Total cross sections for electron capture and transfer ionization by highly stripped, slow Ne, Ar, Kr, and Xe projectiles on helium

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Experimental cross sections are reported for electron capture and transfer ionization for Ne, Ar, Kr, and Xe projectiles on He for charge states (q) between 2 and 13 and for projectile energies between (250 and 1000 eV)q. The double-electron capture is found to be typically an order of magnitude weaker than single-electron capture, and to be dominated in most cases by the transfer ionization channel. While gross features of the cross sections can be qualitatively understood, quantitative agreement with available theoretical model calculations is poor.

I. INTRODUCTION

The capture of outer-shell electrons from neutral targets by slow-moving highly stripped projectiles has become the object of numerous experimental programs in recent years. Reasons for this interest include not only the relevance of the process to understanding and diagnosing magnetic fusion energy plasmas[1,2] and to astrophysics,[3] but also the still embryonic stage of our understanding of how best to treat such systems theoretically.[4] Several generalized models have been proposed and have met with some success,[5–8] while specific treatments of particular systems have been limited almost entirely to the case of an atomic hydrogen target. Such calculations are rendered complex by the large number of basis states necessary to describe the high-n systems populated in the capture.

Total cross-section data, with which this paper is concerned, exist for both the one-electron atomic hydrogen target and for multielectron targets. The former case is the cleaner one theoretically and probably the most relevant one from the applications points of view. However, it excludes the possibility of studying multiple capture and transfer ionization (TI). This latter process involves capture accompanied by release of electrons from the target-projectile system.[9–18] For collisions involving heavy targets of Ne and beyond, experiments show that both processes are quite important.[15–22] Such complex multielectron systems are almost certainly intractable in any detailed model calculation, although recent statistical treatments of the processes are quite promising.[23]

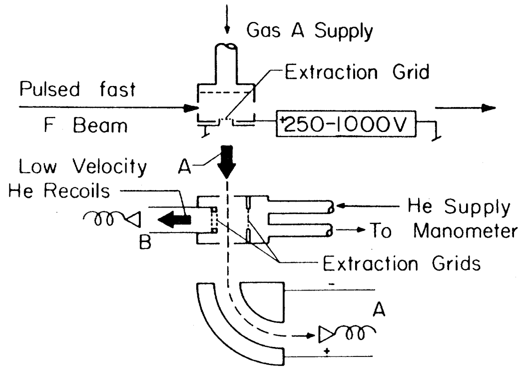

FIG. 1. Schematic of apparatus. Detectors A and B are channeltrons.

The He target case lies intermediate between the simple and complex. Both double-electron capture and TI are possible, yet the existence of only two target electrons should make it possible to treat the system theoretically using eigenfunction-expansion techniques. While few such treatments presently exist in the literature, the increasing availability of data on this target may stimulate work in this direction.

Several previous measurements of total cross sections for electron capture from He by slow, high-q projectiles have been reported.[19–22,24–28] The results reported here lie in a lower energy regime than do most of these, although this is not found to be crucial. In the measurements reported here, we have distinguished experimentally, using coincidence techniques, between normal double capture and transfer ionization, and give separate cross sections for each of these channels. We find the TI to dominate the normal double-capture process in most cases.

II. EXPERIMENT

The experimental apparatus used in the present measurement is shown schematically in Fig. 1 and in more detail in Ref. 29. Pulsed 19-MeV F^{+4} beams from the Kansas State University EN tandem Van de Graaff accelerator, poststripped to charge state 8, and 160-MeV Br^{+12} beams from the Max-Planck-Institut für Kernphysik in Heidelberg, poststripped to charge state 26, were used as "pump" beams in this experiment. The collisions that take place in gas cell A between the fast projectiles and a

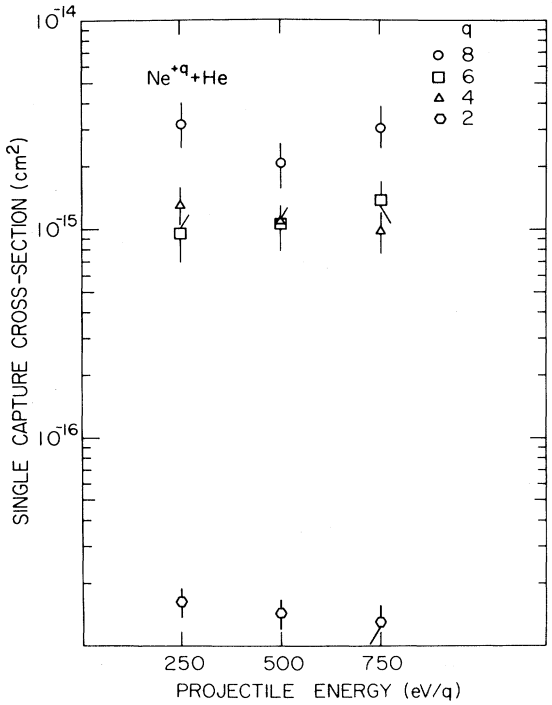

FIG. 2. Representative cross sections for single (p) capture for Ne on He.

