Electron impact ionization-excitation and double-ionization dynamics of He at large momentum transfer

N. Watanabe and M. Takahashi
Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan
noboru@tagen.tohoku.ac.jp

Abstract. We report a collision dynamics study on electron-impact ionization-excitation and double-ionization of He at large momentum transfer. The symmetric noncoplanar (e,2e) and (e,3-1e) cross sections have been measured for transitions to the He⁺ n=2 excited state and He²⁺ doubly ionized state at impact energies of 1.2, 2.1, and 4.3 keV. An investigation of impact energy dependence of the cross sections provides definite evidence that noticeable higher-order effects are involved even at the rather high impact energies used. It has been found that second Born approximation calculations satisfactorily account for the experimental results, indicating that the second-order two-step mechanisms play crucial roles in the ionization-excitation and double-ionization processes under the kinematical conditions considered here.

1. Introduction
Binary (e,2e) spectroscopy [1-4] is an electron-impact ionization experiment performed under the high energy Bethe ridge conditions, where large momentum is transferred from the fast projectile to the target, and an electron with large kinetic energy is emitted into the direction of the transferred momentum. Under these conditions, it is generally believed that electron-impact ionization is dominated by a clean binary knock-out collision between the projectile and a target electron, as in X-ray Compton scattering, and that the process can be well described by the plane wave impulse approximation (PWIA) [3]. Helium is one of the most thoroughly explored targets in binary (e,2e) studies, and the above picture is well confirmed for the primary ionization process, which leaves the residual He⁺ ion in the n=1 ground state; it has been reported that the PWIA provides a good description of the n=1 transition at impact energy above 200 eV [3, 5-9]. By contrast, such studies on two-electron processes of He, i.e. simultaneous ionization-excitation and double-ionization, are scarce [7-9] although the cross sections are expected to provide direct information on electron correlation in the initial target state [7, 10]. The scarcity of such studies arises from extremely small cross sections of the two-electron processes at large momentum transfer and leads to much less being known about the collision dynamics.

In this paper, we report a collision dynamics study on ionization-excitation and double-ionization of He in the high-energy Bethe ridge kinematics. (e,2e) and (e,3-1e) experiments on He have been performed using a symmetric noncoplanar geometry, where two fast outgoing electrons produced by electron impact single- or double-ionization are detected [11, 12]. The cross sections were measured over a wide impact energy range from 1.2 to 4.3 keV using a multichannel (e,2e) spectrometer [13] with a high collection efficiency. We have also developed a method to evaluate symmetric
noncoplanar \((e,2e)\) and \((e,3-1e)\) cross sections based on the second Born approximation (SBA), and have applied it to the ionization-excitation and double-ionization processes of He [12, 14]. The SBA calculations are compared with the experimental results to elucidate the roles of the second-order two-step (TS) mechanisms [15], which are neglected within the first-order PWIA.

2. Experiment
For electron-impact single-ionization and double-ionization of He, conservations of linear momentum and energy require:

\[
\begin{align*}
\mathbf{p}_{\text{he}^+} &= \mathbf{p}_0 - \mathbf{p}_1 - \mathbf{p}_2, \\
E_{\text{bind}} &= E_0 - E_1 - E_2,
\end{align*}
\]

and

\[
\begin{align*}
\mathbf{p}_{\text{he}^2+} + \mathbf{p}_3 &= \mathbf{p}_0 - \mathbf{p}_1 - \mathbf{p}_2, \\
E_3 &= E_0 - E_1 - E_2 - \text{IP}^{2+},
\end{align*}
\]

