Evidence for Correlated Double-Electron Capture in Low-Energy Collisions of O^{6+} with He

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Production of *LMM*-Auger and $L_1L_{23}M$-Coster-Kronig electrons in 60-keV O^{6+} + He and 40-keV C^{4+} + He collisions was measured with high resolution by the method of 0° Auger spectroscopy. Oxygen impact is found to create intense Coster-Kronig lines attributed to the configurations $1s^22pnl$ with $n \geq 6$ which are produced by double-electron capture. Strong evidence is provided that the double-capture process involves electron-correlation effects whose analysis leads beyond the independent-electron model.

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Studies of electron-capture processes in collisions of slow, multicharged ions with atoms are currently receiving considerable attention. This interest has been generated to a large degree in the fields of plasma physics, thermonuclear fusion research, and astrophysics where the collisional properties of highly stripped ions play an important role. Most of the experiments have been devoted to the study of low-energy *single*-electron capture[1,2] which is characterized by the selective population of only a few final states of the projectile. This result has been found to agree well with, for example, the classical over-barrier model,[1,2] showing that the essential features of the single-capture mechanism are now understood.

Less information is available about the mechanisms involving *double*-electron capture.[3-7] Crandall *et al.*[3] have shown that double-electron capture can be an important process in low-energy, highly ionized collision systems. Under the assumption that the electrons are transferred independently from each other, both electrons are expected to be captured in essentially the same shell of the multicharged ion where configurations of equivalent (or nearly equivalent) electrons are created. The picture of producing equivalent electron configurations, however, does not involve electron-correlation effects which may cause deviations from the predictions of the independent-particle model.[8]

The search for experimental evidence for correlated two-electron processes has recently received considerable attention. In high-energy ion-atom collisions, Tanis *et al.*[9] and Anderson *et al.*[10] have studied electron-correlation effects in transfer-excitation and transfer-ionization collisions at incident energies of several megaelectronvolts. At the low energies more relevant for our study, the concept of correlated two-electron processes has been outlined by Brenot *et al.*[11] within the framework of diabatic potential curves. In that framework transitions caused by electron correlation occur at crossings of potentials curves which differ by *two* spin orbitals. The problem of correlated electron transitions in highly ionized systems has been theoretically treated by Grozdanov and Janev,[12] and by Kimura and Olson.[13]

In the present work we provide strong evidence for correlated double capture in slow, highly ionized collision systems. It is shown that at low collision energies there is a direct method to observe the correlation effects in two-electron transfer processes, i.e., by analysis of the production of nonequivalent electron configurations in which one or more electrons are in high Rydberg states. The principles of the present method are illustrated in Fig. 1 which shows the orbital energies of the O^{6+} + He system. In the incident channel two electrons occupy the He $1s$ orbital whose energy decreases strongly as the internuclear distance to the oxygen ion decreases. At ~ 4 a.u. the uncorrelated double-capture process may occur by two sequential single-electron transitions. As the collision partners continue to approach each other, resonance conditions are created for the correlated double-capture process where one electron is transferred into the $2p$ state and another electron is transferred into a Rydberg state. After the collision the doubly excited projectile decays by autoionization, ejecting an electron.

It is emphasized that the *LMM*-Auger electrons (Fig. 1) can arise either from two sequential single-electron transitions or from a correlated two-electron transfer.[3] On the other hand, the production of the $L_1L_{23}M$-Coster-Kronig electrons is expected to result only from correlated double-electron capture. The observation of these Coster-Kronig electrons produced in slow, highly ionized collision systems is reported here for the first time. Bordenave-Montesquieu *et al.*[5] have made extensive measurements of the *K*-Auger series of multicharged nitrogen where double capture into the configurations $2lnl'$ was observed for $n \leq 5$. The corresponding Auger lines were found to be relatively weak. In the present work it is shown that for $n \geq 6$ the correlated double-capture process is at least as important as the sequential single-capture mechanism.

