Effect of angular electron correlation in He: Second-order calculations for transfer ionization

A. L. Godunov, Colm T. Whelan, and H. R. J. Walters

1Department of Physics, Old Dominion University, Norfolk, Virginia 23529-0116, USA
2Department of Applied Mathematics and Theoretical Physics, The Queen’s University of Belfast, Belfast BT7 1NN, United Kingdom

(Received 6 May 2008; published 28 July 2008)

Fully differential cross sections for the transfer-ionization process \( \text{H}^+ + \text{He} \rightarrow \text{H}^0 + \text{He}^{2+} + e^- \) are calculated. The theoretical model is an expansion of the transition amplitude through the second order in the projectile-target interaction with highly correlated wave functions for the target. The theoretical results exhibit a strong sensitivity to angular electron correlation in both first- and second-order calculations. The results of the calculations are in sensible agreement with recent COLTRIMS observations.

DOI: 10.1103/PhysRevA.78.012714

I. INTRODUCTION

The problem of electron-electron correlation is fundamental to understanding atomic and molecular systems with more than one electron. Considering multiple-electron transitions is a direct way to study the electron correlation problem. Over the past several decades there have been numerous experimental and theoretical studies of double-electron transitions in few-electron atoms by photons, electrons, ions, and antiparticles [1–4].

While the total cross sections of double-electron processes give insight into global trends, multiple differential cross sections provide detailed information about collision processes and electron correlation dynamics. Unfortunately, the multiple differential cross sections of the double-electron transition processes, which involve ionization of one or two electrons, though sensitive to electron correlation in targets, are hugely dependent on postcollisional interactions between the charged particles in the final state [5,6]. However, in transfer-ionization processes by proton impact the projectile captures one electron to become atomic hydrogen and one could therefore expect that postcollisional Coulomb interactions would be neutralized and a high sensitivity to target effects would become apparent.

The fully differential cross section for the transfer-ionization process

\[ \text{H}^+ + \text{He} \rightarrow \text{H}^0 + \text{He}^{2+} + e^- \]

has been measured using the COLTRIMS technique [7–9]. These experiments reveal that the ejected electron is predominantly emitted into the backward direction. Until recently, theory was unable to reproduce the general features of these experiments. Godunov et al. [10] produced a simple theoretical model which explained the observed qualitative features in terms of target correlation and gave quantitative predictions for triple-differential cross sections which explicitly demonstrated the sensitivity to terms beyond the (ns\(^2\)) in a multiconfiguration Hartree-Fock description of the target, confirming a suggestion first made by Schmidt-Böcking [11].

The quantitative predictions given in [10] have been confirmed by COLTRIMS experiments [12,13]. The model [10] is a first-order model and given the kinematics of the experiment it is reasonable to ask if the agreement with the experiment is purely fortuitous and the explicit target dependence would be swamped in a higher-order model. In this paper we address the question by extending the previous model to the second order in the projectile target interaction. We show that while the inclusion of the second order does indeed affect the magnitude of the triple-differential cross section, its shape remains sensitive to the target description for the kinematics and dynamics explored in the previous papers [10,12,13]. We present results in both the first- and second-order calculations.

II. THEORETICAL MODEL FOR THE TRANSFER IONIZATION

A. First Born amplitude

We consider a structureless projectile incident upon a two-electron target in the ground state \( \Phi_i(\mathbf{r}_1, \mathbf{r}_2) \). The first Born amplitude for the transfer ionization can be written as [10]

\[
\psi_{\text{BI}} = -(2\pi)^3 \sqrt{\mu_i \mu_f} \langle \Psi_f | V | \Psi_i \rangle,
\]

where the initial channel wave function \( \Psi_i \) is

\[
\Psi_i = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{K}_i \cdot \mathbf{R}_i) \Phi_i(\mathbf{r}_1, \mathbf{r}_2),
\]

and the wave function \( \Psi_f \) in the final channel is

\[
\Psi_f = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{K}_f \cdot \mathbf{R}_f)[\varphi_{\text{he}}(\mathbf{r}_1 - \mathbf{R}) \varphi_{\text{he}}(\mathbf{r}_2)],
\]

where \( \mathbf{r}_1 \) and \( \mathbf{r}_2 \) are the position vectors for the electrons relative to the target nucleus, the vectors \( \mathbf{R}_i \) and \( \mathbf{R}_f \) describe the position of the center of mass of the projectile system relative to that of the target system before and after the collision, while \( \mathbf{K}_i \) and \( \mathbf{K}_f \) are the associated relative momenta, and \( \mu_i \) and \( \mu_f \) are the corresponding reduced masses defined as

