Evidence for Significant Target Outer-Shell Excitation in Multiple-Electron Capture Collisions of Slow Highly Charged Ions with Many-Electron Atoms

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Unequivocal evidence for significant target outer-shell excitation accompanying multiple-electron capture, in slow collisions of highly charged ions with many-electron atoms, has been obtained by means of simultaneous Auger-electron and cold-target recoil-ion momentum spectroscopic measurements. For the 28 keV $^{15}N^{7+}$ + Ar collision system, it is found that target excitation accompanies about 40% of all double-electron capture collisions. The evidence supports the predictions of the molecular classical overbarrier model by Niehaus [A. Niehaus, J. Phys. B **19**, 2925 (1986)].

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In slow collisions ($v < 1$ a.u.) of highly charged ions (HCI) with many-electron atoms, a large number of electrons can become active. It is well known that the dominant target electron removal mechanism is electron capture, and that multiple-electron capture (MC) results in the formation of projectile multiply excited states. In addition to their fundamental scientific significance, both the dynamics of many-electron reactions and the properties (binding energies, lifetimes, relaxation pathways, and branching ratios) of multiply excited many-electron systems have strong practical impact on various applications such as the characterization of fusion and astrophysical plasmas.

Multielectron (≥ 3) processes in such collisions have been investigated for over two decades, and most of the experimental methods have been reviewed by Barat and Roncin [1]. Since quantum mechanical treatment of such collisions is prohibitively difficult, extended classical overbarrier models have been developed [2,3] to account for MC processes. It is well known that target excitation (TX) is important in singly charged ion-atom collisions, but is negligible in most single-electron capture (SC) processes in slow HCI-atom collisions. A key prediction of the molecular classical overbarrier model (MCBM) by Niehaus [3] concerns target outer-shell excitation in MC processes. The model describes a collision by a string j whose number of elements indicate the number of electrons molecularized during the collision. The elements are either 1 or 0 indicating capture by the projectile or recapture by the target, respectively, and whose positions label the electrons in order of increasing ionization potentials. TX is realized whenever initially tightly bound target electrons are captured by the projectile while some loosely bound electrons are recaptured by the target.

The MCBM prediction of TX in MC collisions has been controversial for over a decade, however. Differential cross section measurements were carried out for a number of collision systems by Danared *et al.* [4] and Guillemot *et al.* [5]. Both groups criticized the model for overestimating TX. Similar measurements were carried out by Ali *et al.* [6] who found no evidence for overestimation of TX. In addition, Ali *et al.* [7] reported cross sections that seemed to support the prediction of TX. The $^{15}N^{7+}$ + Ar collision system studied in this work has been investigated by several groups. At 70 keV, Benoit-Cattin *et al.* [8] obtained a singles' Auger-electron spectrum, and argued that double-electron capture (DC) accompanied with TX (DCX) is not important. de Nijs *et al.* [9] obtained partial Auger-electron spectra in coincidence with target ions at the same energy, and concluded that DCX is important. Roncin *et al.* [10,11] studied the same system at 10.5 keV using coincident energy gain spectroscopy (CEGS). They observed DCX [11] and TX with SC [10], but they were attributed to postcollision interactions (PCI). They claimed that direct DCX as suggested in [9] is much weaker than the PCI TX at 10.5 keV [11]. Martin *et al.* observed a wealth of target emission lines in the visible range [12–14] following SC and MC in a number of systems, and argued in support of the PCI mechanism. For the 14 keV $^{15}N^{7+}$ + Ar system in particular [12], they observed target emission lines in SC but did not observe expected 348 nm Ar^{2+} emission lines and concluded that DCX should be very low, in contradiction with the conclusion of de Nijs *et al.* [9]. Recently, Moretto-Capelle *et al.* [15] performed coincident measurements similar to those of de Nijs *et al.* [9] for the 70 keV $^{15}N^{7+}$ + Ar system and discussed DCX. Definite conclusions were not made, however, and they stated that DC is not yet a solved problem. Finally, Cederquist *et al.* [16] compared SC and DC cross sections with the predictions of the model by Bárány *et al.* [2], and suggested a TX mechanism that is effective for two-electron but not for many-electron targets. Later, they showed that this mechanism is not as important as suggested [17].

