Transfer excitation in slow collisions between ions of very high charge and two-electron targets

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We argue that a three-step transfer-excitation (capture and target excitation) mechanism enhances (depletes) the cross section for one- (two-) electron removal from a two-electron target. This mechanism, which becomes very important at high projectile charge (q), is mediated through quasimolecular couplings between diabatic double-capture and transfer-excitation channels, in which the inner of two active electrons is transferred to an excited target state. The inclusion of this process in the extended classical over-barrier model gives excellent agreement with unexplained experimental cross sections for one- and two-electron removal from He in slow collisions with Xe ions of high charge ($q \leq 31$).

I. INTRODUCTION

Charge transfer in slow ($v \ll 1$ a.u.) collisions between highly charged ions and atoms may be described in terms of transitions between quasimolecular adiabatic energy levels. The investigations of electron capture during the last decade have led to a fair understanding of transfer mechanisms involving one active electron, while processes with rearrangements of two electrons still need considerable exploration.[1,2] It is usually taken for granted that single-electron capture is dominated by one-active-electron-transfer mechanisms. This seems, however, not to be true for highly charged projectiles colliding on He. In this paper we argue that the relative contribution from a three-step transfer excitation process with two active electrons increases rapidly with the projectile charge q and becomes highly significant for $q > 20$. Here, the term transfer excitation refers to the process of electron capture to the projectile and excitation of the target,

$$\mathrm{Xe}^{q+} + \mathrm{He} \rightarrow \mathrm{Xe}^{(q-1)+} + (\mathrm{He}^{+})^{*} \; . \qquad (1)$$

Experimental data on electron capture from two-electron targets are of special interest for interpretations of spectroscopic data from astrophysical and fusion plasmas due to the abundance of He in, e.g., stellar atmospheres and tokamak plasmas.[3]

In the extended classical over-barrier model[4] it is assumed that electrons are transferred consecutively from the target to the projectile at the internuclear distances where the outermost remaining target electron is at the top of the internuclear barrier. This assumption is justified by the high density of capture states in highly ionized projectiles.[4] Here we will show that energy resonances between that particular state, which describes the quasimolecule after transfer of the second electron, and a whole set of states dissociating through transfer excitation channels occurs within very narrow regions of internuclear separations. This leads us to associate the corresponding potential crossing radii with minimum impact parameters for removal of two electrons from a two-electron target. Smaller impact parameters will accordingly lead to single-electron capture and target excitation, i.e., to one-electron removal. The incorporation of this three-step transfer-excitation mechanism thus increases (depletes) the total model cross sections for one- and (two-) electron removal from a two-electron target. The extended classical over-barrier model modified by inclusion of this process agrees excellently with recent experimental absolute cross sections[5] for removing one and two electrons from He by 4q-keV Xe^{q+} projectiles ($q \leq 31$), while the original model is more than a factor of 2 off for higher q. We will show that the importance of the transfer-excitation cross section increases rapidly with q, and around $q = 50$ it approaches the total cross section for two-electron removal from He.

In this paper we thus propose a modification of the extended classical over-barrier (ECB) model, which takes the transfer-excitation mechanism into account. The outlines of the original ECB model are given in Sec. II together with the details of the modification. Comparisons between the original and modified versions of the model and experimental results[5] are given in Sec. III. In the discussion, we point out the difference between the present transfer-excitation mechanism and the one from the model by Niehaus.[6] The cross section for the latter process goes to zero at large q, while the cross section for the present transfer-excitation process increases monotonically with q.

II. MODEL

A. Outline of the original ECB model

The Landau-Zener,[1] classical over-barrier,[3] and extended classical over-barrier models[4,6,7] are rather successful in predicting absolute cross sections and final capture states for single-electron capture. The first model yields velocity-dependent single- and multiple-electron capture cross sections for any projectile-target combination, but is mostly limited to one-electron processes since it requires a detailed knowledge of the quasimolecular potential-energy diagram. The static classical over-barrier model is developed for one-electron transfer to a bare nucleus from a hydrogenlike target. This model gives a cross section of $\frac{1}{2}\pi R^2$ for capture at the internuclear distance R of the outermost populated energy resonance between Stark-shifted projectile and target states, which surmounts the internuclear potential barrier. The replacement of $\frac{1}{2}$ by different factors ranging up to unity was made[8] in order to conform to experimental data for partially stripped ions and multiple-electron targets. Absorbing-sphere models[9] assume a quasicontinuum of single-capture states and that impact parameters smaller than a certain value R_1 add up to a single-capture cross section of πR_1^2. It was soon realized, however, that multiple-electron processes play an important role and the extended classical over-barrier[4] and related[6,7] models were developed.