\[
\mu_i = \frac{M_p (M_i + 2)}{M_p + M_i + 2}, \quad \mu_f = \frac{(M_p + 1) (M_i + 1)}{M_p + M_i + 2},
\]

where \( M_p \) is the mass of the projectile and \( M_i \) is the mass of the target nucleus. In Eq. (3) \( \varphi_{\text{he}}(\mathbf{r}_1 - \mathbf{R}) \) is the hydrogen-like wave function of the captured electron, and \( \varphi_{\text{he}}(\mathbf{r}_2) \) is the Coulomb wave function for the ionized electron with momentum \( \mathbf{k}_2 \) in the field of the target nucleus [normalized as \( \langle \varphi_{\text{he}}(\mathbf{r}_2) \varphi_{\text{he}}(\mathbf{r}_1) \rangle = \delta(\mathbf{k} - \mathbf{k}') \)]. The interaction potential \( V_f \) between the
projectile of charge $Z_p$ and a two-electron atomic system
with nuclear charge $Z_t$ is given by

$$V_t = -\frac{Z_p}{|\mathbf{r}_1 - \mathbf{R}|} - \frac{Z_t}{|\mathbf{r}_2 - \mathbf{R}|} + \frac{Z_p Z_t}{R},$$  

(5)

where $\mathbf{R}$ is the coordinate of the projectile respective to the

target nucleus.

In our actual calculations we include symmetrization over space
coordinates, however, it is conceptually useful to present the theory as if the particles were distinguishable. We

use index 1 for the “transferred” electron and index 2 for the

“ionized” electron. The transition amplitude (1) contains three
terms arising from the three terms in the potential $V_t$. The

first term may be considered as one electron of the target

being captured by the interaction with the projectile and the

second is ejected because of correlation (“transfer first”
transition). The second term describes the picture when one
electron is initially ionized and then the second is captured due to

correlation (“ionization first” transition). The last term in Eq. (5)
takes into account the heavy particle interaction in ion-atom
collision.

In order to evaluate the first Born amplitude (1) it is inhibitive to introduce the Fourier transform of the hydro-

genic wave function $\varphi_{nl}(r)$,

$$\varphi_{nl}^p(s) = \int \exp[+i\mathbf{s} \cdot (\mathbf{r}_1 - \mathbf{R})] \varphi_{nl}(\mathbf{r}_1 - \mathbf{R}) \, d\mathbf{r}_1 \cdot d\mathbf{R},$$  

(6)

$$\varphi_{nl}(\mathbf{r}_1 - \mathbf{R}) = \frac{1}{(2\pi)^3} \int \exp[-i\mathbf{s} \cdot (\mathbf{r}_1 - \mathbf{R})] \varphi_{nl}^p(s) \, ds.$$  

(7)

Then the integration over the internuclear coordinate $\mathbf{R}$ in the

first Born amplitude can be easily performed to yield the following form for the transfer first amplitude

$$f_n = -\sqrt{\frac{\mu \mu_j}{(2\pi)^3}} \int ds \frac{4\pi Z_p}{|\mathbf{s} - \mathbf{K}|} \varphi_{nl}^p(s) \mathcal{T}(\mathbf{k}_2; \mathbf{Q} - \mathbf{Q}_t),$$  

(8)

and for the ionization first amplitude

$$f_{ion} = -\sqrt{\frac{\mu \mu_j}{(2\pi)^3}} \int ds \frac{4\pi Z_p}{|\mathbf{s} - \mathbf{K}|} \varphi_{nl}^p(s)$$

$$\times \mathcal{T}(\mathbf{k}_1; \mathbf{Q} - \mathbf{K} + \mathbf{K}, \mathbf{Q} - \mathbf{Q}_t),$$  

(9)

where the momenta transfer $\mathbf{Q}$ and $\mathbf{K}$ are defined by

$$\mathbf{Q} = \frac{M_f + 1}{M_f + 2} \mathbf{K}_1 - \mathbf{K}_f, \quad \mathbf{K} = \mathbf{K}_1 - \frac{M_p}{M_p + 1} \mathbf{K}_f,$$

(10)

$$\mathbf{Q}_t = \mathbf{Q}/(M_f + 1).$$  

$T$ denotes the two-particle transition matrix element

$$T(\mathbf{k}_2; \mathbf{s}_1, \mathbf{s}_2) = \int d\mathbf{r}_1 d\mathbf{r}_2 \varphi_{k_2}^e(\mathbf{r}_2) \Phi_1(\mathbf{r}_1, \mathbf{r}_2) \exp[i\mathbf{s}_1 \cdot \mathbf{r}_1 + i\mathbf{s}_2 \cdot \mathbf{r}_2].$$  