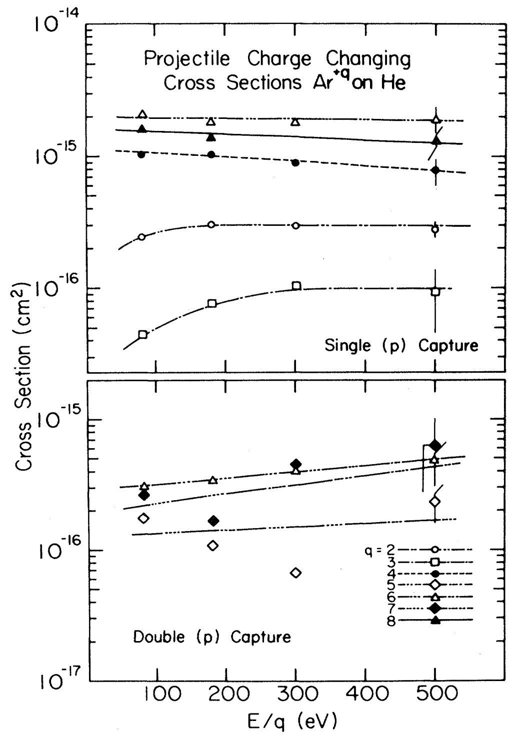

FIG. 3. Representative cross sections for single (p) and double (p) capture for Ar on He.

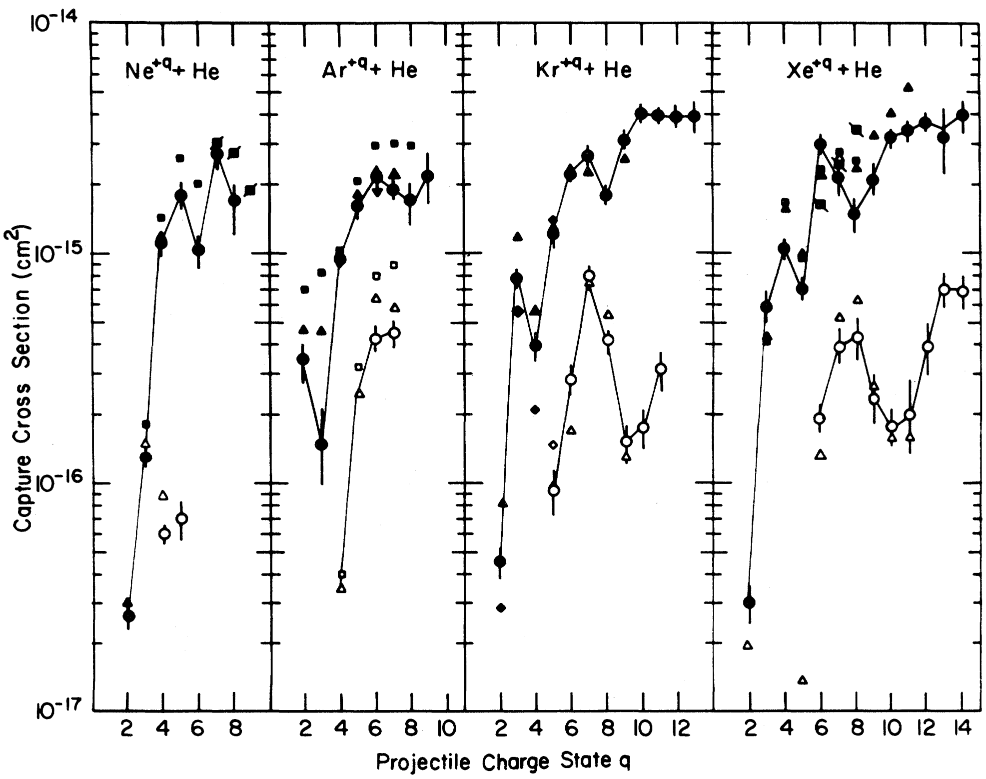

FIG. 4. Cross sections for single (p) (filled symbols) and double (p) (open symbols) capture by rare-gas projectiles on He. Data sources and projectile energies are ●,○, present, $E=(500\text{ eV})q$ for Ne, Ar, and Xe and $(1000\text{ eV})q$ for Kr; ■□, Salzborn *et al.*, Refs. 19 and 20, $E=30$ keV; ▲△, Kusakabe *et al.*, Ref. 21, $E=(1500\text{ eV})q$ (Ne), $(286\text{ eV})q$ (Ar, Kr, and Xe); ■, Iwai *et al.*, Ref. 24, $E=(1500\text{ eV})q$; ◆, Huber and Kahlert, Ref. 26, $E=(1000\text{ eV})q$; ■, Crandall *et al.*, Ref. 25, $E=(10\text{ keV})q$. The solid lines are drawn through the present data to guide the eye.

