Investigation of Multi-Electron Processes in 60 keV O^{6+} + Ar Collisions Using a Triple Coincidence Technique

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Multiple electron capture processes in 60 keV O^{6+} + Ar collision system have been investigated by means of time-of-flight triple-coincidence measurements of Auger electrons, scattered projectile and target ions. This technique is particularly suited for the investigation of electron capture processes involving capture of three or more electrons. From the measured sub-partial Auger spectra, it can be seen that double electron capture dominantly populates the configurations (3,n) with n=3-5. Triple electron capture is found to dominantly populate the configurations (3,3,3) and (3,3,4), while quadruple electron capture populates the (3,3,3,3) configuration.

INTRODUCTION

Studies of multi-electron capture processes in highly charged ion-atom collisions have attracted considerable interest during the last two decades. Significant progress has been made in investigating single and double electron capture processes using the methods of translational energy gain spectroscopy (1-3), recoil ion momentum spectroscopy (4,5), photon spectroscopy (6,7) and Auger electron spectroscopy (8,9). On the contrary, little is known about multi-electron processes. Indeed, from a theoretical point of view, understanding multi-electron processes is a two-fold problem. Firstly, the different mechanisms involved in the collision process that lead to the production of multiply excited states must be recognized and described. Secondly, the radiative and non-radiative properties of the resulting multiply excited states must be known. Moreover, quantum mechanical or semiclassical treatments of collisions involving more than two electrons are prohibitively difficult due to the large number of channels involved.

Although the classical over-barrier model has been extended (10) to account for multiple electron capture, the extended model (ECB) does not take into account the electronic correlation which has been found to play an important role in many cases for double electron capture (11,12). In fact, the ECB model cannot accommodate many of the possible reaction channels such as the simultaneous capture of two or more electrons, and the population of highly Rydberg states (12). This model stops at giving the final capture state distribution on the projectile and possible target excitation. In order to further account for the final collision products, relaxation schemes for the multiply excited states must be invoked.

During the last six years, the Groningen group has made significant contributions (13,14) toward understanding Auger electron spectra obtained in multiple electron capture processes by means of the coincident detection of Auger electrons and target ions. They demonstrated that many of the Auger lines attributed to doubly excited states must have been derived from multiply excited projectile states through successive autoionization processes.

It is important to note that Auger electron spectroscopy in singles mode does not give adequate information on multiple electron capture processes. This is due to the fact that the spectra would contain contributions from doubly, triply, quadruply and possibly quintuply excited states that make the analysis of the spectra rather difficult. However, the partial Auger spectra corresponding to the different target ion charge states are much easier to analyze than singles spectra. One should also realize that these spectra can be further simplified if the final projectile charge state is also determined.

In the present paper we report triple-coincidence measurements of Auger electrons, scattered projectile, and target recoil ions in 60 keV O^{6+} + Ar collisions. Such measurements provide sub-partial Auger spectra corresponding to specific final projectile and recoil ion charge states.

EXPERIMENTAL SETUP

The measurements were performed at the multicharged ion research facility at the University of Nevada, Reno (UNR). The 60 keV O^{6+} ion beam was provided by the UNR 14 GHz electron cyclotron resonance (ECR) ion source, and guided to the collision chamber where it crossed a supersonic Ar gas jet at 90°. The jet furnished a well-localized target with an effective target density of about 0.1 m Torr. After the collision, the target recoil ions were

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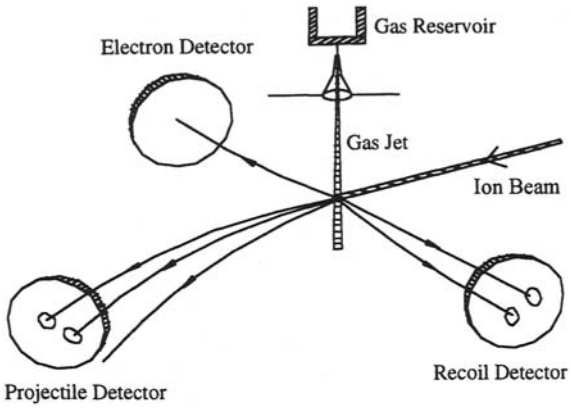

FIGURE 1: A schematic of the experimental setup.

extracted transversely to the ion beam direction by a uniform electric field (≈ 10 V/cm), traveled through a time-of- flight (TOF) spectrometer and were then detected by a microchannel plate detector. The charge exchanged projectiles were charge analyzed downstream from the collision chamber by a parallel-plate electrostatic deflector and detected by a two-dimensional position sensitive microchannel plate detector (2D-PSD) (Fig. 1).

