Energy Correlation among the Three Electrons Emitted in Triple Photoionization

The four-body Coulomb problem arising from the triple photoionization of atoms and molecules is a new research topic in the field of atomic and molecular physics. We have investigated the energy correlation among the three electrons emitted in valence triple photoionization of Ar, using a state-of-the-art multi-electron coincidence method. The differential cross-section for direct triple photoionization, to produce one slow electron with a kinetic energy of a few eV, was measured at about 150 eV above threshold, and is found to have a profile suggesting a "double shake-off" process.

The multi-body Coulomb interaction in multiple photoionization of atoms and molecules has become a arowing field of research in recent years. Double photoionization (DPI) of two-electron atomic systems leads to the three-body Coulomb problem, and is the simplest case of multiple photoionization. Due to the massive experimental and theoretical efforts exerted over the past two decades, our understanding of this prototype multiple photoionization process has improved very significantly [1]. However comparatively little attention has been paid so far to the four-body Coulomb problem which arises in the triple photoionization (TPI) of atoms. Deep insights into TPI dynamics can be gained from the energy correlation among the three outgoing electrons. While some theoretical studies on this topic have already been reported, to the best of our knowledge no experimental study has vet succeeded in observing energy correlation in TPI. This may be related to the fact that such studies require a highly sensitive multi-electron coincidence experiment, because of the extremely small TPI cross-sections.

At the undulator beamline BL-16A, we have succeeded in the first experimental observation of energy correlations among three valence electrons emitted in TPI [2]. The observations were achieved for the TPI of Ar using a state-of-the-art multi-electron coincidence method [3, 4]. Figure 1 shows the energy correlation

maps among the three electrons emitted at a photon energy of hv=234.7 eV (about 150 eV above the TPI threshold), for the formations of (a) $\text{Ar}^{3+}(3\text{p}^{3-2}\text{D})$ and (b) $\text{Ar}^{3+}(3\text{p}^{3-4}\text{S})$. Each map is deduced from the triple coincidence events which fulfil the restriction hv- $(E_1+E_2+E_3)=$ (binding energy of the corresponding Ar^{3+} level). Here E_1 , E_2 and E_3 denote the kinetic energies of the first (fastest), second (medium energy) and third (slowest) electrons to arrive at the detector.

It is expected that direct TPI should appear as smooth distributions in the maps of Fig. 1. because the available energy for the formation of the individual Ar³⁺ state is shared continuously by the three photoelectrons. However the maps in Fig. 1 are dominated by horizontal stripes which result from sequential TPI processes. While the most prominent horizontal stripe is visible in Fig. 1(b) at $E_{2}=2.2$ eV, similar, but weaker horizontal stripes can be identified at many other E_2 in both maps. The stripe at $E_3=2.2$ eV in Fig. 1(b) can be assigned to DPI into an Ar2+(3s-2) satellite state and its subsequent autoionization. The energy distribution of the two photoelectrons emitted in the initial DPI process has a continuous profile increasing gradually toward the minimum and maximum energies, as shown in Fig. 2(a). Such a U-shape profile is expected in direct DPI: the profile thus confirms that the Ar²⁺(3s⁻²) state is produced essentially by direct DPI.



Figure 1

Two-dimensional maps showing the energy correlations among the three electrons. The sums of the energies of the electrons are restricted to the ranges for the formation of (a) $Ar^{3+}(3p^{-3} D)$ and (b) $Ar^{-3+}(3p^{-3} S)$. Intensities are plotted on a common linear scale. The areas of $E_1 - E_2 < 14$ eV are blind due to the detection dead time arising from inseparable electron signals.



Figure 2

(a) Kinetic energy distribution of the two photoelectrons emitted in DPI into an Ar^{2*}(3s⁻³) satellite state of $3p^{-3}d^2$ configuration, which reflects the coincidence yields for the horizontal stripe at $E_3=2.2$ eV in Fig. 1(b). (b) Kinetic energy distributions of the two electrons emitted along with electrons with $E_3=1-2$ eV in TPI into Ar^{3*}(3p^{-3*}D). The gaps seen in the middle and at both extremes of the curves are due to the detection dead time arising from inseparable electron signals.

In Fig. 2(b) are plotted the energy distributions of the two faster electrons emitted in formation of Ar³⁺(3p⁻³ ²D). for events selected by restricting the slowest electron energy to $E_2=1-2$ eV. Because horizontal stripes are scarcely discernible in the selected E_2 range, we believe that most of the selected intensity can be attributed to direct TPI. The distribution again shows a U-shaped profile, but the profile is more enhanced around the maximum and minimum energies than the profile in Fig. 2(a). The observed distributions show that for the emission of a low energy electron, a highly unequal energy sharing is favored for the other two electrons. One possible interpretation of this is that the emission of a high energy photoelectron is accompanied by the emission of two slow electrons in a "double shake-off" process.

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