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Double and Single Electron Capture
in Slow Collisions of Ar$^{9,8+}$ Ions with He Atoms

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Abstract

The technique of zero degree Auger electron spectroscopy has been used to study Na-like Ar\(^{7+}\) excited states populated in double and single capture collisions by 90 keV Ar\(^{9+}\) and 80 keV Ar\(^{8+}\) projectiles from He target atoms. Metastable Ne-like Ar\(^{8+}\) 2p\(^{5}\)3s\(^{-}3\)P\(_{0,2}\) states play an important role in populating several Ar\(^{7+}\) final states. Careful He target pressure dependence studies were performed to establish the excitation/capture mechanism for the Na-like core excited levels. Highly non-equivalent 2p\(^{5}\)3lnl' configurations, with n values peaking at n=8 but extending to near the series limits, were produced in double electron capture onto Ar\(^{9+}\) projectiles. The spectra have been identified and compared with calculated (MCDF) Auger energies and previous measurements. The Auger decay of the Ar\(^{7+}\) 2p\(^{5}\)s3p \(^{4}D_{7/2}\) metastable level has been observed and the energy of the line from the Breit induced decay has been measured. Allowed doublet lines from the 2p\(^{5}\)3s\(^{2}\) \(^{2}P_{1/2,3/2}\) states were also measured. The experimental values are in strong agreement with the results of the MCDF calculations.
Introduction

Electron capture into low velocity, highly charged projectiles from collisions with target atoms has been extensively studied, (see for example ref. 1). The semi-classical over-barrier models\(^2\) describe many aspects of the observed behavior (e.g. predict final quantum states) especially when applied to single capture from simple targets such as He. The process of double electron capture in a single collision is the subject of a great deal of current interest, both theoretically\(^3\) and experimentally\(^4,5,6\). This interest derives, in part, from the existence of competing processes; namely: i) sequential capture (during a single collision) of two electrons, one at each of two avoided crossings of the relevant molecular orbital energy levels, and ii) the process of correlated two electron capture at a single avoided crossing. A number of different channels appear to be open for the two electron correlated capture process, however, it is clear that dynamical or scattering correlation is important in such ion-atom collisions. Much of the experimental data supporting the concept of correlated double capture have been obtained using He-like projectiles. In contrast, this work reports experimental studies with F-like (9 electron) \(\text{Ar}^{9+}\) ions which provide evidence of a double capture processes similar to those observed with He-like projectiles (e.g. \(\text{O}^{6+}\)).

Interest in capture processes arises from the pure physics of many body interactions, and from the need for cross-sections for fusion plasmas. The ion-energies used here (80-90 keV) are higher than the ion temperatures (12 - 20 keV) available in modern Tokamak devices, however, the capture cross-sections in this low energy regime are nearly velocity independent, at least for one electron transfer\(^7\). This work is also related to short wavelength laser research; X-ray laser action using the core excited system
of Na I has been recently described by Harris. From a pure atomic structure point of view, very little data exist for the core-excited states of Na-like ions. The exceptions being Na I, and Mg II, both observed via the UV photon decay channels, and X-ray spectra obtained for Ti XII and Fe XVI. Recently the UV transition has been studied along the Na I sequence, and Auger electron emission following excitation of MeV Ar\textsuperscript{7+} in collisions with He has been reported. In this work, core-excited states of Na-like Ar are populated by two electron capture onto Ar\textsuperscript{9+} and one electron capture onto Ar\textsuperscript{8+} at much lower impact energies. Studies of the optical spectra of Na-like Ar\textsuperscript{7+} produced by electron capture are also being undertaken by Druetta et al.

**Experimental Method**

These experiments were performed using ion beams produced by the ECR (Electron Cyclotron Resonance) ion source at the Lawrence Berkeley Laboratory. A general description of the atomic physics facilities at the LBL ECR source has been recently given by Clark. The experimental arrangement is briefly described here, but is given in more detail in ref 20. A schematic diagram of the apparatus is shown in figure 1. The method uses the technique of zero-degree (forward direction) electron spectroscopy, incorporating two electron spectrometers in tandem. The first small 45 degree analyzer serves primarily to direct the forward emitted electrons into the high resolution, hemispheric analyzer, which provides an energy resolution of about 0.2%. The two sets of 4-jaw collimating slits are used to define the beam and insure that it does not strike any surfaces on the gas cell or 45 degree analyzer, this reduces background electron rates to minimal (often negligible) levels. After charge analysis and collimation, Ar\textsuperscript{8+} or
Ar⁹⁺ beam currents were typically about 50 nA as measured with the Faraday cup located behind the 45 degree analyzer. A beam current integrator connected to this cup is used to normalize the dwell time in any channel of a spectrum sweep to the same amount of beam charge. Typical background pressure in the target chamber is about 3 \times 10^{-7} \text{Torr} and gas cell pressures ranged from 0.1 to 1.5 mTorr; pressure in the beam transport lines were usually about 5 \times 10^{-8} \text{Torr}. Spectra were obtained over the energy range 0-400 eV in the projectile frame, for both Ar⁸⁺ and Ar⁹⁺ on He target atoms. Pressure in the target cell was monitored by a capacitance manometer and regulated by an automatic leak valve.