where \(\mathbf{p}_i\)'s and \(E_i\)'s (\(j = 0,1,2,3\)) are momenta and kinetic energies of the incident and outgoing electrons, respectively. \(\mathbf{p}_{\text{he}^+}\) and \(\mathbf{p}_{\text{he}^2+}\) indicate the recoil momentum of the residual ion \(\text{He}^+\) and that of \(\text{He}^{2+}\), respectively, whilst \(E_{\text{bind}}\) is the ionization energy and \(\text{IP}^{2+}\) is the double-ionization threshold of He (79 eV). Since \((e,2e)\) experiments involve coincidence detection of two outgoing electrons, \(\mathbf{p}_{\text{he}^+}\) and \(E_{\text{bind}}\) are fully determined for single-ionization. On the other hand, for the \((e,3-1e)\) process the obtainable quantities are \(\langle \mathbf{p}_{\text{he}^2+} + \mathbf{p}_1 \rangle\) and \(E_3\). For simplicity both \(\mathbf{p}_{\text{he}^+}\) and \(\langle \mathbf{p}_{\text{he}^2+} + \mathbf{p}_1 \rangle\) are denoted as momentum \(\mathbf{q}\) here, and in the same sense \(\mathbf{p}_{\text{he}^+}\)-dependent \((e,2e)\) cross sections and \(\langle \mathbf{p}_{\text{he}^2+} + \mathbf{p}_1 \rangle\)-dependent \((e,3-1e)\) cross sections are referred to as \((e,2e)\) and \((e,3-1e)\) momentum profiles, respectively.

In this study we measured \((e,2e)\) and \((e,3-1e)\) cross sections in the symmetric noncoplanar geometry. In this kinematic scheme, two outgoing electrons having equal energies \((E_1=E_2)\) and making equal polar angles \((\theta_1 = \theta_2 = 45^\circ)\) with respect to the incident electron beam axis are detected in coincidence. The magnitude of momentum \(\mathbf{q}\) is then expressed by

\[
q = \sqrt{(p_0 - \sqrt{2}p_1)^2 + (\sqrt{2}p_1 \sin(\Delta \phi/2))^2},
\]

where \(\Delta \phi (= \phi_2 - \phi_1 - \pi)\) is the out-of-plane azimuthal angle difference between the two outgoing electrons. If the incident electron energy and momentum are fixed, the single-ionization transition with binding energy \(E_{\text{bind}}\) can be simply selected by the choice of the detection energy and \(q\) is purely determined by \(\Delta \phi\). The same is true for \((e,3-1e)\) experiments, if we detect two fast outgoing electrons having equal energies in the symmetric noncoplanar geometry while leaving one slow ejected electron undetected.

The experiment was performed at incident electron energies, 1.2, 2.1, and 4.3 keV using a multi-channel \((e,2e)\) spectrometer. Details of the spectrometer have been described elsewhere [13]. Briefly, electron impact ionization occurs where an incident electron beam collides with a gaseous target. Scattered and ejected electrons passing through a pair of apertures are energy analyzed by a spherical analyzer and are then detected by a pair of position-sensitive detectors. Since the spherical analyzer maintains azimuthal angles of the outgoing electrons, both the energies and momenta can be determined from their arrival positions at the detectors. Thus the joint use of a spherical analyzer and position-sensitive detectors makes it possible to sample the \((e,2e)\) and \((e,3-1e)\) cross sections over a wide range of binding energy and momentum \(q\) simultaneously. This greatly improves the collection efficiency.

3. Theory
Under the kinematical conditions considered here, energies of the incident electron and two-outgoing electrons detected are high \((E_0=1.2-4.3\text{ keV}, E_{1,2} = E_0/2)\), which enables the projectile-target interaction and the ejected-electron-residual-ion interaction to be treated perturbatively. In our scattering model, the interactions are taken into account up to second-order in the basis of the SBA. Details of the SBA calculations were reported elsewhere for ionization-excitation [12] and double-
ionization [14]. Briefly, however, we describe our SBA method for evaluating the symmetric noncoplanar (e,2e) cross section below.

Within the SBA the scattering amplitude \( f \) is given by

\[
f = f_{SU} + f_{TS1} + f_{TS21} + f_{TS22},
\]

(6)

Here the first-order amplitude, \( f_{SU} \), represents the shake up (SU) mechanism in which the sudden change of potential due to the departure of one of the target electrons results in the simultaneous excitation of the other electron. On the other hand, \( f_{TS1}, f_{TS21}, \) and \( f_{TS22} \) are the second-order scattering amplitudes. \( f_{TS1} \) is responsible for the so-called two-step 1 (TS1) mechanism [15], in which one of the two target electrons is ejected due to an interaction with the incident electron and the other electron is transferred to the target.

