Polarization Dependence of Laser Induced inner-shell excitations

Yunpei Deng, Zhinan Zeng, Pavel Komm, Yinghui Zheng, Wolfram Helm, Xinhua Xie, Zoltan Filus, Mathieu Dumergue, Roland Flender, Maté Kurucz, Ludovit Haizer, Balint Kiss, Subhendu Kahaly, Ruxin Li and Gilad Marcus

1SwissFEL, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
2Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, 201800, China
3Department of Applied Physics, Faculty of Science, Hebrew University of Jerusalem, Jerusalem 91904, Israel
4Zentrum für Synchrotronstrahlung, Technische Universität Dortmund, 44227 Dortmund, Germany
5ELI-ALPS, ELI-HU Non-Profit Ltd., Wolfgang Sandner uet 3, Szeged 6728, Hungary

Abstract: Laser induced x-ray fluorescence were observed against laser polarization ellipticity. While emission from krypton peaks at linear polarization, a signature of recollision, emission from neon shows opposite trend. We attribute it to two competing processes. © 2021 The Author(s)

Correlations among multiple electrons constitute a cornerstone in physics, chemistry, and technology. Electron correlations are inherently involved in atomic relaxation processes such as autoionization, Auger decay and multiple-excitation states. Multi-electron processes in atoms, such as autoionization and Auger decay, involve a cascaded relaxation through many channels, of which not all need necessarily be radiative. Detailed information about the temporal order of these relaxation processes and the coupling between the different states is accessible only through time-domain observations, in which a first “pump pulse” initiates the process and a second “probe pulse” interrogates the system state at a later time. Since the relevant time scale for such dynamics spans from attoseconds to femtoseconds and the relevant energy scale for excitation spans from $10^{-2}$ to $10^{-5}$ eV, x-ray attosecond bursts may be the choice to serve as the pump and the probe events. However, due to the low photon flux of state-of-the-art HHG soft x-ray attosecond sources (for $h\omega > 300$ eV) and the low absorption cross sections in this spectral range, it is currently impossible to both pump and probe these processes with HHG soft x-ray sources. To probe dynamics involving valence electrons, an ultrashort infrared pulse is often used to initiate the process, and the inherently well synchronized XUV attosecond pulse probes it. It is difficult to extend this scheme to excite more strongly bound inner-shell processes, because of the large energy difference between inner-shell excitations and the infrared photon energy. Excitation of inner-shell dynamics by laser-induced electron re-collision might be the key for a solution to this experimental challenge.

Here, we present experimental and theoretical results [1] on soft x-ray emission due to laser-induced inner-shell excitation. This work summarizes the collected results from two different laser systems: The first laser has a pulse duration of 12 fs at a central wavelength of 1800 nm, a repetition rate of 1 kHz and a pulse energy of up to 0.8 mJ. The second system works with a pulse duration of 60 fs at a central wavelength of 3200 nm, at a repetition rate of 100 kHz and a pulse energy of up to 80 μJ. We measured the x-ray emission at right angle to the IR beam propagation direction. Neon and krypton gases were used as targets for the inner-shell ionization process. The gas was injected into the vacuum chamber from a nozzle with orifice of ~100 μm diameter at backing pressures ranging from a few mbar to ~100 mbar.

![Fig. 1.](image)

Fig. 1. (a) The x-ray spectrum generated after inner-shell excitation of neon atoms as a function of the QWP angle (linear polarization approximately at 0, 90, 180 & 270 degree, marked as white solid lines; circular polarization at 45, 135, 225 & 315 degree, marked as white dashed lines) with estimated peak intensity on target $2.8 \times 10^{15}$ W/cm². (b) The corresponding x-ray spectrum for krypton atoms as a function of the QWP angle. Same angle markings and laser parameters as (a), except for estimated peak intensity $2.3 \times 10^{15}$ W/cm².
A common practice to separate re-collision related processes, such as high harmonic generation (HHG), from other competing processes is to check the signal against the driving laser polarization ellipticity. As the polarization changes from linear to elliptical, the electrons’ trajectories quickly run away from the parent ion and the re-collision related process ceases to exist. Hence, to verify the re-collision mechanism for the case of inner-shell electrons, we checked the fluorescence yield against the laser polarization ellipticity.

The results presented in Fig. 1(b) are similar to those we got in our previous work [2], i.e. the yield of the characteristic line in krypton peaks at linear polarization and drops to a minimum at circular polarization, though it does not completely disappear. This picture is more detailed than the previous one and we can see a slight shift between the peak position of the characteristic line and the peak position of the continuum. Contrary to Fig. 1(b), Fig. 1(a) shows a completely opposite behavior: both the characteristic K line and the continuum emission from the neon atoms peak around the circular polarization of the IR laser and reduce to a much lower yield near the linear polarization. To explain this, we resort to a competing excitation mechanism. We suggest that at high gas densities the probability of exciting neighboring atoms by laser-released high-energy electrons is not negligible and has to be considered. Here we present detailed calculation of the probabilities for the tunnel-ionized electron to either re-collide with the parent ion and initiate a core-hole excitation, or to drift away from the parent ion and collide with one of the surrounding atoms, again, with high enough kinetic energy to excite a core-shell electron. (Hereafter we term these electrons “drift away electrons”). Our calculations and experimental results are inline with this assumption. Figure 2. shows the x-ray emission from krypton target at different laser intensities as a function of the laser ellipticity (QWP angle). It clearly shows that at low intensities the x-ray characteristic line (1590 eV) peaks at linear polarization, but when the laser intensity grows, the contributions from elliptical polarization become more pronounced. Our calculations clearly show this transition from re-collision domination at lower intensities to excitation by the drifting electrons at higher intensities.

![False color plots of x-ray emission spectra after ionization of krypton atoms at different laser intensities and for different polarization states. The vertical scale shows the QWP angle. White solid horizontal lines mark the approximate linear polarization, while dashed white lines mark approximately the circular polarization. Laser parameters: \( \lambda = 1800 \text{ nm}, \) pulse duration 12 fs. The estimated laser intensities are (a) \( 5.1 \times 10^{15} \text{ W/cm}^2 \), (b) \( 2.3 \times 10^{15} \text{ W/cm}^2 \), and (c) \( 2.0 \times 10^{15} \text{ W/cm}^2 \).](image)

In conclusion, while the mechanism of laser-induced electron re-collision remains the same for HHG and for electron impact ionization/excitation of valence electrons, the extension towards keV energies and interactions with inner-shell electrons is not trivial. Several factors are involved in these differences: first, because of the high ponderomotive potential needed to reach inner-shell electrons, the atom experiences a series of sequential tunnel ionization events towards a higher ionization state, as opposed to “low energy” HHG (< 100 eV), where we usually have a single ionization state. Second, since the inner-shell electrons are closer to the atom nuclei and partially screened by the valence electrons, their interaction cross section with the fast re-colliding electron is significantly smaller than the recombination cross section of the slower electrons with the parent ion that usually happens in the “low energy” HHG. As a result, other competing processes are brought to the front of the stage which make it more difficult to study the inner-shell excitations. One of the main competing processes is electron collision with neighboring atoms.

References