tenuous (≈ 0.4 mTorr) rare-gas target produce low-energy highly charged recoil ions that are extracted from the collision region by an electric field applied at right angles to the pump beam direction. The recoil ion source as well as the physics involved in recoil ion production are described elsewhere in the literature.[29,30] With the setup shown in Fig. 1 two types of measurement were performed.

(a) *Configuration I.* In the first case a charge-transfer event was characterized by the simultaneous determination of two parameters, the initial and final charge states of the low-energy projectiles. The initial charge state q of each projectile was obtained from the time of flight of the ion from its production in the primary gas cell to its detection by the channeltron A, which is proportional to $\sqrt{m/q}$, where m is the mass of the slow recoil ion. The final charge state q' of the projectile ion, after colliding with the He atoms in the second cell, was determined by sampling, for each ion detected in channeltron A, the electrostatic analyzer voltage at which it was passed. This voltage can be shown to be proportional to q/q', therefore constituting a measure of q', since the initial projectile charge can be inferred from the time-of-flight information. For each event the two parameters described above were recorded by a PDP 11/34 computer with multiparameter data-acquisition capabilities. For normalization purposes, the singles time-of-flight spectra were simultaneously stored by an ungated multichannel analyzer whose dead time could be kept below 3%.

The data obtained under configuration I conditions were analyzed following the procedure described in Ref. 29 and absolute cross sections for single- and double-electron capture by the rare-gas projectiles from the He target and their dependence on projectile energy and charge state were determined. The results thus obtained are presented in Figs. 2—4. In these figures the notation single (p) and double (p) capture cross sections indicate that in this type of experiment only the initial and final projectile charge states were measured.

(b) *Configuration II.* The two-parameter experiment described above cannot distinguish between contributions arising from normal electron capture and those coming from other reaction mechanisms such as direct ionization (DI) and transfer ionization. In the following discussion we will refer to all events which leave the projectile charge unchanged ($q \rightarrow q$) as DI, and to those for which the projectile charge is reduced by one unit ($q \rightarrow q-1$), but He^{+2} is produced, as TI. Events in which the projectile charge is reduced by two units and He^{+2} is produced is referred to as "normal" double capture, although it proves in most cases to be more the exception than the rule. In order to investigate the role played by these processes, a second time-of-flight spectrometer was added to the secondary gas cell (see Fig. 1), and the time of flight of the ionized He target was measured, giving its final charge state q''.

The absolute cross-section scale for the configuration II data was obtained by normalizing these data, for each projectile type, to the configuration I cross section for single capture for a case where this channel was dominant (typically for $q=5$, $q'=4$). This procedure is described further in Ref. 29. For the case of single capture, a single normalization constant proved adequate for placing all of the configuration II single-capture cross sections from a given run on an absolute scale.

For the case of double capture for Kr and Xe projectiles an additional complication arose in that the normalization constant obtained by comparison of configuration I and configuration II data was found to be a monotonically decreasing function of projectile charge state. This comparison is unambiguous for the case of normal ($q \rightarrow q-2$) double capture, since the reduction of the projectile charge by two units must necessarily be accomplished by the loss of both electrons from the two-electron target, and thus produce a He^{+2} ion. We attribute the decrease in the normalization constant to the loss of the He^{+2} ions from the He-extractor system due to the finite energy imparted to these ions in the capture process. Such an effect is expected to be quite small for the extractor geometry used for Ne and Ar projectiles and for single capture, but not for the more slowly moving Xe and Kr ions. It is expected to