Coincident TOF measurements provided the recoil ion charge states while the impact positions on the projectile 2D-PSD provided the final projectile charge states. Auger electrons ejected at 90° relative to the incident beam traveled through a TOF electron spectrometer, located opposite to the recoil ion TOF spectrometer with its entrance aperture close to the interaction region, and were detected by another microchannel plate detector. TOF coincidence measurements of the electrons and the charge exchanged projectiles provided the TOF of the electrons (Fig. 1).

The collision chamber was differentially pumped such that high ion beam purity was maintained prior to entering the chamber and after exiting it and entering the electrostatic analyzer. Typical pressures of about 5×10^{-9} and 1×10^{-8} Torr were maintained up- and downstream of the collision chamber, respectively, while the collision chamber residual pressure was about 1.2×10^{-7} Torr during the experiment. Double collisions were estimated to be less than 0.5 %. A fast timing signal derived from the electron detector was used to start a time-to-digital converter (TDC) which was stopped by a signal from the projectile detector. Another TDC was started by a fast signal from the recoil detector and stopped by the projectile signal. Analog-to-digital (ADC) converters read the position signals from the projectile detector. The time and position information were then read by a computer and stored in list mode for further processing. The true triple-coincidence rate was about 0.65 Hz for a primary ion beam current of 10 pA.

RESULTS AND DISCUSSION

Fig. 2(a) is a scatter plot representing coincidences between recoil ions and Auger electrons or photons. Since the electron detector views the interaction region, any photons with energy higher than about 12 eV that are emitted toward the detector will be detected. The projection of Fig. 2(a) onto the vertical axis gives the recoil ion TOF spectrum shown in Fig. 2(c). The equivalent of a singles Auger electron spectrum results from the projection of Fig. 2(a) onto the horizontal axis as shown in Fig. 2(b).

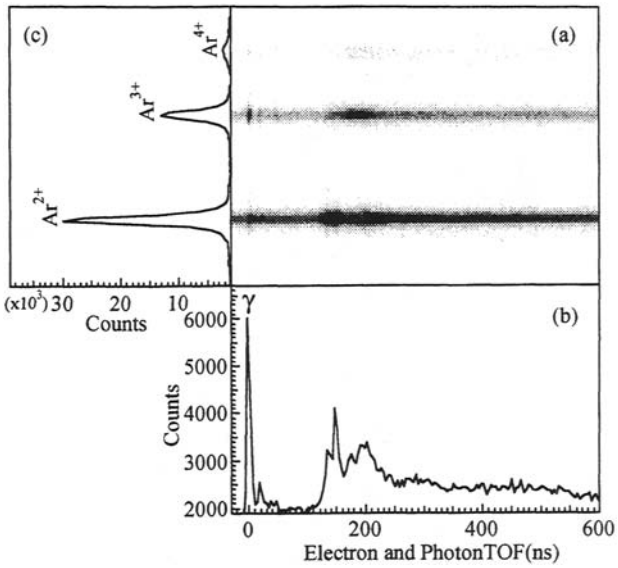

FIGURE 2: (a) A scatter plot representing coincidences between recoil ions and Auger electrons or photons, (b) singles Auger electron/ photon spectrum (the label γ indicates photons), and (c) recoil ion TOF spectrum.

It is evident from Fig. 2(a) that the singles Auger electron spectrum resulted from processes involving loss of two to four electrons by Ar, thus rendering the interpretation of the singles electron spectrum a difficult task. However, partial Auger electron spectra specific to each recoil ion charge state can make the analysis much easier. Such spectra are obtained by placing 2D-windows corresponding to the different recoil ion TOF in Fig. 2(a), and projecting the events within each window onto the horizontal axis. Partial spectra so obtained are shown in Fig. 3. Clearly, the partial spectra present noticeable differences. Fig. 3(a) shows the partial Auger electron spectrum corresponding to double electron capture. The Auger line identification is carried out using the Hartree-Fock code by Cowan (15). According to the ECB model, the reaction window associated with capturing the two outermost electrons overlaps the configurations with principal quantum numbers (3,4) and (3,5) and therefore should be dominantly populated. Their subsequent autoionization gives rise to *L*-Auger lines. Indeed, we observe appreciable populations of these two configurations. There is, however, a substantial population of the (3,3) configuration, which can be accounted for within the framework of the ECB model only if the assumption of double electron capture accompanied by target excitation is invoked. Such a process is a three-electron process whereby the three outermost electrons are assumed to have been molecularized. The projectile then

captures the third and either the first or the second electrons, while the residual target ion recaptures the other