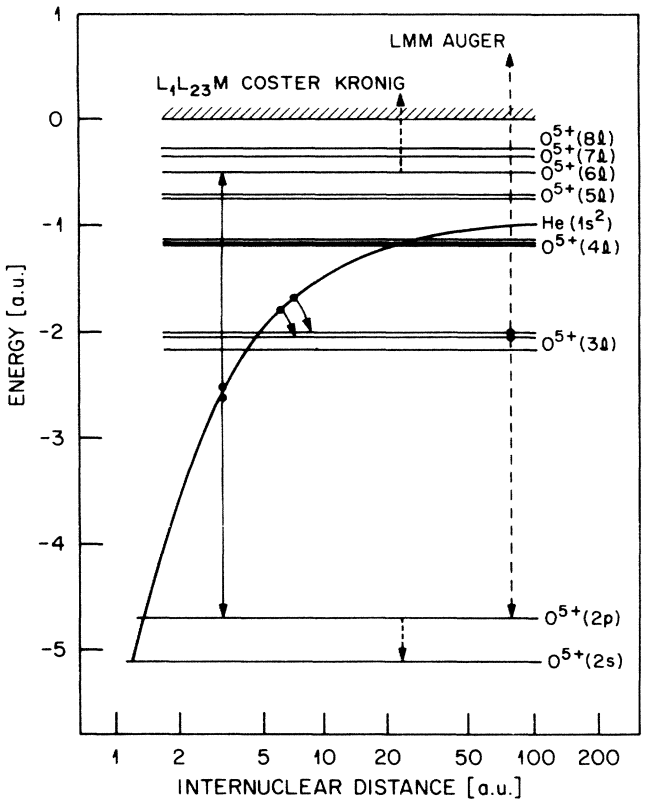

FIG. 1. Diagram of orbital energies for the system $O^{6+}+He$. Note that in the Coster-Kronig transitions an electron is ejected from a higher n state due to an electron transition from one of the $2p$ states to the $2s$ state. In *LMM*-Auger decay, an electron is ejected from the M (or higher) shell due to an electron transition from the M shell to the L shell. The electron binding energies in O^{5+} are obtained from the Rydberg formula $B_n = \mathcal{R}\, Q^2/n^{*2}$ where n^* involves an appropriate quantum defect (see also text).

The experiments were carried out at the Oak Ridge National Laboratory Electron Cyclotron Resonance Ion Source[14] by use of the 0° Auger spectroscopy apparatus which was temporarily transported from the Hahn-Meitner-Institut, Berlin. The apparatus has been described before[15] so that only a few details are given here. Ions of 60-keV O^{6+} and 40-keV C^{4+} extracted from the ion source were magnetically analyzed and directed into a He gas cell located in the scattering chamber. After collimation of the beam to a diameter of 2 mm, currents of typicallly 10 nA were obtained. Electrons produced in the gas cell were measured at a 0° observation angle by a tandem electron spectrometer consisting of two electrostatic parallel-plate analyzers. The entrance analyzer was used as a deflector to steer the electrons out of the ion beam, and the exit analyzer determined the energy of the electrons with high resolution. In this experiment the resolution was typically 0.3 eV (FWHM), obtained by deceleration of the electrons before entering the exit analyzer.

Care was taken to maintain single-collision conditions. The pressures in the 5-cm-long target cell and in the scattering chamber were about 5×10^{-4} and 5×10^{-6} Torr, respectively. The base pressure was below 5×10^{-7} Torr. The fraction of charge states other than the primary one present in the incident beam was estimated to be smaller than 5%. Measurements made over a range of gas pressures verified a linear pressure dependence of the electron yield.

From the observed electron yield, corresponding absolute cross sections were determined by methods described previously.[14] In particular, cross sections were measured for the production of Coster-Kronig electrons whose laboratory-frame energies were as low as 5 eV. Since the low-energy electrons are readily disturbed by spurious instrumental effects, the gas cell was biased by −50 V to accelerate the electrons out of the target region. The focusing effects possibly produced by the electron acceleration were investigated by a change in the bias voltage and found to be negligible within the experimental uncertainties of 30% determined for the measured cross sections.