None of the reports supporting TX [7,9] as predicted by the MCBM produced direct evidence by detecting decay products or by measuring Q values that can be unambiguously associated with TX. They were based only on comparisons with the MCBM. On the other hand, in the measurements where evidence was produced [10–14], TX was argued to be weak and due to PCI. Clearly, the importance of, and mechanisms leading to, TX have remained controversial. In this Letter, we report simultaneous Auger-electron and cold-target recoil-ion momentum spectroscopic (COLTRIMS) studies of the 28 keV $^{15}N^{7+}$ + Ar collision system. The studies employed a cold target, position imaging detectors, and time-of-flight (TOF) triple-coincidence detection of Auger electrons, scattered projectile, and target recoil ions. This powerful combination enables the isolation of events giving rise to individual Auger lines and obtaining the corresponding Q values. The question of TX is closely examined.

The measurements were performed at the University of Nevada, Reno, Multicharged Ion Research Facility which houses a 14 GHz electron cyclotron resonance ion source. The TOF triple-coincidence measurements have been described in detail elsewhere [18]. This part provides subpartial Auger-electron spectra corresponding to specific final projectile and recoil-ion charge states. The new feature in the present measurements is the ability to simultaneously perform COLTRIMS measurements, and therefore obtain Q values. Experimental details will be given in forthcoming papers, while details on how to obtain Q values from the measured recoil-ion momenta can be found in a number of papers [19–23].

We show in Fig. 1 the subpartial Auger-electron spectrum in coincidence with the (Ar^{2+}, N^{6+}) ion pair. This channel is known as autoionizing DC (ADC). The Auger lines were identified using the Hartree-Fock atomic structure code by Cowan [24]. The main populated configuration complexes are the (3, 3), (3, 4), (3, 5), (4, 4), (4, 5), and some $(3, n \geq 6)$. Since the electron detector views the interaction region, some photons are also detected. The spectrum closely resembles that obtained by Emmons *et al.* [18] at 70 keV, and only slight changes in relative intensities are observed. A similar conclusion can be reached from the partial spectra obtained by de Nijs *et al.* [9] at 35 and 70 keV. These measurements, and those by Moretto-Capelle *et al.* [15], reveal significant populations of the (3, 3), (3, 4), and (3, 5) configurations. Some MCBM reaction windows for DC are shown in the inset of Fig. 1. The reaction windows for the strings j1 and j2 overlap the (4, 4), (4, 5), and $(3, n \geq 6)$ configurations. The window for j3 overlaps (3, 4) and (3, 5), while (3, 3) can be accounted for only by a four-electron string such as j4. According to the MCBM, the population of (3, 3), (3, 4), and (3, 5) must be DCX.

Contrary to previous studies, the present one can directly test the TX assumption. The shaded regions in Fig. 1 are Auger-electron gates representing electrons that originate in the different initial configurations. Gated Q-value spectra can be obtained by demanding that events give rise to electrons in the different gates. Figure 2 shows such Q-value spectra. We note a nearly perfect match between the experimental and the Q-value ranges for a pure population,

