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Production and relaxation pathways of multiply excited states in slow highly charged ion-atom collisions

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We report triple-coincidence measurements of Auger electrons, scattered projectile, and target recoil ions in slow collisions of highly charged ions with many-electron atoms. Subpartial Auger-electron spectra corresponding to specific final projectile and recoil-ion charge states have been obtained by means of time-of-flight and position imaging techniques for the 60-keV $O^{7+}+Ar$ collision system. The spectra exhibit marked differences and provide insights into the production and relaxation pathways of multiply excited states populated during the collisions. [S1050-2947(99)51105-0]

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When a slow multiply charged projectile ion moving at a velocity below that of typical outer-shell atomic electrons collides with a many-electron neutral target, a large number of electrons can become active during the collision. Such collisions are dominated by the transfer of a number of target electrons to the projectile, resulting in the formation of projectile multiply excited states. Substantial progress has been made toward understanding one- and two-electron processes in slow ion-atom collisions during the past three decades (see, e.g., Ref. [1], and references therein). Although slow collisions involving more than two active electrons have also been investigated for over two decades as well, our understanding of multielectron processes is by no means comparable to that of one- and two-electron processes. The vast majority of experimental studies of multielectron processes involved measurements of cross sections for projectile charge-change and recoil-ion production, both in a singles' and in coincidence modes, and a limited number involved energy gain and visible photon spectroscopy (see, e.g., [2–5], and references therein). Moreover, since autoionization is a main decay mode of multiply excited states, Auger-electron spectroscopy has been employed in a singles' mode [6,7] to study such collisions. While such measurements have played a significant role in understanding two-electron processes (see, e.g., Ref. [8], and references therein), the situation is drastically different when many electrons are involved. For example, Benoit-Cattin *et al.* [6] obtained a singles' electron spectrum for the 70-keV $N^{7+}+Ar$ collision system. The analysis of the spectrum was rather difficult since it contained contributions from doubly, triply, quadruply, and quintuply excited states that rendered the interpretation nontrivial. During the last six years, however, Morgenstern and co-workers have made significant contributions [9–12] toward understanding Auger-electron spectra obtained in multiple-electron capture processes by means of the coincident detection of Auger electrons and target ions. They obtained partial Auger spectra corresponding to the different target ion charge states that are much more informative than singles' spectra. These spectra can provide further information if the final projectile charge state is also determined.

From a theoretical point of view, understanding multielectron processes is a twofold problem. First, the different mechanisms involved in the collision process that lead to the production of the multiply excited states must be recognized and described. Second, the radiative and nonradiative properties of the resulting multiply excited states must be known. Concerning the first problem, quantum-mechanical or semiclassical treatment of collisions involving more than two electrons is prohibitively difficult due to the large number of channels involved. Therefore, extended classical overbarrier (ECB) models have been developed [13,14] to account for multiple-electron capture processes. These models are limited to giving the capture state distribution on the projectile and possible simultaneous target excitation. There has been, until recently [15–17], a severe lack of theoretical work on the radiative and nonradiative properties of multiply excited states, due in part to the extremely large number of states that need to be taken into account, and in part to the lack of experimental data to which the calculations can be directly compared. Therefore, relaxation schemes [6,12] based on simple arguments, such as autoionization to the nearest continuum limits and minimum electron rearrangement (two-electron transitions), have been invoked. This Rapid Communication reports triple-coincidence measurements of Auger electrons, scattered projectile, and target recoil ions in slow multiply charged ion-atom collisions. The measurements provide insights into the relaxation pathways of multiply excited states populated in such collisions.

The 60-keV O^{7+}-ion beam was provided by the University of Nevada, Reno, 14-GHz electron cyclotron resonance (ECR) ion source, and guided to the collision chamber where it crossed a supersonic Ar gas jet at 90°. After the collisions, the target recoil ions were extracted transversely to the ion beam by a uniform electric field, traveled through a time-of-flight (TOF) spectrometer, and were then detected by a microchannel plate detector (MCP). The charge-changed projectiles were charge analyzed downstream from the collision chamber by a parallel-plate electrostatic deflector and detected by a two-dimensional (2D) position-sensitive MCP.

