Close-coupling study of K-shell-vacancy production in near-symmetric collisions

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The impact-parameter-dependent K-shell-vacancy-production probability for $F^{8+}+Ne$ and $S^{15+}+Ar$ collisions is studied with use of the modified atomic-orbital (AO+) expansion model. It is shown that the AO+ model predicts results in good agreement with experimental data. The discrepancies at low energies are discussed and further experimentation is suggested.

The main features of inner-shell excitation in ion-atom collisions are generally well described within the molecular-orbital (MO) expansion model for slow collisions and within the two-center atomic-orbital (AO) expansion model for collisions at intermediate velocities. In the transitional energy region between the conventional MO and AO domains, it is less clear that a simple physical model should be appropriate.

Recent experiments with highly ionized projectiles have explored this transitional energy region.[1] In addition to total vacancy-production cross sections, detailed information such as the impact-parameter-dependent electron-transfer probabilities out of the K shell of the target to the projectile orbitals are now available.[2,3] These measurements naturally call for a more sophisticated theoretical description.

In this Brief Report, we demonstrate that the modified atomic-orbital (AO+) expansion description[4,5] constitutes a powerful means to describe details of the collision dynamics in the transition energy region. The conventional AO expansion method is frequently regarded as appropriate for collisions at intermediate velocities but not for slow collisions since, in the method, relaxation of the electronic orbitals during the collision is not taken into account. Noting, however, that MO's at large internuclear separations can be represented by two-center AO expansions [linear-combination-of-atomic-orbitals (LCAO) method] with AO's of the separated atoms, it is obvious that conventional AO expansions are, in fact, adequate for describing processes at virtually any velocity at large impact parameters. On the other hand, MO's at small internuclear separations are not adequately represented by the LCAO method. There, however, MO's can be expanded in a set of united-atom orbitals. Therefore, in a modified atomic orbital (AO+) expansion model, we have included the AO's of both the separated atoms and the united atom, the latter positioned at the collision centers. With such expansion sets, the low-energy molecular features of the collision system are indeed well represented, as has been demonstrated[4] by constructing molecular correlation diagrams from AO+ expansion sets. In dynamical calculations within the AO+ expansion model, therefore, a broad range of energies from the intermediate- to low-velocity region can be covered, as was demonstrated in calculations for several collision systems.[5] We point out that, in atomic-orbital expansion calculations, the formal difficulties of using molecular electron translational factors are avoided.

To date, the AO+ expansion method has been applied mainly to outer-shell processes, and details of the description have been presented elsewhere.[4,5] To apply this method to $F^{8+}+Ne$ and $S^{15+}+Ar$ collisions, we need to make a further assumption by adopting the active electron approximation. We will consider transfer of the K-shell electron of the target to the shells of the projectile and assume that all the other electrons remain inactive except for providing a constant screening. The validity of this assumption will be discussed below in conjunction with the comparison of calculated results with experimental data.

In this study the collision Hamiltonian is derived from the atomic potential V_i ($i=T,P$) of the target and the projectile, respectively, as

$$H=T+V_T+V_P$$

in a single-electron potential model (T, the electronic kinetic energy operator). In the following the results of the calculations are presented for $F^{8+}+Ne$ and $S^{15+}+Ar$ collisions and compared with recent experiments.

(i) $F^{8+}(1s)+Ne$. The impact-parameter dependence of Ne K-shell-vacancy-production probability by $F^{8+}(1s)$ projectile has been measured by Hagmann *et al.*[2] at 4.4 and 10 MeV. With the orbital speed of the K-shell electron defined by $v_K=\sqrt{-2E_K}$, where E_K is the neon $1s$ orbital energy in atomic units, these two collision energies correspond to $v/v_K=0.38$ and 0.57, respectively.

In Fig. 1 we show the K-shell vacancy production probability versus impact parameters as measured by Hagmann and co-workers and compare with the results of close-coupling calculations using the conventional two-center two-state AO expansion and the present AO+ expansion. The potential V_T is the Hermann-Skillman[6] potential fitted to an analytical form. In the conventional AO expansion, only the $1s$ orbital on each center has been retained. In the AO+ expansion, additional $1s$, $2s$, and $2p$ orbitals of the *united atom* (UA) are included. For this quasiresonant system, the dominant inelastic process is the K-shell to K-shell charge transfer, and the probability for this process extends over a range much greater than the neon K-shell radius $r_K\approx 0.1$ a.u. The overall agreement of the two calculations with experiment is quite good, especially for collisions at large impact parameters where the two calculations coincide (the UA orbitals have no effect for collisions at large impact parameters). A discrepancy between the two calculations occurs only at small impact parameters, particularly for $E=4.4$ MeV. At this latter energy, the oscillation of experimental $P(b)$ vs b is better reproduced by the AO+ model