Transfer ionization and true double-electron capture involve two active electrons, initially transferred to the projectile. Both electrons become bound in true capture, while the intermediately formed doubly excited projectile state autoionizes in the transfer-ionization process.[10] The two-step electron-transfer mechanism, i.e., two consecutive well-separated one-electron transfers,[11] has been found to dominate in numerous experimental investigations where the densities of final capture states are sufficiently high. The ECB model assumes straight-line projectile trajectories, quasicontinua of capture states, and that m-electron capture proceeds through m consecutive one-electron transfers. The second assumption locates the positrons of the two outermost one-electron transfers to energy resonances between the top of the internuclear potential barrier and the Stark-shifted ionization energies of the neutral and singly ionized target at R_1 and $R_2 < R_1$, respectively. Collisions with impact parameters b between R_1 and R_2 contribute to the cross section $\sigma_1 = \pi(R_1^2 - R_2^2)$ for removing one electron from the target through transitions at R_1, while $b < R_2$ collisions make transitions at R_2, giving a cross section for removing two electrons from a two-electron target of $\sigma_2 = \pi R_2^2$. The encounter with a quasicontinuum of channels leading to capture of a third electron at R_3 prohibits contributions from $b < R_3$ and $\sigma_2 = \pi(R_2^2 - R_3^2)$ for multielectron targets. Both σ_1 and σ_2, which scale linearly[4,12] with q, have been found to describe the gross behavior of experimental data in the $q < 20$ charge-state regime.[5,13,14] The accuracy of the model is, however, limited by the quasicontinuum approximation, since in this approximation it is assumed that there always is a projectile capture state in resonance with the initial target state when the electron reaches the top of the barrier.[15,16] This is not true, but the separation between model crossings and the first real over-barrier crossing generally gets smaller with increasing q and decreasing target-ionization potentials. The uncertainties in σ_1 and σ_2 due to finite-capture-state densities have been estimated by Andersson *et al.*,[12] who assumed evenly spaced angular momentum l states within n manifolds of projectile states. For a He target the uncertainties in σ_2, larger than the ones in σ_1, are estimated[12] to be less than 50%, 15%, and 7% for $q = 10$, 20, and 30, respectively. A recent comparison between cross sections from the original ECB model and experimental Xe^{q+} He $(11 \le q \le 31)$ data[5] revealed systematic over-estimates and underestimates of σ_2 and σ_1, respectively, which were far too large to be accounted for by finite-capture-level densities.

B. Inclusion of transfer excitation in the ECB model

Collisions with two-electron targets seem to be unique in that one of the two active electrons may be promoted effectively to an excited, singly ionized, target state at an internuclear distance smaller than R_2. This mechanism is mediated through interactions between quasimolecular channels associated with transfer to the projectile of two target electrons and a near continuum of potential curves leading to single-electron capture and target excitation, i.e., to transfer excitation (TE). Thus intensity is redistributed from the sum of the cross sections for transfer ionization and true double capture (σ_2) to single-electron capture (σ_1) relative to what is expected from the original ECB model.[4] We modify the cross sections for removing one and two electrons from a two-electron target to

$$\sigma_1 = \pi(R_1^2 - R_2^2 + R_{\mathrm{TE}}^2) \tag{2}$$

and

$$\sigma_2 = \pi(R_2^2 - R_{\mathrm{TE}}^2) , \tag{3}$$

respectively. The internuclear distance at which the transfer excitation channel is populated is

$$R_{\mathrm{TE}}(n) = \frac{R_2}{1 + E_{\mathrm{ex}} R_2/(q-3)} , \tag{4}$$

where all entries are in atomic units (a.u.), n is the principal quantum number of the singly-charged target ion, and $E_{\mathrm{ex}} = E_{\mathrm{ex}}(n)$ is the corresponding target excitation energy. R_{TE} is defined by the crossing between the internuclear potential traversed after the transfer of the second electron to the quaismolecule at R_2 and a single-capture potential with an asymptotic energy given by $Q_1 - E_{\mathrm{ex}}$, where $Q_1 = (q-1)/R_1$ is the inelasticity of single-electron capture without target excitation.[4] R_{TE} assumes a maximum value when E_{ex} equals the $n = 2$ excitation energy of the target ion, while the minimum R_{TE} radius is given by the $n \to \infty$ excitation. The radii R_1 and R_2 in (2)–(4) are from the original ECB model, where pure Coulomb repulsion between the projectile and the target and full (no) screening of the projectile (target) by the active electrons are assumed. The total charge-exchange cross section $\sigma_1 + \sigma_2$ is unaffected by the inclusion of this target-electron promotion mechanism, which may be trivially generalized to m- and $(m-1)$-electron removal from m-electron targets.

