PCI Recapture of Photoelectrons: Conjugate Processes and Angular Correlations

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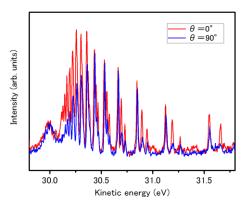
We conducted high resolution Auger electron spectroscopy measurements for gas phase Kr and Xe close above their photoionization thresholds. The Auger electron peaks manifest Rydberg series structures due to the Post Collision Interaction (PCI) induced recapture of the photoelectron into Rydberg orbitals of the final ion. The angular momentum states of the Rydberg series were clearly resolved. Surprisingly, conjugate lines seemingly defying dipole selection rules were found to be dominant in the Kr Auger spectra. Also, very conspicuous angular dependence was found in the intensity pattern of the Rydberg series structure in the Xe Auger electrons.

In case of photoionization close above threshold followed by an Auger process, Post Collision Interaction (PCI) may take place. It results in the shift, broadening and continuous tailing of the photoelectron peak towards lower energy, as well as the shift, broadening and continuous tailing of the Auger electron peak towards higher energy. With the photon energy tuned even lower, very close to the threshold the photoelectron slows down enough to be recaptured by the remaining ion into one of the Rydberg orbitals. This manifests as the Rydberg series structure imprinted as detailed structure on the Auger line. With our high electron energy resolution, we manage to resolve further sub-structure on each Rydberg line due to the angular momenta of the final ionic states [1].

The Kr $(M_5N_{2,3}N_{2,3})$ Auger electron spectrum originating from $3d_{5/2}^{-1}$ photoionization was measured with the photon energy tuned very close above the ionization threshold. In the Kr⁺3 $d^{-1}_{5/2} \rightarrow 4p^4 ({}^{1}S_0)$ ml spectrum, this can be clearly seen with each principal quantum number leading to the appearance of four fine structure component lines. The four components converge and form a single peak within the resolution limit for higher m. The energy order from low to high allows the assignment of the four conspicuous components as the $3d^{-1}_{5/2} \rightarrow 4p^{-2}({}^{1}S_{0})$ ms, mp, (m-1)d and (m-2) f states. Unexpectedly, conjugate peaks $3d^{-1}_{5/2} \rightarrow 4p^{-2}({}^{1}S_{0})$ (m-1)d due to the exchange of angular momentum between the photoelectron and the Auger electron through Post-Collision-Interaction were found to dominate the spectrum. Comparison with valence photoelectron spectrum of Kr via direct photoionization confirms this new assignment. It was also found to be in accord with the quantum defect values obtained for the high Rydberg series $Kr^{+}4p^{4}({}^{1}S_{0}, {}^{1}D_{2}) ml$.

The conjugate component is the strongest among the four, instead of the $3d^{-1}_{5/2} \rightarrow 4p^{-2}({}^{1}S_{0})$ mp component, for all principal quantum numbers that have been identified. This is in conflict with assignments in previous investigations giving the mp assignment for the most conspicuous component lines. In previous PCI theories, such an exchange of angular momentum taking place with low photon energy excess had not been accounted for.

In the case of Xe $(N_5O_{2,3}O_{2,3})$ Auger electron spectrum originating from $4d^{-1}_{5/2}$ inner-shell hole created by photoionization close above the photoionization threshold, very rich Rydberg structures quite similar to the case of Kr were observed. However, after similar analysis, "regular" *mp* peaks instead of the conjugate *md* peaks were found to be dominant in contrast to the Kr case. Nevertheless, angle resolved Auger electron measurements for Xe demonstrate very direct manifestation of strong dynamic angular correlations, as shown on the figure below.



Our measurements were made at the soft x-ray beamline BL6U of the Ultraviolet Synchrotron Orbital Radiation Facility (UVSOR) at IMS in Okazaki. The UVSOR BL6U beamline, was equipped with an undulator and employed a variable-included-angle varied-line spacing plane-grating monochromator. The photon energy resolution $E/\Delta E$ was set at 10000 through the photon energy range of 90 - 100 eV. Kinetic energies of the emitted electrons were measured by a hemispherical electron energy analyzer (MBS-A1)

References

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