XUV Emission from Autoionizing Hole States Induced by Intense XUV-FEL at Intensities up to $10^{17}$ W/cm$^2$

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Abstract. Aluminium targets were irradiated with 92 eV radiation from FLASH Free Electron Laser at DESY at intensities up to $10^{17}$ W/cm$^2$ by focussing the beam on target down to a spot size of ~1 μm by means of a parabolic mirror. High resolution XUV spectroscopy was used to identify aluminium emission from complex hole-states. Simulations carried out with the MARIAS code show that the emission characterizes the electron heating in the transition phase solid-atomic. The analysis allows constructing a simple model of electron heating via Auger electrons.

1. Introduction
The study of dense strongly coupled plasmas DSCP and warm dense matter WDM is of intense interest because such matter is difficult to handle theoretically, with electrostatic and thermal energies being of equivalent importance, rather than one being a perturbation of the other as in solid state physics or classical plasma physics. Moreover in this regime experimental data is not easy to obtain and one of the key issues is the ability to create uniform well-defined samples of matter near solid density but with temperatures of ~ 1-100 eV. A possible way forward is the use of the new generation Free Electron Lasers (XUV-FEL and X-FEL) to irradiate solid samples [1, 2].

Free electron XUV- and X-ray lasers allow volumetric heating because the plasma frequency of the solid is smaller than the laser frequency. The radiation penetrates therefore deep into the solid and...
the analysis of the data does not suffer from critical surface effects. Moreover, in regimes where thermal energies are too low to create short wavelength radiation that can exit a high density sample without reabsorption, the emission from photoionized core states, allows to get information on high energy density matter at low temperatures [3].

We report on one of the first experiments with a high-intensity 4th generation light source FLASH (XUV-Free-Electron-Laser at DESY in Germany) where the solid is effectively heated and destroyed after intense photoionization of the target. Finally we present an analysis of the characteristic XUV transitions from the target.

2. Experimental setup
The present experiment was carried out using the FLASH XUV-Free Electron Laser Facility at DESY in Germany. Solid Al-foils were irradiated with intense pulses of λ = 13.5 nm radiation operating at 5 Hz. The pulse length was τ ≈ 15 fs and the beam was focussed down to about 1 μm diameter with a multilayer parabolic optic (multilayer Mo-Si) with a focal length of 269 mm to achieve intensities up to 10^17 W cm^{-2}. A 3 mm aperture in the beam limited its size and the pulse energies varied between 5 and 10 μJ on target. The best focus was established by analysis of PMMA (Polymethylmethacrylate) ablation using a range of target positions relative to the focus optic and a range of pulse energies. This analysis established that the best focus was ~ 1 μm FWHM. Further, the reflectivity of the optic was measured after the experiment and found to be 48%. The spectrometer employed a 1200 lines/mm Hitachi grating with variable line spacing to create a flat spectral focal plane on a CCD camera positioned on a vacuum flange, providing a spectral coverage from ~ 10-30 nm. An Al edge filter was used to establish that the spectral resolution achieved was ~ 0.1 nm thus providing an effective spectral resolution of about ∆λ/λ~150 in the relevant spectral range. The samples were composed of 10 μm thick Al foils that were continuously moved transverse to the FEL beam across the focus to expose a fresh surface each shot. Figure 1 shows an experimental spectrum obtained by integrating 2000 shots at irradiation intensities of about 10^16 W/cm^2. The dominant spectral feature (Figure 1) is identified as atomic Al IV lines: 1s2s2p5-1s2s2p6 (3P°)3s 1P and 3P at 16.1 nm and 16.2 nm, respectively by comparison with transition energies obtained from Hartree-Fock calculations [4]. The transitions at shorter wavelengths are related to transitions 1s2s2p5-1s2s2p3d 1P, 3P and 3D at 13.0 nm, 13.1 nm and 13.2 nm respectively.