(11)

The amplitude corresponding to the interaction between heavy particles [the last term in the interaction potential (5)]
may be written using the Fourier transform as

$$f_{pn} = -\sqrt{\frac{\mu \mu_j}{(2\pi)^3}} \int ds \frac{4\pi Z_p Z_t}{|\mathbf{s} - \mathbf{K}|} \varphi_{nl}^p(s) T(\mathbf{k}_2; \mathbf{s} + \mathbf{Q} - \mathbf{Q}_t).$$  

(12)

One may see from Eq. (8) that the transfer and ionization transitions in the transfer first amplitude are separable and can be written as

$$f_n = -\sqrt{\frac{\mu \mu_j}{2\pi}} V_{nl}(\mathbf{K}_2; \mathbf{Q}_t - \mathbf{Q}_f),$$  

(13)

where $V_{nl}$ is proportional to a matrix element for the capture of a free electron into the bound state of the hydrogenlike
target atom

$$V_{nl}(\mathbf{K}) = -\frac{4\pi Z_p}{(2\pi)^3} \int ds \frac{\varphi_{nl}^p(s)}{|\mathbf{K} - \mathbf{s}|^2}$$

$$= \int d\mathbf{r} \varphi_{nl}(\mathbf{r}) \left( -\frac{Z_p}{r} \right) \exp(i\mathbf{Kr}).$$  

(14)

This fact essentially simplifies the calculations of the amplitude $f_n$ which can be performed analytically by means of standard methods. The evaluation of the amplitudes $f_{ion}$ and $f_{pn}$ requires numerical three-dimension integration over $s$.

### B. Second Born amplitude

The amplitude for the transfer-ionization in the second Born approximation may be written in the form similar to a single capture transition, i.e.,

$$f^{\beta 2} = -\sqrt{\mu \mu_j/\Psi_j} V_j \hat{G}^{\beta 2}(E) \frac{v_j}{\Psi_j},$$  

(15)

where $V_i$ is the interaction potential in the incoming channel (5), $V_f$ is the interaction potential in the outgoing channel,

$$V_f = -\frac{Z_j}{r_2} + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} - \frac{Z_j}{|\mathbf{r}_2 - \mathbf{R}|} + \frac{Z_j Z_t}{R},$$  

(16)

and $\hat{G}^{\beta 2}$ is a Green operator. The spectral form of the Green operator can be written as

$$\hat{G}^{\beta 2}(E) = \lim_{\varepsilon \rightarrow 0_+} \sum_{n} \int d\mathbf{k} \frac{|\Phi_n(E)|}{E_k - E - E_n - K_j^2/2\mu_j + i\varepsilon}$$

$$\times \exp[i\mathbf{k}(\mathbf{R} - \mathbf{R}')]$$  

(17)

Here the sum on $n$ is over all discrete and continuum states $\Phi_n$ of the target, and $E_n$ is the energy of the state $\Phi_n$.

Exact calculation of the second-order terms is a very challeng- ing computational problem. One of the simplest but ef-

ficient methods for performing the sum over $n$, and the one

adopted here, is to use the closure approximation [14,15]. In

this approximation $E_n$ is set to an average value $E_{av}$ so that

the denominator becomes independent on $n$. Using the comple-

teness of the states.
and integrating over \( R' \), one would easily get the second Born amplitude of transfer-ionization in the closure approximation

\[
\sum_{n} |\Phi_{m}(\Phi_{n}| = 1, \tag{18}
\]

Numerical integrations in the amplitude (19) is a cumbersome computational task, particularly for \( 1/|r_{1}-r_{2}| \) electron-electron interaction term in \( V_{f} \) potential. On the other hand, the first two terms in the potential \( V_{f} \) (16) describe the interaction of the transferred electron with the residual ion of the target. We may consider that as an interaction with a particle with a net charge \( (Z_{t}-1) \). In this way the potential \( V_{f} \) can be approximated as

\[
V_{f} = -\frac{(Z_{t}-1)}{r_{1}} - \frac{Z_{p}}{|r_{2} - R|} + \frac{Z_{t}Z_{t}}{R}. \tag{21}
\]

