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Ar L Auger-electron emission in 0.4-, 0.8-, and 1.6-MeV Ar⁺-Kr and Kr⁺-Ar collisions

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The electron-energy spectra for electrons emitted from Ar^+ -Kr and Kr^+ -Ar collisions have been measured for projectile energies of 0.4, 0.8, and 1.6 MeV. The spectra show a peak, or peaks, attributable to *L*-vacancy production in the Ar ions superimposed on a continuous portion of the electron spectra. This continuous portion, which decreases with increasing electron energy, extends to electron energies in the 600-800-eV range. The data show some events for which electrons with more than twice the normal $L_{2,3}$ -MM Auger energies are emitted, and their origin is not clear. They might be due to the decay of L_1 vacancies or to the filling of a vacancy in a molecular orbital during the collision. An Auger process in which two 2*p* vacancies are filled with the subsequent emission of only one electron, an $L_{2,3}L_{2,3}$ -MMM transition, might contribute to the spectra above 400 eV. The association of the major peak with Ar *L*-vacancy production is in contrast to a recent paper in which the peak, together with the higher energy electrons, was attributed to quasimolecular electron emission, specifically, filling of $4s\sigma$ and $4p\pi$ molecular-orbital vacancies during the collision.

I. INTRODUCTION

The ionization processes that give rise to multiple ionization in slow heavy ion-atom collisions are not well understood. It is clear that molecular-orbital (MO) processes provide the primary mechanism by which translational energy is converted to electronic energy in these collisions. 1^{-5} However, the specific means by which this energy transfer results in ionization for a given collision system is not always clear. Investigation of the resulting electron-energy spectra is an invaluable aid in interpreting the results. For example, if well-resolved Auger lines are observed, it is known that the collision produced the corresponding inner-shell vacancy configuration and that portion of the decay process is well understood. On the other hand, a number of experiments have demonstrated that for collisions of very heavy ions, such as the Kr-Kr and Kr-Xe systems, 3.6-10 the ionization is dominated by processes which give rise to a continuous range of electron energies which often extend from near zero energy to beyond 500 eV. The higher-energy portions of such spectra might be due to a variety of processes. Woerlee et al.⁷ argue that the higher-energy portion of the spectra (above 300 eV) resulting from Kr-Kr collisions is due to the radial and/or rotational coupling of highly excited MO's with the continuum. Another possibility suggested for either high- or low-energy electron emission in the Kr-Ar, Kr-Kr, and Kr-Xe systems is Auger-type decay during the

collision, ${}^{3,6-13}$ giving rise to what have been termed "MO electrons." Molecular-orbital calculations^{14,15} for these systems show that certain MO's are momentarily lowered in energy during the collision and it would be energetically possible for the collisional filling of a vacancy to result in electrons with energies greater than 500 eV.¹⁰ There are a number of processes which might contribute to the lower-energy portion of the spectra. These include a multiplicity of outer-shell processes, overlapping satellite lines from decay of ions having different states of excitation and ionization, and electron shakeoff. Also possible are Auger-type processes in which one electron drops into an inner shell and the excess energy is shared by two or more outgoing electrons.^{16,17} Another suggestion for electrons in the 0–110-eV range from Ar-Ar collisions is the promotion of electrons directly into the continuum.¹⁸

Electron data from the Ar-Kr system, similar to those presented here, have been obtained by several groups;^{8,9,11-13} however, the earlier papers failed to include the possibility of Ar L Auger electrons as a contributing factor in analyzing their spectra. For example, recently Shanker and co-workers obtained spectra similar to the present ones using 0.7-MeV collision energies, but they interpret the peak not as being due to Auger excitation but solely to quasimolecular electron emission.¹² Specifically, they attribute the peak herein associated with Ar L Auger production to the filling of vacancies in the $4s\sigma$ and $4f\pi$ MO's during the collision. The present investigation measures this peak for both the Ar^+ -Kr and the Kr⁺-Ar collisions, and through analysis of the Doppler shifts shows its origin to be associated with L vacancy production in the Ar ion.

II. EXPERIMENTAL RESULTS

The projectile ions, either Ar^+ or Kr^+ , are produced in the terminal of a 2-MV Van de Graaff accelerator, mass analyzed, and directed into a 42 cm-diam scattering chamber through 0.76-mm collimation. The ion beam passes approximately 1 mm above a 0.75-mm-i.d. needle from which the target gas, Ar or Kr, effuses. Electrons from this target region are detected after passing through a cylindrical electrostatic analyzer. The exit slit of the analyzer provides 5% energy resolution and the electrons are detected with a channeltron-type electron multiplier. The data presented here were obtained at angles of 70°, 90°, and 110° with respect to the beam direction. Further experimental details and the criteria used to ensure single collisions may be found in Refs. 19 and 20.

Electron spectra from Ar^+ -Kr collisions obtained at 90° with respect to the ion beam and for 0.4-, 0.8-, and 1.6-MeV incident ion energies are shown in Fig. 1. These data have been corrected for the 1/E dispersion of the cylindrical analyzer and show the same general features as reported in earlier investigations at different energies.^{8,11,12} These are an exponential decrease with increasing electron energy between 50 and 100 eV, a shoulder between 100 and 500-600 eV, followed by an exponential tail above the 500-800-eV region. In order to determine the origin of the electrons contributing to the shoulder, spectra were measured at different electron emission angles for Ar^+ -Kr and Kr⁺-Ar collisions. The



FIG. 1. Differential cross sections for electron emission from Ar^+ -Kr collisions are plotted as a function of electron energy for collision energies of 0.4, 0.8, and 1.6 MeV. These data were obtained for electron emission angles of 90° with respect to the ion beam.