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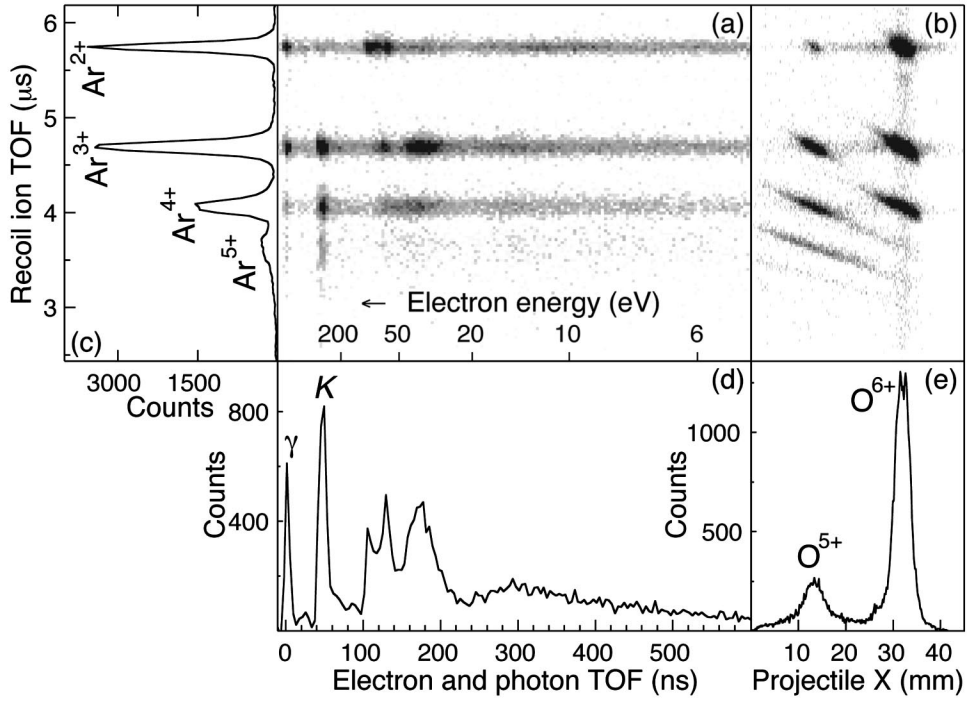

FIG. 1. (a) Coincidences between recoil ions and Auger electrons or photons. (b) Coincidences between projectile and recoil ions. (c) Recoil-ion TOF spectrum. (d) Singles' Auger-electron/photon spectrum. The labels K and γ indicate K-Auger electrons and photons, respectively. (e) Final projectile charge-state distribution.

Auger electrons ejected at 90° relative to the incident ion beam traveled through a TOF electron spectrometer, located opposite to the recoil-ion spectrometer, and were detected by another MCP. The impact positions on the projectile detector provided the final projectile charge states, while coincident TOF measurements between projectile and recoil ions provided the recoil-ion charge states and between projectiles and electrons provided the TOF of the electrons. The major advantage of the TOF electron spectrometer is that it simultaneously accepts electrons of all energies, so that no scanning is needed. If high resolution is desired, the electrons may be decelerated using a grid assembly. The true triple-coincidence rate was about 0.16 Hz for a primary ion-beam current of 2 pA. The data presented in this paper were collected in 48 h. More experimental details will be given in forthcoming articles.

Coincidences between recoil ions and Auger electrons or photons are represented by the scatter plot of Fig. 1(a). Since the electron detector views the interaction region, photons with energy higher than 12 eV that are emitted toward the detector will be detected. Figure 1(b) represents coincidences between projectile and recoil ions. Projections onto the appropriate axes provide the recoil-ion TOF spectrum [Fig. 1(c)], the equivalent of a singles' Auger-electron spectrum [Fig. 1(d)], and the final projectile charge state distribution [Fig. 1(e)]. It is evident from Fig. 1(a) that the singles' Auger-electron spectrum resulted from processes involving from two to five active electrons, and the interpretation of this spectrum would be a formidable task. However, partial spectra corresponding to the different recoil-ion charge states can be obtained from the current measurements and are shown in Fig. 2. Clearly, the partial spectra exhibit marked differences. In particular, K-Auger electrons are nearly absent in the Ar^{2+} spectrum. This indicates that high-lying doubly excited states are populated that preferentially autoionize to the nearest continuum limits, thus rendering the K-shell vacancy passive. Examination of Fig. 1(b) shows that Ar^{q+} $(q=3-5)$ recoil ions are found in coincidence with

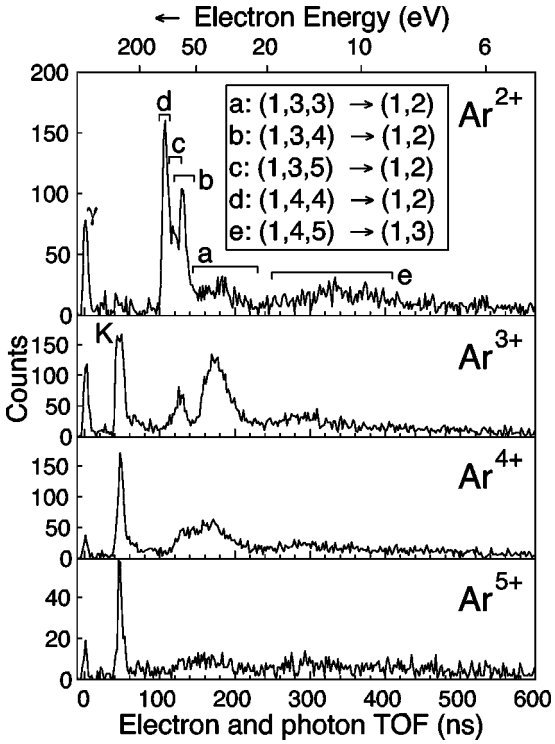

FIG. 2. Partial Auger-electron/photon spectra corresponding to the different recoil-ion charge states.