Two target electrons is ejected due to an interaction with the incident electron and the other electron is transferred to the target.

\[
\text{where } \Phi = \phi \chi
\]

(7)

with \( \chi^{(-)}(r) \) denotes the continuum distorted wave of the ejected electron, and \( \Phi(r_1, r_2) \) represents the initial He state having the energy \( E_0 \). The second-order amplitudes are given by

\[
f_{TS1} = -\frac{4}{K^2} \sqrt{2} \sum_{j} \int dp_a \left\langle \chi^{(-)}(r_1) \phi_j(r_2) \right| V_{ij} \left| \chi^{(+)}(r_1) \phi_a(r_2) \right| e^{iK_1r_1} \Phi(r_1, r_2)
\]

(8)

\[
f_{TS21} = -\frac{2\sqrt{2}}{\pi^2} \sum_{j} \int dp_a \left\langle \chi^{(-)}(r_1) \phi_j(r_2) e^{iK_1r_1} - \left\langle \Phi(r_1, r_2) \right| \sum_{i} \langle \chi^{(+)}(r_1) \phi_a(r_2) \rangle \delta(E_i - E_j) - i \eta \right| K_1^2 K_2^2
\]

(9)

\[
f_{TS22} = -\frac{2\sqrt{2}}{\pi^2} \sum_{j} \int dp_a \left\langle \phi_j e^{iK_1r_1} \phi_a \right| \chi^{(-)}(r_1) \phi_j(r_2) e^{iK_1r_1} - \left\langle \Phi(r_1, r_2) \right| K_1^2 K_2^2
\]

(10)

with \( K_i = p_0 \phi_0 \) and \( K_f = p_a \phi_a \). Here \( \phi_0(r) \) denotes the wave function of the He\(^+\) ion with the energy \( E_0 \), and \( \Phi(r_1, r_2) \) represents the intermediate target state. \( V_{ij} \) is the Coulomb potential between the two target electrons. Because of high energy of the ejected electron under the present experimental conditions, the corresponding continuum distorted waves in equations (7)-(10), \( \chi^{(-)}(r) \) and \( \chi^{(+)}(r) \), can be approximated by plane waves. It is worthwhile to note that each of the matrix elements in the second-order amplitudes describe an (e,2e) ionization or single-electron excitation process involved in the TS mechanisms.

When \( \chi^{(-)}(r) \) in equation (7) is replaced with a plane wave and the impulse approximation is used, \( f_{SU} \) is equivalent to the PWIA scattering amplitude. Within the PWIA the symmetric noncoplanar (e,2e) cross section is given by

\[
\frac{d^3 \sigma_{PWIA}}{d \Omega_1 d \Omega_2 dE_1} = \frac{p_1 p_2}{p_0} \frac{2 \pi \kappa}{\exp(2 \pi \kappa) - 1} \frac{4}{K^2} G_f(q),
\]

(9)

\[
G_f(q) = \left\langle \int \frac{d^3 \Phi}{(2 \pi)^3} \sqrt{2} \int \phi_j(r_1) e^{iK_1 r_1} \Phi(r_1, r_2) dr_1 dr_2 \right\rangle^2,
\]

(10)

where \( \kappa = \frac{1}{(p_1 + p_2)} \). The PWIA cross section is expressed as a product of kinematical and structure factors, i.e. \( d^3 \sigma/d \Omega_1 d \Omega_2 dE_1 = F_f \times G_f(q) \). The structure factor \( G_f(q) \) is independent of \( E_0 \) and is a function of \( q \) only. Furthermore, under the present experimental conditions the kinematical factor \( F_f \) is
practically constant over the range of $q$ and $E_{\text{bind}}$ covered by the measurements, such that within the PWIA the shape and relative intensity of the momentum profiles are solely determined by $G_d(q)$’s.

4. Results and Discussion

4.1. Binding energy spectra

Figure 1 shows the $\Delta\phi$-angle-integrated binding energy spectra of He measured at $E_0 = 2.1$ and 4.3 keV. Vertical bars indicate the ionization energies, showing transitions to the He$^+$ n=1, 2, 3, and 4 states as well as the double ionization threshold. For ease of comparison, the data at $E_{\text{bind}} > 45$ eV are scaled by a factor of 50.