Schematics of internuclear potentials are shown in Fig. 1 for A^{q+} projectiles of (a) $q = 6$, (b) $q = 15$, and (c) $q = 30$ colliding on He. The incident channel $A^{q+} + \mathrm{He}$ (denoted by IN in Fig. 1) couples to the single-capture channel (SC) $A^{(q-1)+}(n') + \mathrm{He}^+(n=1)$ at R_1. The second electron makes a transition to the double-capture channel (DC) $A^{(q-2)+}(n',n'') + \mathrm{He}^{2+}$, when the separation between the nuclei is R_2. This channel crosses with

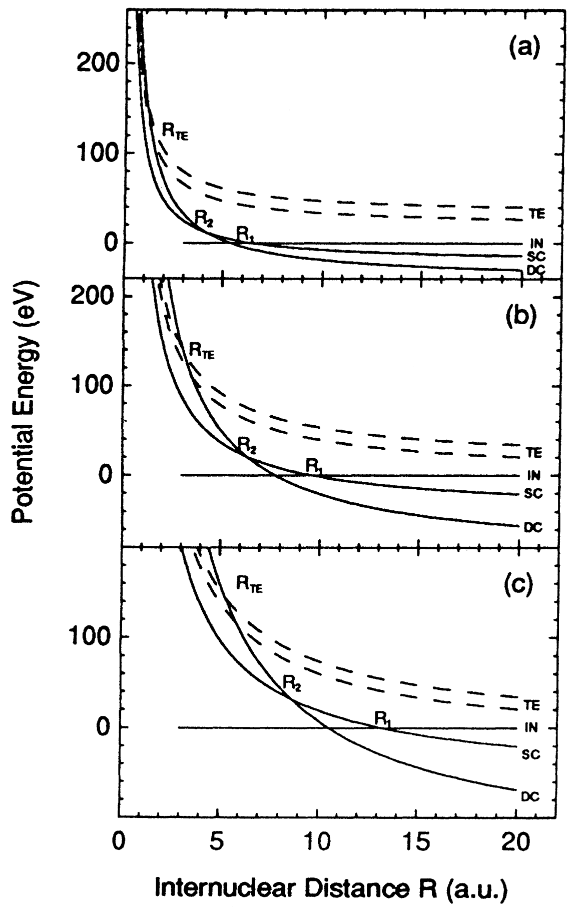

FIG. 1 Potential-energy diagrams describing the three-step transfer-excitation mechanism in A^{q+}+He collisions at (a) $q=6$, (b) $q=15$, and (c) $q=30$ (cf. text).

the curve for $A^{(q-1)+}(n')+\mathrm{He}^{+}(n=2)$ (lower dashed curves of Fig. 1) at the maximum R_{TE} radius and shortly afterwards with potential curves dissociating to $A^{(q-1)+}(n')$ and $\mathrm{He}^{+}(n>2)$. As can be seen from Fig. 1, there is an infinite number of crossings corresponding to He^{+}- excitations all the way up to the ionization limit $\mathrm{He}^{+}(n=\infty)$ (upper dashed curves), within very small intervals of internuclear separations. The density of $\mathrm{He}^{+}(n)$ energy levels that is encountered when the system follows the DC channel is obtained from (4) as

$$\left|\frac{dn}{dR}\right|=(q-3)n^3/4R^2\ , \tag{5}$$

while the density of DC energy levels encountered when traveling on one of the TE channels is given by

$$\left|\frac{dn''}{dR}\right|=(q-3)n''^3/q^2R^2\ . \tag{6}$$

In (6) we have assumed that both electrons (n',n'') can be described by hydrogenlike energy levels and that only one-electron transitions are active.[4] The densities of TE and DC channels are obtained from (5) and (6) by multiplications by the degeneracies n^2 and n''^2, respectively, and the probability ϕ_{TE} to stay on the TE channel can be estimated by

$$\phi_{\mathrm{TE}}=\frac{|dn/dR|n^2}{|dn/dR|n^2+|dn''/dR|n''^2} =[1+(2/q)^2(n''/n)^5]^{-1}\ . \tag{7}$$

