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Multiphoton Ionization of Xenon at the LCLS Free-Electron Laser

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Synopsis With the first X-ray free-electron laser (FEL), the Linac Coherent Light Source (LCLS), multiphoton ionization has been pushed to a new regime, where atoms and molecules are not just ionized by a series of valence ionizations but "from the inside out". At unprecedented high intensities and short pulse durations in the soft X-ray regime, a series of inner-shell photoionizations followed by cascades of Auger decays was observed to lead to highly charged final states in rare gases such as Ne, Ar, Kr, and Xe. Ion time-of-flight and fluorescence spectra were recorded for different FEL pulse energies and pulse lengths and compared to theoretical models to explain the underlying processes that lead to unexpectedly high charge states in Xe.

Experiments at the vacuum-ultraviolet FEL FLASH have marked the transition from strong-field ionization of valence electrons towards multiphoton ionization of inner-shell electrons [1]. While the most significant effects in the multiphoton ionization of xenon at VUV energies have been attributed to resonances in the 4d shell, ionization at soft X-ray energies is governed by the competition of photoemission and Auger and fluorescent decay in the 3p and 3d shells. By measuring both, the ion and the fluorescence spectrum, new insights into multiphoton ionization processes at soft X-ray energies and into the ultrafast decay mechanisms can be obtained.

Our experiments were performed in the CAMP instrument [2] at the LCLS AMO beamline with photon pulses between 3 and 300 fs duration and irradiance levels up to $\sim 10^{16} \frac{W}{cm^2}$. For simultaneous detection of ion and fluorescence spectra, an ion time-of-flight spectrometer equipped with position-sensitive delay line detectors was employed together with large-format, high-speed, single-photon counting X-ray pnCCDs. The shot-by-shot ion and fluorescence data can thus be correlated with each other and with the shot-by-shot pulse energy provided by LCLS.

Our measurements reveal the production of unexpectedly high charge states with ionization potentials beyond the chosen FEL photon energy along with a strongly enhanced fluorescence yield which increases non-linearly with the FEL pulse intensity. Comparisons with recent calculations performed with the XATOM package [3] explain some of the observed features but also emphasize the important of additional, most likely resonant effects not included in the calculations.

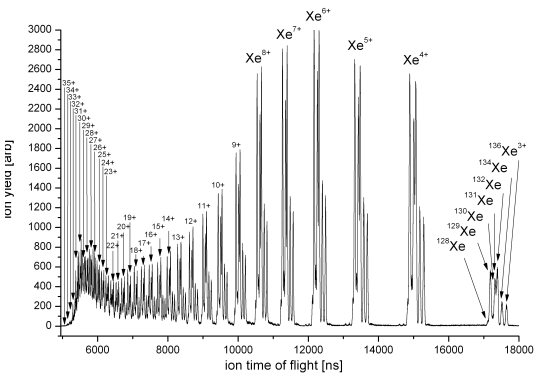

Figure 1. Xenon ion time-of-flight spectrum; FEL beam parameters: photon energy 1.5keV, pulse length 85fs, nominal pulse energy 2.4mJ, focus $\sim 10 \mu m^2$.

References

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