**Results and Discussion**

\[ \text{Ar}^{8+} + \text{He} \]

\text{Ar}^{8+} is a Ne-like ion with a 2p⁶ ¹S₀ ground state. For both single and double electron capture there is little possibility of producing states with Auger decay channels. However, the Ne-like system has two long lived metastable levels, namely 2p⁵3s ³P₀ and ³P₂, situated about 250 eV above the ground state. Either one or two electron capture onto these metastable core levels will produce states which autoionize. The two electron capture process will lead to core-excited levels of the Mg-like ion (Ar⁶⁺). Lines originating from the decay of such states fall outside the energy range studied, or are too weak to interfere with the analysis presented here. Fig. 2a shows a spectrum produced when Ar⁸⁺ collides with He. The measured electron spectra have been transformed into the center of mass system of the projectile. All of the lines in fig. 2a can be attributed to the Auger decay of core-excited Na-like levels, and show a linear dependence on gas pressure. This spectrum can be compared to the similar work of Bordinave-Montesquieu et
al.\textsuperscript{18}, where it was shown, using the theoretical data of Folkmann et al.\textsuperscript{19}, that the most probable principal quantum number populated by single electron capture (at this energy) is \( n = 4 \). This is in agreement with the predictions of the semi-classical over-barrier model for single capture.

\textbf{Ar}^9^+ + He

\( \text{Ar}^9^+ \) is F-like with a \( 2p^5 \, {}^2P \) ground configuration. Single electron capture will, at these energies, produce a Ne-like singly excited system and consequently no Auger electrons. Two electron capture can produce Na-like core-excited states. These states, in general, decay via three modes: i) by UV photon emission (to a lower lying core excited level), ii) by X-ray photon emission and iii) by Auger decay. The Z scaling of the probabilities for these individual channels is discussed in (11). The configurations are of the form \( 2p^5nln' \, l' \, {}^2L \) and \( {}^4L \). Fig. 2b shows a spectrum from \( \text{Ar}^9^+ \) following collision with He. It can be seen by comparing figs. 2a and 2b that the spectra are very similar up to energies near 200 eV. The differences in the spectra above 200 eV suggested that careful pressure dependence studies for the \( \text{Ar}^9^+ + \text{He} \) case be carried out and spectra at pressures ranging from 0.1 to 1.5 mTorr were recorded. The results of plotting integrated line intensities vs. pressure showed that lines below 200 eV were populated in two separate collisions. Above 200 eV the lines appeared to come from levels populated by single collision, double capture events. This is demonstrated in a more immediate way in fig. 3a which was constructed by subtracting two spectra, such as shown in Fig. 3b, taken at different pressures, after linear scaling by the pressure ratio. Lines from single collision events should not appear in such a plot, and one notes the disappearance of the structures above 200 eV.
The probable explanation of this pressure behavior, and the similarity of the spectra from $\text{Ar}^{8+}$ and $\text{Ar}^{9+}$ projectiles below 200 eV electron energy, is the fact that most of the lines below 200 eV can be obtained from capture to the metastable $2p^53s^3P_{0,2}$ levels. For $\text{Ar}^{8+}$ projectiles these metastable states are present in the beam and the doubly excited Na-like states can be populated by single capture. For $\text{Ar}^{9+}$ projectiles the metastable core is formed by one single capture with a second single capture yielding the Na-like Auger states. Thus one observes a quadratic pressure dependence for these lines, and they survive the linearly scaled subtraction process. Lines with energies above 200 eV come from levels such as $2p^53snl$ where $n$ is greater than 4. Such levels are not populated by single electron capture to the metastables. For a more complete understanding of the spectrum we have used MCDF calculations for the $2p^53l3l'$ and $2p^54l4l'$ configurations. The published values for the $2p^53snl^2L'$ levels have also been used. Fig. 4 shows the possible Auger decay channels available for these configurations and the energies associated with them. The energy data are also summarized in fig. 2b. Comparing the possible Auger lines with the $\text{Ar}^{9+} + \text{He}$ spectrum shows that the single collision double electron capture appears to preferentially populate configurations with $2p^53lnl'$. The $n$ value showing the highest cross section is around $n = 8$, although the distribution is very wide and appears to show population to states near the $2p^53s$ limit (250 eV). The mechanism for the production of these states may be similar to the correlated capture processes described by Stolterfoht et al and Winter et al which produced LLX Coster-Kronig transition lines.

