Coincidence Measurements of Electron Capture and Ionization in Low-Energy Ar^{+q} + (He, Ne, Ar, Xe) Collisions

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(Received 6 March 1981)

We have used a secondary ion recoil source to study electron capture and ionization in collisions of Ar^{+q} with He, Ne, Ar, and Xe at projectile energies between 250 and 660 eV per charge. By analyzing outgoing projectiles and target ions in coincidence we have measured cross sections for ionization of the system with and without simultaneous electron capture, as well as for normal electron capture only. Cross sections for direct ionization (no electron capture) are found to be small, whereas those for ionization accompanied by electron capture are often quite important.

PACS numbers: 34.70.+e, 34.50.Hc

In recent years considerable interest has been shown in electron capture by slow, highly charged projectiles from both single-electron and multielectron targets.[1] At least part of the stimulus for such work is the role such reactions play in sapping energy from hot (e.g., tokamak) plasmas,[2] and in the possible application to x-ray lasers of the inverted populations generated by the capture to excited states of the projectiles. Most previous work has studied only the capture in such reactions. In this Letter we address the problem of ionization as well, in collisions with multielectron targets. {In our discussion the term ionization is restricted to processes for which the projectile-target system *loses* electrons. The two classes of ionization reported here are direct ionization (no electron capture) and transfer ionization [ionization accompanying electron capture (Ref. 3)]}.

Salzborn and co-workers[4] have studied collisions between highly charged rare-gas projectiles and neutral rare-gas targets, primarily at energies higher than those used here, and have found large cross sections for multiple electron capture. In those experiments, as indeed is the case for the great majority of data on electron capture, only the final projectile charge state was measured. Here we report results of measurements in which the final charge states of both projectile and target are distinguished. We find that transfer ionization (TI) can be the dominant channel in many cases and can feed a large fraction of the double-capture process back into channels characterized by a change of projectile charge by only one unit. We also present experimental evidence that direct ionization, in which the target is ionized at the expense only of the projectile kinetic energy, is negligibly weak.

Evidence of a different nature that TI can be important for capture involving highly charged projectiles has been previously reported by Flaks, Ogurtsov, and Fedorenko,[5] who measured integral electron production in Ne^{+q} on Xe, and by Woerlee *et al.*,[6] who found autoionization lines from doubly excited Ne for Ne^{+q} ($0 < q \leq 4$) on Ar. Considerable work on the TI process involving lower-charged projectiles has been carried out by Niehaus and collaborators,[3] and theoretical analyses have been presented.[7,8] Related coincidence measurements of target-projectile charge-state spectra were made by Kessel and Everhart.[9] We believe ours to be the first of this type of measurement to be applied to the high-q capture problem for projectile velocities in the region of 10^7 cm/sec.

Argon ions with charges between 1 and 10 were generated in a secondary ion recoil source by bombarding a tenuous (<1 mTorr) argon gas with a 19-MeV F^{+4} beam from the Kansas State University tandem Van de Graaff accelerator (Fig. 1). This ion source for slow, highly charged ions is rather novel in itself. The idea has been discussed in the literature[10-12] but only recently used in the study of low-energy capture processes.[13-15] The present work presents the first case in which the secondary ion beam has been extracted for use in cross-section measurements. In this application the slow recoil ions were extracted by an electric field at right angles to the F beam and accelerated to an energy $V_a q$ between 250 and 660 eV per charge into a second gas cell, 2 cm long, containing He, Ne, Ar, or Xe at a pressure typically below 5×10^{-4} Torr. The argon projectiles emerge with charge q', enter a double-focusing spherical-sector electrostatic analyzer, and are detected by a channel electron multiplier (CEM). Ionized target reaction products produced in the second cell are collected by a second electric

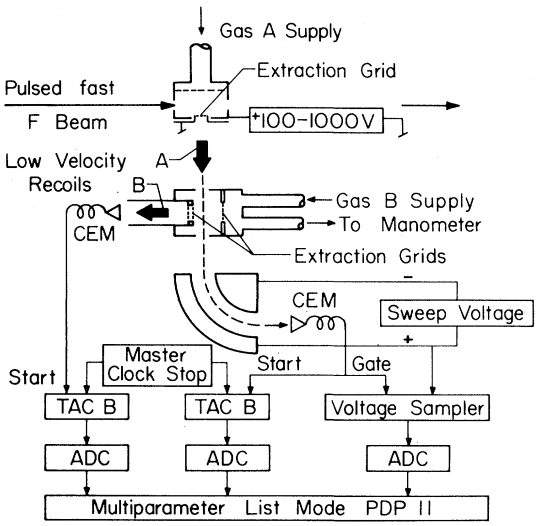

FIG. 1. Schematic of experimental apparatus.

