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Ionization excitation of helium by the $(e,2e)$ reaction

X. G. Ren, C. G. Ning, J. K. Deng,* G. L. Su, S. F. Zhang, Y. R. Huang, and G. Q. Li
Department of Physics and Key Laboratory of Atomic and Molecular NanoSciences of MOE, Tsinghua University,
Beijing 100084, People's Republic of China

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The $(e,2e)$ cross sections for transitions to the $n=1$, 2, and 3 final state of He^+ have been measured at impact energies of 1000 and 1600 eV by using a newly developed energy and momentum dispersive spectrometer. Binding energy spectra in the range of 8 to 86 eV and momentum profiles for the transitions to the ground and excited ion states of He^+ are reported at different impact energies, and the experimental results are compared with plane wave impulse approximation calculations. The impact energy dependence of cross section ratios for the He^+ $n=1$, 2, and 3 final states are obtained. Some discrepancies are observed between experimental data and theoretical calculations.

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I. INTRODUCTION

Electron momentum spectroscopy (EMS), or binary $(e,2e)$ spectroscopy, has played an important role in providing unique and versatile information for understandings of the electronic structure and electron correlation effects [1,2]. It can provide electron momentum profiles of individual molecular orbitals in momentum space and yield valuable quantum chemical information such as orbital symmetry, ordering, and spectroscopic factors. EMS is based on an electron impact ionization $(e,2e)$ process in which kinematics of all the electrons are fully determined by coincident detection of the two outgoing electrons. Within the plane-wave impulse approximation (PWIA) [1], the EMS cross section for a gaseous target is proportional to the spherically averaged square of the momentum space overlap of the initial many-body target and final-ion wave functions.

For the target helium the final-ion wave functions are known exactly, and hence the overlap provides a sensitive measure of the helium ground state wavefunction. The cross section leading to excited ion states are particularly sensitive to ground-state correlations. In the case of simultaneous ionization-excitation processes of helium the excitation of the ns ion states can occur, since the atom $1s$ and ion ns orbitals $(n \geqslant 2)$ are not quite orthogonal, but the shape of the cross section to all ion states is given simply by the square of the $1s$ atom momentum space wave functions and is independent of ion state. However, ground-state correlation allows excitation of higher l ion states and severely affects both the magnitude and shapes of cross sections to excited ion states. The electron correlation in the target ground state can be directly probed by the ionization excitation of helium, because the electron correlation is absent in the one electron final ion state, any ionization-excitation processes do not occur unless correlation is included in the target initial wave function. Moreover, helium is the simplest nontrivial target and accurate many-body wave functions are available for helium, which provides an important and accurate test case for the $(e,2e)$ reaction [3–6], both in regard to its sensitivity for measuring correlation effects and to its accuracy in obtaining orbital electron momentum distributions.

Despite the importance of the ionization-excitation process of helium, however, most of the previous EMS experiments have been limited to the primary ionization process that leaves the residual He^+ ion in the $1s$ ground state. The rarity of studies for transitions to excited ion states [4–6] can be accounted for by the experimental difficulties which include the extremely small cross sections and the limited sensitivity of the spectrometers. The EMS experiments of helium ionization and excitation process were performed by Cook *et al.* [4] and Lermer *et al.* [5] at an impact energy of 1200 eV under noncoplanar symmetric $(e,2e)$ kinematics. And the nonsymmetric $(e,2e)$ experiment was carried out by Lahmam-Bennani *et al.* [6] at scattered and ejected electron energies of 5500 and 75 eV, respectively. Highly accurate theoretical calculations of helium have been made and compared with these experimental data, showing remarkable discrepancies between experiment and theory. Further investigations of the ionization-excitation of helium at different impact energies and at higher statistical precision would be desired to resolve these issues, though the extremely small cross sections of the processes make such studies difficult. Recently, a high-resolution and high-sensitivity energy- and momentum-dispersive EMS spectrometer has been developed at Tsinghua University [7], which allows studies for transitions to excited ion states at the proper time of the measurements. In our EMS experiment we developed a double toroidal analyzer (DTA) and a pair of wedge-strip-anode (WSA) position sensitive detectors (PSDs) with a USB multiparameters data-acquisition system. Moreover, a conical deceleration lens system is equipped to achieve higher energy resolution. The typical energy resolution of 1.2 eV, the θ and ϕ angle resolution $\pm 0.7^\circ$ and $\pm 1.9^\circ$ are achieved from the measurements of argon and helium. Another advantage of this spectrometer is its ability to measure the $(e,2e)$ cross sections over a wide range of experimental impact energies, which is especially important for some accurate and convincing investigations, such as distorted wave effects [8–11], spectroscopic factors, and the validities of

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*Electronic address: djk-dmp@mail.tsinghua.edu.cn

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some approximations in theoretical calculations [12].

