

Double and transfer ionization in collisions of He with bare ions

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Double and transfer ionization processes are theoretically investigated for collisions of He atoms with bare energetic projectile ions. Two-electron transition amplitudes are described by linear combinations of products of one-electron transition amplitudes, which enables one to incorporate the effects of electron correlation in the asymptotic channels. A collision mechanism in which the projectile interacts separately with both electrons gives a realistic account of the processes only at medium impact energies, while the final state correlation and the shake-off mechanism together with correlations in the initial state are found to be important for the lower and higher collision energies, respectively.

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Collisions between He atoms and bare ionic projectiles are the simplest systems to investigate two-electron transitions. Among the various two-electron phenomena, double ionization (DI) and transfer ionization (TI), where both electrons leave the target, have received continuous interest over decades [1]. The most addressed question in these studies is concerned with the role of electron-electron ($e-e$) correlations which can be traced both in the dynamical and in the asymptotic regimes of the collisions [1,2]. With the advent of the cold target recoil ion momentum spectrometry (COLTRIMS) technique more accurate total and fully differential measurements have become available over the last 20 years [3]. Fully differential measurements provide the most detailed information on the collision processes and on the electron correlation dynamics. However, when the transfer of more than one electron is considered the fully differential cross sections are often too complicated to analyze, especially when one is only interested in the driving dynamics of the collisions. Instead, general trends in the collision dynamics can, for example, be identified by studying total cross sections (TCSs).

In the case of DI three collision mechanisms have been identified in energetic collisions [1]. Two of them are the *two-step-1* (TS1) and the *shake-off* (SO) mechanisms, where the projectile interacts with only one of the target electrons and the other electron is ejected due to electron-electron interaction and due to rearrangements in the final state, respectively. In the third mechanism referred to as *two-step-2* (TS2), electrons are ejected in two independent interactions with the projectile. Despite the integrated contribution of the $e-e$ interaction a marked role of the final-state correlation has been identified in the total cross sections for DI of He by impact of H^+ and He^{2+} ions [4,5].

In TI a less pronounced role is expected for the $e-e$ interaction in the final channel as one of the electrons is captured by the projectile. However, in this process a definite role of the initial-state correlation and a competition of the TS2 and SO or TS1 mechanisms have been reported at medium and

high impact energies [1,2,6]. Furthermore, other correlated processes have been identified: (i) a double-scattering Thomas mechanism, in which the incoming projectile hits one electron, and this electron is scattered off the second target electron thereby acquiring the velocity and direction of the projectile [7]; and (ii) a first-order mechanism, in which the electron-electron interaction mediates the two-electron transition in a fashion similar to that in an Auger process [8].

Theoretical descriptions of the DI and TI processes are based on three- and four-body treatments and all of them rest on some sort of approximation in the numerical evaluations [6,9,10]. Some of these approximations are within the framework of the *independent electron model* (IEM) where the main focus is a more accurate computation of the one-electron transitions [9]. In four-body treatments a further difficulty appears due to the explicit inclusion of the $e-e$ interaction whose role can be revealed in both the static and dynamical regions of the collision [6,8,10,11]. Despite great advances over the last decades, a better understanding of the TI and DI processes still remains a challenge for theory.

In this Brief Report we investigate TCSs for the DI and TI processes. TI is treated as a special case of DI where one of the electrons is emitted with the same velocity as the projectile. The two-electron amplitudes are evaluated in both the IEM and the *frozen correlation approximation* (FCA) [12]. The latter enables an explicit inclusion of the electron correlation in the asymptotic states. Dynamic correlation is neglected. While this implies that we cannot describe the physics at play in full detail, it turns out that TCSs can be calculated with some accuracy over a wide range of impact energies.

The simplest description of DI can be given within the framework of the IEM where the transition amplitude at a given impact-parameter vector \mathbf{b} is expressed as [1]

$$a_{\text{DI}}^{\text{IEM}}(\mathbf{b}) = a_{i \rightarrow \mathbf{k}_1}^{1e}(\mathbf{b}) a_{i \rightarrow \mathbf{k}_2}^{1e}(\mathbf{b}), \quad (1)$$

with $a_{i \rightarrow \mathbf{k}_j}^{1e}(\mathbf{b})$ being the ionization amplitude of a single electron from a specific initial state i into a final state

characterized by momentum \mathbf{k}_j . In the description of a^{1e} , the role of the other electron is taken into account only by a (static) mean field. Therefore, electron correlation effects are excluded both during the collision and in the asymptotic regions.

