectroscopy (XANES).

The ultrashort broadband x-ray probe was created with a synchronized (with a main laser pulse), 5 J energy, 1.4 ps duration, 0.53 µm wavelength beam focused on an erbium (Er) target. To collect the transmitted spectra through the heated sample and the direct emission of the x-ray source (and deduce absorption curves), a specific x-ray absorption spectrometer (presented in figure 6) composed of two distinct conical KAP crystals was used. This configuration releases the results from shot-to-shot fluctuations of the x-ray source. The thickness of the Al sample (d=0.5 or 1.6 µm) was chosen to optimize the signal-to noise ratio in the absorption spectra. Experimental constraints required averaging the x-ray data radially over 170 µm in the heated target, starting from the axis of the proton beam irradiation (with an error of +/- 30 µm). As it was shown, a maximum temperature of 15 eV at solid density could be reached, however the mean temperature integrated over the radial dimension (normal to the axis of irradiation) of the heated area is in the eV range [29].

Controlling the delay between the two laser pulses, we probed the sample in different states. Due to shot-to-shot fluctuations, the use of two different Al sample thicknesses and proton beam variations, we collected the XANES spectra for different temperatures. Some results are presented in figure 7(a).

The averaged temperatures of the sample are estimated using a simple model based on Fermi-Dirac statistics where a one-to-one correspondence between the $K$ edge slope and the temperature is found [29].

![Figure 6. Schematic of the double x-ray spectrometer.](image)

![Figure 7.](image)

(a) Experimentally measured XANES spectra near the K-edge. The spectra are normalized to the mean value without the XANES structure. The ‘cold shot’ corresponds to a shot without heating proton beam; (b-c) Calculated K-edge XANES spectra (normalized to the mean value without XANES structures) of aluminum as a function of the temperature (solid density): (b) (M)HNC-NPA model, (c) QMD data.
The measured spectra were compared to two theoretical approaches of modelling. The first one is a liquid-metal approach based on the Neutral Pseudoatom (NPA) concept of dense matter coupled with a specific finite difference modeling of XANES structures [14]. The second one uses QMD simulations in the framework of density functional theory [16, 17]. The absorption spectra calculated for different temperatures using these two approaches are given in figure 7(b) and (c). Both the experimental results and the calculations exhibit a K edge broadening with increasing temperature and the progressive smoothing of the absorption spectra just after the K edge. According to the theory of XANES [30], the modulation observed in the spectrum is directly related to short-range order of the probed material. Consequently, the smoothing of the XANES structures is due to a progressive loss of ion-ion correlation.

![Figure 8.](image)

To quantitatively compare experimental and calculated XANES profiles, we have calculated a ‘contrast’ \( C \) for each profile as a function of temperature: \( C(\%) = (I_{\text{max}} - I_{\text{min}})/(I_{\text{max}} + I_{\text{min}}) \), where \( I_{\text{max}} \) and \( I_{\text{min}} \) are average values of the absorption signal around the first XANES maximum and minimum, respectively. Results are reported in figure 8(a). The evolution of both experimental and calculated contrasts is similar. The information about the temporal dynamics of the observed ordering loss we obtain by following in time the simulated target conditions (using the ESTHER code) and use them to compute averaged XANES spectra similarly as in the experiment. For this, we exploit the QMD spectra calculated at solid-density for various temperatures and calculate the contrast of the simulated average spectrum at each time (details are given in [29]), as shown in figure 8(b). The contrast falls rapidly, in 10 ps, while the average temperature increases. At this point, according to the theoretical models, the medium becomes structure-less and we can put an upper bound of 10 ps on the loss of short range ordering [29].

5. Conclusion

In summary, we have presented the results of the study on characterization of the warm dense aluminum plasma created by laser-accelerated proton beam. We have demonstrated that we can produce large volumes (~ 10^6 µm^3) of well defined warm dense matter as well as that it can be used to study the ionic structure of this state by x-ray absorption measurements.

Using a previously characterized proton source as input, the target heating and expansion were simulated self-consistently, allowing a direct comparison with experimental measurements. In this way, we have minimized the number of free parameters in the modeling of the heating. We have shown that heating was isochoric and almost uniform along the target thickness.

Using ultrafast x-ray probing, we have experimentally observed a progressive loss of ordering within solid density sample as the temperature rises from 300 K to > 104 K. The loss of short-range
ordering was detected through the progressive smoothing of the time-resolved X-ray Absorption Near Edge Spectroscopy (XANES) structure. The observation were correlated to Quantum Molecular Dynamic calculation as well as a simpler liquid description and good agreement, qualitative as well as quantitative was found. Such comparison allowed to conclusively relate the observed XANES spectral modifications to structural disordering of the heated material.

The next step would be to study the stopping power of ions in WDM which is still not well known and it is very important for the inertial fusion research. As the electron structure is known to play an important role in SP, we expect to observe variations in the stopping power.

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