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

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We inspect the first-order electron-electron-capture scenario for transfer ionization that has been recently formulated by Voitkiv and Ma [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-Brinkman-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 and co-workers published a series of papers [1–4] putting forth a new first-order capture mechanism that can be called electron-electron Auger (eeA) [3]. According to this mechanism, the electron undergoes a nonradiative transition from the target atomic state to the bound state of the projectile, transferring the energy excess to another atomic electron which is emitted from the atom. This scenario resembles a kind of Auger decay and is to be contrasted with the first-order radiative capture [5], which is accompanied by the emission of a photon instead of an electron. A clear signature of the eeA mechanism, according to Voitkiv and co-workers, is the emission of the electron in the direction opposite to the projectile motion (in the rest frame of the target atom).

Indeed, 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. The first quantum-mechanical explanation on how a target electron can be captured into a bound state of a fast moving projectile (proton) was given by Oppenheimer, Brinkman, and Kramers (OBK) [6]. 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 and transfer ionization (TI) processes.

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- [7] and ee- [8] 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 *prior* terms (see below). The SBA contains 12 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 and co-workers 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 within the FBA, the electron-electron interaction is, to a large degree, consumed by the electron correlation in the initial state of the electrons. The intermediate and residual effect of this interaction is certainly physically relevant, but at first sight, it appears to be a part of a higher-order approach, and we, thus, question the validity of the first-order eeA mechanism. Moreover, the main formula, employed by Voitkiv *et al.* for the transition amplitude, seems to contain flaws.

In their papers [1–4], Voitkiv and co-workers use a time-dependent approach. In this Comment, we consider a time-independent formulation, noting that both treatments are equivalent at high projectile velocities [9]. Atomic units [(a.u.), $\hbar = e = m_e = 1$] are used throughout, unless otherwise specified.

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II. ELEMENTS OF MULTICHANNEL SCATTERING THEORY

In this section, we remind the reader about the basic formulas of quantum scattering theory for many-body systems. Particularly, for the case involving charged fragments, one can find more mathematical details in the review articles [9,10]. A set of relative momenta defining the motion of n_α fragments colliding in the asymptotic channel is denoted by $\vec{p}_\alpha$. In turn, ket vector $|\phi_\alpha\rangle$ stands for a product of bound- (spectral-) state wave functions, which define channel α. Hence, ket vector $|\phi_\alpha,\vec{p}_\alpha\rangle$ is the eigenfunction of the asymptotic Hamiltonian H_α: $(E-H_\alpha)|\phi_\alpha,\vec{p}_\alpha\rangle=0$. The total Hamiltonian is $H=H_\alpha+V_\alpha=H_\beta+V_\beta$, where V_α (V_β) 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 channel α to channel β can be presented using two forms. These are the *post* form

$$T_{\beta\alpha}(E)=\langle\phi_\beta,\vec{p}_\beta|V_\beta|\Psi^+_\alpha(\vec{p}_\alpha)\rangle, \tag{1}$$

and the *prior* form

$$\tilde{T}_{\beta\alpha}(E)=\langle\Psi^-_\beta(\vec{p}_\beta)|V_\alpha|\phi_\alpha,\vec{p}_\alpha\rangle, \tag{2}$$

where $(E-H)|\Psi^\pm_{\alpha(\beta)}(\vec{p}_{\alpha(\beta)})\rangle=0$. It is straightforward to show that (see, for instance, Refs. [11,12])

$$T_{\beta\alpha}(E)=\tilde{T}_{\beta\alpha}(E). \tag{3}$$

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 Sommerfeld parameter of the channel,

$$\eta_\gamma=\sum_{i<j}\frac{Z_i^{(\gamma)}Z_j^{(\gamma)}}{v_{ij}^{(\gamma)}}\quad(\gamma=\alpha,\beta),$$

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 Sommerfeld parameter differs from zero, Eqs. (1) and (2) become more complicated [9,10] 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 $H^+ + He \to H + He^{2+} + e$ discussed in the papers of Voitkiv and co-workers [1–4]. The authors utilize the *post* amplitude, which, in the nonsymmetrized FBA, can be written as

$$T^{\rm FBA}_{fi}(E)=\langle\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H|V_{N1}+V_{p2}+V_{12}+V_{Np}|\Phi_0,\vec{p}_0\rangle. \tag{4a}$$

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

$$\tilde{T}^{\rm FBA}_{fi}(E)=\langle\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H|V_{p1}+V_{p2}+V_{Np}|\Phi_0,\vec{p}_0\rangle. \tag{4b}$$

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, $Z_{\rm He}=0$). It is also fulfilled in the final channel because $Z_{\rm H}=0$, and in Eq. (4), we use 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

$$\langle\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H|V_{p1}|\Phi_0,\vec{p}_0\rangle=\langle\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H|V_{N1}+V_{12}|\Phi_0,\vec{p}_0\rangle. \tag{5}$$

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. [6]. The matrix element on the right-hand side is the same OBK but in its *post*-form representation. It is important to note that, within the 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 and this interaction includes not only the ee, but also the Ne potential. This means that the eeA mechanism, which is attributed by Voitkiv and co-workers to the V_{12} contribution on the right-hand side of Eq. (5), is not independent and is included in the FBA scenario. It has been repeatedly shown (see, for instance, Ref. [13]) 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 at equal footing, and no one can be lost.