Beyond the IEM, the framework of the FCA enables one to include electron correlations in the asymptotic initial and final states, while they are still neglected in the course of the collision [12]. As in our earlier application of the FCA [13], we use the configuration interaction (CI) wave function as given by Silverman *et al.* [14] to describe the initial ground state of the He atom. In the final state, the e - e correlation is accounted for by the so-called Gamov factor $\varphi_{k_{12}} = e^{-\pi Z_{12}/(2k_{12})} \Gamma(1 - iZ_{12}/k_{12})$, with $Z_{12} = 1$ and $k_{12} = |\mathbf{k}_1 - \mathbf{k}_2|$. These forms of the asymptotic wave functions allow one to write the correlated transition amplitude for two-electron emission as [12,13]

$$a_{\text{DI}}^{\text{FCA}}(\mathbf{b}) = \frac{1}{\sqrt{2}} \sum_{j_1 j_2} C_{j_1 j_2} [a_{j_1 \rightarrow \mathbf{k}_1}^{1e}(\mathbf{b}) a_{j_2 \rightarrow \mathbf{k}_2}^{1e}(\mathbf{b}) + a_{j_1 \rightarrow \mathbf{k}_2}^{1e}(\mathbf{b}) a_{j_2 \rightarrow \mathbf{k}_1}^{1e}(\mathbf{b})] \varphi_{k_{12}}, \quad (2)$$

where the $C_{j_1 j_2}$ coefficients are due to the CI wave function for the initial state [14].

The cross section differential in the momenta of the ejected electrons is defined as

$$\frac{d\sigma_{\text{DI}}^X}{d\mathbf{k}_1 d\mathbf{k}_2} = \int d\mathbf{b} |a_{\text{DI}}^X(\mathbf{b})|^2, \quad (3)$$

where X stands for IEM or FCA. The total DI cross section σ_{DI}^X is obtained when Eq. (3) is integrated over the coordinates of both electrons. In the present work, the one-electron ionization amplitude $a_{j_i \rightarrow \mathbf{k}_i}^{1e}$ for Eqs. (1) and (2) is evaluated with the *continuum distorted wave with eikonal initial state* (CDW-EIS) approximation [15]. For more details on the theory, with particular attention to the present application, the reader is referred to Refs. [12,13].

The TI process can be considered and described on the same footing as the DI process, which is based on a specific ionization mechanism where the velocity of the emitted electron (\mathbf{v}_e) is equal to the velocity of the projectile ($\mathbf{v}_e = \mathbf{v}_P$). This process is known as *electron capture into continuum states* of the projectile (ECC) [16]. Due to the continuity of the transition amplitude above the ionization limit, ECC relates to the capture to highly excited (n) levels just below the ionization threshold [16]. The DCS for TI (e.g., capture of electron 1 into the projectile's bound state (n) and the ejection electron 2 with \mathbf{k}_2) can be related to that of the DI process with $\mathbf{k}_1 = \mathbf{v}_P$ and \mathbf{k}_2 :

$$\begin{aligned} \frac{d\sigma_{\text{TI}}^n}{d\mathbf{k}_2} &= \lim_{\substack{n \rightarrow \infty \\ E'_1 \rightarrow 0}} \frac{d\epsilon'_1}{dn} \left(\frac{d\sigma_{\text{DI}}}{dE'_1 d\mathbf{k}_2} \right) \\ &= \frac{q^2}{n^3} \int \left(k'_1 \frac{d\sigma_{\text{DI}}}{d\mathbf{k}'_1 d\mathbf{k}_2} \right)_{E'_1 \rightarrow 0} d\Omega'_1 \\ &= \frac{4\pi q^2}{n^3} \left(|\mathbf{k}_1 - \mathbf{v}_P| \frac{d\sigma_{\text{DI}}}{d\mathbf{k}_1 d\mathbf{k}_2} \right)_{\mathbf{k}_1 \rightarrow \mathbf{v}_P} \end{aligned} \quad (4)$$

