Comment on “Dynamics of transfer ionization in fast ion-atom collisions”

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Abstract

We inspect the first-order electron-electron capture scenario for transfer ionization that has been recently formulated by Voitkiv et al. (Phys. Rev. A 86, 012709 (2012) and references therein). Using the multichannel scattering theory for many-body systems with Coulomb interactions, we show that this scenario is just a part of the well-studied Oppenheimer-Brinkmann-Kramers approximation. Accurate numerical calculations in this approximation for the proton-helium transfer ionization reaction exhibit no appreciable manifestation of the claimed mechanism.

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I. INTRODUCTION

Recently Voitkiv et al. published a series of papers [1–3] putting forth a new first-order capture mechanism that can be called electron-electron Auger (or ee-Auger) [3]. According to this mechanism, the electron undergoes a nonradiative transition from the atomic state to the bound state of the projectile, transferring the energy excess to the another atomic electron which is emitted from the atom. This scenario resembles a kind of Auger decay and to be contrasted with the first-order radiative capture [4] which is accompanied by emission of a photon instead of an electron. A clear signature of the ee-Auger mechanism, according to Voitkiv et al., is emission of the electron in the direction opposite to the projectile motion (in the rest frame of the atom).

A quantum mechanical explanation how a target electron can be captured into a bound state of a fast moving projectile (proton) was given by Oppenheimer, Brinkmann and Kramers (OBK) [5]. In the OBK scenario the electron transfer proceeds via an overlap of initial and final wave functions of the projectile-target system. This so-called kinematical capture relies strongly on the radial and angular electron correlations in the target, if we consider transfer excitation (TE) and transfer ionization (TI) processes. It must be noted that in quantum mechanics the transition of an electron to the projectile bound state can be nonradiative, and the energy excess can be carried away by a third body that participates in the reaction. It is the first-order, Born term.

In quantum mechanics fast processes are usually treated within Born approximations. This framework is directly applicable in the case of two-body scattering, but requires additional careful considerations in the many-body case. The situation of particular importance is when the entrance channel of the reaction is different from its exit channel, for example, as it is in capture processes. Within the multichannel scattering theory, the OBK mechanism can be attributed to the first Born approximation (FBA), whereas the Ne- [6] and ee- [7] Thomas mechanisms can be described using the second Born approximation (SBA). Any Born approximation is a sum of matrix elements. Each of them corresponds to a particular interaction that enters a total perturbation potential. For example, the OBK matrix element is one of the three FBA terms (see below). The SBA contains twelve different terms, and only two of them correspond to the Ne- and ee-Thomas mechanisms.

In this Comment, we examine the mechanism suggested by Voitkiv et al. on the basis of
consistent multichannel scattering theory. We show that, in contrast to the claim of Ref. [1], it is not new and previously undiscussed. Namely, it is just a part of the usual kinematic capture in the OBK approximation, and the correlated electron-electron emission is nothing else but a result of the electron-electron correlations in the target atom. Moreover, the main formula, employed by Voitkiv et al. for the transition amplitude, contains apparent flaws.

Voitkiv et al. use in their works [1–3] a time-dependent approach. In this Comment, we consider a time-independent formulation, noting that both treatments are equivalent at high projectile velocities [8]. Atomic units (a.u., \( \hbar = e = m_e = 1 \)) are used throughout unless otherwise specified.

II. ELEMENTS OF MULTICHANNEL SCATTERING THEORY

In this section we remind basic formulas of quantum scattering theory for many-body systems. More mathematical details, particularly for the case involving charged fragments, one can find in the review articles [8, 9]. A set of relative momenta defining motion of \( n_\alpha \) fragments colliding in the asymptotic channel is denoted by \( \vec{p}_\alpha \). In turn, the ket vector \( |\phi_\alpha \rangle \) stands for a product of bound (spectral) state wave functions, which define the channel \( \alpha \). Hence, the ket vector \( |\phi_\alpha, \vec{p}_\alpha \rangle \) is the eigenfunction of the asymptotic hamiltonian \( H_\alpha \):

\[
(E - H_\alpha)|\phi_\alpha, \vec{p}_\alpha \rangle = 0.
\]

The total hamiltonian is \( H = H_\alpha + V_\alpha = H_\beta + V_\beta \), where \( V_\alpha \) (\( V_\beta \)) is a sum of two-body interaction potentials, which we consider as perturbation, and they define the terms of the Born series: FBA, SBA, and so on.

