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Published in:
Journal of Physics: Conference Series

Document Version:
Publisher's PDF, also known as Version of record

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XFEL resonant photo-pumping of dense plasmas and dynamic evolution of autoionizing core hole states

F B Rosmej¹,², A Moinard¹,², O Renner³, E Galtier⁴*, J J Lee⁴, B Nagler⁴, P A Heimann⁴, W Schlotter⁴, J J Turner⁴, R W Lee⁵, M Makita⁵, D Riley⁶, J Seely⁶
¹ Sorbonne Universités, Pierre and Marie Curie, UMR7605, LULI, case 128, 4 place Jussieu, F-75252 Paris Cedex 05, France
² LULI, Ecole Polytechnique, CEA, CNRS, Physique Atomique dans les Plasmas Denses, Palaiseau, France
³ Institute of Physics, Academy of Sciences CR, Prague, Czech Republic
⁴ SLAC Accelerator National Laboratory, California, USA
⁵ Queen’s University of Belfast, Centre for Plasma Physics, UK
⁶ Naval Research Laboratory, Washington, DC, USA
* part of the work was done being a post-doc at LULI-PAPD

E-mail: frank.rosmej@upmc.fr

Abstract. Similarly to the case of LIF (Laser-Induced Fluorescence), an equally revolutionary impact to science is expected from resonant X-ray photo-pumping. It will particularly contribute to a progress in high energy density science: pumped core hole states create X-ray transitions that can escape dense matter on a 10 fs-time scale without essential photo-absorption, thus providing a unique possibility to study matter under extreme conditions. In the first proof of principle experiment at the X-ray Free Electron Laser LCLS at SLAC [Seely, J., Rosmej, F.B., Shepherd, R., Riley, D., Lee, R.W. Proposal to Perform the 1st High Energy Density Plasma Spectroscopic Pump/Probe Experiment”, approved LCLS proposal L332 (2010)] we have successfully pumped inner-shell X-ray transitions in dense plasmas. The plasma was generated with a YAG laser irradiating solid Al and Mg targets attached to a rotating cylinder. In parallel to the optical laser beam, the XFEL was focused into the plasma plume at different delay times and pump energies. Pumped X-ray transitions have been observed with a spherically bent crystal spectrometer coupled to a Princeton CCD. By using this experimental configuration, we have simultaneously achieved extremely high spectral (λ/Δλ = 5000) and spatial resolution (δx=70 µm) while maintaining high luminosity and a large spectral range covered (6.90 - 8.35 Å). By precisely measuring the variations in spectra emitted from plasma under action of XFEL radiation, we have successfully demonstrated transient X-ray pumping in a dense plasma.

1. Introduction
Laser Induced Fluorescence “LIF” [see, e.g. 1] is a well-known method of laser spectroscopy in the optical wavelength range that has received worldwide attention, e.g., in physics, biology, medicine, and technology. Its physical principle is based on the stimulated photo-absorption of an electric dipole transition by a laser photon that matches the energy difference of the upper and lower atomic states (i.e., fulfills conditions of the resonant photo-pumping). The pumped upper states can decay via spontaneous radiative emission, the so-called fluorescence radiation. Here, we report on the laser-
induced fluorescence in the X-ray spectral range that could be realized only with the emergence of the 4th generation of light sources, X-ray Free Electron Lasers. LCLS at SLAC in United States [2] has been the first installation available for the scientific community. Although the principle of the X-ray LIF can also be approximated by an effective two level system, the X-ray spectral range shows several important differences. First, radiative transition probabilities scale as $Z^4$ (where $Z$ is the effective charge of the ion) and transition energy as $Z^2$, i.e., the laser intensity requested to efficiently pump X-ray transitions scales as $I \propto Z^6$ [3]. Second, in most of the cases X-ray pumping creates core hole states that are autoionizing. To induce effective changes in X-ray energy levels, X-ray pumping must therefore be realized in a time interval corresponding to the characteristic time scale of autoionization. As autoionization rates are very large (characteristic time scales are of the order of 1-100 fs), efficient pumping in the X-ray range is a great challenge: extremely high X-ray photon intensities are requested on a 10 fs time scale to “beat the Auger clock”. Moreover, X-ray fluorescence may be seriously reduced by the non-favorable branching ratio $A/(A+\Gamma)$ where $A$ is the spontaneous X-ray transition probability and $\Gamma$ the autoionization rate. We note that these constraints are also strongly related to the production of “hollow” ions in intense radiation fields [3-5], a subject that became recently also of interest in the high-energy laser community [6].

2. Experimental setup

Figure 1 (left) shows the scheme of the experimental setup. A dense plasma was created by a YAG laser ($\lambda=800$ nm, $E=24$ mJ, $\tau=120$ ps, $I=4 \cdot 10^{12}$ W/cm$^2$) irradiating solid aluminium and magnesium that was mounted on a rotating cylinder (to provide fresh target surfaces for each shot at repetition frequency of 120 Hz). At 11.5° relative to the optical laser beam, the XFEL (about $10^{13}$ photons/shot, pulse duration 250 fs) was focused to a diameter of 20 μm in the dense plasma plume at different delay times and X-ray pump energies. Pumped X-ray transitions have been observed with a Bragg crystal coupled to a Princeton CCD (2048x2048 pixels). The spherically bent crystal of mica (004) set at the distance of 57 cm from the plasma source provided a magnification factor of 0.21 and a high spectral resolution at the level of $\lambda/\delta\lambda = 3500$ while maintaining a large spectral range (6.90 - 8.3 Å).