projectile ions that changed their charge states by one or two units. This implies that the corresponding partial spectra can be further reduced to subpartial spectra associated with the different final projectile charge states.

For double-electron capture, the ECB model by Niehaus [14] predicts that the two most weakly bound target electrons will be captured by the projectile with a total binding energy of 71 eV. The reaction window for this process overlaps the configurations (1,4,4) and (1,4,5), which therefore should be dominantly populated. Indeed, this is the case as seen in the partial spectrum for Ar^{2+}. The Auger line identification was carried out using the Hartree-Fock atomic structure code by Cowan [18]. On the other hand, the observed $(1,3,n)$, $n=3-5$, configurations can be accounted for within the ECB model only if the assumption of double-electron capture accompanied by target excitation is invoked. Such an assumption has been invoked by de Nijs *et al.* [11] in the case of the $N^{7+}+Ar$ collision system; however, no direct experimental evidence in support of this assumption has been reported.

We now turn our attention to the next most challenging problem of three active electrons. The ECB model predicts that the three most weakly bound target electrons will be captured with a total binding energy of 120.9 eV. The reaction window overlaps the configurations (1,3,4,4), (1,3,4,5), and (1,4,4,4). The model also predicts a population of the configurations (1,3,3,4), and (1,3,3,5) via four-electron processes where the target may be left in an excited state. Judging from the double-electron capture population, however, and the fact that with an increasing number of captured electrons the population shifts toward lower-lying levels on the projectile, we believe that the (1,4,4,4) is an unlikely configuration that will therefore be disregarded. The subpartial Auger spectra corresponding to the O^{6+} and O^{5+} final projectile charge states are shown in Fig. 3. A striking feature is that *K*-Auger electrons are nearly absent in the (Ar^{3+},O^{5+}) channel. Instead, a strong photon peak is observed, signalling the important role played by radiative stabilization in retaining two electrons, and the fact that *K*-Auger electrons are not emitted in the first autoionization step. Another difference is the near absence in the (Ar^{3+},O^{5+}) channel of electrons resulting from the autoionization of the (1,3,4,4) and (1,3,4,5) configurations to the (1,3,3) configurations, which in turn autoionize to the (1,2) continuum limits, thus leading to the retention of one electron only.

Auger electrons resulting from the autoionization of the (1,3,3,4) and (1,3,3,5) configurations to the (1,2,3), (1,2,4), and (1,2,5) configurations are common to both channels. Whether an event results in the combination (Ar^{3+},O^{6+}) or the combination (Ar^{3+},O^{5+}) is then determined by the competition between the radiative and autoionization decay modes of the doubly excited states (see, e.g., Ref. [19], and references therein). The (Ar^{3+},O^{5+}) spectrum is therefore a pure spectrum involving one autoionization step only, while the (Ar^{3+},O^{6+}) spectrum is still a composite one resulting from the autoionization of the triply and subsequent doubly excited states. We did not include in the labeling possible lines resulting from the autoionization of the (1,3,4,4) and (1,3,4,5) configurations to doubly excited configurations other than (1,3,3). This is because all states belonging to these configurations are energetically allowed to autoionize

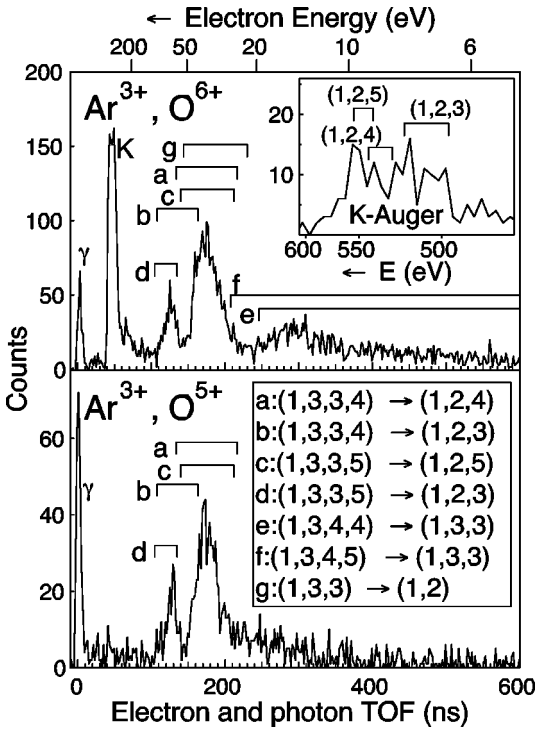

FIG. 3. Subpartial Auger/photon spectra corresponding to triply ionized recoil ions and projectiles that retained one (top spectrum) or two (bottom spectrum) electrons. Inset: a moderate resolution *K*-Auger-electron spectrum in coincidence with the combination (Ar^{3+},O^{6+}).