where $\frac{d\sigma_{\text{DI}}}{d\mathbf{k}_1 d\mathbf{k}_2}$ is defined in Eq. (3), $\epsilon'_1(n) = -q^2/(2n^2)$, $E'_1 = k'_1{}^2/2$, $d\mathbf{k}_1 = d\mathbf{k}'_1 = k'_1 dE'_1 d\Omega'_1$ and $\mathbf{k}'_1 = \mathbf{k}_1 - \mathbf{v}_P$.

The TCS for TI is obtained by

$$\sigma_{\text{TI}} = \sum_n \int d\mathbf{k}_2 \frac{d\sigma_{\text{TI}}^n}{d\mathbf{k}_2} = 1.202 \times \int d\mathbf{k}_2 \frac{d\sigma_{\text{TI}}^{n=1}}{d\mathbf{k}_2}. \quad (5)$$

The latter expression is based on the $1/n^3$ rule (see Eq. (4) and Refs. [11,17]) which enables one to express the cross sections summed over n . Similarly to DI, TI cross sections are also evaluated in both the IEM and FCA treatments.

Using the theoretical methods described above, we evaluated total cross sections of DI and TI processes for X^{q+} -He collisions with $q = 1-3$ in the impact energy range of 0.1–10 MeV/amu. Calculations performed within the IEM for the one-electron transition amplitudes (1) apply the same static target core (He⁺ ion) potential [18] for both the initial and final asymptotic states. In the case of the FCA (2) the electrons in the outgoing channel are represented by Coulomb waves corresponding to the effective charge $Z = 1.67$.

As mentioned earlier, DI and TI processes may proceed via different channels that present different contributions to the total yield of electrons at intermediate collision energies [1]. These can be specified in the many-body perturbative formulation and our treatment considers only a single term of this expansion, which is of second order in the projectile-electron interaction and corresponds to the TS2 mechanism. Therefore, in the following, calculations based on Eq. (1) or (2) are referred to as contributions of the TS2 mechanism to the DI or TI processes within the framework of the IEM or the FCA, respectively.

Other mechanisms that contribute to the TI process are the SO and various correlated mechanisms. As a general trend, their roles were found to increase with increasing impact energy [1,6]. A simple modification of the TS2 amplitudes allows one to account for the SO process: we evaluated the DI and the TI transition amplitudes [see Eq. (1) or (2)], where one of the one-electron amplitudes, e.g., $a_{i \rightarrow \mathbf{k}_i}^{1e}$ in Eq. (1), is replaced by the $\langle \varphi_{\mathbf{k}_i}^{\text{He}^{2+}} | \varphi_i \rangle$ overlap integral, with $\varphi_{\mathbf{k}_i}^{\text{He}^{2+}}$ being a continuum orbital in the field of the He²⁺ ion. The overlap integrals and therefore the one-electron SO probabilities are independent of the impact parameter. Therefore, the two-electron cross sections for SO are proportional to the one-electron cross sections [1]. This can be clearly seen in Fig. 1(a), where the TCSs for single ionization (SI, one of the electrons remains in the ground state of the target) and the contributions of TS2 and SO mechanisms to DI are presented. These SI and DI cross sections are evaluated within the framework of the IEM and it is clearly seen that the SO mechanism becomes important at high projectile energies.

When the TI process is considered the emission energy and the angle of one of the electrons is fixed, $\mathbf{k}_e = \mathbf{v}_P$ [see Eq. (4), ECC kinematics]. Therefore, the SO contribution exhibits a much stronger decrease with the increase of E_P than for DI. This is clearly seen in Fig. 1(a), which for comparison shows the TS2 cross sections for TI also evaluated within the IEM. Interestingly, in the case of the TI process the contribution of the SO mechanism compared to that of the TS2 mechanism is negligible in the whole range of impact energies studied.

