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One- and two-step double-electron capture in slow Ar^{6+}-He collisions

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Double-electron capture by slow (15–30 eV/u) Ar^{6+} ions from He has been studied by simultaneous measurements of postcollisional projectile energy-gain and angular distributions. The recorded spectra are interpreted with the aid of a semiclassical collision model. We find evidence for distinguishable one- and two-step double-electron capture in the energy region investigated.

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I. INTRODUCTION

We recently [1] reported on measurements and calculations of very slow (1.6–13 eV/u) *single*-electron charge-exchange collisions of Ar^{6+} and He. It was found that the experimentally measured angular-differential cross sections could not be reproduced by multichannel Landau-Zener (MCLZ) calculations unless single-capture channels correlating to core-excited states of Ar^{5+} were included. The reason for the relatively large importance of such states is due to a significant mixing of the dominantly populated $3s^24p\ ^2P$ and $(3s3p\ ^{1,3}P)3d\ ^2P$ Ar^{5+} configurations.

In this paper we report corresponding measurements and theoretical calculations for *double*-capture channels at slightly higher energies.

Two-electron capture in multicharged systems can take place via both one- and two-step processes [2]. In the one-step process the capture of the two electrons takes place at one single well-localized potential-energy curve crossing, while the two-step process goes via two consecutive one-electron transitions at separated crossings, and requires a single-capture channel as mediator. These two cases can be distinguished in an angular spectrum when the two-step process gives rise to larger scattering angles [2, 3]. Recent studies [4] of the relative importance of one- and two-step processes have stimulated much discussion of the role of electron-electron correlation in two-electron transfer. As will be seen, the Ar^{6+}-He collision system has proved to be very helpful in clarifying several important issues raised [4], since it turns out that the relative strength of one- and two-step processes can be controlled by varying the collision energy in the 15–30-eV/u region, and because distinctive signatures of each process can easily be separated in the simultaneously recorded energy-gain and angular-differential spectra.

One-electron charge transfer to moderately charged projectiles ($q < 10$) usually populates a limited number of states of the ion. The diabatic potential-energy curves correlating to these states cross the initial potential curve in a range of internuclear distances R, which depends on the collision energy and the details of the collision system. The range of these favored crossing distances (the reaction window [5]) shifts to smaller R when the collision energy increases. The reaction window for the kind of collision system we are considering here is typically situated in the R =3–10-a.u. range. Channels with crossings outside the reaction window are not appreciably populated. In the present collision system and energy range, the single-capture channel correlating to $Ar^{5+}(3s^24s\ ^2S)$ (cf. Fig. 1) is on the inner edge of the reaction window. The probability for capture to this channel is therefore fairly sensitive to the collision energy. When a crossing starts to change its behavior from completely adiabatic to more active, new reaction paths open up. The flux in the incident channel can pass through the crossing, which at lower energies acted as a repulsive wall, and reach the region inside this crossing. Referring to Fig. 1, this corresponds to a situation where the part of the initial channel between the $3d$ and $4s$ crossings can be traversed with non-negligible probability. If a

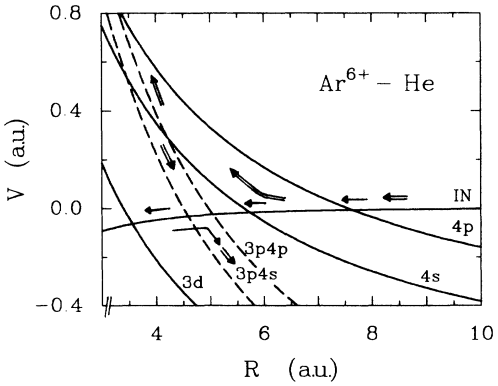

FIG. 1. Section of the relevant diabatic potential-energy curves of the $(ArHe)^{6+}$ system. The principal one-electron capture curves labeled according to their dissociation limit on Ar^{5+} ($3d, 4s$, and $4p$) (see also Ref. [1]) are shown together with the two-electron capture curves correlating to $Ar^{4+}(3p4s)$ and $(3p4p)$, respectively.

double-capture channel crosses the initial channel in this region, as drawn in Fig. 1, this means that a one-step two-electron transition in principle becomes possible. At lower energies, when the $4s$ crossing is strongly adiabatic, the double-capture channel can only be reached by two consecutive one-electron transitions. By a simultaneous measurement of scattering angle and energy gain we can clearly distinguish between one- and two-step double capture, and thereby we are able to reconstruct parts of the detailed collision dynamics.