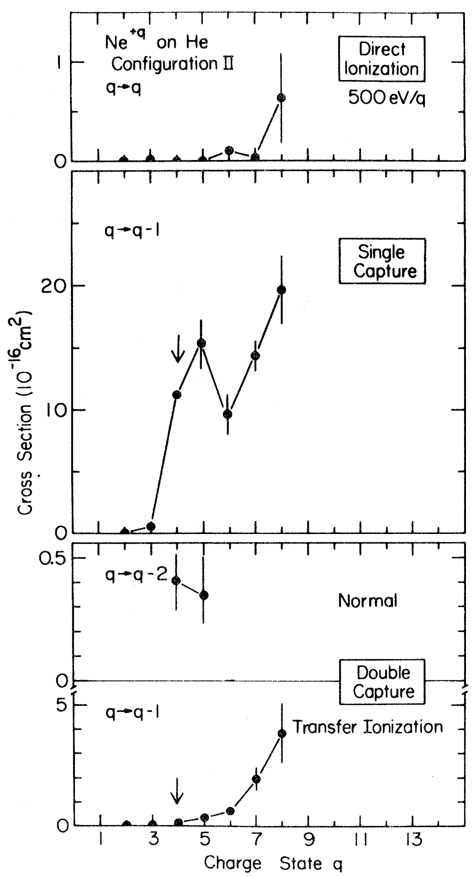

FIG. 5. Configuration II cross sections for Ne^{+q} on He at $E=(1500\text{ eV})q$. "Direct ionization" refers to events for which He^{+} is produced in coincidence with projectiles whose charge is unchanged, and is attributed here to a metastable beam component (see text). "Single capture" and "double capture" refer to events for which He^{+} and He^{+2} are produced, respectively. For the latter case, "transfer ionization" refers to events for which the coincident projectile charge q was reduced by only a single unit. Arrows in the single capture and TI parts of the figure indicate where single capture to the first fully vacant projectile shell and transfer ionization becomes exoergic, respectively.

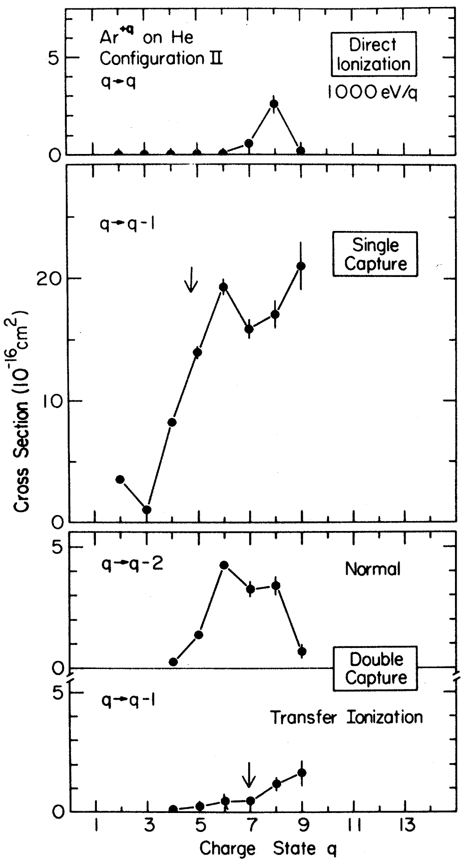

FIG. 6. Similar to Fig. 5, but for Ar^{+q} on He at $E=(1000\ \mathrm{eV})q$.

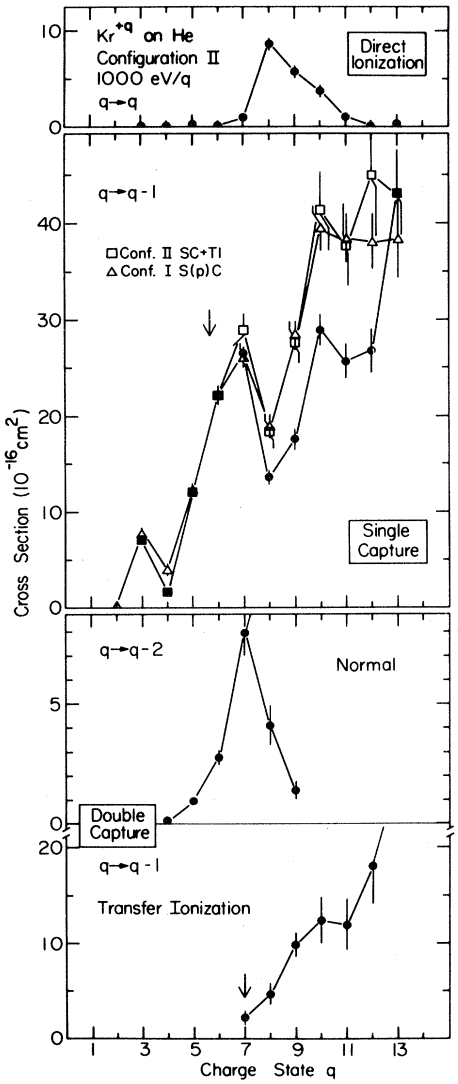

FIG. 7. Similar to Fig. 5, but for Kr^{+q} on He at $E=(1000\ \mathrm{eV})q$.

be especially severe for the case of double capture, for which small cross sections indicate close crossing radii and large energy releases.