A more realistic description for the two-electron transitions can be given within the FCA, which provides a simple tool to

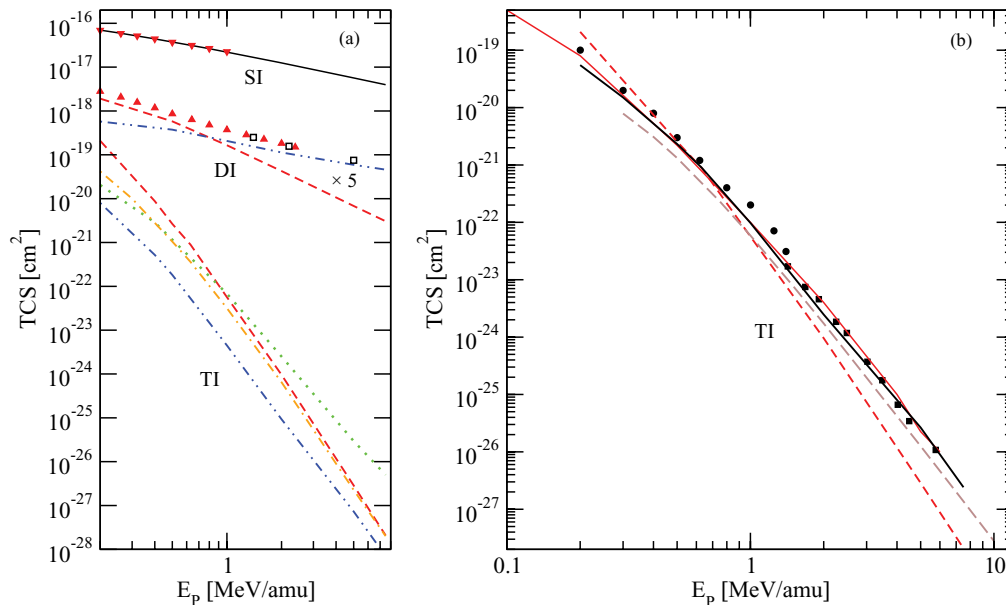


FIG. 1. (Color online) Total cross sections for SI, DI, and TI processes as functions of the projectile energy for H^+ -He collisions. (a) Present results: SI in IEM (thin solid line); SO for DI or TI in IEM (double-dot-dashed line); SO for TI in FCA (dotted line); TS2 for DI or TI in IEM (dashed line); TS2 for TI with inclusion of the Gamov factor for correlation in the final state in IEM (dot-dashed line). Experiment: \blacktriangle , Ref. [19]; \blacktriangledown , Ref. [20]; \square , Ref. [21]. (b) Present results: TS2 for TI in IEM as in panel (a) (dashed line); TI including contributions of TS2 and SO mechanism within FCA when correlations both in initial and final channels are taken into account (thick solid line). Other calculations: BDW-4B from Ref. [10] (long-dashed line) and 2nd Born from Ref. [6] (thin solid line). Experiment: \blacksquare , Ref. [22]; \bullet , Ref. [23] (see the text).

include correlations in the asymptotic channels [see Eq. (2)]. The FCA has already been applied in our earlier studies of DI [13] and here we extend those treatments to the TI process as described by Eqs. (4) and (5).

Let us first consider results when the correlation is included only in the initial state. When the Gamov factor is set equal to one in the FCA calculation [see Eq. (2)], the TS2 cross section for TI (not shown in the figure) is in close agreement with that of the IEM in Fig. 1(a). However, the SO cross sections in the FCA and IEM treatments are quite different: at high impact energies the SO cross section in the FCA dominates over the SO or TS2 cross sections obtained in the IEM owing to the electron correlation included in the asymptotic initial state. Therefore, it can be stated that the TI process is predominantly due to the TS2 mechanism at low impact energies and to the SO mechanism at high energies.

Now we turn our attention to the correlation in the final state. The emission of electrons with low velocities gives the main yield to the total DI cross section. Therefore, the main contribution to the final-state correlation is expected to originate from those electrons which are ejected to this emission regime. Since in the case of the TI the velocity of one of the electrons is fixed by the ECC kinematics, the role of the e - e final-state interaction is expected to be important mostly at low impact energies. This is confirmed by our results presented in Fig. 1(a), which shows that the e - e correlation plays an important role at low impact energies and becomes negligible for E_p above 1–2 MeV (see results for the TS2 contribution to TI with and without the Gamov factor, dot-dashed and dashed curves, respectively).

