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Atomic Processes in Plasmas Created by an Ultra-short Laser Pulse

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Abstract. Point projection K-shell absorption spectroscopy has been used to measure absorption spectra of transient aluminum plasma created by an ultra-short laser pulse. 1s-2p and 1s-3p absorption lines of weakly ionized aluminum were measured for an extended range of densities in a relatively low-temperature regime. Independent plasma characterization was obtained from frequency domain interferometry (FDI) diagnostic and allows the interpretation of the absorption spectra in terms of spectral opacities. The experimental spectra are compared with opacity calculations using the density and temperature inferred from the analysis of the FDI data.

The radiative opacities of dense plasmas are important for stellar interior physics [1] and inertial confinement fusion plasmas [2]. Experimental investigation of plasma photoabsorption coefficients has been an active field of research for many years. In order to test theoretical models, measurements of hot dense plasma absorption at well defined temperature and density were obtained using volumetrically radiatively heated foils [3]. This method allows uniform heating to temperatures up to 80 eV and for densities between 0.001 and a few 0.01 g/cm³ in thermodynamic conditions near local thermodynamic equilibrium (LTE) [3, 4].

Higher density plasmas with minimized gradients can be created by intense laser irradiation of solids. When a sub-picosecond laser pulse irradiates a very thin foil (∼tens of nm) with intensities in the range of 10¹⁵ – 10¹⁶ W/cm², impulse heating followed by a rapid heat conduction produces a high-density, moderate-temperature plasma before any hydrodynamic expansion [5]. Short-pulse x-ray sources of a few picosecond duration emitting in the sub-5-keV range has proven to be feasible by irradiating high-Z materials with a sub-picosecond laser pulse [6]. This offers the possibility to use point-projection time-resolved absorption spectroscopy to study the spectral opacities of dense plasmas.

We present quantitative data of transmission spectra in the warm dense matter regime. 1s-2p and 1s-3p absorption lines of aluminum were measured for densities up to 0.3 g/cm³ and temperatures varying from 12 to 25 eV. In the warm dense regime studied, the aluminum plasma is weakly ionized and density effects are not negligible. The continuum lowering modify
the ionization balance and pressure ionization tends to delocalize atomic orbitals. In the studied spectral range, many 1s-np absorption lines merge in the K-shell photoionization. Therefore our experimental data allow us to test the accuracy of approximations that have been proposed to take into account density effects in spectral opacity calculations.

The experimental setup uses two ultrafast lasers to produce both the plasma and the x-ray probe. The first ultra-short laser pulse created a thin, high-density plasma slab fairly uniform in temperature. An ultra-short x-ray pulse, produced by the second ultra-short laser, was used to backlight the slab.

The experiment was performed at the 100 TW LULI facility, using two 300-fs laser beams. The experimental setup has been described in details in previous papers [7]. A 0.3-J (1.06-μm) heated a 45-nm thick aluminum layer, deposited on a self-standing 25-nm silicon nitride (Si$_3$N$_4$) substrate. The heating beam was focused on the Si$_3$N$_4$ side of the target, limiting the longitudinal gradients in aluminum. A 8-J energy, (0.53-μm) beam focused on a samarium sample with an off-axis parabola to a spot size of 20-μm, produced the backlighter. The time-delay between the x-ray pulse and the heating laser at the target was measured before each shot with an uncertainty of 1 ps. Diagnostics viewed the rear side of the target, i.e., the thin Al layer side. A single-shot FDI diagnostic system [8] monitored the phase of a chirped probe laser beam (1.06-μm) of 35-ps duration, reflected at a 47° incidence angle on the rear surface of the plasma. X-ray transmission spectra in the 1470-1610 eV spectral region were recorded with a conically-bent potassium-hydrogen-phthalate crystal spectrometer coupled to a cooled 1024×1024 16-bits CCD camera. For the spectral region studied here, the Sm plasma provided a 4-ps duration quasicontinuum backlighter spectrum [9].

Fig. 1a shows an example of a space-resolved absorption spectrum measured 12 ps after the heating laser at a laser intensity of $1.5 \times 10^{15}$ W/cm$^2$. The spectra covered the 1s-2p absorption lines of Al$^{4+}$ and Al$^{5+}$, the K-edge position at 1560 eV and the Al$^{3+}$ and Al$^{4+}$ 1s-3p absorption lines. The cold Al K-edge is visible outside the focal spot. The initial areal density of the Al layer was measured at each shot by comparing the tabulated cold Al and Si$_3$N$_4$ transmission near the K-edge with the measured transmission (Fig. 1b). We found that the areal density of the aluminum layer is equal to $12 \pm 4 \, \mu g/cm^2$.