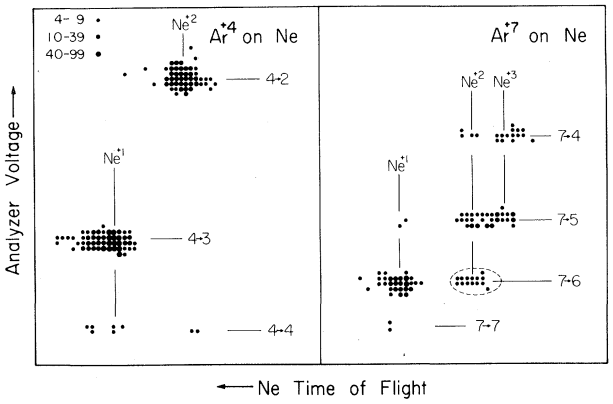

FIG. 2. A two-dimensional V vs T_B slice of the data for 500-eV/q Ar^{+q} on Ne. Final charge states of Ar and Ne are dispersed vertically and horizontally, respectively. The dashed circle indicates the TI discussed in the text.

field, at right angles to the slow argon beam, which sends them to a second CEM 4 cm away.

The F beam is pulsed, with an 8 μsec repetition rate and <5 nsec pulse width. Since argon ions with different q have different velocities, measurement of the flight time (T_A) to the first CEM gives q. The analyzer voltage V, which is swept by a triangle wave generator, analyzes ions at an energy E such that $E/q = kV$. Thus $q/q' = kV/V_a$ and V provides a measure of q'. Meanwhile the flight time (T_B) of the He, Ne, Ar, or Xe recoil to the second CEM distinguishes different charges of that reaction product. A data acquisition system based on a Digital Equipment Corp. PDP-11/34 computer is used to record T_A, T_B, and V for each detected event. The data-collection rate is slow in the three-parameter mode, a 5-h run for a given collision system being typical.

In Fig. 2 we show two-dimensional slices of V vs T_B, with windows set on T_A so as to select argon +4 and +7 projectiles. The target was neon with $E/q = 500$ eV. Events in which single and double electron capture from the neon occur without accompanying ionization dominate the +4 spectrum. Direct ionization of the target, accompanied by no projectile-charge change, is weak in both spectra, and even those events seen are due to random coincidences between any target-ionizing event and direct argon ions which underwent *no* collision in the second cell. Events in which the overall system appears to have *lost* charge are due to charge-state impurities caused largely by capture collisions by the argon beam leaving the first cell. In the case of the 7+ beam, strong production of Ne^{+2} ions in coincidence with singly capturing argon ions is seen, providing clear evidence for the occurrence of the TI process.

In Fig. 3, we show cross sections for direct ionization, single capture, double capture ($q \rightarrow q - 2$), and double-capture $q \rightarrow q - 1$ TI. We take "single" and "double" capture here to mean those events in which singly and doubly charged Ne, He, Ar, or Xe recoils are produced. The random coincidences were measured separately and subtracted before extracting the cross sections of Fig. 3; they are negligible except at $q = q'$. Absolute scales were assigned by measuring projectile-charge-changing fractions, for low q, in targets of known thickness. For example, the open circles for He are from such singles measurements without coincidence with He recoils. Error bars indicate reproducibility.

The direct ionization is undetectably small for all cases, at a level less than 2% of the single capture, except for the case of an Ar^{+8} projectile, for which the direct-ionization cross section appears consistently to be about 14% of that for single capture. We believe that this curious behavior is due to the presence of a 14% metastable beam component. For example, neonlike $2p^5 3s\ ^3P_{2,0}$ ions could be present, and would autoionize, after capturing a single $n = 3$ electron, to generate an event resembling direct ionization.

The TI, on the other hand, is quite strong, es-

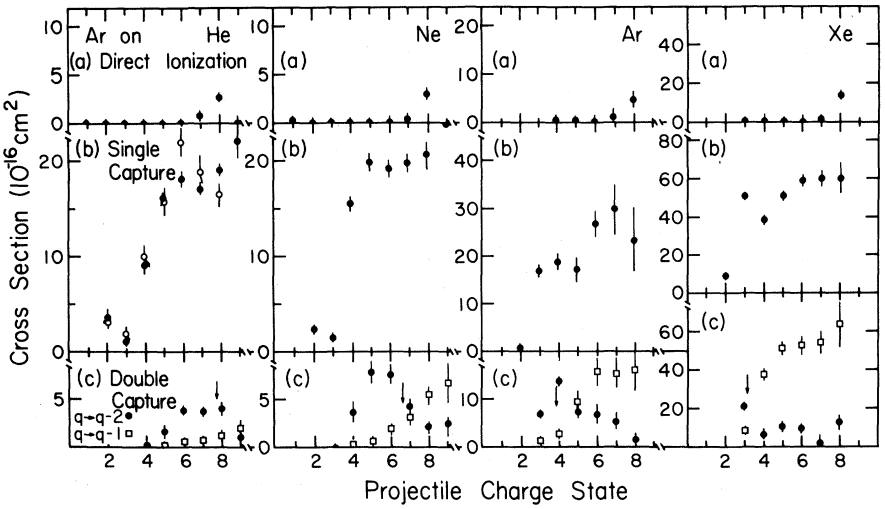

FIG. 3. A summary of data for 500-eV/q Ar^{+q} on targets of He, Ne, Ar and Xe. Direct ionization means ionization of the target with no projectile-charge change. "Single" and "double" capture here mean events in which singly and doubly charged target ions are produced, respectively. The arrows on (c) indicate values of projectile-charge state at which capture to doubly excited projectile states may commence.