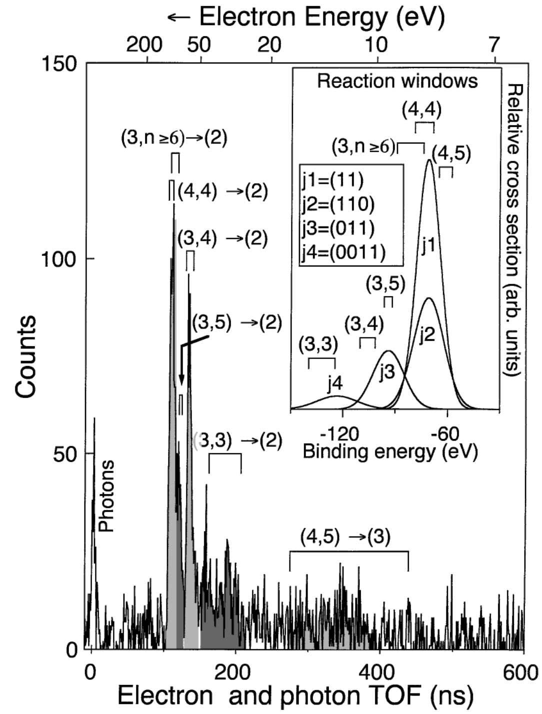

FIG. 1. The subpartial Auger-electron spectrum in coincidence with the (Ar^{2+}, N^{6+}) ion pair. The shaded regions are gates used to obtain gated Q-value spectra. Inset: MCBM reaction windows for some strings j giving rise to DC. Other windows that lie in between the shown reaction windows are not shown. Binding energies for various N^{5+} doubly excited configurations are also shown.

i.e., no TX, of (4, 5), (4, 4), and $(3, n \geq 6)$, in agreement with the MCBM predictions. The Q-value spectra for the $(3, n = 3–5)$ configurations, however, exhibit significant shifts toward smaller Q values. These shifts constitute a direct and unequivocal evidence for outer-shell TX, again in agreement with the MCBM predictions and the conclusion of de Nijs *et al.* [9]. For each populated configuration, we have estimated the approximate fraction f_{TX} of DCX, and the average TX energy E_{TX} for that fraction. Table I summarizes our findings for ADC. Note that the relative intensities (I_{rel}) of the populated configurations assume isotropic electron emission. Clearly, DCX is a significant fraction of DC into $(3, n = 3, 4)$, but most dramatic is the population of (3, 3) where essentially all events are DCX. Using Table I, it can be shown that about 54% of all ADC events involve TX. Determining the fraction of all DC with TX requires knowledge of true DC (TDC). Since no electrons are ejected in TDC, we have performed COLTRIMS only measurements under the same conditions. We show in Fig. 3 the Q-value spectra for SC, ADC, and TDC. Using Table I and the intensity of TDC relative to total DC in Fig. 3, we find that DCX is about 40% of all DC, which is indeed significant.

The E_{TX} values in Table I provide hints about the levels to which the Ar^{2+} ions are excited. The 17 eV accompanying DC into (3, 5) is typical of a $\Delta n = 0$ single TX to

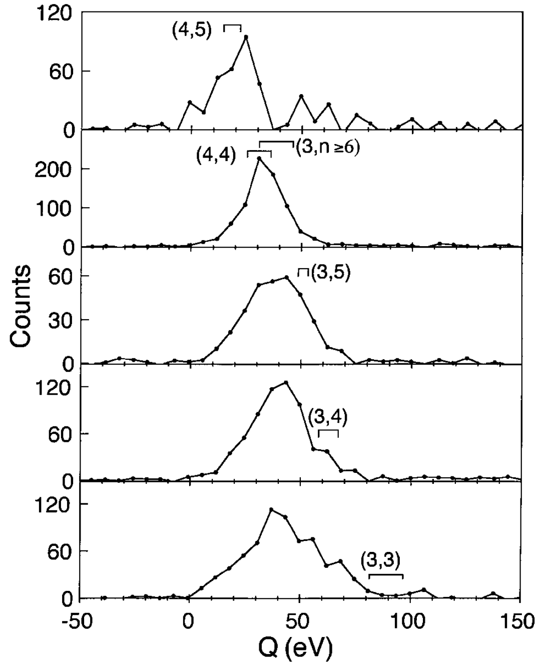

FIG. 2. Gated Q-value spectra corresponding to the different gates in Fig. 1. The spectra are identified by the initial populations of the gates. The labeled ranges assume DC without TX.