![Figure 1. $\Delta\phi$-angle-integrated binding energy spectra of He at $E_0 = 2.1$ keV (a) and 4.3 keV (b).](image)

Although the instrumental energy resolution does not allow a complete separation of the n=2 and adjacent n=3 transitions, it is possible to extract the individual n=2 band contribution by a deconvolution procedure. Here a Gaussian curve with the width of the instrumental energy resolution, which was estimated from the width of the n=1 transition peak, is attributed to each transition band. The fitting curves and their sum are represented by broken and solid lines, respectively. The fitting procedure was applied to the binding energy spectra obtained at each $\Delta\phi$, with the area under the Gaussian curve representing the transition spectral intensity. ($e,2e$) momentum profiles of the n=1 and 2 transitions were subsequently produced by plotting their spectral intensities as a function of the ion recoil momentum. Similarly, the ($e,3-1e$) momentum profiles at $E_3 = 20$ eV were generated by plotting the intensity summed over the energy range of $94 < E_{\text{bind}} < 104$ eV as a function of $q$ and by multiplying a factor of $27.2 \text{ [eV/Hartree]} / 10 \text{ [eV]}$ [11].

4.2. ($e,2e$) momentum profiles

In figure 2 we show the experimental ($e,2e$) momentum profiles of transitions to the n=1 and 2 ion states at $E_0 = 2.1$ and 4.3 keV, together with the results at $E_0 = 1.2$ keV. Also illustrated in the figure are associated PWIA and SBA calculations obtained using the configuration-interaction wave function generated by Mitroy et al [16], which reproduces 98.6 % of the correlation energy of He. All the theoretical momentum profiles were folded with the experimental momentum resolution. In order to compare the results obtained at different $E_0$’s, a common intensity scale is generated where all the experimental and theoretical n=1 momentum profiles were individually normalized so that their areas in a momentum range of 0.1 < $q$ < 1.7 a.u. are equal to unity. The resultant scaling factors were subsequently applied to the corresponding n=2 momentum profiles, since the relative intensity between individual transitions is maintained in the measurements.

Figure 2 (a) shows that the experimental n=1 momentum profiles exhibit no variation with $E_0$ except for small changes due to the effects of the instrumental momentum resolution, and are
satisfactorily reproduced by the PWIA. It can also be seen that the SBA calculations are almost indistinguishable from the PWIA ones, indicating that the second-order contributions are practically negligible for the n=1 transition at $E_0 \geq 1.2$ keV. These findings are consistent with the results of the earlier binary ($e,2e$) studies on He [3, 5-9] in that the PWIA provides a very good description of the n=1 transition at $E_0 > 200$ eV.

![Figure 2. (Color online). Comparison of experimental and theoretical momentum profiles of He for transitions to the n=1 state (a) and n=2 state (b). All the results are normalized so that the area of the n=1 momentum profile becomes equal to unity. See text for details.](image)

By contrast, for the n=2 transition there are substantial differences between the experimental and PWIA profiles. It is immediately clear from figure 2 (b) that the PWIA calculation underestimates the experimental results at all of the impact energies considered. Furthermore, the intensity of the experimental n=2 momentum profile decreases with the increase in $E_0$. Recall that within the PWIA, the relative intensities of momentum profiles are determined only by structure factors $G_f(q)$’s, and are essentially independent of $E_0$. The observed $E_0$ dependence of the n=2 momentum profile therefore provides strong evidence that noticeable higher-order contributions are involved in the experiment.

The most striking result seen in figure 2 (b) is that the SBA achieves significant improvements over the PWIA in reproducing the experimental momentum profile. The SBA calculation exhibits higher intensity than the PWIA for the n=2 transition, and as a result, the differences between experiment and theory are almost resolved; in particular at $E_0 = 4.3$ keV, the SBA calculation shows excellent agreement with the experiment. These findings clearly reveal that the second-order TS mechanisms play prominent roles in the n=2 transition under the kinematical conditions considered here.