The Auger line labeled $\beta$ in Figures 2 and 3 is of some special interest because it comes from the decay of the $2p^53s3p^4D_J$ levels, one of which, $4D_{7/2}$, is metastable.
This state is forbidden to decay by Coulomb auto-ionization or electric dipole (E1) photon emission because it is essentially a pure state of total spin 3/2 (quartet). There are no lower energy, opposite parity states with the same total spin to which it might decay by E1 photon emission and, similarly, the Auger final state 2p^6e, with spin 1/2, cannot be reached because Coulomb auto-ionization preserves spin. The state does autoionize slowly via the spin-spin part of the Breit interaction to the final state 2p^6eg (J=7/2).

We established the metastability of a component of line $\beta$ by biasing the He gas cell to -100 V, which shifts all prompt lines by 100 eV, but leaves lines from metastable states which decay between the gas cell and the 45 degree analyzer unshifted. From such biased spectra we measured 114.10±0.33 eV for the $^4D_{7/2}$ Auger line energy; the MCDF calculation predicts 114.09 eV. From high resolution scans we also measured the prompt lines from 2p^53s^2 ^2P_{3/2} and ^2P_{1/2} states (these are blended within the line labeled $\alpha$ in Figures 2 and 3) to be 100.6±0.33 eV and 102.8±0.33 eV in excellent agreement with the MCDF values of 100.58 and 102.78 eV. The metastable $^4D_{7/2}$ level, as demonstrated by the pressure studies, was populated by two collisions and thus its observation in this work does not indicate spin re-orientation in a single collision double capture event.

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Figure Captions

Figure 1. Experimental arrangement for zero-degree electron spectroscopy using a collimated beam from the LBL ECR ion source. The beam passes through the gas cell and 45 degree parallel plate analyzer into the Faraday Cup. Forward emitted electrons are deflected by the 45-degree analyzer into the high resolution hemispherical instrument.

Figure 2. Auger electron spectra from (a) 80 keV Ar$^{8+}$ and, (b) 90 keV Ar$^{9+}$ collisions on a He gas target (pressure 0.4 mTorr). Electron energies are given in the projectile frame. The location of lines (see text) which originate from several initial configurations are indicated. In all cases the final state is 2p$^6$e$^1$ except for the lower energy 2p$^5$4l4l$'$ group where the final state is 2p$^5$3s$^1$. The lines labeled $\alpha$, $\beta$ are from 2p$^5$3s$^2$ $^2P_J$ and 2p$^5$3s3p $^4D_J$ states. The latter contains the metastable $^4D_{7/2}$ component.

Figure 3. Spectrum (a) is synthesized from two spectra taken at pressures $P_1$ and $P_2$ (with $P_2$ about twice $P_1$) according to the scaling, $S=(P_1/P_2)S_2 - S_1$, where $S$, $S_1$, and $S_2$ are the intensities at each electron energy. (b) shows the spectrum taken at $P_1$ and, for comparison, (c) shows a spectrum from Ar$^{8+}$ incident projectiles. The scaling process removes structure linearly dependent upon pressure. The structures in (b) above about 200 eV are absent in (a) and are produced by single collision double electron capture. The lines in (a) labeled $\alpha$ and $\beta$ are the same as those shown in Fig. 2 (b). All structures in (a) arise from two separate single capture collisions, the first of which populates one of the metastable Ar$^{8+}$ 2p$^5$3s $^3P_J$ states.

Figure 4. Energy level diagram showing the allowed electron decay channels for the
core-excited levels of Na-like Ar. The energies of the various decay channels divide almost without overlap into distinct energy regions. These energy regions are displayed in Fig. 2 (b).
Adjustable slit collimators

Inner and outer Gas Cell

45° Parallel Plate Analyzer

Figure 1.
Figure 2.
Figure 3.
Figure 4.