the lower levels of the [Ne]$3s^2 3p^3 3d$ electronic configuration. The 24 eV in the case of $(3,4)$ suggests that both $\Delta n = 0$ single TX to the upper levels of [Ne]$3s^2 3p^3 3d$, and $\Delta n = 1$ single TX to the lower levels of [Ne]$3s^2 3p^3 4l$ electronic configurations are important. The 47 eV accompanying DC into $(3,3)$ is higher than the 40.74 eV ionization energy of Ar^{2+} and cannot be associated with single TX. It can be associated, however, with some doubly excited states of Ar^{2+}. These states must be metastable against autoionization; otherwise they will result in Ar^{3+} ions. Likely candidates are the [Ne]$3s^2 3p^2 3d^2\ {}^5S^e$, ${}^5D^e$, the [Ne]$3s^2 3p^2 3d4s\ {}^5D^e$, the [Ne]$3s^2 3p^2 3d4p\ {}^5P^o$, ${}^5F^o$, and the [Ne]$3s^2 3p^2 3d4d\ {}^5S^e$, ${}^5D^e$, and ${}^5G^e$ states. These states span an excitation energy range of 41–57 eV and can give rise to an E_{TX} of 47 eV. Since there is no reason to expect that only metastable states are produced, we believe that some of the Ar^{3+} recoil ions produced during the collisions are due to DC and simultaneous double TX of the Ar^{2+} ions which then autoionize. This implies that the previously quoted DCX fraction of 40% is a lower limit, since some of the DC events appear in the Ar^{3+} channel. The MCBM needs at least four-electron strings to fully account for DC. Limiting the discussion to the six four-electron strings that give rise to DC, only the string (1100) does not involve TX. An upper limit of 60% for the DCX fraction is obtained by assuming no target autoionization for strings where the target is left in doubly excited states. This upper limit is in good agreement with the experimental lower limit of 40%. The MCBM predicts average E_{TX} for the five four-electron strings involving TX ranging from 7.1 eV for the (1010) string to 42.8 eV for the (0011) string. This range compares well with the experimental E_{TX}. Indeed, the MCBM predictions are impressive.

The present measurements also provide new insights into the PCI mechanism known as autotransfer to Rydberg states (ATR) [10,25] and its role in realizing TDC. The spectra shown in Fig. 3 are the equivalent of the energy gain spectra obtained by Roncin *et al.* [10]. SC populates the same $n = 4,5$ levels in N^{6+}, but with different relative intensities due to the difference in collision energies. The ADC and TDC Q-value spectra are very similar to their spectra [10]. The TDC profile is narrower and matches well with the $(4,4)$ configuration. The centroid of the ADC profile also matches well with $(4,4)$. They assumed a dominant $(4,4)$ population for ADC, and attributed the wider profile to kinematic broadening caused by autoionization. In our case, the ADC profile is free of any kinematic broadening since the Ar^{2+} recoil ions do

TABLE I. Populated configurations in ADC and their relative intensities (I_{rel}), approximate fraction accompanied with TX (f_{TX}), and average TX energy (E_{TX}).

Configuration	$I_{rel}(\%)$ [a]	$f_{TX}(\%)$	E_{TX} (eV) [b]
$(4,5)$	9	0	0
$(4,4) + (3, n \geq 6)$	27	0	0
$(3,5)$	12	65	17
$(3,4)$	26	77	24
$(3,3)$	26	100	47

[a] Relative to total ADC.
[b] Calculated for f_{TX}.

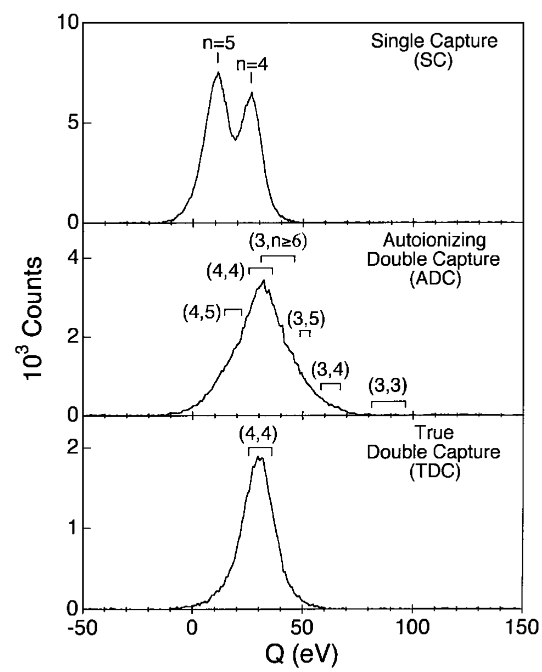

FIG. 3. Q-value spectra for SC, ADC, and TDC. The resolution is 11.5 eV (FWHM).

not autoionize, and the wider profile must clearly be due to the population of other configurations. Under the assumption that DC dominantly populates $(4,4)$, Roncin *et al.* [10] obtained an apparent radiative stabilization probability $P_{\rm rad}(4,4) \approx 40\%$ (ratio of TDC to total DC). This value is much higher than expected since states belonging to $(4,4)$ are expected to dominantly autoionize.