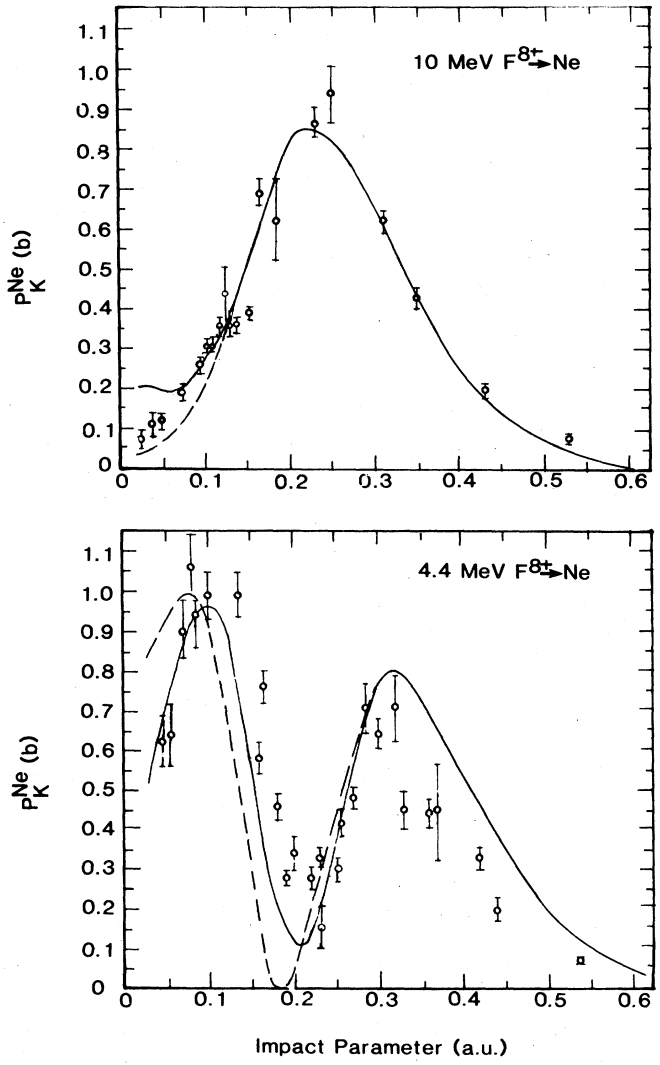

FIG. 1. Neon K-shell vacancy production probabilities vs impact parameters for F^{8+} collisions at 10 and 4.4 MeV. Experimental data are from Hagmann *et al.* (Ref. 2). The dashed lines are from the two-state, two-center atomic expansion calculations and the solid lines are from the present 10-state AO+ model calculations. At large impact parameters the two results coincide.

where the important relaxation effect of the electronic charge cloud is accounted for by the UA orbitals included. Another noticeable feature of the AO+ result is that the maximum and the minimum of the vacancy production probability no longer reach unity and zero, respectively. This is due to the effect of $2p\sigma - 2p\pi$ rotational coupling which is included in the AO+ model through the united-atom $2p$ orbitals. This rotational coupling also gives a small shoulder at $b \sim 0.03$ a.u. at $E = 10$ meV, but this is not evident in the experimental data.

The present situation is not unlike that of resonant charge transfer in $H^+ + H(1s)$ collisions.[7] There the two-state AO expansion is quite adequate for describing charge transfer at impact parameters $b \geq 1$ a.u. Since total electron capture is dominated by contributions from large impact parameters, the total electron capture cross section can be accurately predicted by the simple two-state AO expansion model. On the other hand, collisions at small impact parameters require proper account of electron relaxation for $E \leq 25$ keV. Conventional AO expansion models fail to explain excitation and charge transfer to $2s$ and $2p$ states for $v \leq 1$ a.u. because these processes occur mostly at small impact parameters.[4(a)] The experiment of Helbig and Everhart[8] where charge transfer probability versus collision energies for $H^+ + H(1s) \rightarrow H(1s) + H^+$ at the fixed scattering angle of 3° was measured cannot be explained quantitatively in a conventional AO expansion model because of the failure of considering the relaxation effect at small impact parameters. Such a relaxation effect cannot be accounted for by including more excited separated-atom orbitals,[9] but is well described by the AO+ model[4(b)] (as well as triple-center expansions[7]).