II. EXPERIMENTAL TECHNIQUE

The experimental setup has been described in detail earlier [3,6], and we give only a short review here. A fast heavy-ion beam collides with an Ar-gas target to produce very slow Ar recoil ions. After extraction from the collision cell and acceleration, the recoil ions are mass-to-charge-state analyzed by a Wien filter, retarded, collimated, and directed onto an effusive gas-jet target. Post-collisional analysis of the slow Ar^{6+} beam is performed by a cylindrical electrostatic analyzer. The charge states, scattering angles, and energies of the ions are registered simultaneously on a two-dimensional position-sensitive detector. The locations of the registered ions on the detector are linearly related to the energy gain ΔE of the scattered projectile in the plane of deflection and to the scattering angle θ in the perpendicular direction. Thereby it is possible to connect distinct features in the two-dimensional scattering distribution to a certain θ and the corresponding ΔE. In the present work this information was used to establish a relation between ions scattered into small angles with large energy-gain values and, on the other hand, large scattering angles with small energy gains.

III. RESULTS AND DISCUSSION

The angular distributions of double capture are shown in Fig. 2. The common features are a forward peak separated from a distribution at much larger angles. Using the simultaneously recorded energy-gain spectrum, we can relate the small angles to energy gains of $\Delta E \sim 40$ eV and the large scattering angles to energy gains in the region $\Delta E \sim -5$ to 20 eV, depending on the collision energy. Due to the kinematics of the collision system (heavy projectile, light target) and the low collision velocity, projectiles scattered through large angles are recorded at small energy-gain values. In Fig. 3 the energy-gain spectra corresponding to Fig. 2(b) are plotted for projectiles scattered into angles (a) between 1.5° and 5° and (b) into the forward direction within ±1°. By inverting the kinematical relation [7] connecting the energy gain to the scattering angle, we find that both the small- and large-angle peaks derive from capture into states having Q values of about 44±4 eV. Translated to binding energies, this means that states bound with approximately 30 eV with respect to the ionization limit of Ar^{4+} are populated. To our knowledge, the only tabulated Ar^{4+} configuration in this energy range is $3p4s$ [8]. We therefore use an extended Rydberg formula [9] in order to

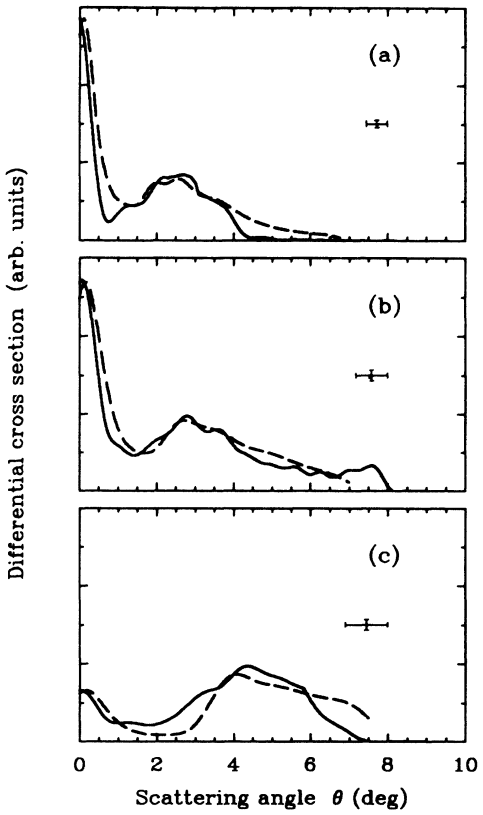

FIG. 2. Angular-differential cross sections of two-electron capture in Ar^{6+}-He collisions at (a) 30, (b) 22.6, and (c) 15.1 eV/u. The solid line represents the experimental two-electron capture distributions and the dashed line represents a MCLZ calculation using the potential curves of Fig. 1. Horizontal and vertical bars mark the angular width of the primary projectile beam and the statistical error, respectively.