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. For example, in Ref. [9], the eikonal approximation was derived, which, in the *prior*-form FBA matrix element (4b), introduces a distorting phase factor,

$$|\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H\rangle\to e^{(i/v_p)\hat{\delta}_f}|\phi_{p1},\varphi^-_{N2}(\vec{k}),\vec{p}_H\rangle.$$

One can find the details concerning its derivation in Ref. [14]. We note that it is the asymptotic form of the product,

$$e^{(i/v_p)\hat{\delta}_f}\to\Lambda^-_f=\Lambda^-_{p2}\Lambda^-_{pN}\Lambda^-_{N1}\Lambda^-_{12}, \tag{6}$$

with

$$\Lambda^-_{Z_1Z_2}=\exp\left(-\frac{\pi Z_1Z_2}{2v_p}\right)\Gamma\left(1-i\frac{Z_1Z_2}{v_p}\right)\times{}_1F_1\left[i\frac{Z_1Z_2}{v_p},1;-i(v_pr_{\rm rel}+\vec{v}_p\vec{r}_{\rm rel})\right],$$

where $\vec{r}_{\rm rel}$ is the relative 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 [15]).

We can utilize the same procedure in the case of the FBA matrix element in the *post* form, replacing, in (4a),

$$|\Phi_0,\vec{p}_0\rangle \to e^{(i/v_p)\hat{\delta}_i}|\Phi_0,\vec{p}_0\rangle.$$

Here, again,

$$e^{(i/v_p)\hat{\delta}_i} \to \Lambda^{+}_{i} = \Lambda^{+}_{p1}\Lambda^{+}_{p2}\Lambda^{+}_{pN}, \tag{7}$$

and

$$\Lambda^{+}_{Z_1Z_2} = \exp\left(-\frac{\pi Z_1Z_2}{2v_p}\right)\Gamma\left(1+i\frac{Z_1Z_2}{v_p}\right) \times {}_1F_1\left[-i\frac{Z_1Z_2}{v_p},1;i(v_pr_{\rm rel}-\vec{v}_p\vec{r}_{\rm rel})\right].$$

Each factor $\Lambda^{+}_{Z_1Z_2}$ in (7) describes the distortion due to interactions between the projectile proton and the 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 [16]. The main requirement of this model is to obey the correct Coulomb asymptotic conditions in the initial and final channels of the reaction [9]. These conditions are given by ($\gamma=\alpha,\beta$) [17],

$$e^{-iH_\gamma t}|\Psi^{\pm}_{\gamma}(\vec{p}_\gamma)\rangle \to e^{-iE_\gamma t\pm i\eta_\gamma\ln|t|\pm iA_\gamma(\vec{p}_\gamma)}|\phi_\gamma,\vec{p}_\gamma\rangle, \quad t\to\mp\infty, \tag{8}$$

where $A_\gamma(\vec{p}_\gamma)$ is the so-called Dollard phase. Representations (6) and (7) are not unique, and other forms are available (see, for instance, Refs. [9,18] and references therein).

IV. *eeA* 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 kinematical capture), 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 and Ma use the following formula for the *eeA* amplitude (see Eq. (7) of Ref. [3]):

$$T^{\rm EEA}_{fi}(E) = \langle\phi_{p1},\varphi^{-}_{N2}(\vec{k}),\Lambda^{-}_{N1},\vec{p}_H|V_{12}|\Phi_0,\Lambda^{+}_{p1},\vec{p}_0\rangle. \tag{9}$$

Thus, when summing the contribution calculated in this way with that of the OBK, Voitkiv and Ma 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. [1–4]. It should also be 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 $-\vec{\nabla}_{N1}\cdot\vec{\nabla}_{p1}$ (see details in Refs. [9,18] also in Ref. [19]), which is clearly not equivalent to V_{12}.

In Ref. [3], the final distortion factor was neglected in calculations $\Lambda_f = 1$ as being not very significant. As remarked in Ref. [3], without the initial distortion factor, i.e., when $\Lambda_i = 1$, the contribution of the *eeA* mechanism calculated there becomes much larger, whereas, 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 *eeA* mechanism. Thus, if conclusions of Voitkiv and co-workers 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. [1–4] is the double differential cross section (DDCS),

$$\frac{d^2\sigma}{dk_\perp dk_z} = \frac{2k_\perp}{(2\pi)^5v_p^2}\int_0^{2\pi}d\varphi_k\int d^2q_\perp|T_{fi}|^2, \tag{10}$$

which describes a two-dimensional 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. 1(a). In these calculations, an accurate highly correlated trial helium function from Ref. [20] 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. Higher Born approximations are expected to contribute to the forward lobe ($k_z > 0$).