Although estimates of the size of this effect can be made, no definite efficiency can be calculated without knowledge of the actual exoergicities of the reaction of interest. We chose instead to adopt empirical normalization constants, for Kr and Xe, for the double-capture channels (i.e., those producing He^{+2}) by requiring agreement between configuration I and configuration II data for the normal $(q \rightarrow q-2)$ channel. Thus the configuration II data were used to measure the ratio between TI and normal double capture for each projectile type and charge state under the tacit assumption that the collection efficiencies for the He^{+2} products in these channels are nearly the same. We note that this efficiency loss is a result of the extremely light He target, and can be estimated to be small for heavier targets.[29]

The results thus obtained are shown in Figs. 5–8. In these figures the labels single and double capture are used to designate events for which He^{+} and He^{+2} are produced, respectively. The three-parameter data were subjected to a consistency test (see Figs. 6 and 7) by comparing the sum of the measured cross sections for single capture and TI to the single (p) capture cross sections of the configuration I data. The agreement between the two procedures is found to be good.

Possible energy dependences of the TI cross sections for systems where this process plays an especially important role (Kr^{+q} and Xe^{+q} on He) were also investigated. The results obtained are shown in Fig. 9 where TI cross sections for the systems mentioned above are plotted versus the projectile charge state q for two collision energies (500 and 1000 eV per projectile charge). No appreciable change of these cross sections with impact energy was observed.

III. DISCUSSION

A. Velocity dependences

Cross sections for single (p) and double (p) capture, plotted versus projectile energy per unit charge, are shown in Figs. 2 and 3. Except for the lowest projectile charges (Ne^{+2}, $Ar^{+2,3}$) they are nearly velocity independent. This behavior is now well documented in the literature for

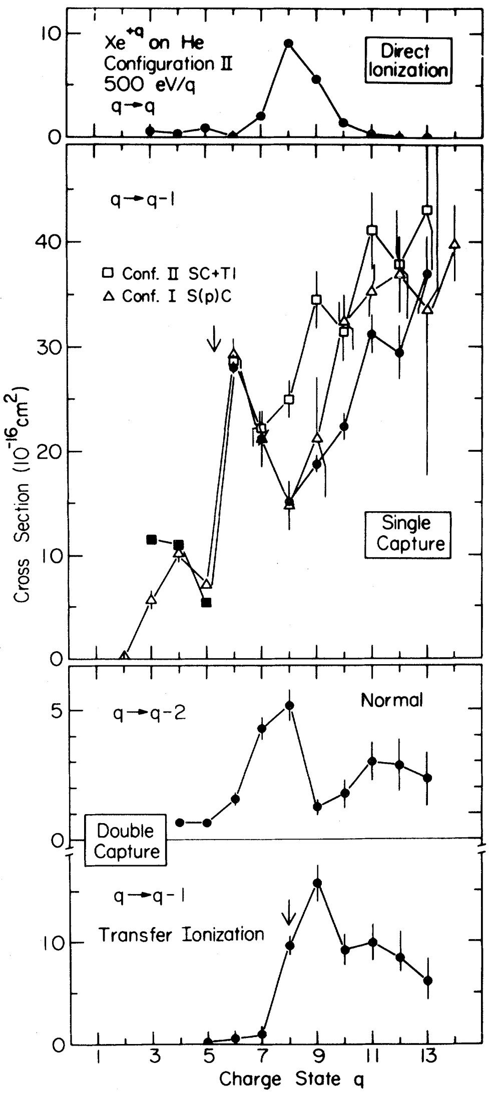

FIG. 8. Similar to Fig. 5, but for Xe^{+q} on He at $E=(500\ \text{eV})q$.

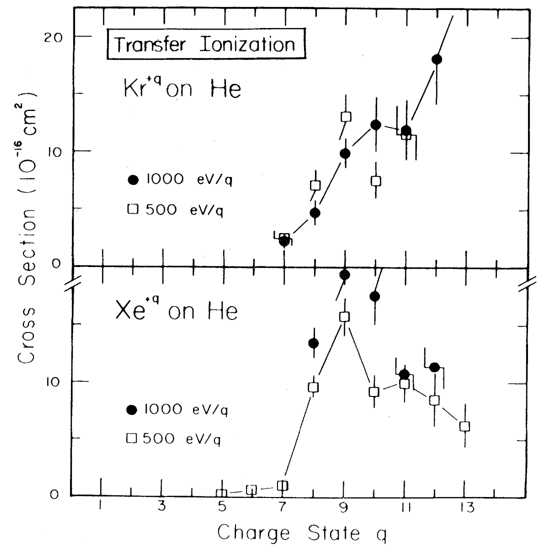

FIG. 9. Cross sections for transfer ionization by Kr and Xe ions on He at energies per charge of 500 and 1000 eV.