The amplitude of the transition from the channel \( \alpha \) to the channel \( \beta \) can be presented using two forms. These are the post-form

\[
T_{\beta\alpha}(E) = \langle \phi_\beta, \vec{p}_\beta | V_\beta | \Psi^+_\alpha(\vec{p}_\alpha) \rangle,
\]

and the prior-form

\[
\tilde{T}_{\beta\alpha}(E) = \langle \Psi^-_\beta(\vec{p}_\beta) | V_\alpha | \phi_\alpha, \vec{p}_\alpha \rangle,
\]

where \( (E - H)|\Psi^\pm_{\alpha(\beta)}(\vec{p}_{\alpha(\beta)}) \rangle = 0 \). It is straightforward to show that (see, for instance, Refs. [10, 11])

\[
T_{\beta\alpha}(E) = \tilde{T}_{\beta\alpha}(E).
\]

Moreover, since \( V_\alpha = H - H_\alpha \) and \( V_\beta = H - H_\beta \), the relation (3) holds true in the FBA
case as well, that is, on the energy shell the FBA post- and prior-amplitudes coincide,

\[ \langle \phi_\beta, \vec{p}_\beta | V_\beta | \phi_\alpha, \vec{p}_\alpha \rangle = \langle \phi_\beta, \vec{p}_\beta | V_\alpha | \phi_\alpha, \vec{p}_\alpha \rangle. \]

The above formulas are valid only in the case where colliding fragments do not interact via long-range, Coulomb-like potentials at asymptotically large separation distances. This can be formulated using the Zommerfeld parameter of the channel

\[ \eta_\gamma = \sum_{i<j} Z_i^{(\gamma)} Z_j^{(\gamma)} v_{ij}^{(\gamma)}, \]

where \( Z_i^{(\gamma)} \) and \( Z_j^{(\gamma)} \) are the total charges of the colliding fragments \( i \) and \( j \), and \( v_{ij}^{(\gamma)} \) is their relative velocity. If the Zommerfeld parameter differs from zero, Eqs. (1) and (2) become more complicated [8, 9], because the asymptotic states \( |\phi_\alpha(\beta), \vec{p}_\alpha(\beta)\rangle \) do not obey the correct asymptotic conditions anymore.

Let us apply the above general formulas to the fast TI reaction \( \text{H}^+ + \text{He} \rightarrow \text{H} + \text{He}^{2+} + e \) discussed in the papers of Voitkiv et al. [1–3]. The authors utilize the post-amplitude, which in the nonsymmetrized FBA can be written as

\[ T_{fi}^{FBA}(E) = \langle \phi_{p1}, \varphi_{N2}(\vec{k}), \vec{p}_H | V_{p1} + V_{p2} + V_{12} + V_{Np} | \Phi_0, \vec{p}_0 \rangle. \]  \hspace{1cm} (4a)

In Eq. (4a), electrons are labelled by “1” and “2”, whereas “p” labels the fast proton projectile, and “N” the target nucleus. The wave function \( |\phi_{p1}\rangle \) is the bound (ground) state of atomic hydrogen, \( |\varphi_{N2}(\vec{k})\rangle \) the continuum state of the He\(^+\) ion, \( |\Phi_0\rangle \) the helium wave function, \( \vec{p}_0 \) the proton momentum, \( \vec{p}_H \) the hydrogen momentum, and \( \vec{k} \) the momentum of the emitted electron. This amplitude is equal to that in the prior-form

\[ \tilde{T}_{fi}^{FBA}(E) = \langle \phi_{p1}, \varphi_{N2}(\vec{k}), \vec{p}_H | V_{p1} + V_{p2} + V_{Np} | \Phi_0, \vec{p}_0 \rangle. \]  \hspace{1cm} (4b)

It should be noted that Eqs. (1) and (2) are applicable because there is no long-range asymptotic interaction in the initial and final channels. In the present case, it is clearly fulfilled in the initial channel (the He atom is neutral, \( Z_{He} = 0 \)). It is also fulfilled in the final channel, because \( Z_H = 0 \), and we use in Eq. (4) the spectral Coulomb functions \( \varphi_{N2}(\vec{k}) \) instead of plain waves, so that the neutral hydrogen subsystem does not asymptotically interact with the He\(^+\) subsystem.