When R decreases on the way into the radial turning point, $n\to\infty$ as the R_{TE} region is passed, while n'' is limited to values given by the original ECB model[4,12] (e.g., $n''\sim 10$ for $q=30$). Thus it is reasonable to assume a unit probability for transfer to the TE channel in this region. As the projectile and the excited He^{+} ion separate the TE channel crosses with double-capture channels that are associated with progressively larger n'' for increasing R. However, n'' can only match large n at large R ($n''\to\infty$ when $R\to\infty$) where couplings are expected to be weak because of the increase in the radial velocity. In the present evaluation of the model cross section we thus use R_{TE} $(n=\infty)$ as the minimum impact parameter for removing two target electrons. This means that collisions with $b<R_{\mathrm{TE}}$ $(n=\infty)$ are prohibited from contributing to σ_2 and instead contribute to σ_1. We wish to stress, though, that the choice of the R_{TE} radii (between $n=2$ and $n=\infty$) in (2) and (3) only has a minor influence on the numerical values for model cross sections.

We assume that the transitions between relevant channels around the radii R_1, R_2, and R_{TE} are neither too adiabatic nor too diabatic, but that transition probabilities are close to $\frac{1}{2}$. With the aid of semiempirical formulas for adiabatic energy splittings,[9,17] Landau-Zener calculations have confirmed this assumption through comparisons with ECB capture radii.[2,15,17,18] Coupling strengths decay rather rapidly on both sides of the model radii[19] and the vicinity of R_{TE} to R_2 is essential for a high transition probability at R_{TE}. In fact, R_{TE} and R_2 become equal at infinite q according to (4). The quasimolecular state (n',n''), populated when the second electron becomes active at R_2, is at first strongly localized to the projectile and is therefore likely to dissociate to a doubly excited projectile state and a bare target nucleus. At R_{TE} $(n=\infty)$, however, the n'' electron can become strongly localized to the target through couplings to a quasimolecular state that dissociates into singly excited projectile and target states with high probability.

III. RESULTS

The original and modified ECB cross sections and experimental $4q$-keV Xe^{q+}-He data[5] for removal of one and two electrons from He are shown as functions of q in Fig. 2. The upper (lower) solid line shows cross sections from the modified ECB model for removing one (two) electrons from the He target, while the upper (middle) dashed curve shows cross sections for removing one (two) target electrons due to the original model. The lowest dashed curve shows the TE cross section from the present model. The upper (lower) set of data points shows experimental results from Ref. 5 for one- (two-) electron removal from He. As can be seen from Fig. 2, the agreement between

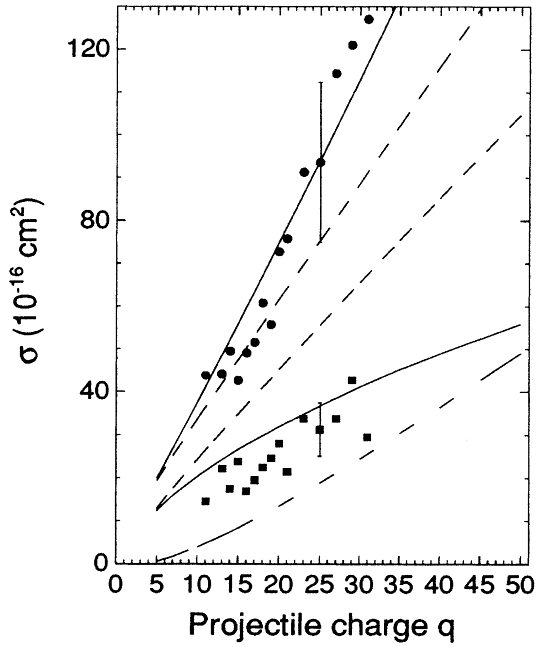

FIG. 2 Experimental and model cross sections for removal of one and two electrons from He as functions of the projectile charge q. Experimental σ_1 and σ_2 cross sections (Ref. 5) are indicated by circles and squares, respectively. The error bars show systematic errors which are $\sim 30\%$ for all q, while relative uncertainties are a few percent (Ref. 5). The upper and lower of the solid curves are the cross sections for one- and two-electron removal, respectively, i.e., $\sigma_1=\pi(R_1^2-R_1^2+R_{\rm TE}^2)$ and $\sigma_2=\pi(R_2^2-R_{\rm TE}^2)$ with $R_{\rm TE}(n=\infty)$ (cf. text). The dashed lines indicate $\sigma_1=\pi(R_1^2-R_2^2)$ (upper line) and $\sigma_2=\pi R_2^2$ (middle line) of the original ECB model and the cross section for transfer excitation $\sigma_{\rm TE}=\pi R_{\rm TE}^2$ from the modified ECB model (lower line).

the experimental data and the modified ECB model is excellent, while the original model is off by more than a factor of 2 for higher q. The relative importance of the target-electron promotion mechanism increases rapidly with q in both σ_1 and σ_2, as is evident by the increasing discrepancies between the modified and the original ECB cross sections of Fig. 2. At $q=30$, e.g., more than 20% of the total single-capture cross section is due to transfer excitation, while the corresponding number for $q=15$ is $\sim 15\%$. The present TE mechanism thus explains the stronger-than-linear q dependence of σ_1 as well as the weaker-than-linear one for σ_2 in Xe^{q+}-He collisions.

