Multiple ionization in single, low-MeV collisions of Kr ions with Kr atoms

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The energy and relative intensities of *L-MM* Auger lines show that for low-MeV Kr^{+}-Kr collisions most of the ionization takes place during the collision, due to the prompt emission of as many as 12 electrons per atom. The later decay of *L*-shell vacancies contributes in only a minor way to the final ionization state. These results provide insights into a variety of collision phenomena, including shifts in Auger and x-ray energies, fluorescence yields, and equilibrium charge distributions in high-*Z* foils.

The phenomenon of multiple ionization in atomic collisions has generated a great deal of interest. As long ago as 1912, Thomson observed the loss of as many as eight "negative corpuscles" from mercury atoms in his electric discharges.[1] (As he noted, "Eight is a very large number of negative corpuscles to lose. . . .") Ionization states in excess of $+25$ have been observed following single collisions of heavy ions[2,3] and similar charge states may be observed following the passage of heavy ions through solid targets.[4] For the case of solid targets, efforts have been made to identify the relative importance of inner- and outer-shell ionization to the final charge state.[4,5] In the present investigation of electron production in single collisions of Kr ions with Kr atoms, we have found that ionization in the outer molecular orbitals of the quasimolecule formed during the collision account for most of the ionization and contribute a low-energy continuum to the energy spectrum of the emitted electrons; while decay of *L*-shell vacancies results in a broad, well-defined peak in the spectrum at an energy characteristic of *L* Auger decay in Kr ions that are already 10–12 times ionized.

Figure 1 shows doubly differential measurements of electron production in Kr^{+}-Kr collisions plotted versus the electron energy for several collision energies. The predominant feature of the spectra is the electron continuum extending from 100 eV out to about 800 eV, produced by the decay of quasimolecular states during the collision. The continuum has been investigated by several researchers,[6–10] but comparatively little attention has been paid to *L* Auger processes in these collisions, although above a collision energy of 0.35 MeV, *L* Auger transitions are significant and are responsible for the broad peak at 1100 eV in Fig. 1.

The 1100-eV peak is almost 300 eV lower than the centroid of *L* Auger spectra obtained by bombarding Kr with protons[11] or electrons.[12] Auger transition energies depend upon the state of ionization,[13,14] and the lines that contribute to the *L* Auger peak in Fig. 1 are not characteristic of singly ionized Kr. This makes good physical sense if we assume that the *L*-shell decay occurs after the ionizations contributing to the low-energy continuum have taken place. In order to determine quantitatively the effects of prior ionization, we used the one-electron binding energies of multiply ionized Kr to estimate the Auger energies for decay to various charge states. The calculations were performed in the Dirac-Fock formalism using a program written by Desclaux.[15] The results of these calculations are summarized in Fig. 2, which shows that Kr *L* Auger energies are shifted down in energy by as much as 300 eV for final charge states of $+14$. An initial double *L*-shell vacancy (not shown in the figure) would shift the *L-MM* lines up about $+35$ eV. We conclude from the Dirac-Fock calculations that the 1100-eV peak in Fig. 1 is attributable to *L* vacancy decay in ions that are already 10–12 times ionized. We note that the Auger energies and associated charge states appear to be relatively insensitive to varia-

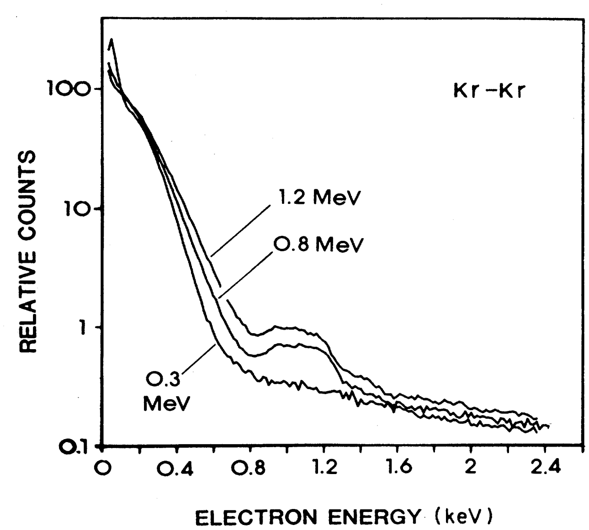

FIG. 1. Doubly differential cross sections for production of electrons in Kr-Kr collisions. The continuum from 100–800 eV is due to ionization processes in the transient Kr-Kr quasimolecule. The structure in the 800–1300-eV portion of the spectrum is attributable to *L*-shell vacancy production.

tions in collision energy from 0.4–1.6 MeV, indicating that most L vacancies are produced in collisions for which outer-shell processes remove a nearly constant number of electrons.

Direct measurements of the final charge state have been performed earlier.[16] It was found that high charge states were obtained before the distance of closest approach threshold for L-shell vacancy production is reached at about 0.1 a.u. These charge states increase only gradually for smaller distances of closest approach, demonstrating that Auger decays remove relatively few additional electrons. For Kr-Kr collisions, then, electron shake off and vacancy cascading induced by decay of the L-shell vacancy contributes little to the overall postcollision charge state. It is possible that deeper interpenetration of the shells will produce further inner-shell excitation; however, the charge state data shows that the effect on the final ionization state remains less than that of the quasimolecular processes which remove the bulk of the electrons during the collision. It is not clear how many electrons will be available to participate in cascade processes if a K vacancy is produced or for L vacancy production in heavier systems, as in Xe-Xe.[3]