to the (1,3,3) continuum limits, whereas the other $(1,2,n),n=3-5$, limits are substantially distant in energy. While autoionization of the $(1,2,n),n=3-5$, configurations involves the emission of *K*-Auger electrons that are well separated in energy from the autoionization electrons of the triply excited configurations, the (1,3,3) configurations autoionize with the emission of electrons that overlap those from the $(1,3,3,n)$ configurations, thus further complicating the analysis of the (Ar^{3+},O^{6+}) spectrum.

Information on the properties of the $(1,3,3,n),n=4,5$, configurations can now be extracted. We start by examining the (Ar^{3+},O^{5+}) spectrum. It is clear that autoionization of these configurations to the associated $(1,2,n),n=4,5$, continuum limits is more probable than to the (1,2,3) limits. This spectrum should, in principle, provide branching ratios for autoionization to the different continuum limits. One complication, however, arises from the fact that these configurations give rise to electrons that overlap in energy, thus rendering the relative initial populations impossible to obtain from this spectrum. Examination of the inset of Fig. 3, which is a moderate resolution *K*-Auger-electron spectrum obtained in coincidence with the combination (Ar^{3+},O^{6+}), reveals that both the (1,2,4) and (1,2,5) configurations have been populated. However, due to the overlap in the *K*-Auger-electron energies and the limited statistical precision, we refrain from attempting to obtain the relative initial populations using simple arguments such as the n^{-3} scaling law. Instead, we will give experimental branching ratios for the unknown combination of the $(1,3,3,n),n=4,5$, configurations. By taking the ratio of the Auger line intensities under the labels ''(a) and (c)'' and ''(b) and (d),'' in the

(Ar^{3+},O^{5+}) spectrum, to the total intensity under ''(a), (b), (c), and (d),'' we obtain autoionization branching ratios of ≈ 0.8 to the $(1,2,n)$ continuum limits, and ≈ 0.2 to the $(1,2,3)$ limits. These branching ratios are weighted according to the unknown initial populations. Since the energy gap between the $(1,2,5)$ and $(1,2,3)$ continuum limits is larger than that between the $(1,2,4)$ and $(1,2,3)$ limits, we can conclude that the branching ratio for autoionization of the $(1,3,3,5)$ configurations to the $(1,2,5)$ limits is larger than 0.8, while that of the $(1,3,3,4)$ to the $(1,2,4)$ limits is less than, but probably not far from, 0.8. It should be noted here that while the absence of *K*-Auger electrons in the (Ar^{3+},O^{5+}) spectrum is consistent with the widely used assumption of the dominance of autoionization to the nearest continuum limits, the data show clear deviations from this assumption when several other continuum limits are available.

The subpartial spectra should also contain information on the branching ratios for radiative and autoionizing decays of the doubly excited states. Taking the ratio of the Auger line intensities under the labels ''(b) and (d)'' in the (Ar^{3+},O^{5+}) spectrum to the total intensity under the same labels in both spectra, one obtains a fluorescence yield of ≈ 0.3 for the $(1,2,3)$ doubly excited configurations. The fluorescence yields for the $(1,2,4)$ and $(1,2,5)$ configurations are larger than 0.3. However, more accurate yields are harder to obtain due to the strong overlap in electron energies between processes ''(a), (c), and (g).'' Discussion of four- and five-electron processes is reserved for a more extensive report.

In summary, we have reported triple-coincidence measurements of Auger electrons, scattered projectile, and target recoil ions in slow highly charged ion-atom collisions. The subpartial Auger spectra provide an opportunity to better understand the production and the subsequent relaxation of multiply excited states. The spectra reveal clear deviations from the widely adopted criterion of autoionization to the nearest continuum limits, and provide information on branching ratios for autoionization to the different continuum limits, and on fluorescence yields for some intermediate states. The ECB model predictions are reasonably well supported by the measurements; however, the prediction of target excitation is yet to be experimentally confirmed. To further understand multielectron processes in such collisions, a number of similar measurements are planned using bare ions, for which the Auger lines will be narrower and easier to separate, and theoretical branching ratios are easier to obtain for purposes of comparison.

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