However, although 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 and Ma, 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 [21,22]. And we see a manifestation of their effect in the region $k_z < 0$. However, it is quite different from the manifestation of the *eeA* mechanism claimed by Voitkiv and co-workers. According to Eq. (5), the effect of V_{12} found in Refs. [1–4] (see Fig. 1(b) in this paper and compare with Fig. 1 in Ref. [3]) is clearly canceled 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.

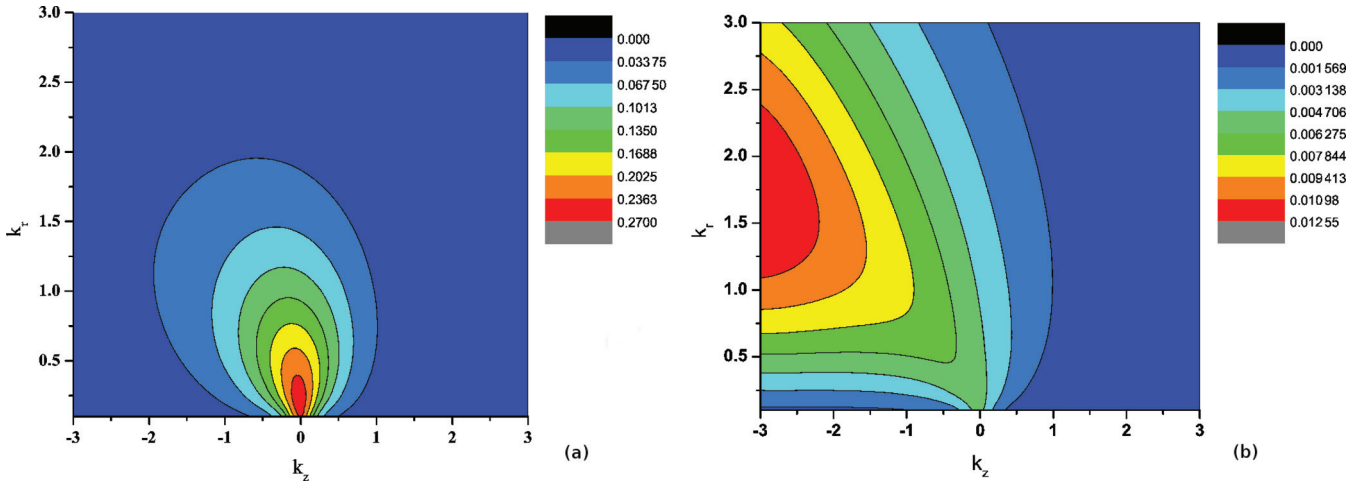

FIG. 1. (Color online) DDCS (10) in b/(a.u.)^2, $E_p = 3.6$ MeV. (a) $T_{fi} = prior$ OBK (5), (b) $T_{fi} = \langle \phi_{p1}, \varphi^-_{N2}(\vec{k}), \vec{p}_H | V_{12} | \Phi_0, \vec{p}_0 \rangle$.

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:

$$\left(\varepsilon_0^{\mathrm{He}} - h_{10} - h_{20} - V_{N2}\right)|\Phi_0\rangle = (V_{N1} + V_{12})|\Phi_0\rangle. \quad (11)$$

It can be readily shown, using energy conservation and properties of the final asymptotic state, that the projectile-electron potential V_{p1} on the left-hand side of Eq. (5) can be replaced with the operator $(\varepsilon_0^{\mathrm{He}} - h_{10} - h_{20} - V_{N2})$ occurring on the left-hand side of Eq. (11). Thus, the left- and right-hand sides of the Schrödinger equation (11) correspond to the *prior*- and *post*-matrix elements, respectively, 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 and Ma, using the correlated and uncorrelated helium functions in their *post* amplitude, are similar (see Fig. 7 of Ref. [3]).

V. SUMMARY AND CONCLUSIONS

We theoretically considered 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 and co-workers [1–4]. We have shown that the *eeA* term considered by Voitkiv and co-workers appears to be already included in the OBK transfer mechanism. This indicates that the heuristic approach of Voitkiv and co-workers should be analyzed in a more formal way, despite some potentially coincidental agreement with the experiment in Ref. [1].

The formula employed by Voitkiv and co-workers for the transition amplitude is found to be unjustified and contradicting the CDW model. The full OBK calculations with an accurate trial helium function exhibited no signature of the *eeA* process. The authors did not account for the matrix element $\langle \phi_{p1}, \varphi^-_{N2}(\vec{k}), \vec{p}_H | V_{N1} | \Phi_0, \vec{p}_0 \rangle$ in (5), which cancels the *eeA* mechanism.

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 and co-workers 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. [19]) since the physics of the process does not depend on the form of the matrix element. Interaction of the transferred 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 articles of Belkić *et al.* [9].

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