ELECTRON ENERGY (eV)

FIG. 2. Uncorrected electron emission intensities for emission angles of 70°, 90°, and 110° with respect to the beam axis, plotted vs electron energies for 1.6-MeV collisions. The peak locations are indicated by arrows. (a) For the Kr^+ -Ar collisions: these data show no Doppler shift in the peak attributed to Ar L Auger electrons. (b) For the Ar^+ -Kr collisions: the peaks in these spectra are shifted in energy with respect to the data in (a). These shifts correspond to those expected for the Doppler shifting of the energies of electrons emitted from the 1.6-MeV Ar ions.

results for 1.6-MeV collisions are shown in Fig. 2 for data obtained at angles of 70°, 90°, and 110° with respect to the ion beam. These data are plotted on a linear scale and are *not* corrected for analyzer dispersion (in order to display the peak more clearly). Figure 2(a), for the Kr^+ -Ar collisions, shows peaks that do not vary with the detection angle while those in Fig. 2(b), for the Ar^+ -Kr

collisions, show peaks which do. This latter case indicates the electrons are emitted from the fast-moving scattered Ar ion. That is, the unshifted data in Fig. 2(a) correspond to the electrons being emitted from the relatively slow moving target Ar ions while in Fig. 2(b) the lines are Doppler shifted by +49 and -32 eV with respect to the expected shift for 90°. The corresponding Doppler shifts calculated for these angles are +45 and -34 eV, respectively, well within the estimated 20% error associated with the experimental determination of these values. This is in contrast to the shifts expected if the electrons contributing to the peak are emitted from the moving center of mass (c.m.) of the collision system, an appropriate assumption for the emission of MO electrons. For MO electrons from the Kr⁺-Ar collisions, c.m. emission would result in the spectra of Fig. 2(a) being Doppler shifted (+21 eV for the 70° detection angle and -18 eV at 110°, with respect to the shift for 90°), and no Doppler shift is observed for these data. For the Ar⁺-Kr data the corresponding shifts for c.m. electron emission would be +14 and -13 eV, about one-third of the shifts displayed in Fig. 2(b). In summary, the observed shifts are those expected for L Auger emission from the Ar ion and are not consistent with MO electron emission from the center of mass of the collision.

To better display the nature of the peaks contributing to the shoulder in Fig. 1 (and following the procedure used by Shanker *et al.*¹²), Fig. 3 shows these same data, but after subtraction of exponential functions fitted to both the low- and high-energy portion of the spectra. The prominent peak between 125-170 eV (corresponding to 150-195 eV in the rest frame of the scattered Ar ion) is consistent with electron emission $(L_{2,3} MM)$ from an Ar ion that is already up to six times ionized.²¹ This would result in final charge states up to seven and is consistent with the corresponding charge state data.^{21,22} There is evidence in Fig. 3 for a high-energy shoulder due to the emission of electrons in the 250-500-eV energy range; however, it is not possible from the present data to determine their origin. This is not a likely range in which to find Auger electrons from Kr, but it is the same range of energies as found for MO electrons from Kr-Kr and Kr-Xe collisions.¹⁰ (It is such a process, the filling of a MO during the collision, to which Shanker et al. attribute all the electrons of Fig. 3, including the prominent peak which this paper shows to be due to Ar L Auger emission after the collision.) Auger electrons created by the filling of 2s vacancies in up to six times ionized Ar would have energies in the range of 180-285 eV.²¹ For electron energies above 400 eV, electrons from the filling of two 2p vacancies with the subsequent ejection of a single electron might be a contributing factor. The analogous process for photon emission is well known. 23,24

III. DISCUSSION

In light-ion-atom collisions the electron emission spectra generally show distinct and well-resolved Auger peaks, while for the very heavy-ion-atom systems such as Kr-Kr and Kr-Xe, the spectra are dominated by a band of elec-



FIG. 3. The data of Fig. 1 are shown after subtraction of separate exponential functions which were fitted to the low- and high-energy portions of the spectra. The prominent peak is attributed to Ar L Auger electrons from ions that are up to six times ionized prior to the Auger decay.

trons with energies extending to as high as 1000 eV.¹⁰ For these very heavy systems, Auger transitions in the separated atoms following the collision contribute relatively little to the final ionization state.²⁵ Nearly all the ionization occurs during the collision through some presently unknown or unknown combination of rapid processes. The Ar-Kr system was chosen for this investigation as an intermediate case and the spectra show Auger peaks superimposed on an electron continuum. The origin of the continuous portion of the spectra is uncertain. As noted in the Introduction, there are several mechanisms which might contribute to it, especially for electrons with energies of less than 100 eV. Molecular-orbital electrons are likely to contribute to the continuum, but do not appear to be the source of the peak in the 125-200-eV range. The origin of the high-energy tail above 500-800 eV is not known; the counting rates were very low and they could be partially due to beam-associated background counts.

It is clear for this intermediate case that the spectra are not dominated by the continuous portion of the spectra to the degree found for the heavier Kr-Kr and Kr-Xe systems. For the Kr-Kr system, analysis of the Kr L Auger energies showed the Auger transition took place after the ions were already 10-12 times ionized. In other words, 20-24 electrons, presumably those comprising the continuous portion of the electron emission spectra, were removed from the collision complex before Auger decay took place in the separated ions.²⁵ The Ar-Kr data presented here are similar in that the Auger peak is shifted to lower energies by an amount that corresponds to the Ar ion being up to 6 times ionized when the Ar L Auger decay occurs. 670

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