The spatial resolution in a direction perpendicular to the expanding plasma (about 64 μm) was essentially determined by the CCD pixel size of 13.5 μm. The crystal was protected with a 2 μm thick polycarbonate ($C_6H_{14}O_3$) foil (effectively resulting in 4 μm thick foil due to transmission of incoming and outgoing rays), the CCD was protected with a 1 μm polypropylene ($C_3H_6$) coated with 0.1 μm aluminum from each side to suppress the visible light. Spectra have been corrected for crystal reflectivity, CCD response and filter transmission.

The right part of Fig. 1 shows the Mg spectrum obtained when irradiating the rotating Mg cylinder by optical laser only, i.e., without XFEL interaction. The spectrum shows the He-like resonance line.
series ($\text{He}_\beta = 1s3p \, ^1P_1 - 1s^2 \, ^1S_0$, $\text{He}_\gamma = 1s4p \, ^1P_1 - 1s^2 \, ^1S_0$, $\text{He}_\delta = 1s4p \, ^1P_1 - 1s^2 \, ^1S_0$, $\text{He}_\epsilon = 1s5p \, ^1P_1 - 1s^2 \, ^1S_0$) and associated dielectronic satellite emission, $\text{He}_\beta$ - satellites $1s2l3l' - 1s^22l''$ and $\text{He}_\gamma$ - satellites $1s2l4l' - 1s^22l''$. The spectrum demonstrates excellent quality and resolution of the implemented X-ray diagnostics: a) high signal to noise ratio, b) large spectral coverage 6.9-8.3 Å, c) extremely high spectral resolution - note that even single LSJ-split configurations in the $\text{He}_\beta$-satellites $1s2l3l' - 1s^22l''$ are resolved (for further details on $\text{He}_\beta$ - satellites see [7,8]).

3. Resonant X-ray pumping

Figure 2 shows the X-ray spectral distributions observed at solid Al targets. The upper spectrum was obtained when irradiating the Al cylinder with the optical laser only. The strongest emission is the He-like resonance line $\text{He}_\alpha = W = 1s2p \, ^1P_1 - 1s^2 \, ^1S_0$ and the intercombination line $Y = 1s2p \, ^3P_1 - 1s^2 \, ^1S_0$. Li-like satellite emission $1s2l2l' - 1s^22l''$ (indicated by $1s2l2l':=K \, ^1L_2$ in Fig. 2) is rather weak. The spectral distributions indicated with $\text{He}...\text{C}$ designate pumping of the $\text{He}_\alpha$ transition (pump photon energy set to 1597 eV), pumping of the Li-like configuration $K \, ^1L_2$ (pumping $K \, ^1L_1 + h\nu_{\text{XFEL}} \rightarrow K \, ^1L_2$ at 1587 eV), Be-like configuration $K \, ^1L_3$ (pumping $K \, ^2L_2 + h\nu_{\text{XFEL}} \rightarrow K \, ^1L_3$ at 1570 eV), B-like configuration $K \, ^1L_4$ (pumping $K \, ^2L_3 + h\nu_{\text{XFEL}} \rightarrow K \, ^1L_4$ at 1551 eV) and C-like configuration $K \, ^1L_5$ (pumping $K \, ^2L_4 + h\nu_{\text{XFEL}} \rightarrow K \, ^1L_5$ at 1531 eV).

The pumping at 1587 eV (curve labeled as “Li” in Fig. 2) is of particular interest. This photon energy corresponds to the transitions $1s^22l + h\nu_{\text{XFEL}} \rightarrow 1s2l2l'$. The pumped states are therefore the autoionizing Li-like levels of the type $1s2l2l'$. Although the spontaneous electric dipole radiative transitions that originate from these states are of very low intensity for the case without pumping (see upper spectrum with transitions labelled $1s2l2l'$ in Fig. 2), the X-ray pump creates an impressive intensity increase. It is important to emphasize that emission lines, being entirely absent in the dense plasma produced by the optical laser only (upper spectrum in Fig. 2, emission labelled “Be”, “B” and “C”), have been pumped to intensities that exceed those of the resonance line W (usually the most intensive X-ray emission line) by a factor of about 100. This is an extremely important demonstration of X-ray pumping: ion charge state distributions of dense matter can be probed via X-ray transitions that have been inaccessible up to present due insufficient signal/noise, background level. This is particular important for high energy density science: one of the most challenging plasma regimes are strongly coupled plasmas where the coupling parameter is proportional to $n^{5/2}/T$ with “n” being the density and “T” the temperature. High coupling parameters request therefore high density and low temperatures, but low temperatures usually prohibit X-ray transitions.

![Figure 2: X-ray spectral distribution of Al pumped at different XFEL energies.](image-url)
Our final concern has been the demonstration that X-ray pumping also provides a possibility to study the evolution of transient dense matter. Figure 3 shows its successful demonstration via a time dependent pumping of the Be-like configurations (right part of Fig. 3) which was realized by setting different delay times between the maximum of the optical laser pulse and the XFEL onset. As can be seen from the spectra, the spectral distribution changes in time, thus reflecting the dense plasma evolution. The left part of Fig. 3 shows the MULTI-fs simulations of aluminum plasmas created by the optical YAG laser that provides insight in the transient evolution of temperature and density for different target distances.

4. Conclusion
In the first proof of principle experiment at the LCLS X-ray Free Electron Laser, we have successfully demonstrated transient X-ray pumping of the dense plasma. The application of curved Bragg crystal X-ray spectroscopic technique provided high-resolution spectral distributions that permitted interpretation that goes much beyond the hydrogenic approximations (via resolving even LSJ-split levels in complex autoionizing configurations that permits in turn to separate the different excitation channels). These results open new horizons in X-ray pumping/X-ray fluorescence XFEL physics.

5. References