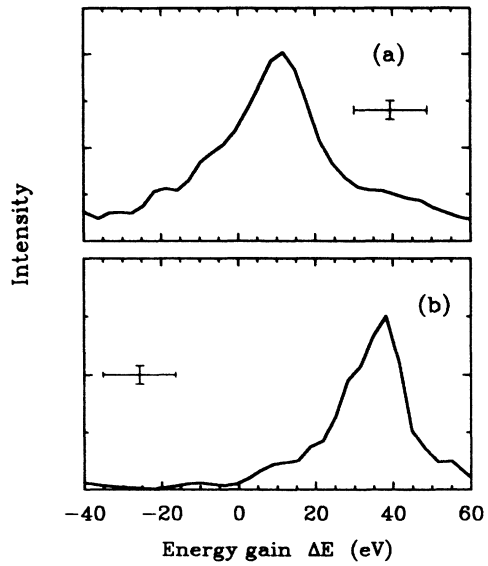

FIG. 3. Energy-gain spectra for two-electron capture in 22.6-eV/u Ar^{6+}-He collisions at (a) large scattering angles (1.5° to 5°) and (b) forward angles within ±1°. The energy resolution and the statistical error are indicated by bars.

assign further possible double-capture states. We find that double capture to the $3p4s$ and $3p4p$ levels of Ar^{4+}, having Q values of 48.3 and 43.6 eV, respectively, are the most likely double-capture channels. These are consistent with an independent-particle picture, in which the first transferred electron goes into the $4s$ or $4p$ orbital and the second electron is captured into the $3p$ orbital without affecting the first electron.

Transfer ionization, which in many cases accompanies double capture (as, e.g., in Ar^{4+}-Ar collisions [3]), is *not* seen in the present energy range for Ar^{6+}-He collisions. This is consistent with the true double-capture states observed (which are well below the ionization limit of Ar^{4+}), and is also expected for capture from a tightly bound target like He.

The two double-capture potential curves (correlating to the $3p4s$ and $3p4p$ configurations of Ar^{4+}) cross the initial potential curve inside the $4s$ crossing (~ 5.8 a.u.), cf. Fig. 1. The collision-energy behavior of the single-capture channel correlating to $Ar^{5+}(3s^2 4s\ ^2S)+He^+$ is very important for the understanding of the recorded double-capture spectra. At the very low energies considered in our previous report [1], this channel is strongly adiabatic and is negligibly populated compared to the active $3s^2 4p\ ^2P$ and $(3s3p\ ^{1,3}P)3d\ ^2P$ channels. The $4s$ channel serves then as the "adiabatic termination" of the initial channel, preventing the probability flux from following the initial diabatic channel inside the $4s$ crossing. At higher collision energies, the $4s$ channel will become increasingly populated and a larger fraction of the flux in the initial channel is thereby also allowed to pass diabatically through the $4s$ crossing. This is the case for the energy region shown in Fig. 2 (15–30 eV/u). The crossing of the double-capture channel with the initial channel can then be reached and one-step two-electron transitions are made possible. The classical trajectory corresponding to simultaneous two-electron capture during the receding stage of the collision results in a rainbow peak at small angles [3, 10].

For the probability flux which has been transferred to the $4s$ potential curve on the way in, a second transition at the crossing between the $4s$ and one of the double-capture curves is possible. This two-step sequential electron-capture process results in large scattering angles. The mechanisms discussed above qualitatively explain the structure of the angular spectra in Fig. 2. The forward peak diminishes as the energy is lowered, consistent with increasing adiabaticity of the $4s$ crossing at lower energies. The probability flux is thereby prevented from reaching the crossing between the initial and the double-capture potentials.

A mechanism for one-step double capture has been suggested by Laurent *et al.* [2]. Close to a strongly active single-capture curve crossing (as the $4s$ crossing in the present collision system), the initial diabatic state contains in reality an admixture of the single-electron capture state. Since the initial channel already contains components of single-capture states, the transfer of an additional electron can proceed in a single step by a higher-order one-electron transition [2] into a double-capture state which crosses the initial channel in the neighborhood of the single-capture channel crossing. This type of mechanism requires thus a certain topology of the potential curve diagram, in which a double-capture channel crosses the initial channel close to a single-capture crossing which is quite adiabatic. The difference between this mechanism for double-capture and our suggested mechanism for transfer excitation [1] is that the former is due to mixing of the initial state with a single-capture state induced by the interaction of states correlating to different centers when the collision partners approach, while the latter involves a mixing of final states on the same center due to configuration interaction present already in the asymptotic region. Both types of mechanisms require, however, that a suitable single-capture state is available. If the one-step process proceeds via the mechanism proposed by Laurent *et al.* [2], it most likely populates the $3p4s$ state. However, other one-step mechanisms, for example, electron-electron interaction [4], cannot be definitively ruled out.