(ii) $S^{15+}(1s) + Ar$. The impact parameter dependent K-shell vacancy production probability for this system has been measured recently by Schuch *et al.*[3] The experimental collision energies correspond to $v/v_K = 0.16$, 0.21, 0.34, 0.41, and 0.69, thus covering a broader range of velocities than the study for the $F^{8+} - Ne$ system. In order to be able to represent the $S^{15+} + Ar$ system over this energy range, a larger AO+ expansion set has been employed than in the former system, consisting of the Ar $1s$, the S^{14+} $n = 1, 2$ shell orbitals, and the $n = 1, 2$ united-atom orbitals at both collision centers. With this basis set, transitions from the target K shell into the projectile K and L shells are taken into account (the latter corresponding to $2p\sigma - 2p\pi$ rotational coupling transitions in the MO expansion description). In test calculations for the model system $S^{16+} + Ar^{17+}$ with an analog of this basis set, we have closely reproduced the results[10] of the direct numerical solution of the Schrödinger equation for that system. It appears, therefore, that the basis set should lead to near-converged transition probabilities for the $S^{15+} + Ar$ system, too. As potentials V_P and V_T, we have chosen the atomic potentials by Garvey, Jackman, and Green,[11] where the η parameter is adjusted to reproduce the Hartree-Fock $1s$ binding energy in the Ar case.

In Fig. 2 we show the measured Ar K-shell excitation probabilities (full circles) of Schuch and co-workers[3] and compare with various calculations. (1) The AO+ model. (2) The two-state AO model. Only the $1s$ orbitals of Ar and S^{14+} are included in this expansion. (3) The two-state model with empirical matrix elements.[12] In this approach, the AO matrix elements are fitted to reproduce static MO potential curves. Therefore, the Hamiltonian used in this calculation is different from the Hamiltonian used in calculations (1) and (2). (4) The direct integration of the time-dependent Schrödinger equation[10] (open circles). This calculation is performed for the one-electron system $S^{16+} + Ar^{17+}$.

By comparing the calculated results with experimental data, we notice that the AO+ description gives the best overall agreement with experiment at the three energy points shown. The simple two-state AO results are also in reasonable agreement with experimental data, but the positions of the maxima and minima are slightly shifted. The calculation based upon model matrix elements[12] is not very satisfactory, especially at 32 MeV. In this model, the other electrons are assumed to be relaxed so that a "molecular" Hamiltonian was employed. The discrepancy at higher energies may be considered an indication that relaxation of outer electrons is not present then.[13]