Figure 1(b) shows present results for TI in a calculation which includes contributions of the TS2 and SO mechanisms

within the framework of the FCA where correlations in both the initial and final states are taken into account. The cross sections of Fig. 1(a) for the contribution of TS2 within the IEM are also shown for comparison. Figure 1(b) shows that cross sections based solely on the TS2 mechanism within the IEM are larger and smaller than those observed experimentally at low and high impact energies, respectively. Including the contribution from the SO mechanism and a proper account of the e - e correlation in the initial and final asymptotic channels are essential for a better account of the TI process. This is clearly shown by the nice agreement between experiment and our results for TI which incorporate the TS2, SO, and asymptotic e - e correlation mechanisms. This nice agreement indicates that only relatively small room is left for contributions from the other correlated mechanisms mentioned above, which accordingly might be relevant only at higher impact velocities. We note that previous studies for DI have found that the Gamov factor overestimates the role of the final-state e - e correlation [5,24].

Figure 1(b) also presents the results of two recent calculations by other authors [6,10]. In Ref. [6], in agreement with our FCA results, a dominant role of the initial-state correlation at high impact energies was reported in a second-order Born description. These results show also nice agreement with the experiment in a wide range of impact energies. Correlation in the final state was excluded, but strong effects of the *off-shell* terms were reported at low impact energies in this application, where the interelectronic interaction and the summation over the intermediate states are evaluated with the help of further approximations. In the four-body Born distorted wave (BDW-4B) calculation of Ref. [10] uncorrelated wave functions were applied in both the initial and final states and the best agreement with the experiment was reported

at high impact energies. Like the present calculations, the models [6,10] are based on high-energy approximations for the electron-projectile interactions whose accuracies decrease with decreasing impact energies. This might also be responsible for the fact that these studies do not attribute the same roles for the different transition and correlation mechanisms at low impact velocities (approximations applied on the dynamics of electron transitions might artificially increase or decrease the role of e - e correlation or vice versa). At this point we must also refer to the systematic studies of DI by high energy electron impact [$(e,3e)$ process], where the computed cross sections were found to be quite sensitive to the amount of correlation included in the bound state wave function [25,26]. The present study deals with the total TI cross sections, which appear to be less sensitive to these details of the wave functions. This might explain the good results obtained with the relatively simple CI wave function in the present study.

The present description of TI relies on a treatment of DI in which the contribution of the SO mechanism depends only on properties of the target atom, whereas the cross sections attributed to the TS2 mechanism increase with increasing projectile charge. As a result, the relative contribution of the SO mechanism become less important when projectiles with larger q are considered. This can be observed in Figs. 2 and 3, where TI cross sections for He^{2+} and Li^{3+} projectiles are presented. As for the case of proton impact the dominant contributions are provided by the TS2 process at lower impact energies. Initial- and final-state correlations present also a negligible role in the TS2 mechanism at asymptotic impact velocities. At low impact energies the correlation in the final state has a determining role as, similar to the H^+ impact, the inclusion of the e - e interaction reduces the cross sections drastically. However, at the lowest impact energies, this effect is overemphasized, which probably signals the shortcoming