The potential corresponding to the first term in the potential above can be written as

\[
f^{B2}(E_{av}) = -\frac{\sqrt{\mu_{i}\mu_{f}}Z_{t}^{2}}{(2\pi)^{3}} \lim_{E \to 0} \int \frac{dK_{a}}{E - E_{av} - K_{a}^{2}/2\mu_{i} + i\epsilon p_{a}^{2}} \frac{4\pi}{p_{a}^{2}} \frac{d\phi_{m}^{a}}{d\epsilon_{m}}(q_{a})
\times [\tau^{1}(k_{1}; Q - q_{a}) + T^{1}(k_{2}; Q - p_{a} - q_{a} - Q_{s})]
\times [\tau^{1}(k_{s}; Q - q_{a}) + T^{0}(k_{s}; Q - p_{a} - Q_{s})], \tag{22}
\]

where

\[
\tau(k_{2}; s_{1}, s_{2}) = \int d\mathbf{r}_{1}d\mathbf{r}_{2} \phi_{m}^{1}(r_{2}) \frac{1}{r_{1}} \phi_{m}^{1}(r_{1}, r_{2}) \exp(is_{1}r_{1}) + is_{2}r_{2}). \tag{23}
\]

The contribution of the last two terms of the final interaction potential \( V_{f} \) after introduction of the Fourier transform (6) and integration in Eq. (19) over coordinate \( R \) may be represented in the following form:

\[
f^{B2}(E_{av}) = -\frac{\sqrt{\mu_{i}\mu_{f}}Z_{t}^{2}}{(2\pi)^{3}} \lim_{E \to 0} \int \frac{dK_{a}}{E - E_{av} - K_{a}^{2}/2\mu_{i} + i\epsilon p_{a}^{2}} \frac{4\pi}{p_{a}^{2}} \frac{d\phi_{m}^{a}}{d\epsilon_{m}}(q_{a})
\times [\tau^{1}(k_{1}; Q - q_{a} + s.q_{a} - Q_{s})
+ T(k_{1}; Q - K + s.K - Q_{s})
- Z_{t}T(k_{2}; Q - K + s.q_{a} - Q_{s} - s)], \tag{24}
\]

C. Cross sections

The triple-differential cross section for the transfer ionization in the perturbative model through the second order is given by

\[
\frac{d^{3}\sigma}{dE_{\text{res}}d\Omega_{\epsilon}d\epsilon_{f}} = \frac{K_{i}}{K_{s}} [f^{B1} + f^{B2}]^{2}, \tag{26}
\]

where \( k_{s} \) and \( \Omega_{\epsilon} \) are the momentum and the solid angle for the ionized electron, and \( \Omega_{\epsilon} \) is the scattered angle. The cross section is the coherent sum of transition amplitudes and thus depends on their interference. The factor of 2 in the equation above is a result of spatial antisymmetrization over electron coordinates in the initial and the final state wave functions.

III. CALCULATION METHOD

We calculated the transfer- ionization cross sections in various approximations to study the effects of first- and second-order terms, and the electron correlation. The first Born calculations (Born 1) include a coherent sum of three first-order amplitudes, namely, \( f^{B1} = f_{f} + f_{ion} + f_{pn} \). For calculating the second-order terms we use the Sokhotsky theorem [16]

\[
\lim_{\epsilon \to 0} \int \frac{f(x) dx}{x - x_{\pm} \pm \epsilon} = \pi \int \frac{f(x)}{x - x_{0}} dx \pm i\pi f(x_{0}), \tag{27}
\]

where \( P \) stands for the Cauchy principal value integral. Thus, a second-order term can be written as \( f_{f}^{B2} = f_{f}^{B1} \text{off} + f_{f}^{B2} \text{on} \). In atomic and nuclear scattering theory terms corresponding to the Cauchy principal value integral are usually called “off-shell” terms. The second term in Eq. (27) is usually called as an “on-shell” term. The calculations of the off-shell terms take the most computational time. Comparing to the \( f_{f}^{B2} \) (22) term, calculations for \( f_{f}^{B2} \) (24) and \( f_{f}^{B2} \) (25) terms require additional three-dimensional integration, thus increasing considerably the computational time. However, it is the amplitude \( f_{f}^{B2} \) that corresponds more closely to the concept of the \( V_{f}G_{0}V_{f} \) expansion. In the following, we denote the second-order calculations in the on-shell approximation as “Born 2 on”, that is, \( f = f^{B1} + f_{f}^{B2} \text{on} \). Born 2 stands for calculations with both on-shell and off-shell terms, i.e., \( f = f^{B1} + f_{f}^{B2} \text{on} + f_{f}^{B2} \text{off} = f^{B1} + f_{f}^{B2} \).

