Nonlinear atomic processes in intense EUV-FEL fields studied by multielectron-ion coincidence spectroscopy

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Abstract: Simultaneous detections of electrons and counterpart ions allow us to identify two-photon formation of Xe^{4+} through the Xe 4d double-core-hole states.

Understanding of nonlinear optical responses of materials in the extreme ultraviolet (EUV) and X-ray regions is of great importance for various applications of intense ultrashort free-electron lasers (FELs). Intensive studies on isolated atoms and molecules have revealed that multiphoton multiple ionization of valence as well as inner-shell electrons is a typical nonlinear optical response [1]. When inner-shell electrons are ionized, the created core-holes relax through Auger processes, producing electrons with kinetic energies depending on the final electronic states. Since the Auger relaxation will compete with the subsequent photoabsorption in multiphoton processes, identification of electronic states related to each ionization pathway is necessary for understanding details of nonlinear ionization mechanism.

In this contribution, we will present our recent work on nonlinear ionization of Xe in intense EUV laser fields using a multielectron-ion coincidence technique[2]. By taking an advantage of ion charged states which carry information on how many photons are absorbed, we show that weak nonlinear electron signals can be extracted from widely distributed one-photon (linear) signals.

All the experiments were carried out at SACLA BL1 of RIKEN Harima institute in Japan. Irradiation with EUV-FEL pulses (91 eV, 30 fs, 60 Hz, 1.6×10^{12} W/cm^2) produced Xe^{Z+} ions ($Z \leq 4$). Among them, two-photon absorption processes are necessary for the Xe^{4+} formation while the other ions can be formed by single-photon ionization. Figure 1 shows spectra of (a)the total electrons, and electrons detected in coincidence with (b)Xe^{2+}, (c)Xe^{3+}, and (d)Xe^{4+} ions, respectively. The doublet peak at ~22 eV is assigned to the spin-orbit splitting of $4d^{-1}$ photoelectrons while sharp peaks at 8-20 and 30-35 eV are attributed to Auger electrons resulting from the 4d core-hole decays to Xe^{2+}. These spectral features are identified in the electron spectrum for Xe^{4+} (Fig. 1(d)), suggesting that Xe^{4+} ions are formed by sequential ionization through Xe^{2+} states. In addition, a broad peak distribution is observed at 25-38 eV, which are not explained by the sequential ionization via the Xe^{2+} states. This energy region agrees well with energy differences between the $Xe^{2+}(4d^{-2})$ and Xe^{3+} $(4d^{-1}5p^{-2})$ states [3], which indicates two-photon ionization through the 4d double-core-hole (DCH) states in the formation of Xe^{4+}.

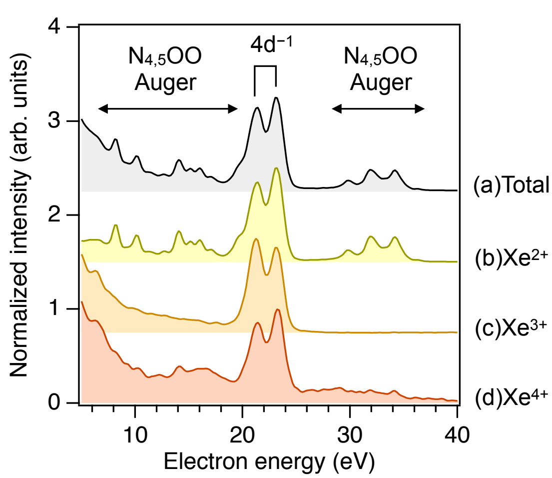

Figure 1 : Electron spectra of Xe in EUV-FEL laser fields (91 eV, 30 fs, 60 Hz, 1.6×10^{12} W/cm^2). (a) Total electrons, electrons detected in coincidence with (b)Xe^{2+}, (c)Xe^{3+}, and (d)Xe^{4+} ions.

By comparing integrated peak intensities of Auger signals associated with the two pathways, we found that the contribution of the Xe 4d DCH pathway is comparable to that of the sequential one [2] even though EUV-FEL pulse duration (30 fs) is longer than lifetime (~6 fs) of the $Xe^{+}(4d^{-1})$ states. Simulation based on coupled rate equations taking into account two-photon absorption processes revealed that the efficient two-photon formation of the Xe 4d DCH states can be made when the ionization cross section σ_{DCH} of $Xe^{+}(4d^{-1})$ states, $Xe^{+}(4d^{-1}) + h\nu \rightarrow Xe^{2+}(4d^{-2}) + e^{-}$, falls in the range between 27 and 64 Mb. The obtained results suggest that σ_{DCH} is considerably larger than the normal 4d cross sections (~20 Mb), which could be attributed to the resonance excitation from the $4d^{-1}$ to $4p^{-1}$ states in Xe [2].

References

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