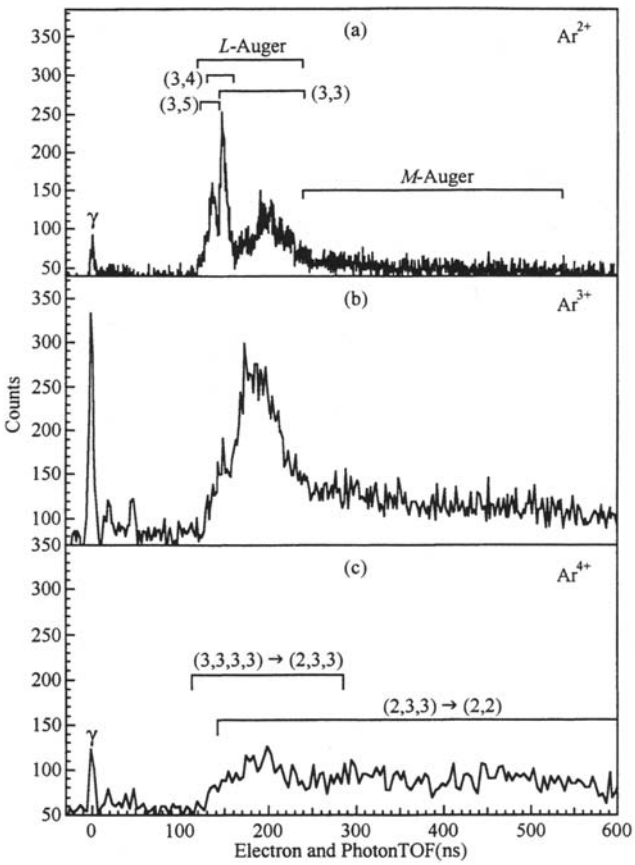

FIGURE 3: Partial Auger electron/photon spectra corresponding to the different recoil ion charge states.

electron into an excited level. de Nijs *et al.* (14) argued in favor of such a process in the case of the 60 keV C^{6+} + Ar collision system. While this assumption explains the presence of Auger lines derived from the (3,3) configuration, a comparison of the ECB predicted relative intensities of the (3,3) configuration to the (3,4) and (3,5) configurations with the experimentally determined ones shows that the ECB model underestimates the importance of the (3,3) configuration by about a factor of four.

In addition to the *L*-Auger lines, we also observe weak *M*-Auger lines in Fig. 3(a), an indication that the configurations (4,n) with $n \geq 5$ are weakly populated. In general, our findings concerning double electron capture processes for this collision system are similar to those of de Nijs *et al.* (14) for the C^{6+} + Ar collision system. It seems that the electron capture process is dominated by the incoming projectile charge state in both systems, while the projectile core effect is negligible; at least in the case of O^{6+}.

The partial Auger spectrum corresponding to triple electron capture is shown in Fig. 3(b). Inspection of Fig. 4, however, shows that the triply ionized recoil ions are found in coincidence with projectiles that changed their charge state by one or two units. Hence, the partial Auger spectrum is still a composite one, and therefore can be further reduced according to the final projectile charge states. This

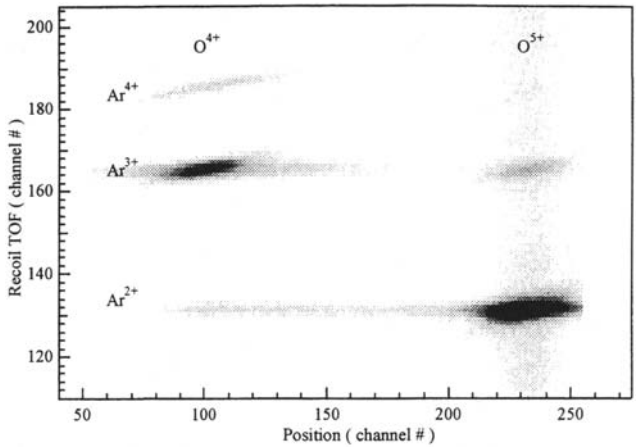

FIGURE 4: A scatter plot representing coincidences between scattered projectiles and recoil ions.

reduction results in the sub-partial Auger spectra shown in Fig. 5. These sub-partial Auger spectra provide further insights into the population and relaxation pathways of triply excited states. It is important to note here that the small concentration of events in Fig. 4 corresponding to the combination (Ar^{2+}, O^{4+}) represents true double electron capture events, where both electrons stabilize radiatively, and are then found in coincidence with photons only.

The ECB model predicts a reaction window for the capture of the three outermost electrons that overlaps the configurations (3,3,3), (3,3,4), and (3,4,4). The autoionization of the first two configurations results in singly excited daughter configurations, thus leading to the retention of two electrons by the projectile. Auger lines derived from these configurations should then be found in the sub-partial spectrum corresponding to the (Ar^{3+},O^{4+}) combination. Substantial population of these configurations can indeed be inferred from Fig. 5(a). The quasi-continuum electron distribution beyond about 260 ns can be accounted for using the ECB model only if the triple electron capture is assumed to be accompanied by target excitation. The capture may then proceed into the configurations (2,3,3) and (2,3,4) resulting in the observed distribution.