The average energy \( E_{av} \) in the closure approximation for the second-order terms can be chosen arbitrarily. In practice, this parameter should correspond to the energy of the most
important intermediate states. Since our results were quite insensitive to variations of \( E_{av} \) within a 20–60 eV interval, the calculations presented below were carried out with \( E_{av} = 32.5 \) eV.

The wave function \( \Phi_{f} (\vec{r}_1, \vec{r}_2) \) for the helium, ground state was calculated in the multiconfigurational Hartree-Fock approximation (MCHF) [17]. The full set of calculations with both radial and angular correlation include \((ns)^2\), \((ps)^2\), and \((nd)^2\) terms with \( n \) through 4. These configurations yield about 97% of the correlation energy. The same correlated wave function was used in both the first- and second-order calculations.

The calculation model has been recently tested [18] on total cross sections of the transfer ionization in \( \text{H}^+\), \( \text{He}^+\), and \( \text{Li}^+\) collisions. The theoretical cross section has shown good agreement with available experimental data for all three projectiles.

**IV. RESULTS AND DISCUSSION**

Using the theoretical model above, we calculated triple-differential cross sections of transfer ionization for \( \text{H}^+\) + He collisions as a function of the electron emission angle for the scattering angle of \( \theta_s = 0.10 \) mrad, the impact energy of 630 keV, and the electron emission energies of 5 and 20 eV. These conditions match the recent COLTRIMS measurements [19]. The theoretical results were averaged over the scattering angle (\( \pm 0.05 \) mrad) and electron energies (\( \pm 2.5 \) eV and \( \pm 5.0 \) eV) to compare with the COLTRIMS data.

In Fig. 1, we present multiple-differential cross sections of transfer ionization for \( \text{H}^+\) + He collision calculated in various approximations for the first- and second-order transition amplitudes. The first-order calculations with only \( f_{\text{tr}} + f_{\text{ion}} \) amplitudes correspond to our old model [10]. That model gives two peaks in the angular dependence of the differential cross sections. The first peak corresponds to the ionization-first mechanisms, and the second peak matches to the angular correlation effect in the transfer-first mechanism. For a quantitative comparison with other calculations the results were multiplied by a factor of 0.3. Compared to the \( f_{\text{tr}} + f_{\text{ion}} \) model, the full first-order calculations \( f_{\text{tr}} + f_{\text{ion}} + f_{\text{pm}} \) [the amplitude \( f_{\text{pm}} \) (12) describes the interaction between heavy particles] demonstrate a decrease in the magnitude of the cross section but preserve the same angular behavior with the two peaks. Calculations with both first- and second-order amplitudes demonstrate the same two-peak structure in the differential cross sections like we observe in the first-order calculations. The positions of both peaks are practically the same. However, in calculations with the second-order terms, the peak in the backward direction has a few times larger amplitude compared to the first peak. Thus the second-order terms magnify the effect of angular correlation in the target.

We calculated the second-order amplitudes with and without the off-shell terms. From the results of calculations it follows that the off-shell terms decrease the magnitude of differential cross sections but do not change the shape. Calculations in the first + second order without angular correlation in the target wave functions do not reproduce the large peak in the backward direction, confirming one more time the strong effect of angular electron correlation in differential cross sections of transfer ionization.
EFFECT OF ANGULAR ELECTRON CORRELATION IN He

Theoretical results compared to recent COLTRIMS experimental data are normalized in the maximum to analyze the shape of the non-$s^2$ peak. Our calculations with both first- and second-order amplitudes reproduce the angular behavior of the differential cross sections observed in the experiment. Moreover, the inclusion of the second-order amplitudes reproduces the relative intensity of the two peaks, where the second (angular correlation) peak in the backward direction is about four to five times stronger compared to the first (ionization) peak in the forward direction. The first-order calculations with correlated functions give nearly the same intensity for the peaks.

The shape of the differential cross sections calculated through the second order with and without the off-shell terms is very similar. However, as we saw in Fig. 1 before, the off-shell term reduces the magnitude of the differential cross sections.

V. CONCLUSIONS

In summary, we have evaluated the fully differential cross sections of transfer ionization in a theoretical model that included both the first- and second-order terms in the transition amplitude with highly correlated wave functions of the target. The inclusion of the second-order terms in the transition amplitude preserved the angular correlation peak in the backward direction, thus supporting conclusions made in previous papers [10,12,13] regarding the nature of the peak. The theoretical results are in sensible agreement with the latest COLTRIMS experimental data.

REFERENCES