many cases,[19–22,24–26] and is attributed to the availability of many slightly exoergic final channels. While the population of a single final state may be quite velocity dependent, the internuclear radius (R), for which the coupling matrix element between incident and final channels becomes sufficiently strong that transitions between them occur, is roughly localized between 3 and 10 Å. This localization is due to the steep dependence of the coupling matrix element on R. Thus as long as there are enough final channels that curve crossings can occur near the optimum R, the cross sections are nearly geometrically determined and only the distribution of the capture among the final channels is v dependent. In cases where a comparison between our low-energy cross sections and those at much higher energy can be made, remarkably good agreement is seen (Fig. 4), confirming this velocity independence. For the low-projectile charge states, e.g., Ne^{+2}, only a single channel (or bunched group of channels) can be exoergically fed, and thus the velocity dependence characteristic of this single crossing is seen.

Similar velocity-independent cross sections are seen for double capture and TI (Figs. 2, 3, and 9), for which q is necessarily high. Presumably the same reasons as discussed above are responsible. We believe the TI is likely to be proceeding through doubly excited states formed by double capture to autoionizing states on the projectile,[13,17] and thus should really be thought of as another manifestation of double capture. Experimental evidence for this process for N^{+7} and O^{+7} on He has been seen in energy-gain spectra by Tsurubuchi *et al.*[18] Other forms of TI are certainly possible, which would involve electron emission during the collision.[13] We would expect the latter process not to be velocity independent, however, even in the total cross sections. Direct experimental clarification of this point awaits further electron or energy-gain spectroscopy on these systems at low projectile velocities.

B. Charge-state dependences

1. *Direct ionization*

True direct ionization at collision energies as low as ours is expected to be quite small, since it involves endoergic reactions for which no curve crossings are possible. The direct ionization cross sections shown in Figs. 5–8 are observed to be undetectably small except for a few ($q=8$, 9, and 10) projectile charge states. This rather peculiar behavior can be explained on the basis of the presence of metastable beam components. A small fraction of the Ar^{+8} beam, taken here as an example, may be in the $(2p^5 3s)\,^2P_{0,2}$ metastable states. Such a projectile can undergo a single-capture collision to form a doubly excited state which may promptly autoionize, thus giving rise to Ar^{+8} ions exiting the collision region. For Kr^{+8} and Xe^{+8} the same argument holds since configurations of the type $3d^9 4s$ and $4d^9 5s$, respectively, for which electric dipole relaxation is parity forbidden, may be present in the beam. Similar metastable states are expected even in the slightly more complicated cases of $Kr(Xe)^{+7,+9,+10}$ projectiles. While the present experiment does not exhibit direct evidence supporting this interpretation, the circumstantial evidence based on the systematic behavior observed for the various collision systems studied is strong. In particular, the metastable beam fraction, deduced to be the ratio between apparent DI cross sections and total single-capture cross sections, is found to be independent of the target used, as would be expected in this interpretation. Our interpretation is thus that there is no experimental evidence for any true DI in our data, as would be expected. The effect of the metastable beam is to take apparent events out of the single-capture channel and cause them to appear in the DI channel. Thus the DI cross sections should be added to the single capture ones before any interpretation of single capture is made, with the tacit assumption being that the capture cross section is not sensitive to the core configuration but only to its charge.

2. *Capture: General features*

All charge-state dependences are characterized by small, velocity-dependent cross sections for low q, followed by a rapid rise to the velocity-independent region. The rapid rise occurs near that value of q for which the population of the first completely empty shell on the projectile becomes exoergic. The minima in the cross sections seen for $q=2$–3 (Ne), 3 (Ar), 4 (Kr), and 5 (Xe) probably occur because these are the last charge states for which the capture proceeds to partially filled valence shells of the projectiles. These last charge states will have the smallest crossing radii for coupling to available exoergic channels, and thus small cross sections. This behavior is illustrated

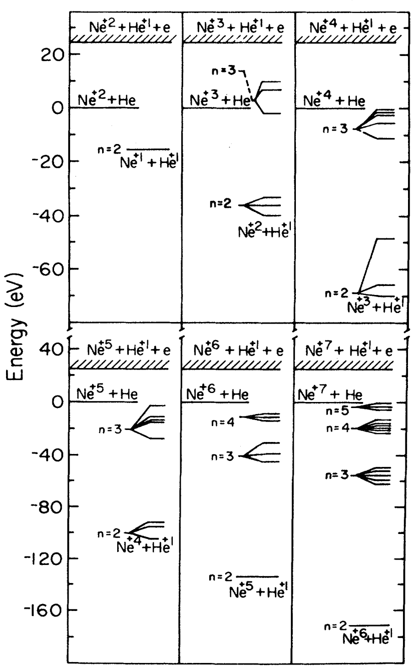

FIG. 10. Energy-level diagram for Ne^{+q} on He.

by the energy-level diagram in Fig. 10. For Ne^{+2} and Ne^{+3}, only $n=2$ can clearly be fed exoergically. In fact, the subshell splitting for $n=3$ in the Ne^{+3}-He case makes a small number of these states exoergic and probably is responsible for the slight rise in cross section at this q. The real rise occurs for $q=4$, when $n=3$ becomes exoergic.