On the other hand, the configuration (3,4,4) is expected to predominantly autoionize to the (3,3) doubly excited configuration, which in turn autoionizes, resulting in the retention of one electron only by the projectile. Indeed, the sub-partial spectrum, shown in Fig. 5(b), corresponding to the (Ar^{3+},O^{5+}) combination exhibits Auger lines resulting from the two autoionization steps. Worthy of particular notice is the appearance of a *K*-Auger line in the (Ar^{3+},O^{5+}) sub-partial spectrum. This is attributed to the presence of a small metastable (1s2s) component in the O^{6+} ion beam. One may argue that the relative ratio of the (Ar^{3+},O^{5+}) to the (Ar^{3+},O^{4+}) events is enhanced due to the metastable component, and that the Auger lines in the (Ar^{3+},O^{5+}) channel are not necessarily derived from the initial

population of the (3,4,4) configuration. However, examination of Fig. 3(b) shows that *K*-Auger electrons constitute only about 1.5% of the total electrons detected in coincidence with Ar^{3+}, whereas a separate measurement of the recoil ion charge state fractions in coincidence with the final projectile charge states yielded an (Ar^{3+},O^{5+}) fraction of 25% of the total events leading to the production of triply ionized recoils. A metastable component of a few percent can not account for this fraction. In fact, even assuming a 5% metastable component, we still infer about a 20% fractional population for the (3,4,4) configuration. This calls into question the conclusion by Nakamura *et al.* (16), who also studied the 60 keV O^{6+} + Ar collision system using high resolution Auger electron spectroscopy in singles mode, that the (3,3) doubly excited states are not affected by Auger cascades from triply excited states.

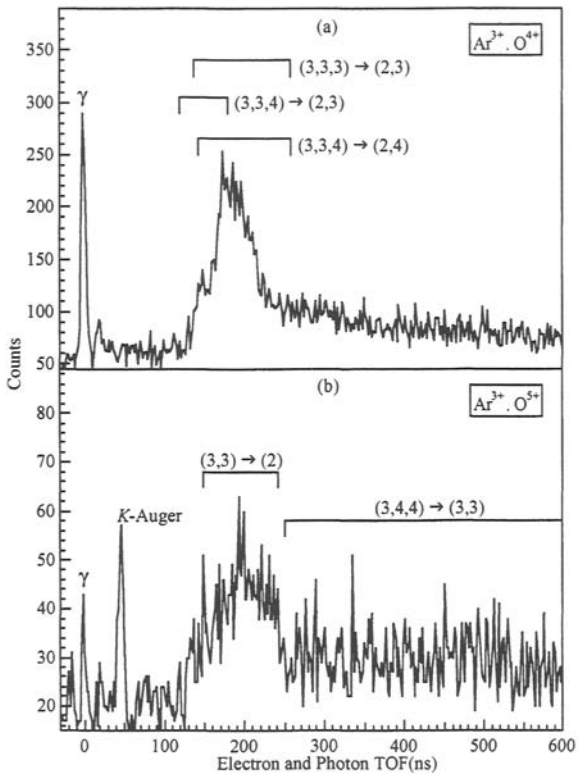

FIGURE 5: Sub-partial Auger electron spectra corresponding to triply ionized recoil ions and projectiles that retained (a) two electrons and (b) one electron.

Quadruple electron capture is predicted by the ECB model to proceed into the (2,3,3,3) and the (3,3,3,3) configurations. Disregarding the metastable ion beam component, the former configuration autoionizes once leading to the retention of three electrons by the projectile. We did not attempt to observe such events. The latter configuration autoionizes to the (2,3,3) continuum limits. The resulting configurations then autoionize to the (2,2) limits, resulting in the retention of two electrons by the projectile. Fig. 4 shows that Ar^{4+} recoil ions are found in coincidence with O^{4+} only. Furthermore, the partial Auger spectrum corresponding to quadruply ionized recoils, shown in Fig. 3(c), does indeed contain Auger lines consistent with the initial population of the (3,3,3,3) configuration .

CONCLUSIONS

We have investigated multi-electron processes in 60 keV O^{6+} + Ar collision system by means of triple-coincidence measurements of Auger electrons, scattered projectile and recoil ions. The measurements provided sub-partial Auger spectra corresponding to specific final projectile and recoil ion charge states. Such spectra provide better understanding of multi-electron processes. The experimental findings were compared with the predictions of the ECB model. While the presence of a large fraction of the observed Auger lines is well accounted for by the model, the predictions concerning the relative intensities are not in good agreement with the experiment.

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