pecially for the Ar and Xe targets. This process is of the form $Ar^{+q}+B \rightarrow Ar^{+(q-1)}+B^{+2}+e^{-1}$, and may occur through several different mechanisms, as discussed by other authors.[4-8] We make a broad separation of these mechanisms into (1) autoionization during the collision from the $(ArB)^{+q}$ molecule, and (2) population of the autoionizing states of one of the collision partners during the collision followed by electron emission from the excited system after separation. The importance of process (1) in very slow collisions has been established by Niehaus and collaborators,[3] while Woerlee *et al.*[6] found clear evidence that process (2) dominated for 100-keV Ne^{+q} projectiles (q = 1–4) capturing from rare-gas targets.

The present data do not allow us to distinguish experimentally between processes of types (1) and (2), since the corresponding final states differ only in the kinetic energy distribution among the reaction products. However, we favor the interpretation that the ionization we see results from the population by double capture of autoionizing states on the argon, with both captured electrons having principal quantum number $n=4$, and present a discussion indicating the consistency of this interpretation.

We note that the experimental data in Fig. 3(c) show that the onset of dominance of TI occurs at decreasing q as we proceed to targets of lower first (and second) ionization potential, and attempt to account for this behavior. We first make use of the treatment of the capture process by a highly charged projectile according to the classical transfer model discussed by Beyer, Schartner, and Folkmann[14] and by Ruyfuku, Sasaki, and Watanabe.[16] This leads to the conclusion that $n=4$ will first be populated for $q=8$, 8, 6, and 6 for Ar^{+q} on He, Ne, Ar, and Xe, respectively. This is qualitatively in the correct direction but quantitatively inadequate.

A better picture should emerge from an examination of the molecular electronic energies at large internuclear separation. The channel through which double electron transfer will occur should be slightly exoergic. Such a situation would allow a curve crossing with the entrance channel at large R. We have calculated the values of q at which the reaction becomes exoergic occur if a doubly excited $Ar^{+(q-2)}$ ion, carrying two 4s electrons, is populated. This occurs at $RE[Ar^{+q}] + RE[Ar^{+(q-1)}] - Ex^{**}[Ar^{+(q-2)}] = IP(B) + IP(B^{+})$, where RE is recombination energy, IP is ionization energy, and Ex** is the double excitation energy of the argon ion. In the absence of experimental values for Ex**, we used the Froese Fisher–Hartree–Fock (FFHF)[17] program to calculate average excitation energies for $1s^2 2s^2 2p^6 3s^l 3p^m 4s^2$ configuration, where the $n=3$ shell was progressively stripped to obtain the q of interest. For neutral and singly charged argon, the excitation energies were estimated on the basis of 4s excitation energies from the tables of Moore,[18] and

$(4s)^2$ interaction energies for higher q from the FFHF program.

The values of q at which this channel opens were found to be 7.6, 6.6, 3.9, and 3.1 for targets of He, Ne, Ar, and Xe, respectively (q has been treated as a continuous variable by linear interpolation). These values are indicated by arrows on Fig. 3(c), and correspond approximately to onset of dominance of the $q \rightarrow q-1$ TI channel in double capture. While this agreement certainly does not prove that population of the doubly excited argon is the only contributor to the TI that we observe, it is quite consistent with this interpretation. We cannot rule out the population of autoionizing target states, although double capture to excited projectile states appears much the more natural process.

In summary, we have shown that, while direct ionization of multiple-electron targets in collision with slow, highly charged projectiles is negligibly small, ionization promoted through the electronic energy afforded by strong binding of the captured electrons by the projectile is important. Independent of the specific interpretation given above, the data show clearly that detecting only the final charge state of the projectile may lead to a seriously inadequate picture of what is occurring in such collisions. For example, for the case of Ar^{+7} on Xe, 50% of the cross section which would be called "single capture" on the basis of exiting Ar^{+6} ions alone comes in fact through the TI channel and is probably more correctly associated with double capture. Although we have discussed only the double-capture case here, copious TI was also observed for Ar and Xe targets for capture of more than two electrons and is much more common than normal capture for these cases. Serious interpretation of both single and multiple capture from such targets should be made with caution in the absence of final channel discrimination of both collision partners.

This work was supported in part by the Chemical Sciences Division, U. S. Department of Energy.

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