Because of the energy degeneracy of the He$^+$ 2s and 2p states, the n=2 momentum profile represents the sum of their respective contributions. Although binary ($e,2e$) experiments can not distinguish these sublevels, theoretical examinations of the individual transitions allow one to shed further light on the ionization-excitation dynamics. The SBA and PWIA momentum profiles of the 2p and 2s channels are separately plotted in figures 3. It is evident from figure 3 (a) that for the 2p channel there are remarkable differences in both shape and magnitude between the SBA and PWIA results. The PWIA momentum profiles have a maximum at $q \sim 1.0$ a.u. and the intensity falls off with the decrease in $q$, while the SBA profiles have maximum intensity near the momentum origin. Furthermore, the SBA predicts much larger cross sections than those obtained using the PWIA. These findings clearly indicate that contributions from the TS mechanisms dominate over those from the SU mechanism for the 2p transition under the kinematical conditions considered here. By contrast, for the 2s channel, though the SBA predicts slightly larger cross sections than the PWIA, the intensity differences between the calculations are small, and it indicates that the 2s transition is dominantly due to the SU mechanism at $E_0 \geq 1.2$ keV. These results reveal that the deviations from the PWIA
observed in the n=2 momentum profiles are mainly attributed to the 2p transition, and the TS contributions are strongly dependent on the final ion states.

\[
\begin{align*}
\text{SBA} & : 1: 1.2 \text{ keV,} & 2: 2.1 \text{ keV,} & 3: 4.3 \text{ keV} \\
\text{PWIA} & : 4: 1.2 \text{ keV,} & 5: 2.1 \text{ keV,} & 6: 4.3 \text{ keV}
\end{align*}
\]

Figure 3. (Color online). The SBA and PWIA momentum profiles of the transitions to the He\(^+\) 2p (a) and 2s (b) states. The results are normalized so that the area of the corresponding n=1 momentum profile become equal to unity. See text for details.

The final-ion-state dependence of the second-order contributions can be accounted for by considering the simplest scenarios of the TS mechanisms, as was highlighted by Takahashi et al. [17]. Briefly, in the TS mechanisms one of the 1s target electrons is ejected by a binary (e,2e) collision either before or after an excitation of the other 1s electron due to a collision with the incoming or outgoing electron. The exciting collision should be dominated by forward scattering or pseudo-photoimpact because of the much higher projectile energy in the collision process compared with the energy loss. By analogy with photon-impact, contributions of the forward-scattering excitation process are much larger for the ‘optically allowed’ transition from the 1s state to the 2p state than for the ‘optically forbidden’ transition to the 2s state. Hence, the TS contributions are much larger for the 2p transition than those for the 2s transition.

Such final-ion-state dependence of higher-order contributions has actually been observed in ionization-excitation of the isoelectronic system H\(_2\) [13, 17, 18]. In contrast to the He\(^+\) 2p and 2s states, the corresponding sublevels of the H\(_2^+\) ion, 2p\(\sigma_u\) and 2s\(\sigma_g\) states, are energetically separated from each other and can thus be experimentally resolved. It has been reported that for the transition to the 2p\(\sigma_u\) state there are remarkable discrepancies between experimental and PWIA momentum profiles in terms of both shape and magnitude, as is similar to the 2p transition of He. On the other hand, for the 2s\(\sigma_g\) channel, which resembles the 2s channel for He, the PWIA momentum profile shows a moderate agreement in shape with the experimental results, and the intensity differences between the experiment and theory are much smaller than those for the 2p\(\sigma_u\) channel. Recently, we have carried out binary (e,2e) experiments on ionization-excitation of H\(_2\) with higher precision having been achieved [19]. These results were compared with the associated SBA calculations [20] to unequivocally confirm that the TS mechanisms serve as a key to understanding binary (e,2e) ionization-excitation of H\(_2\).

4.3. (e,3-1e) momentum profiles

Figure 4 (a) shows the experimental (e,3-1e) momentum profiles at \(E_3 = 20 \text{ eV}\) and \(E_0 = 2.1 \text{ and 4.3 keV}\) [11, 14]. Also plotted in this figure are the associated PWIA and SBA calculated momentum profiles [14]. The normalization procedure used for the (e,2e) momentum profiles was also applied to the (e,3-1e) results. Thus all the (e,3-1e) momentum profiles share a common intensity scale, being normalized relative to the (e,2e) cross section for the n=1 transition. The experiment exhibits higher intensity than the PWIA by factors of about 2.9 and 1.8 at \(E_0 = 2.1 \text{ and 4.3 keV}\), respectively. This
shows a marked impact energy dependence, where the experimental intensity decreases with the increase in $E_0$, definitely indicating that significant higher-order effects contribute to double ionization.