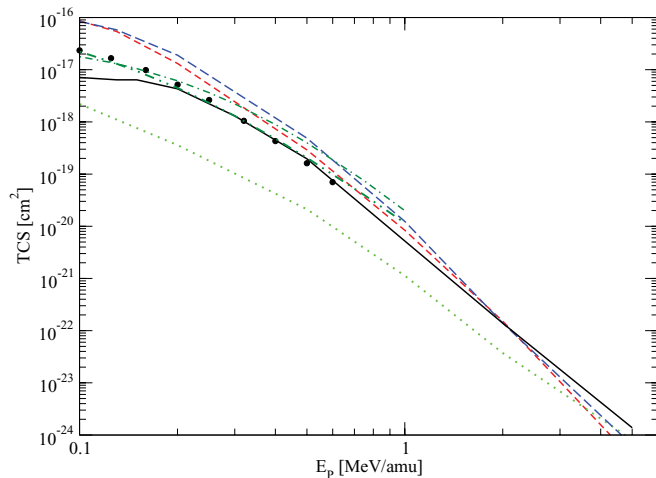


FIG. 2. (Color online) Total TI cross sections as functions of projectile energy for the He^{2+} -He collision. Present results: TS2 in IEM, (dashed line); TS2 in FCA (long-dashed line); SO in FCA (dotted line); including contributions of TS2 and SO mechanisms within FCA when correlations in both initial (CI) and final (Gamov factor) channels are taken into account (thick solid line) (see the text). Other calculations: CDW-4B [27] in prior (dot-dashed line) and in post (dot-dot-dashed) forms. Experiment: ●, Ref. [19].

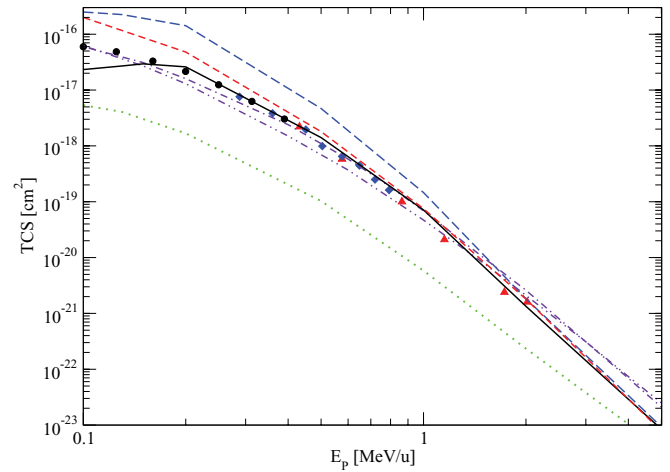


FIG. 3. (Color online) Same as for Fig. 2 but for Li^{3+} -He collisions. CDW-4B is from Ref. [28]; ●, Ref. [19]; ▲, Ref. [29]; and ◆, Ref. [30].

of the Gamov factor for modeling correlation in the final state (2). The contribution of the SO mechanism becomes relevant at above 2 and 5 MeV/amu impact energies for He^{2+} and Li^{3+} projectiles, respectively. Available experimental data are also presented for comparison in Figs. 2 and 3. Except in the low impact energy range ($E_p \leq 0.2$ – 0.3 MeV/amu) the present treatment, which includes contributions of the SO and asymptotic e - e correlation mechanisms, is in good agreement with the measured data.

Figures 2 and 3 also show the theoretical results of the CDW-4B model [27,28]. These results give a better account of the experiments than the present ones at low impact energies, where in our treatment the final-state correlation is overemphasized. Discrepancies appear also at asymptotic impact velocities; however, the lack of experimental data in this region makes it difficult to draw a definite conclusion. Finally the application of the CDW-EIS method for describing TI in He^{2+} -He collisions by Galassi *et al.* [9] should be mentioned. Their treatment was also based on the IEM and they evaluated TI cross sections in the 0.1–0.5 MeV/amu impact energy range. These results, not shown, reveal nice agreement with those of the present TS2 displayed in Fig. 2, which point out that charge transfer reactions can realistically be considered as special cases of ionization processes [see Eqs. (4) and (5)].

In summary, various roles of different transition mechanisms have been identified in the description of DI and TI reactions at different collision energies. Within the framework of the IEM the SO mechanism predominates the TS2 mechanism in the DI process at high impact energies. At the same time the prevailing mechanism in the TI process is the TS2 mechanism in the whole range of impact energies considered in this study. The SO mechanism, as in DI, becomes dominant in TI at high impact velocities when correlation in the asymptotic initial state is taken into account. Furthermore, the e - e correlation in the final channel is relevant at lower impact energies. The present description of TI including these ingredients shows a nice account of the experimental data in a wide range of collision energies for H^+ + He collisions. Similar conclusions can be drawn for the other collisions with He^{2+} and Li^{3+}

projectiles. However, shortcomings of the present treatment, especially in the description of the final-state e - e correlation, become more emphasized at low collision energies. More experimental results for a more profound test of the theories are desirable.

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