The data in Fig. 1 represent total electron production irrespective of impact parameter. Spectra for specific impact parameters may be obtained by selecting out those electron events that occur in coincidence with the detection of ions scattered to known angles. In general, coincidence measurements provide more detail than measurements of total cross sections, but are more difficult and time-consuming to perform. Figure 3 is an electron-energy spectrum obtained by detecting only those electrons which result from collisions in which the incident ion is scattered through 10°. These ion-electron coincidence data are less affected by the low-energy continuum that dominates the spectra in Fig. 1 because the strong contribution from smaller angle collisions is removed. There is enough discrete structure in the spectrum to make a rough comparison of the data with what might be expected from Auger decay in an ion already having many vacancies in its outer shells. The solid curve is the result of a calculation in which it is assumed the 0.9-keV peak is due primarily to L_{23}-$M_{23}M_{23}$ transitions from an ion that is already 12 times ionized (final charge state $+13$). The primary contribution to the two higher-lying peaks would then correspond to the L_{23}-$M_{23}M_{45}$ and L_{23}-$M_{45}M_{45}$ groups. The assignment of the lines to these transitions is not unique. For example, an initial configuration involving a double L-shell vacancy and an overall charge state of $+15$ would yield an equivalent simulation spectrum; however, this would not be consistent with the final charge states observed for this scattering angle.[16]

The relative strengths of the three peaks of the solid curve in Fig. 3 are different from what we would expect from H^+-Kr experiments[11] and from the transition rate calculations performed by Chen for Kr^{+1}.[17] The present simulation was obtained by reducing the $M_{45}M_{45}$ line intensities by 50% and the $M_{23}M_{45}$ intensities by 30%, relative to the theoretical transition rates, in order to provide a reasonable fit to the data. The

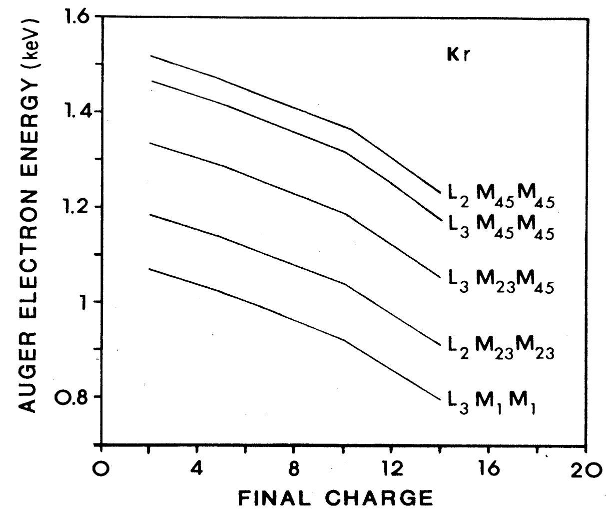

FIG. 2. L-MM Auger-electron energies as a function of charge state. The energies were approximated using the final-state, one-electron binding energies computed in the Dirac-Fock formalism.

reduction in relative intensity of the transition groups that involve the M_{45} subshells (the $3d$ orbitals) suggests these are among those electrons which are removed by the quasimolecular processes earlier in the collision.

Fortner, Woerlee, and Saris,[18,19] and Shanker *et al.*[20,21] have probed L-shell vacancy production in Kr-Kr collisions via the x-ray decay channel. They analyzed their data in terms of a model involving electron promotion facilitated by rotational coupling of one-electron orbitals in the Kr-Kr quasimolecule. The ratio

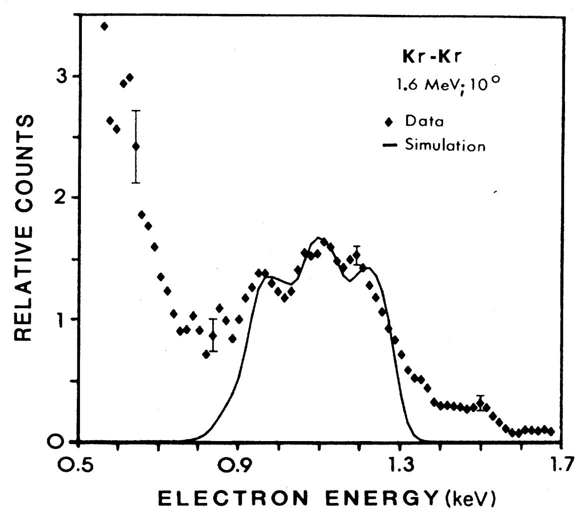

FIG. 3. Triply differential measurements of electron production in Kr-Kr collisions. The projectile ion scattering angle is 10° and the collision energy is 1.6 MeV, corresponding to an impact parameter of 0.9 a.u. The data points represent the average of five spectra. The simulation spectrum is constructed of L_{23}-MM Gaussian peaks shifted in energy to correspond to a post-transition charge state of $+13$ and broadened by 5% to account for the instrumental resolution.

of x-ray production rates to L-shell vacancy rates, the fluorescence yield ω, is necessary to correlate theory and experiment; and Woerlee and Shanker and co-workers assumed that the ω for singly ionized Kr is appropriate for the entire range of collision parameters. In support of this, these researchers have pointed to the apparent invariance of ω in total cross-section measurements over the 0.3–1.6-MeV collision range. Unfortunately, ω for Kr is small and may be subject to fluctuations with changes in charge state. The high charge state existing prior to decay of the L vacancy shows the use of ω for Kr^{+1} is not appropriate and is probably responsible for the discrepancy between their calculated and experimental cross sections.

The tentative analysis of the electron spectra presented herein provides strong evidence, based on the energy shifts in the electron spectra and the changes in relative transitions rates, for a collision picture in which most of the ionization occurs during the collision and that this is followed by Auger decay which contributes relatively little (one charge state) to the overall ionization.

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