IV. DISCUSSION

Target electron promotion has been discussed before in general terms by Janev and Winter[1] and for specific moderate-q collisions by Hoekstra, de Heer, and Winter.[20] The latter authors used the ECB-related model by Niehaus,[6] which excludes projectile screening and treats the active electrons as quasimolecular until, on the way out from the collision, they are consecutively trapped on either the projectile or the target as the internuclear barrier rises above populated molecular energy levels. The capture probabilities are determined by available projectile and target phase-space volumes, taken to be the degeneracies of hydrogenlike states[6] when they are at the top of the barrier. Accordingly,[2,6] the projectile capture probability approaches unity and the transfer-excitation cross section approaches zero at high q. Note that transfer excitation within the Niehaus model most likely involves the outer of two quasimolecular electrons, while the present TE mechanism always proceeds through transfer of the inner electron. Moreover, the present mechanism takes place at crossings above the barrier ($R_{\rm TE}<R_2$), where the principal quantum numbers of excited target states rapidly become larger than those of the projectile capture states, allowing the transfer excitation cross section to increase with q.

Apart from the experiment already discussed, only four[21–24] low-energy experimental studies have been reported for $q>20$. Iwai *et al.*[21] and Tawara *et al.*[22] measured the sum of the cross sections for single-electron capture and transfer ionization for Kr^{q+}-He ($q\le 25$) and for I^{q+}-He ($q\le 41$), respectively. Mann[23] has measured I^{q+}-He, $H_2 \rightarrow I^{(q-1)+}$ ($q\le 27$) cross sections and Cederquist *et al.*[24] performed energy-gain measurements of relative cross sections for true single-electron capture and transfer ionization in 4q-keV Xe^{q+}-Xe ($q\le 35$) collisions. None of these investigations has reported on absolute and resolved values for σ_1 and σ_2. Nevertheless, some very valuable experimental information can be extracted. According to the modified ECB model the transfer-excitation components in single-electron capture should be associated with inelasticities, which are between 41 ($n=2$ in He^+) and 61.5 eV ($n\rightarrow\infty$) lower than those of the main single-capture peaks and with intensities approaching those of transfer ionization as $\sigma_{\rm TE}=\pi R_{\rm TE}^2$ approaches σ_2 (cf. Fig. 2). Low-energy-gain features in accord with these predictions are indeed present in the I^{q+}-He data,[20] while no such features appear in the Xe^{q+}-Xe data,[24] which might indicate that the present three-step transfer-excitation process is hindered in many-electron targets since then $R_{\rm TE}$ lies inside the transfer radius R_3 for the third electron. A few measurements of He^+ Ly-α-emission cross sections ($\sigma_{\rm em}$) have been reported for moderate-q collisions, where transfer-excitation mechanisms involving the outer active electron also may be important. The ECB model is expected to overestimate $\sigma_{\rm TE}$ at moderate q due to relatively low densities of capture states. In addition, a considerable fraction of the He^+ decay may bypass the $He^+(2p)$ state. Therefore we take $\sigma_{\rm TE}$ as upper limits for $\sigma_{\rm em}$. Hoekstra, de Heer, and Winter,[20] Bouchama, Druetta, and Martin,[25] and Roncin *et al.*[26] measured $\sigma_{\rm em}$ to be between 1.4 and 15 times smaller than the $\sigma_{\rm TE}$ values of the present model for charge states ranging between $q=4$ and $q=8$.

V. CONCLUSIONS

We have, for the first time, drawn attention to the significance of a three-step target-electron promotion mechanism in one- and two-electron removal from two-electron targets at high q. Inclusion of this mechanism in the extended classical over-barrier model leads to excellent agreement with, hitherto unexplained, experimental q-dependencies of absolute cross sections for one- and two-electron removals from He. Additional support is found in high-q energy-gain data. The present transfer-excitation process is most likely suppressed for multiple-electron targets since a transition to triply excited quasimolecule states can occur outside R_{TE}. More experimental absolute charge-exchange cross sections, collision inelasticities, and cross sections for target-photon emission at high q are urgently needed for further tests of the ideas put forward here.

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