To explain the high $P_{\rm rad}(4,4)$, the initially populated $(4,4)$ configuration was assumed to undergo a population transfer to the quasidegenerate, and highly asymmetric, Rydberg series $(3, n > 9)$ via the ATR mechanism. The Rydberg states were believed to have large fluorescence yields. Models with different degrees of sophistication have been developed [10,26] to account for the population of the Rydberg states and their subsequent decay. The refined ATR model by Kazansky and Roncin [26] takes into account the postcollisional increase of the angular momentum of the Rydberg electron. It was found that this increase is fast enough to quench the autoionization process, thus increasing $P_{\rm rad}(4,4)$, and that it is strongly velocity dependent. Large average $P_{\rm rad}(4,4)$ values, even larger than the experimental value, have been obtained.

This Letter, however, provides more detail than previously possible. Figure 2 shows significant overlap in the Q-value spectra corresponding to DC into the different configurations. Therefore, it is impossible to accurately identify and obtain the relative intensities of all configurations using CEGS [10,11]. Although COLTRIMS alone provides hints that configurations other than $(4,4)$ are populated, it also cannot accurately identify and provide the relative intensities of all configurations. In fact, based on the ADC Q-value spectrum in Fig. 3, one may conclude that $(3,3)$ is not populated at all, and that $(3,4)$ is weakly populated. Both conclusions cannot be farther from the truth. Assuming that all DC populates $(4,4)$ yields the apparent $P_{\rm rad}(4,4) = 27\%$. This value, being smaller than that reported by Roncin *et al.* [10] at 10.5 keV, is in general agreement with the velocity dependence of $P_{\rm rad}$ [26]. In reality, however, at most 27% of ADC proceeds into $(4,4)$ (see Table I). This yields the true $P_{\rm rad}(4,4) \geq 58\%$. The minimum limit assumes no $(3, n \geq 6)$ population. This value is in very good agreement with the refined ATR model [26]. We do not expect a significant change in population at 10.5 keV, and believe that $P_{\rm rad}(4,4)$ reported by Roncin *et al.* [10] is small by about a factor of 2.

The PCI mechanism for TX suggested by Roncin *et al.* [10,11] is actually rooted in ATR. According to them, DCX requires [11] initial triple-electron capture (TC) into $(3,4,4)$, followed by ATR to $(3,3,n \gg 4)$, and then some of the Rydberg electrons $(n \gg 4)$ are recaptured by the Ar^{3+} target ions, thus resulting in TX. Assuming dominant TC into $(3,4,4)$, they estimated that 18% of ADC (12% of all DC) involves TX. While this PCI mechanism may explain TX in the case of $(3,3)$ in the present measurements, it fails to explain TX in the case of $(3,4)$ and $(3,5)$. Also, the subpartial Auger-electron spectra in coincidence with the (Ar^{3+}, N^{6+}) and (Ar^{3+}, N^{5+}) ion pairs in the present measurements (not shown here), and at 70 keV [18], show that TC into $(3,3,n = 3-5)$ is more important than $(3,4,4)$. While we cannot exclude a possible PCI contribution to DCX in the case of $(3,3)$, we believe that a significant fraction of TX is taking place during the collision as predicted by the MCBM.

In conclusion, we have investigated the 28 keV $^{15}N^{7+}$ + Ar collision system by means of simultaneous Auger-electron and COLTRIMS spectroscopic measurements. Unequivocal evidence for significant TX accompanying the population of the $(3, n = 3-5)$ configurations in DC has been obtained. The MCBM predictive powers have been tested at a finer level than has been possible before and found to be rather impressive. The measurements also revealed very high $P_{\rm rad}(4,4)$ in support of the refined ATR model [26].

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