Figure 2 shows typical electron spectra obtained for the systems 60-keV $O^{6+}+He$ and 40-keV $C^{4+}+He$. The data are transformed from the laboratory into the projectile rest frame. Superimposed on the continuous background which may contain contributions from instrumental effects, the spectra exhibit Coster-Kronig lines. Each line is attributed to a certain quantum number n of the configuration $1s^2 2pnl$. The line energies are obtained from $\epsilon_n = \Delta E_{2s2p} - B_n$, where ΔE_{2s2p} is the energy difference between the $2s$ and $2p$ orbitals and B_n is the binding energy of the Rydberg electron. The binding energy of the Rydberg electron is obtained from the well-known formula $B_n = \mathcal{R}\, Q^2/n^{*2}$ where Q is the effective nuclear charge, n^* involves the quantum defect, and $\mathcal{R}$ is the Rydberg constant. The series limit occurs at ΔE_{2s2p} which is equal to 12.0 eV for oxygen and 8.0 eV for carbon. The cross sections for the production of the Coster-Kronig electrons which are energetically allowed for $n \geq 6$ in oxygen and $n \geq 4$ in carbon were determined to be 3.3×10^{-17} and $\sim 5\times10^{-19}$ cm^2, respectively. These results refer to the projectile frame of reference where the electron emission is assumed to be isotropic.

In addition to the Coster-Kronig electrons we measured L-Auger series between 15 and 75 eV. Oxygen impact yields the cross section of 4.6×10^{-17} cm^2 which represents an upper limit for the uncorrelated double-capture cross section. In the present experiment the corresponding cross section for the C^{4+} impact cannot be accessed since the electrons are transferred into the $n=2$ shell so that no autoionizing state is produced (provided that the ion is incident in its ground state). The double-capture cross section for

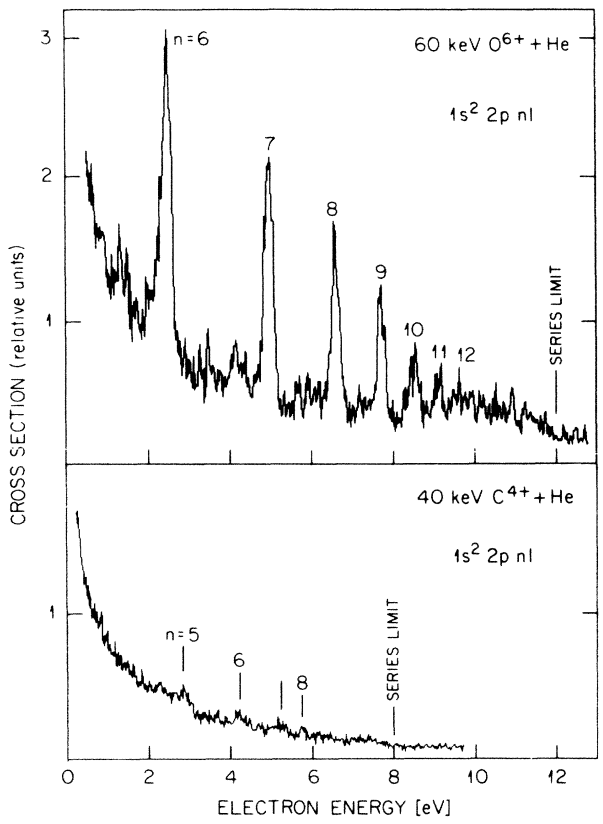

FIG. 2. Spectra of Coster-Kronig electrons produced in 60-keV O^{6+} + He and 40-keV C^{4+} + He collisions. The relative cross sections and the energy scale refer to the projectile rest frame.

the system 40-keV C^{4+} + He obtained from the work by Crandall *et al.*[3] is 16×10^{-17} cm^2. This value is seen to be of the same order of magnitude as the one obtained for oxygen impact. In contrast, the cross sections for producing the Coster-Kronig electrons given above for the two collision systems differ by about a factor of 65.