3. Analysis of the XUV Al target emission
A comparison between the experimental data in Figure 1 (black solid line) and the simulation of the Al IV lines (blue solid line) shows large discrepancies indicated by the hatched area. It is known that transitions located at the red wing of resonance line transitions might be due to screened transitions, so called satellite transitions, firstly investigated in detail for the He-like resonance line of highly charged ions [5]. The observed transitions on the red wavelength wing of the Al IV resonance line transitions might therefore originate from XUV-transitions into the L-shell of heated Al while spectator electrons are present in the M-shell:

![Figure 1. Aluminum emission (solid black line) at best focus. The dominant spectral features are Al IV resonance line emission (blue solid line). The hatched areas indicates potential emission from hole states. The spectral fit (solid green line) has been obtained from MARIA-code simulations [6] and the application of a genetic algorithm [7].](image-url)
In order to understand the effective wavelengths shift due to several spectator electrons, we have performed detailed atomic structure calculations by means of the Multi-Configuration Hartree-Fock method [4]. As the number of line transitions I. and II. is too large to be analysed in table form, we represent the transitions by spectral simulations using

\[ I(\omega) = \sum_{i,j} g_{ji} A_{ji} e^{-\frac{E_j - E_0}{k_B T}} \Phi_{ji}(\omega) \]  

(1)

g_{ij} is the statistical weight, A_{ji} the radiative decay, E_j and E_0 are the energies of level j and the ground state respectively, \( \Phi_{ij} \) is the line profile. Figure 2 shows the spectral distribution of M-L transitions for different number of electrons in the M-shell: x=1, 2, 3. The five vertical dotted lines 1-5 indicate the central positions of dominating emission groups.

The spectral distribution of transitions originating from K^2L^2M^2 and K^2L^2M^3 configuration indicate rather strong shifts to the red for the first 3 groups (dotted lines 1-3 at about 9.4 nm, 13.0 nm and 16.1 nm, respectively). As screening electron and optical electrons are in the same M-shell, the shifts are rather strong (about 0.5-0.6 nm for each additional M-electron). In contrast to the dotted lines 1-3 in Fig. 2 are the dotted lines 4 and 5 (at 26.3 nm and 29.6 nm). Almost all transitions, independent of the number of electrons in the M-shell, are located near these dotted lines. The atomic structure calculations indentifies inner-shell transitions with \( n=1 \):

\[ 1s^22s^22p^6M^2 \rightarrow 1s^22s^22p^5M^3 + h\nu \]

For these transitions, the screening effect from M-electrons is not large and barely visible in the spectral distribution. The simulation presented in Figure 2 (solid curve indicated as K^2L^2M^2 + hole states) show that the discrepancies indicated by the hatched area can be attributed to the transitions I. and II. because the shifted transitions (originating from configurations K^2L^2M^2 and K^2L^2M^3) just fall into the requested wavelength interval.

Figure 3 identifies a large temperature sensitivity of the spectral distribution of hole states from about 1 eV – 50 eV. Based on these sensitivities the electron temperature is estimated from the fit presented in Figure 1 (solid curve indicated as K^2L^2M^2 + hole states) to be about 25±5 eV.

We note that there is a substantial difference in the temporal evolution of the Al IV emission compared to those of Al II and Al III of channels I and II. Transition probabilities of Al IV lines are \( 10^9 \) to \( 10^{11} \) s\(^{-1} \) [4], so that the self-emission occurs about 0.1 ns after excitation (taking into account radiation transport effects). This time is much longer than the 15 fs FEL pulse and corresponds to a recombination phase. As the Al IV resonance lines are sensitive to radiative recombination, a low temperature recombining phase may considerably contribute to the time integrated spectra. The situation for the Al II and Al III emission is dramatically different. The
configurations K2L7M2 and K2L7M3 are autoionizing, their characteristic time scales being ≤ 100 fs [4]. We therefore based our analysis on hole states from Al II, III rather than resonance line emission from Al IV.

A temperature Te, which is much higher than melt, enables one to construct a qualitative model of electron heating. After photoionization of the LII and LIII shells an excess energy of ~19 eV will be distributed amongst the 4 conduction band electrons. Assuming rapid thermalization on sub fs-scale [8] and accounting for the 12 eV Fermi energy for Al, a Te of ~8 eV is obtained. This Te represents an excited transient state that will start relax after ~40 fs by either radiative decay or by autoionization. As the branching ratio favours autoionization by 99.8% the energy of the decaying electron is shared with the three remaining conduction band electrons. Simulations show that the energy difference, e.g. between the configuration K2L8M3 and K2L7M4 is about 70 eV which is equivalent to the kinetic energy of the Auger electrons [4]. Assuming that all the Al atoms in the intense FEL beam are photoionized this excess 70 eV will rapidly thermalize with the 8 eV electrons resulting in Te of ~22 eV. This model is in good agreement with the spectroscopic analysis of channels I and II presented in Figure 1. As saturated absorption has been observed [9] (which means Al becomes transparent to the laser intensity due to the photoionization of all 2p6-electrons) the spectroscopically determined Te of ~25 eV indicates that Auger electron heating plays an important role in the laser-matter interaction.

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