From the equality of the FBA amplitudes (4a) and (4b) we find that

\[ \langle \phi_{p1}, \varphi_{N2}(\vec{k}), \vec{p}_H | V_{p1} | \Phi_0, \vec{p}_0 \rangle = \langle \phi_{p1}, \varphi_{N2}(\vec{k}), \vec{p}_H | V_{N1} + V_{12} | \Phi_0, \vec{p}_0 \rangle. \]  \hspace{1cm} (5)
The matrix element on the left-hand side amounts to the OBK approximation. It can be easily transformed into the overlap of the initial and final wave functions described in Ref. [5]. The matrix element on the right-hand side is the same OBK, but in the post-form representation. It is important to note that within FBA the physical effect of the interaction of the transferred electron with the proton projectile is exactly equal to that of the interaction of the same electron with the residual target ion. This means that the ee-Auger mechanism, which is attributed by Voitkiv et al. to the $V_{12}$ contribution in the right-hand side of Eq. (5), is not independent and is included in the OBK scenario. It has been repeatedly shown (see, for instance, Ref. [12]) that, in the FBA prior-amplitude (4b), even the OBK term is not leading in some kinematical situations. In other words, all four terms in (4a) should be considered in the general case.

III. DISTORTED WAVE APPROXIMATIONS

From Eqs. (1) and (2) one can derive the higher Born terms as well as different versions of the distorted wave Born approximation (DWBA). For example, in [8] the eikonal approximation was derived, which introduces in the prior-form FBA matrix element (4b) a distorting phase factor,

$$|\phi_{p1}, \varphi_{N2}^{-}(\vec{k}), \vec{p}_H) \rightarrow e^{(i/v_p)\delta_f} |\phi_{p1}, \varphi_{N2}^{-}(\vec{k}), \vec{p}_H).$$

The details concerning its derivation one can find in Ref. [13]. We note that it is the asymptotic form of the product

$$e^{(i/v_p)\delta_f} \rightarrow \Lambda_f^- = \Lambda_{p2}^- \Lambda_{pN}^- \Lambda_{N1}^- \Lambda_{12}^-,$$

with

$$\Lambda_{Z_1Z_2}^- = \exp \left( -\frac{\pi Z_1 Z_2}{2v_{rel}} \right) \Gamma \left( 1 - \frac{Z_1 Z_2}{v_{rel}} \right) iF_1 \left[ \frac{Z_1 Z_2}{v_{rel}}, 1; -i(v_{rel}r_{rel} + v'_{rel}r'_{rel}) \right],$$

where $v_{rel}$ and $v'_{rel}$ are the relative velocity and position of the pair of particles. Each factor $\Lambda_{Z_1Z_2}^-$ in (6) describes the distortion of interactions between different constituents of the two final compound subsystems, H and He+. In some sense, it is a 4C model (in analogy with well known 3C and 6C models in the scattering theory [14]).
The same procedure we can utilize in the case of the FBA matrix element in the post-form, replacing in (4a) 

\[ |\Phi_0, \vec{p}_0\rangle \rightarrow e^{(i/\nu_p)|\delta_i|} |\Phi_0, \vec{p}_0\rangle. \]

Here, again, 

\[ e^{(i/\nu_p)|\delta_i|} \rightarrow \Lambda_i^+ = \Lambda^{p_1+}_{p_1^+} \Lambda^{p_2^+}_{p_2^+}, \] \hspace{1cm} (7)

and

\[ \Lambda^{+}_{Z_1Z_2} = \exp\left(-\frac{\pi Z_1 Z_2}{2v_p}\right) \Gamma \left(1 + i \frac{Z_1 Z_2}{v_p}\right) \frac{1}{1} \frac{F_1}{1} \left[-i \frac{Z_1 Z_2}{v_p}, 1; i(v_p r_{rel} - \vec{u}_p r_{rel})\right]. \]

Each factor \( \Lambda^{+}_{Z_1Z_2} \) in (7) describes the distortion due to interactions between the projectile proton and different constituents of the helium atom. This approximation is analogous to the 3C model.