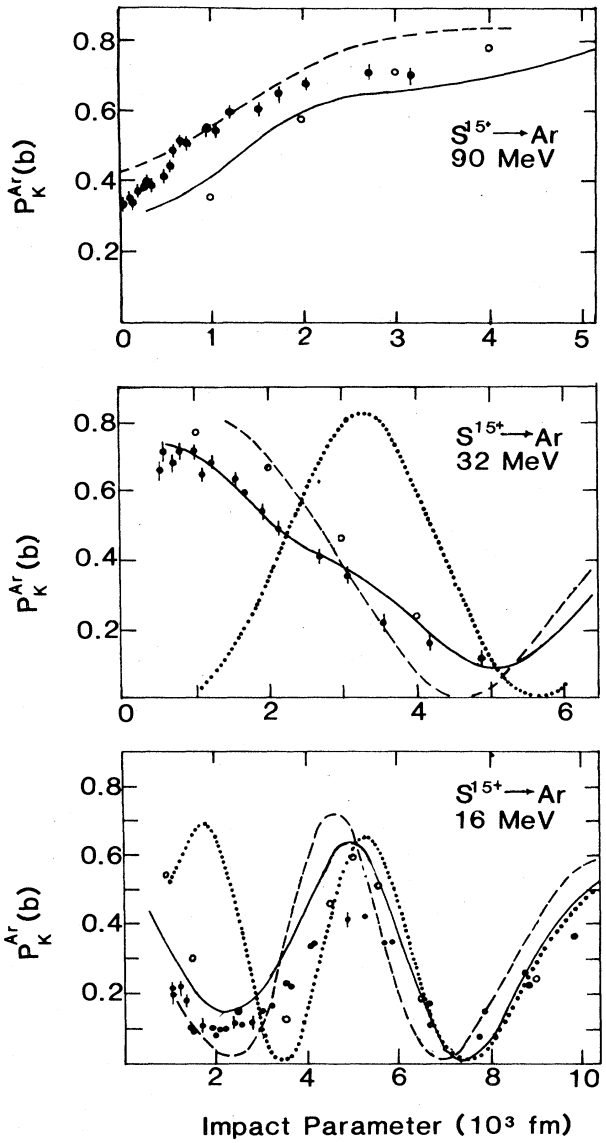

FIG. 2. Argon K-shell vacancy production probabilities vs impact parameters for S^{15+} collisions at 90, 32, and 16 MeV. Experiment: ●, Schuch *et al.* (Ref. 3). Theory: – – –, two-state AO; ——, present 16-state AO+; · · ·, two-state empirical MO (Ref. 13); ooo, TDSE (Ref. 11). (See text.)

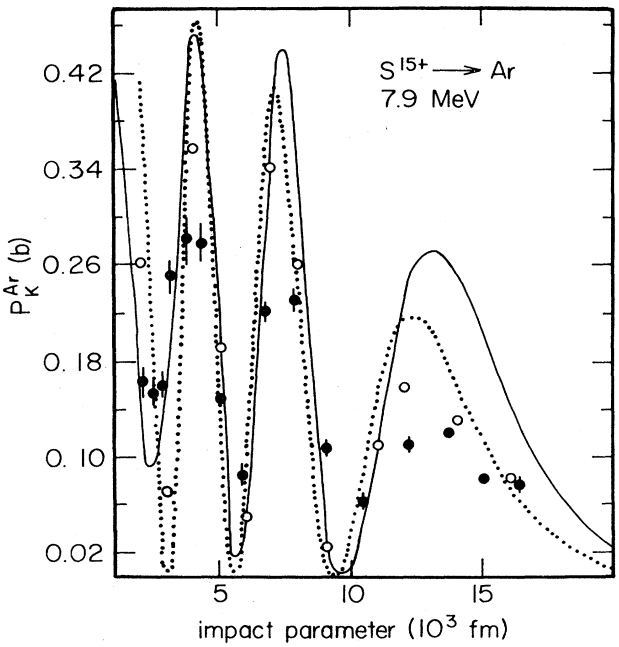

FIG. 3, As in Fig. 2, but for 7.9 MeV S^{15+} ions.

We notice that at 16 MeV, the amplitudes of oscillations in the transfer probability are not well reproduced by any theoretical calculations neither in the maxima nor in the minima. This discrepancy is even more evident at $E=7.9$ MeV, as shown in Fig. 3. At this energy the theoretical results exhibit much more pronounced extrema than the experimental data. Similar discrepancies also appear for $F^{8+}+Ne$ collisions at 2.5 MeV,[14] or $v/v_K=0.29$. It is doubtful that such discrepancies could be understood by changing the model Hamiltonian *alone,* since three of the calculations in Fig. 3 use different model Hamiltonians but each shows similarly sharp structures. In slow, close collisions one might suspect that the one-electron picture is no longer adequate. Particularly since the rotational couplings are important, the K-K transition could be partly blocked by transitions from the target L shell into the projectile K shell. Furthermore, transitions by the second target electron (with spin parallel to the spin of the projectile electron) into the projectile L shell will become important. Furthermore, one might suspect that the reduction of experimental angular distributions to impact-parameter dependences is questionable at low energies. Since multiple ionization and capture of outer-shell electrons are probably quite large for each single K-shell capture event at low collision energies, the relation between scattering angles and impact parameters will depend on the charge state of the final products. Without such an analysis, the superposition of several oscillatory structures from different charge states of the products will tend to smear out the structure. It is desirable to carry out coincidence measurements in which the final charge state of target and projectile is analyzed. We expect that the transfer probability in these coincidence measurements would show sharper oscillatory structure as predicted in theoretical calculations. On the other hand, it is still to be seen whether the coincidence results can be explained in terms of the simple one-electron model employed here.

In summary, we have shown that the AO+ expansion model provides accurate predictions on the impact-parameter-dependent K-shell-vacancy-production probability over a broad velocity region. Remaining discrepancies in slow collisions are discussed and coincidence experiments with charge state analysis of the final products are suggested for resolving these discrepancies.

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