It is also evident from figure 4 (a) that the SBA gives rise to much higher intensity than the corresponding PWIA, and as a result it significantly reduces the observed differences between experiment and theory. Substantial contributions of the TS mechanisms in the $(e,3-1e)$ cross sections are clearly revealed. It is noted that this is similar to that previously observed for the n=2 transition, however, the TS contributions are much larger and more prominent in the double ionization process.

To get a more detailed insight into the double-ionization dynamics, we have examined the $E_3$ dependence of the $(e,3-1e)$ cross section. The experimental and theoretical $(e,3-1e)$ cross sections at $E_0 = 2.1$ keV are plotted in figure 4 (b) as a function of $E_3$. It is noted that the results at individual $\Delta \phi$'s exhibit similar $E_3$ dependence, and that the $\Delta \phi=6^\circ$ result serves as a representative example, corresponds to $q\sim 0.7$ a.u. It can be seen from figure 4 (b) that all the theoretical and experimental cross sections fall off monotonically with the increase in $E_3$. Nevertheless, one may notice that the SBA and experimental cross sections decrease more slowly than the PWIA result. For instance, the ratio of the cross section at a certain $E_3$ value to that at $E_3=5$ eV, $\sigma(E_3)/\sigma(5)$, is $\sigma(20)/\sigma(5)=0.50\pm 0.08$, 0.40, 0.30 and $\sigma(29)/\sigma(5)=0.33\pm 0.08$, 0.26, 0.16 for experiment, SBA, and PWIA, respectively. This observation indicates that the TS mechanisms play more prominent roles in electron-impact double ionization of He as $E_3$ increases. Specially, the double ionization is mainly due to the first-order shake off (SO) mechanism at $E_3\sim$ 0 eV, while at $E_3>\sim$ 40 eV, it occurs predominantly through the TS mechanisms.

It is worthwhile to note that a similar result was also observed in photo double ionization (PDI) experiments of He, by Knapp et al [21]. Here a photon energy of 529 eV was employed to produce PDI with the angular distributions of fast and slow photoelectrons being measured in extremely asymmetric energy sharing conditions. It was found that the PDI process is governed by the SO mechanism with a slow photoelectron energy of 2 eV, but at 30 eV the contributions of the TS1 mechanisms are significant. Interestingly, more prominent roles of the TS mechanisms at higher slow ejected electron energy are found both in electron- and photon-impact studies, though the kinematics employed are fundamentally different from each other. In particular, the momentum transferred to the target is large in the $(e,3-1e)$ reaction under the symmetric noncoplanar geometry, while it is practically zero in PDI.

5. Summary
We have carried out a study on collision dynamics of ionization-excitation and double-ionization of He under the high-energy Bethe ridge conditions. Experimentally, the symmetric noncoplanar \((e,2e)\) and \((e,3-1e)\) cross sections have been measured at \(E_0 = 1.2, 2.1, \) and \(4.3 \text{ keV}\). The \(n=2\) \((e,2e)\) and double-ionization \((e,3-1e)\) momentum profiles, which are normalized relative to the \(n=1\) transition, have been found to exhibit a marked \(E_0\) dependence, which provides clear evidence that noticeable higher-order effects are involved. It has also been shown that the collision dynamics of the two-electron processes does not yet reach the high-energy limit where the PWIA is valid, even at rather high impact energy of \(4.3 \text{ keV}\). Theoretically, we have implemented SBA calculations that incorporate the TS mechanisms. The SBA shows good agreement with the experiments for both the \(n=2\) transition and double-ionization at each impact energy employed, indicating that a principal source of the observed differences between the experiment and PWIA are the TS contributions. Comparisons between the SBA and experimental results have shown that the TS contributions strongly depend upon the final \(\text{He}^+\) ion state or the ejected electron energy \(E_3\). The present work has clarified that the TS mechanisms play crucial roles in two-electron processes under high-energy Bethe ridge conditions.

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