At the laser intensity used in the experiment the energy is delivered to the target in a very short time before any hydrodynamic expansion. The density and temperature during the plasma expansion depend mainly of the maximum temperature achieved at the end of the laser pulse. To know the plasma parameters at the time of the absorption measurements we have used the one-dimensional hydrodynamic MULTI-FS code [10]. The uncertainty in the laser interaction parameters measurements leads to a non-negligible uncertainty in the laser intensity and therefore on the density and temperature calculated with the hydrodynamic code. Nevertheless a precise determination of the plasma parameters is necessary to interpret the absorption spectra in terms of spectral opacities. To overcome this problem, we used the FDI diagnostic system to infer the density and temperature of the expanding plasma during the absorption measurements. Indeed, the phase shift of the reflected light provided by the FDI diagnostic depends mainly on the velocity of the critical surface, which is related to the laser intensity [11]. The Helmholtz equation is solved to calculate the propagation of the probe beam in the subcritical plasma and its reflection near the critical surface. These calculations provide the phase variation of the probe beam as a function of the laser intensity and were used as a postprocessor for the MULTI-FS code. Fig. 1c shows the temporal evolution of the phase of the chirped probe laser beam reflected at the rear critical surface of the plasma. The best agreement between theoretical calculations and experimental data is obtained by adjusting the laser intensity in MULTI-FS. An input laser intensity of $I_L \pm 10\%$ is determined from the best fit of the measured phase shift, whereas $I_L$ and the measured laser intensities are equal within 30%.
To interpret the transmission spectra, a LTE opacity code is used with the density and temperature profiles given at the time of the absorption measurements by the hydrocode reproducing the FDI measurements [12]. The areal densities used to calculate the transmission spectra are between 8.5 and 9.5 $\mu$g/cm$^2$, in the lower limit of the measured areal density. The calculated transmission spectra are convolved with the spectral resolution, i.e., 2 eV.

Spectral profiles in the center of the focal spot were extracted from all the space-resolved spectra, taking into account a 20-µm spatial width compatible with the Sm source diameter. Fig. 2a shows lineouts measured for different laser intensities and different probe-times as well as the theoretical calculations. Fig. 2b shows the corresponding thermodynamic paths inferred from the FDI analysis. The layer temperature is mainly driven by the time-delay between the heating pulse and the backlighter beam whereas the density is mainly dependent on the laser intensity. An overall good agreement is obtained between the experimental spectra and the theoretical transmission. Satellite lines from excited Al$^{3+}$ configurations and negative shifts of photoionization thresholds due to continuum lowering contribute to enhance the absorption of the red wing of the Al$^{4+}$ 1s-3p absorption structure. For a LTE aluminum plasma at temperature 16 eV and density 0.09 g/cm$^3$, close to the conditions obtained at 12 ps, the theoretical photoionization threshold shifts are between -21.9 eV and -8.8 eV for an ionic stage between Al$^{6+}$ and Al$^{+}$. A red wing asymmetry due to unresolved broad resonant and satellite lines of Al$^{2+}$ can be observed for Al$^{3+}$ 1s-3p absorption structure at 12 ps. Calculations indicate that the blue wing of the Al$^{3+}$ 1s-3p absorption structure is unaffected by neighbouring lines. Then we can test our electron impact broadening calculation based on a semi-empirical method [13]. The theoretical half-width at half-maximum of the Al$^{5+}$ 1s-3p line is equal to...
1.5 eV. Taking into account the spectral resolution, this value becomes equal to 1.8 eV, close to experimental value (1.86 eV). At later times, the 1s-3p absorption structures are in good agreement with the experimental absorption structures. At 51 ps the plasma recombination seems to be overestimated, leading to an underestimation of the Al$^{5+}$ population.

In conclusion, absorption spectra of transient Al plasmas have been measured for densities up to 0.3 g/cm$^3$ and temperatures varying from 12 to 25 eV using high-intensity, ultra-short laser pulse. Independent characterization of the plasma parameters allowed an interpretation of the absorption spectra in terms of spectral opacities. Our experimental data are in good agreement with results of our LTE opacity code using a detailed line accounting approach coupled with hydrodynamic simulations. Such agreement contributes to validating the treatment of the density effects occurring in the experimentally explored thermodynamic regime.

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