In Fig. 2 is also shown the results of a multichannel Landau-Zener calculation performed with the potentials plotted in Fig. 1. The coupling strengths between initial and one-electron capture channels are identical to those used in Ref. [1]. Six coupling strengths between initial and double-capture channels, as well as between single- and double-capture channels, were varied within reasonable limits in order to obtain the best possible fit to the experimental spectra. Of the six free coupling strengths, two have only marginal influence over the calculated differential cross sections. The two-step transitions mediated by the $4p$ single-capture curve require relatively small impact parameters, giving small contributions to the cross section at scattering angles beyond the position of the large-angle structure in the experimental distributions. Only variations of the remaining four couplings can appreciably alter the shape of the calculated cross section. As can be seen in Fig. 2 it is indeed possible to reproduce the recorded angular spectra very closely. We note, however, that the same set of coupling strengths work well for all the collision energies employed.

Such close agreement is reached only if both one- and two-step double-capture reaction paths are included in the calculations. It would be impossible to account for the forward peak without allowing for one-step double-capture, and, on the other hand, two-step processes are required to reproduce the large-angle structure. In the course of the calculations it was also found that one double-capture channel alone cannot account for the broadness of the large-angle peak. At a given collision energy the relative importance of the large- and small-angle features is sensitive to variations of the free coupling strengths. However, as a function of collision energy, this relation is not greatly influenced by the exact magnitudes of the free couplings (which are the same at all the energies), but is almost exclusively controlled by the adiabaticity versus impact-energy behavior of the $4s$ single-capture crossing.

IV. CONCLUSION

Double-electron capture collisions between Ar^{6+} and He have been studied by simultaneous translational

energy-gain spectroscopy and registration of the projectile scattering angle for collision energies of 15–30 eV/u. We observe an angular distribution characterized by a peak at angles close to 0° separated from a broader distribution at significantly larger angles. By relating the scattering angles directly to the energy gains, both these distinct features can be attributed to capture to states having exoergicities of 44±4 eV, and are given the assignments $Ar^{4+}(3p4s)$ and $Ar^{4+}(3p4p)$. We interpret the small-angle peak as due to one-step transitions at the crossing between the double-capture channels and the initial channel, while the large-angle distribution derives from two-step transitions mediated by the $4s$ or $4p$ single-capture channels. The theoretical picture we have developed, in which the collision-energy variation of the relative importance of one- and two-step double capture is primarily determined by the adiabaticity of the $4s$ single-capture crossing, is in accordance with the experimental observations.

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[1] L.R. Andersson, H. Cederquist, A. Bárány, L. Liljeby, C. Biedermann, J.C. Levin, N. Keller, S.B. Elston, J.P. Gibbons, K. Kimura, and I.A. Sellin, Phys. Rev. A **43**, 4075 (1991).

[2] H. Laurent, M. Barat, M.N. Gaboriaud, L. Guillemot, and P. Roncin, J. Phys. B **20**, 6581 (1987).

[3] C. Biedermann, H. Cederquist, L.R. Andersson, J.C. Levin, R.T. Short, S.B. Elston, J.P. Gibbons, H. Andersson, L. Liljeby, and I.A. Sellin, Phys. Rev. A **41**, 5889 (1990).

[4] N. Stolterfoht, C.C. Havener, R.A. Phaneuf, J.K. Swenson, S.M. Shafroth, and F.W. Meyer, Phys. Rev. Lett. **57**, 74 (1986); **58**, 958 (1987); H. Winter, M. Mack, R. Hoeckstra, A. Niehaus, and F.J. de Heer, *ibid.* **58**, 957 (1987).

[5] M. Kimura, T. Iwai, Y. Kaneko, N. Kobayashi, A. Matsumoto, S. Ohtani, K. Okuno, S. Takagi, H. Tawara and S. Tsurubuchi, J. Phys. Soc. Jpn. **53**, 2224 (1984).

[6] H. Cederquist, C. Biedermann, J.C. Levin, C.-S. O, I.A. Sellin, and R.T. Short, Nucl. Instrum. Methods B **34**, 243 (1988).

[7] H. Danared and A. Bárány, J. Phys. B **19**, 3109 (1986).

[8] C.E. Moore, *Atomic Energy Levels* Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, DC, 1949), Vol. 1.

[9] F.H. Read, J. Phys. B **10**, 449 (1977).

[10] L.R. Andersson, J.O.P. Pedersen, A. Bárány, J.P. Bangsgaard, and P. Hvelplund, J. Phys. B **22**, 1603 (1989).