Probing the Mechanism of Simultaneous Two-Electron Emission from Atoms

We have studied simultaneous two-electron emission upon 2*p* core-electron excitation in Ar, using a state-of-the-art multi-electron coincidence method. The simultaneous two-electron emission effectively populates Rydberg-excited Ar^{2+} states, in which the excited electron remains a spectator of the direct double Auger decay of the core hole. This observation constitutes experimental evidence that the shake-off mechanism is not sufficient to model the direct double Auger decay of a 2*p* hole in Ar.

Understanding many-body Coulomb interaction is one of the unsolved fundamental problems in atomic and molecular physics. The many-body Coulomb problem arises in emission of two electrons from an atom or molecule after absorption of a single photon. Here, the two-electron emission may occur either as two successive steps or as a simultaneous process. In the stepwise process the many-body problem can be simplified by considering each step individually. Particular interest is attached to the simultaneous process as it requires direct correlation between the motions of the electrons. The mechanism generally invoked for simultaneous two-electron emission is "shake-off" (SO). In this purely quantum mechanical effect, the second electron ejection is the result of relaxation of the initially bound electron into an unbound state, following the sudden change of central potential on ejection of the first electron. In some theoretical frameworks, the introduction of the "knock-out" mechanism in which a primary-outgoing electron knocks out a second electron, has noticeably improved agreement of the calculations with experiments. However, so far no experiment has directly tested the inadequacy of the SO model.

In this study, we demonstrated that the inadequacy of the SO model in double Auger decay can be tested by observing simultaneous two-electron emissions from core-excited atoms. In practice, we investigated the simultaneous two-electron emission in resonant double Auger (RDA) decay from a $2p_{3/2}^{-1}4s$ state in Ar [1], using a state-of-the-art multi-electron coincidence method [2]. Figure 1 displays the energy correlation map for the two Auger electrons emitted from an Ar $2p_{3/2}^{-1}4s$ state. In this plot, coincidence counts associated with the formation of an individual Ar^{2+} state necessarily fall on a diagonal line defined by $E_f + E_s$ = (photon energy) - (binding energy of the Ar^{2+} state), where E_f and E_s are the energies of fast and slow Auger electrons, respectively. Along the diagonal lines corresponding to Ar^{2+} $3p^{-2}/3s^{-1}3p^{-1}$ formation, intense spots are observed in the energy range 180–200 eV for E_f. These spots result from cascade RDA processes, in which both electrons have discrete kinetic energies. On the other hand, the magnified area of the energy correlation map exhibits several diagonal stripes on which no clear spot structure due to the cascade RDA process is present. These stripes originate from direct RDA decay in which the two Auger electrons share the available energy continuously.

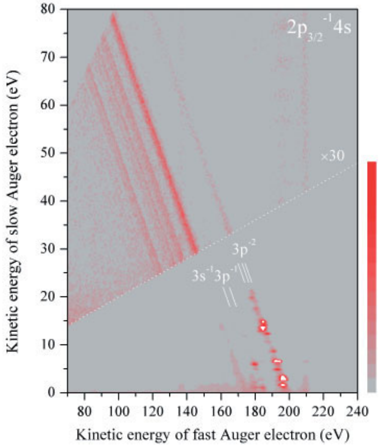

Figure 1
Energy correlation map for two Auger electrons emitted from the Ar $2p_{3/2}^{-1}4s$ state lying at a photon energy of 244.39 eV, where the intensities in the area of (slow electron energy) > (fast electron energy) × 0.2 are magnified by a factor of 30.

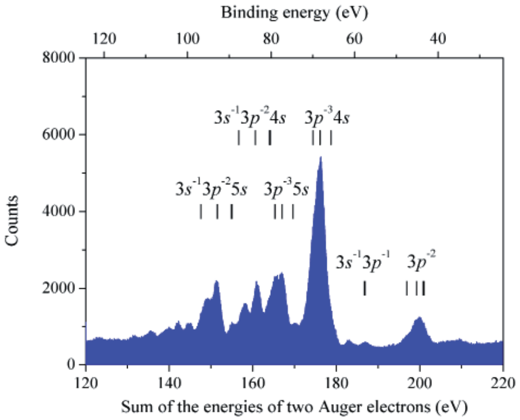

Figure 2
Spectrum displaying the Ar^{2+} states populated via the simultaneous two-electron emission from the Ar $2p_{3/2}^{-1}4s$ state, deduced from Fig. 1 as coincidence yields in the range of (slow electron energy) > (fast electron energy) × 0.2.

To focus attention on Ar^{2+} states populated via direct RDA decay, a histogram of $E_f + E_s$ has been obtained for coincidence yields in the area of $E_s > E_f \times 0.2$ in Fig. 1, thus effectively masking the contribution from the intense cascade RDA processes. The histogram obtained, which is plotted in Fig. 2, shows strong formation of high-lying Ar^{2+} states and only weak Ar^{2+} $3p^{-2}/3s^{-1}3p^{-1}$ state formation. The formation of these Rydberg Ar^{2+} states is most naturally attributed to noninvolvement of the initial Rydberg electron in the direct RDA process, that is, the Rydberg electron behaves as a spectator of two-electron ejection from the ion core.

Let us assume that the direct RDA decay is of pure SO origin. In this framework, the sudden change of the central potential on the primary valence electron ejection, due to single Auger decay, leads to the second valence electron ejection. However, the removal of screening by the primary-ejected electron should have more effect on the Rydberg electron than on another valence electron. In addition, the Rydberg electron is already closer to the electronic continuum than any valence electron. Therefore it is expected that the Rydberg electron will be easily removed, compared with a second valence electron, and the formation of Rydberg Ar^{2+} states will be unlikely, in striking contrast to the observation. The striking contrast demonstrates that the simultaneous two-electron emission observed cannot be fully attributed to SO.

In response to the failure of the pure SO mechanism, we consider electron correlations through the KO mechanism. Here, direct Coulomb interaction (inelastic collision) of the first Auger electron with another electron results in the ejection of a second electron. Interaction with another valence electron must be much more probable than interaction with the distant Rydberg electron. It follows that the simultaneous ejection of two valence electrons, leaving the Rydberg electron in place, is the dominant predicted result for the KO mechanism. This mechanism therefore explains the observed formation of Rydberg Ar^{2+} states; the present observations demonstrate the inadequacy of the pure SO model in double Auger decay.

REFERENCES

[1] Y. Hikosaka, P. Lablanquie, F. Penent, P. Selles, T. Kaneyasu, E. Shigemasa, J.H.D. Eland and K. Ito, *Phys. Rev. A* **80** (2009) 031404(R).
[2] K. Ito, F. Penent, Y. Hikosaka, E. Shigemasa, I.H. Suzuki, J.H.D. Eland and P. Lablanquie, *Rev. Sci. Instrum.* **80** (2009) 123101.

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