To interpret the present observations we use the potential-curve diagrams in Fig. 3. The diagram indicates crossings between states which differ by one spin orbital and by two spin orbitals (denoted diabatic I and diabatic II by Brenot *et al.*[11]). At the first type of crossings, transitions are caused by one-electron interactions such as radial coupling, whereas a transition at the second type of crossing requires a two-electron interaction such as electron correlation[8] (if we assume orthogonal states).

It is important to note that the occurrence of the resonance condition for correlated double capture indicated in Fig. 1 corresponds to a crossing in the corresponding potential-curve diagram (Fig. 3). The system O^{6+} + He is seen to involve an infinite number of such crossings between the entrance channel and exothermic exit channels leading to the nonequivalent electron configurations $1s^22pnl$ in oxygen. In the system C^{4+} + He, on the other hand, the $1s^22pnl$ channels are endothermic, and thus do not lead to crossings. We believe that this difference in the potential-curve structure explains the significant (factor of ~ 65) difference observed in the production of the Coster-Kronig electrons for these two systems. It should be recalled that the Coster-Kronig electrons are

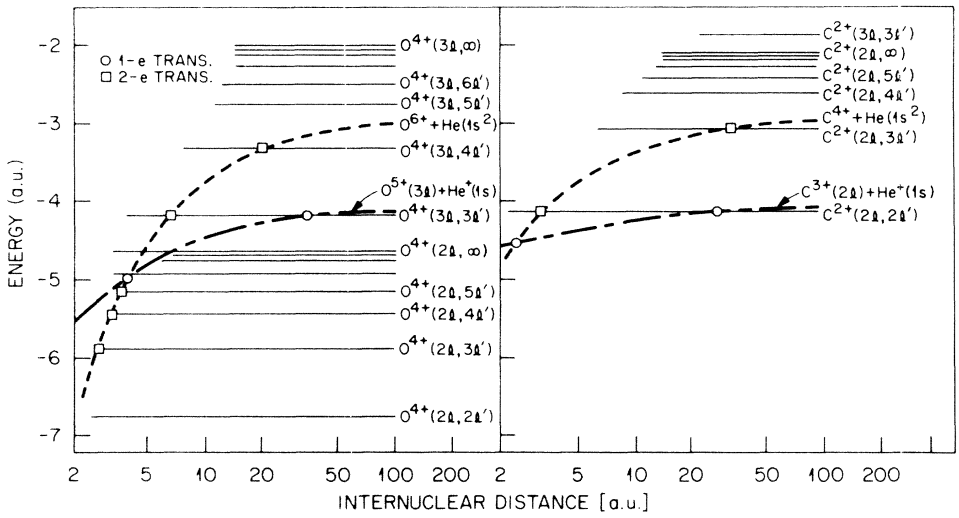

FIG. 3. Potential-curve diagrams for the systems O^{6+} + He and C^{4+} + He. To permit comparison with previous potential-curve plots (Refs. 3, 6, and 7) it is noted that the energy q/R was subtracted from all potentials (where $q=8$ for oxygen and $q=4$ for carbon) to produce horizontal lines for the projectile final-state potential curves.

expected to result only from correlated double-electron capture.

Finally, it should be pointed out that strong Coster-Kronig lines were expected and also observed for the system 40-keV $C^{4+} + H_2$, where, because of the reduced target-ionization potential, crossings leading to the nonequivalent configurations $1s^2 2pnl$ are present. This is consistent with our finding that the production of Coster-Kronig electrons by correlated double capture depends critically on the collision system and that on the basis of the curve-crossing model it is possible to predict those collision systems where they will be produced. Also, it should be recalled that apart from the two sequential single-electron transitions, the equivalent electron configuration $1s^2 3l 3l'$ of oxygen (Fig. 3) may be produced by a correlated two-electron transition. In this case the spatially correlated Wannier states postulated by Datz *et al.*[16] are possibly created.

In summary, we have provided strong experimental evidence for the significant role of correlated two-electron capture during the collision of a highly charged ion with a neutral atom. In view of the present results it would be interesting to verify the relative contributions of the different correlated and uncorrelated capture processes. Explicit calculations of correlated double-electron capture would be desirable in elucidation of the proposed mechanism.

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