The arrows on the single-capture parts of Figs. 6–9 indicate the values of q for which the population of the first fully open shell becomes, on the average, exoergic. The average binding energy was determined here either from experimental values[31] or from a Hartree-Fock calculation.[32] The rise appears to occur (in most cases) for slightly lower q than would be expected from this result, but this we attribute to our neglect of subshell splitting for given n. A certain fraction of levels for each n manifold becomes exoergic before the average energy does.

The arrows in the TI cross sections shown in Figs. 5–8 indicate where, on the basis of the asymptotic energy levels, reactions of the type

$$A^{+q}+\mathrm{He}\to A^{+(q-1)}+\mathrm{He}^{+2}+e \qquad (1)$$

become exoergic. As in the single-capture case, Hartree-Fock calculations were performed for those cases where experimental compilations[31] were incomplete. Good agreement is found between the arrow position and the projectile charge state for which normal double capture begins to fall and TI begins to rise. One also notices that although energetically allowed, the TI cross sections do not go up sharply until one or two charge states later. This is consistent with the picture discussed in previous works[17,29] that in these cases TI proceeds via capture into autoionizing doubly excited states which becomes (as a function of projectile charge state) exoergic later than the direct reaction (1) does. The Xe case is an exception to this and its early rise might indicate that here a different mechanism is responsible for the TI process. Further experiments are necessary to clarify this point.

3. *Capture: Model comparisons*

Several models[5–8] have been proposed for a broad-brush treatment of the capture of electrons by highly charged projectiles. Two which we consider here are the absorbing sphere model of Olson and Salop,[5] and the classical barrier model.[7,8] The tunneling model of Grozdanov and Janev,[6] for which results directly applicable to our cases are not available, is expected to give slightly higher cross sections than the absorbing sphere model.[27,30] Each of these models treats the projectile as structureless and the target as possessing a single electron. While neither requirement is particularly well satisfied by the systems studied here, the absence of detailed calculations on even the two-electron target leads us to see how the models fare. Since the models do not directly treat the possibility of double capture, we choose to compare them with *total* cross sections for capture (Fig. 11) formed from summing all channels for which capture occurs. This includes even the DI, since it is believed to result from a true capture event, but to a metastable core. In most cases, the single-capture channel is by far the dominant one.

Comparisons between the model predictions and the

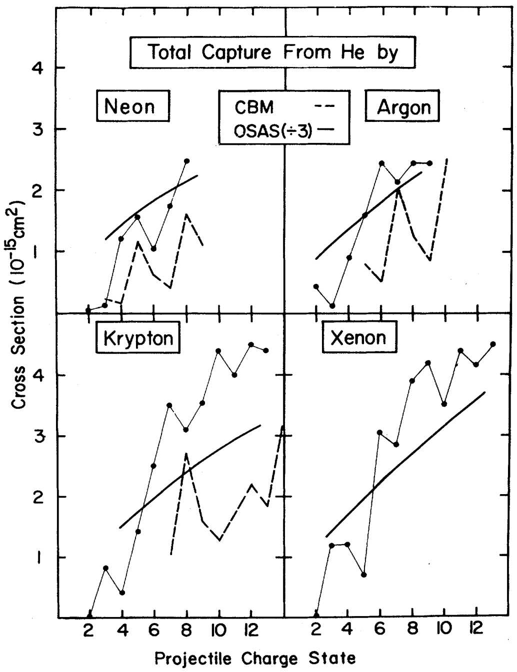

FIG. 11. Cross sections for total capture found by summing all configuration II cross sections from Figs. 5–8. Theory: ——, classical barrier model (CBM) (Refs. 7 and 8); Olson and Salop absorbing sphere (OSAS) (reduced by a factor of 3), Ref. 5. Light solid lines are to guide the eye only.