Distortion factors (6) and (7) are typical of the continuum-distorted-wave (CDW) model. The main requirement of this model is to obey the correct Coulomb asymptotic conditions in the initial and final channels of the reaction [8]. These conditions are given by \( (\gamma = \alpha, \beta) \) [15]

\[ e^{-iH_{\gamma} t} |\Psi_{\gamma}^{+}(\vec{p}_{\gamma})\rangle \rightarrow e^{-iE_{\gamma} t} e^{i \ln |t|} |\phi_{\gamma}, \vec{p}_{\gamma}\rangle, \quad t \rightarrow \mp \infty, \] \hspace{1cm} (8)

where \( A_{\gamma}(\vec{p}_{\gamma}) \) is the so-called Dollard phase. Representations (6) and (7) are not unique, and other forms are also available (see, for instance, Refs. [8, 16] and references therein).

IV. EE-AUGER MECHANISM IN THE RIGOROUS SCATTERING THEORY

In Refs. [1, 3], the authors present calculations of contributions from, as they suppose, different mechanisms. These include the OBK (or capture-shakeoff), two-step (or independent transfer ionization [3]), ee-Thomas, and ee-Auger. The details concerning calculation of these contributions, except that of the ee-Auger, in Refs. [1, 3] are rather scarce. Referring to the CDW model, Voitkiv et al. use the following formula for the ee-Auger amplitude (see Eq. (7) of Ref. [3]):

\[ T_{f_i}^{EEA}(E) = \langle \phi_{p_1}, \Phi_{N_2}(\vec{k}), \Lambda_{N_1}^{-}, \vec{p}_H |V_{12}| \Phi_0, \Lambda^{p_1^+}_{p_1^+}, \vec{p}_0\rangle. \] \hspace{1cm} (9)

Thus, when summing the calculated in this way contribution with that of the OBK, Voitkiv et al. take into account the first-order ee-Auger mechanism twice. Such a conclusion immediately follows from Eq. (5), which states that the ee-Auger contribution, connected with
the $V_{12}$ term on the right-hand side, is already taken into account in the OBK approximation given by the left-hand side. And the presence of the distorting factors does not principally change this apparent flaw in the calculations of Refs. [1–3]. It should be also noted that formula (9) contradicts the CDW model. First, it explicitly violates the correct asymptotic condition (8) both in the initial and in the final channels of the discussed reaction. Second, even if one uses the correct asymptotic factors (6) and (7) in Eq. (9), the post-form of the CDW model assumes that the perturbation is given by the nonorthogonal kinetic energy $-\nabla N_1 \cdot \nabla p_1$ (see details in Refs. [8, 16], also in [17]), which is clearly not equivalent to $V_{12}$. It should be also remarked with respect to Eq. (9) that it violates not only the correct asymptotic conditions, but also the identity of the helium electrons.

In Ref. [3] the final distortion factor was neglected in calculations, $\Lambda_f = 1$, as being not so much significant. As remarked in Ref. [3], without the initial distortion factor, i.e., when $\Lambda_i = 1$, the contribution of the $ee$-Auger mechanism calculated there becomes much larger, while that of $ee$-Thomas vanishes. In view of these remarks, one might expect that neglecting the distortion effects does not reduce the role of the $ee$-Auger mechanism. Thus, if conclusions of Voitkiv et al. are correct, from Eq. (5) it follows that the discussed mechanism must manifest itself in the calculations based on the OBK approximation, because the former is a part of the latter.

The quantity that was studied numerically in Refs. [1–3] is the double differential cross section (DDCS)

$$\frac{d^2\sigma}{dk_x dk_z} = \frac{2k_\perp}{(2\pi)^3v^2} \int_0^{2\pi} d\varphi_k \int d^2q_\perp |T_{fi}|^2, \quad (10)$$

which describes a 2D distribution of the momentum components of the emitted electron ($k_x = k_\perp \cos \varphi_k, \ k_y = k_\perp \sin \varphi_k$). Numerical results for the DDCS using the prior-OBK approximation (5) are shown in Fig. 1a. In these calculations an accurate, highly correlated trial helium function from [19] is employed. The kinematical situation is the same as that of Fig. 1 in Ref. [3]. We see a general tendency for the ejected electron to be preferably emitted in the backward lobe ($k_z < 0$), which is typical for the OBK with highly correlated trial helium wave functions. Next Born approximations are expected to contribute to the forward lobe ($k_z > 0$).