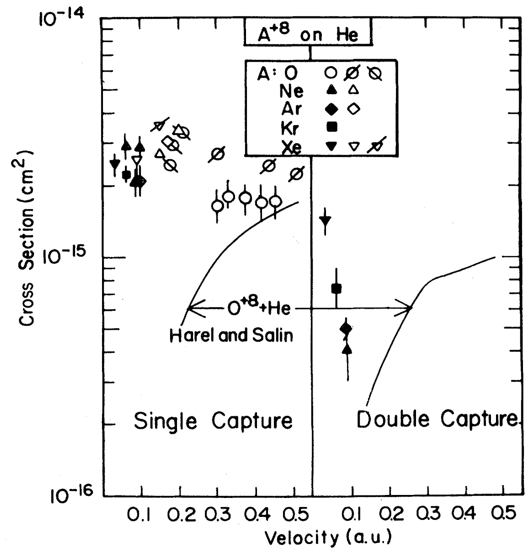

FIG. 12. Cross sections for single and double capture from He by projectiles in charge state +8. Experiment: solid symbols, present results; △,⧅ Iwai *et al.* (Ref. 24); △,◇, Salzborn *et al.* (Ref. 20); ⊽, Crandall *et al.* (Ref. 25); ○, Bliman (Ref. 28); ∅, Afrosinov *et al.* (Ref. 27). Theory: Harel and Salin (Refs. 28 and 35).

data are shown in Fig. 11. In making the classical barrier model calculations, we have taken into account the structure of the projectile by employing, for each projectile and q, an effective charge Z_e in the calculation. The Z_e was adjusted in each case to be that which reproduced the binding energies of the excited states on the projectile which were expected to be primarily populated. Since in many cases these binding energies are not known experimentally, Hartree-Fock calculations were made of the needed binding energies and used to calculate Z_e. In all cases the binding energies of different angular momentum states, at a given n for the captured electron, were averaged to a single binding energy E_B. The effective charge Z_e was obtained from $Z_e=(2E_Bn^2)^{1/2}$ (where E_B is in a.u.).

The results in Fig. 11 show that the absorbing sphere values are too high by a factor of near 3 for Ne, improving somewhat for heavier projectiles. This is in contrast to earlier success of this model with targets of lower ionization potential,[5] and with our recent results for a loosely bound Li target,[33] but is consistent with recent results on atomic hydrogen targets.[34] The model is based on the assumption of a quasicontinuum of final states into which the capture can proceed, an assumption poorly met for the rather tightly bound He target, but better met for the case of a Li target.

The classical barrier results produce roughly the correct magnitude for the cross sections. There is slight evidence for the oscillatory behavior of the cross sections predicted by the model. The oscillations in the model cross sections, which result from the discontinuous rise in cross section each time a new n begins to be fed, should not be taken very seriously here, since subshell splitting is quite appreciable for the states being fed but has been eliminated in the formulation of the model. Were this splitting incorporated into the model, it would almost certainly wash out the oscillatory behavior for higher q. The low cross section for Ne^{+6} on He probably occurs because this is a transition case between the feeding of $n=3$ (for $q=5$) and $n=4$ (for $q=7$). The onset of population of the first vacant shell on the projectile is consistently predicted to begin at larger q than appears to be the case experimentally. This is not unexpected, since neither tunneling through the barrier nor the splitting of different l for each n is taken into account, both of which will allow a given n to be populated earlier than the model predicts.

The case of O^{+8} on He is the only one for which we are aware of a direct treatment of low-energy capture from the two-electron target in our velocity range. Harel and Salin[35,28] have used a molecular-orbital treatment of this system to calculate both single- and double-electron capture. In Fig. 12 we show a comparison between their results and those of several recent experiments for A^{+8} on He. This comparison, for projectiles other than O^{+8}, has validity only to the extent that the charge 8 projectile can be thought of as a point charge, even though it in reality has core structure. While this assumption seems physically reasonable, it is known that the symmetry properties of the pure Coulomb force cause the behavior of the molecular orbitals, at avoided crossings between incident and final channels, to be different for point projectiles and for clothed projectiles. Thus there may be fundamental differences between O^{+8} and A^{+8} as projectiles, and this point remains to be clarified.

The most striking disagreement between theory and experiment lies at low velocity, where theory dives precipitously while experiment remains nearly constant for single capture. At higher velocities the magnitudes of theory and experiment seem to come into somewhat better agreement.

For double capture, we have included only data from the present experiment, since we know from this work that the TI channel is the dominant one and this contribution is lost if only the projectile charge state is measured. That the TI channel should be dominant is in qualitative agreement with Harel and Salin, who find that the double capture feeds (n,n) of (4,4), (4,3), and (3,3), all of which lead to autoionizing states for O^{+8}. The absolute comparison between theory and experiment for double capture is inconclusive, and points to the importance of a direct measurement for O^{+8} on He for which the TI is resolved.

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