However, while there is a common feature such as a maximum located at $k_z = 0$, we find no maximum located at negative $k_z$ values (approximately, at $k_z \approx -3.0$), in contrast to the
results presented in Fig. 1 of Ref. [3]. The latter feature is, according to Voitkiv et al., a clear signature of the ee-Auger mechanism. Thus, our numerical calculations using the accurate, highly correlated wave function of helium do not support the findings of Refs. [1–3]. In this connection, it should be noted that angular correlations due to the $V_{12}$ interaction play a very important role, if being included in the trial helium wave function $\Phi(\vec{r}_1, \vec{r}_2)$. They strongly influence the momentum distribution of the emitted electron in the backward direction, which is due to the shake-off mechanism [18]. And we see manifestation of their effect in the region $k_z < 0$. However, it is quite different from manifestation of the ee-Auger mechanism claimed by Voitkiv et al. According to Eq. (5), the effect of $V_{12}$ found in Refs. [1–3] (see Fig. 1b) is clearly cancelled by the other first-order mechanism, which involves the target nucleus (the $V_{N1}$ term). This finding markedly illustrates the importance of accounting for all the binary interactions between the particles taking part in the reaction.

Some comments should be made with regard to the equivalence of the post- and prior-forms of the transition amplitude. It is realized only if the exact helium wave function is employed. In that case, we have the following equation for this function:

$$ (\varepsilon^H_{0} - h_{10} - h_{20} - V_{N2})|\Phi_0\rangle = (V_{N1} + V_{12})|\Phi_0\rangle. \quad (11) $$

It can be readily shown, using energy conservation and properties of the final asymptotic state, that the projectile-electron potential $V_{p1}$ in the left-hand side of Eq. (5) can be replaced with the operator $(\varepsilon^H_{0} - h_{10} - h_{20} - V_{N2})$ occurring in the left-hand side of Eq. (11). Thus, the left- and right-hand sides of the Schrödinger equation (11) correspond to, respectively, the prior- and post-matrix elements in Eq. (5). This feature explains a well documented fact that Eq. (5) is fulfilled to a good approximation in the case of an accurate trial helium function, which is typically obtained from a variational procedure. But if the trial function is poor, then the right-hand side of Eq. (11), which is related to the post-matrix element in Eq. (5), appears to yield a better approximation to the exact result than in the case of the left-hand side. This observation explains why the results of Voitkiv et al., using the correlated and uncorrelated helium functions in their post-amplitude, are similar (see Fig. 7 of Ref. [3]).
V. SUMMARY AND CONCLUSIONS

We considered theoretically a transfer ionization channel in a fast proton-helium collision, focusing on the so-called first-order electron-electron capture mechanism proposed recently by Voitkiv et al. [1–3]. It was shown, using consistent quantum collision theory, that this specific mechanism is included in a well known OBK scenario. The formula employed by Voitkiv et al. for the transition amplitude is found to be unjustified and contradicting the CDW model. The OBK calculations with an accurate trial helium function exhibited no signature of the $ee$-Auger process.

Neither distorted waves nor different representations of the amplitude should change the basic physics of the process, which is essentially governed by the projectile-target interaction. Voitkiv et al. use the post-formulation, whereas many authors prefer the prior-formulation. However, in approximate treatments, one should try to achieve their convergence (see, for instance, Ref. [20]), since the physics of the process does not depend on the form of the matrix element. Interaction of the captured electron with both nuclei is important. However, if it is accounted for within the distorted-wave treatment, then such a treatment must be carried out in a mathematically correct fashion. To our knowledge, all the requisites for this problem can be found, for example, in the review article of Belkić et al [8].

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Figure 1: (Color online) DDCS (10) in barn/(a.u.)^2, $E_p = 3.6$ MeV. a) $T_{fi} = \text{prior-OBK}$, b) $T_{fi} = \langle \phi_{p1}, \varphi_{N2}(\vec{k}), \vec{p}_H|V_{12}|\Phi_0, \vec{p}_0 \rangle$. 