In the present work, the EMS spectrometer has been applied to the ionization-excitation measurements of helium. The measurements were carried out at impact energies of 1000 and 1600 eV, and binding energy spectra and momentum profiles have been obtained at these different impact energies. The high sensitive measurements of momentum profiles for helium leading to the ground (He$^+$ $n=1$) and excited (He$^+$ $n=2,3$) states of ion are reported. The results are compared with some theoretical calculations. Furthermore, the experimental $(e,2e)$ cross section ratios of the $n=2$ to $n=1$, $n=3$ to $n=1$ and $n=3$ to $n=2$ for He$^+$ are also reported. Momentum profiles and cross section ratios obtained at two different impact energies were compared with each other in order to discuss some similarity and dissimilarity of EMS measurements with the variation of impact energies.

II. THEORETICAL AND EXPERIMENTAL BACKGROUND OF EMS

EMS is an $(e,2e)$ electron impact ionization experiment. A high-energy incident electron knocks out an electron from target atoms or molecules and the two outgoing electrons are subsequently detected in coincidence. Measuring energies and momenta of the incident (E_0,p_0) and two outgoing electrons (E_1, p_1 and E_2, p_2) allows the target electron binding energy ε and target recoil momentum q to be determined by using the conservation of energy and momentum

$$\varepsilon = E_0 - E_1 - E_2, \quad (1)$$

$$q = p_0 - p_1 - p_2. \quad (2)$$

One of the kinematic geometry which has been mostly used for EMS is noncoplanar symmetric kinematics. In this kinematics two outgoing electrons have equal scattering polar angles ($\theta_1=\theta_2=45°$) and energies ($E_1 \approx E_2$). Under high impact energy and high momentum transfer conditions, PWIA provides a good description of the collision [1,2] and the ionized electron essentially undergoes a clean "knock-out" collision, as prescribed by the binary encounter approximation. In PWIA the momentum p of the ejected electron prior to knock-out is equal in magnitude but opposite in sign to the momentum q of the recoiling ion, i.e., $p=-q$. The magnitude of the electron momentum p is related to ϕ

$$p = \left\{ (2p_1 \cos\theta - p_0)^2 + \left[2p_1 \sin\theta \sin\left(\frac{\phi}{2}\right)\right]^2 \right\}^{1/2}, \quad (3)$$

where p_1 and p_0 are the momenta of the outgoing and incident electrons, respectively, and ϕ is the azimuthal angle difference between the two outgoing electrons. Within the PWIA the EMS cross section for randomly oriented molecules can be given by

$$\sigma_{\mathrm{EMS}} \propto \int d\Omega |\langle p\Psi_f^{N-1}|\Psi_i^N\rangle|^2, \quad (4)$$

where p is the momentum of the target electron prior to electron ejection. $|\Psi_f^{N-1}\rangle$ and $|\Psi_i^N\rangle$ are the total electronic wave functions for the final ion ($N-1$ electron) state and the target molecule initial (N electron) state, respectively. The integral represents the spherical average due to the randomly oriented gas phase target in the collision region [1]. The overlap of the ion and neutral wave functions in Eq. (4) is known as the Dyson orbital while the square of this quantity is referred to as an ion-neutral overlap distribution. Thus, the $(e,2e)$ cross section is essentially proportional to the spherical average of the square of the Dyson orbital in momentum space [1,2].

In our EMS experiment an incident electron beam is produced by the electron gun incorporating a tungsten filament. Electron impact ionization occurs where the incident electron beam collides with targets from the gas jet. The two angle-selected electrons can be decelerated with the associated electron optic lenses in order to achieve higher energy resolution. The electrons passing through the apertures are energy analyzed and dispersed by a double toroidal analyzer, and then detected by a pair of two-dimensional PSDs placed behind the DTA analyzer exit. Since the DTA analyzer can well maintain azimuthal angles for the electrons, both energies and angles can be simultaneously determined from their arrival positions at the detectors.

III. RESULTS AND DISCUSSIONS

A. Binding energy spectra

Binding energy spectra of He measured at impact energies of 1000 and 1600 eV are shown in Fig. 1 in the range of 8 to 86 eV, which were obtained by summing up the coincidence counts over the entire azimuthal ϕ angles. The solid line represents the sum of Gaussian peaks fitted to the individual transitions, and the energy scale was calibrated by setting the $n=1$ ionization potential of 24.6 eV. The Gaussian peak width is about 1.2 eV in FWHM, which corresponds to the instrumental energy resolution. With fixed width and positions the determination of the solid curve of Fig. 1 is a fitting in which only the heights of the respective Gaussians were allowed to vary. The resulting peak areas are used to establish a relative intensity scale between the transitions to the ground and the $n=2$ and 3 excited states of He$^+$. A similar curve fitting deconvolution procedure was employed for a series of binding energy spectra at each angle ϕ to produce momentum profiles for the transitions to the $n=1$, 2, and 3 final ion states. Note that the profiles for these transitions share a common intensity scale.

B. Momentum profiles

Experimental $(e,2e)$ cross sections have been obtained at impact energies of 1000 and 1600 eV for the transitions to the He$^+$ $n=1$, 2, and 3 states. Since an absolute cross section cannot be determined with EMS, the He 1s momentum profiles at individual impact energies were normalized to the corresponding theoretical ones. The scaling factors obtained were applied to the momentum profiles for ionization excitation. Hence the transitions to the He$^+$ $n=2$ and 3 excited states exhibit normalized intensities relative to the He 1s primary ionization cross section.

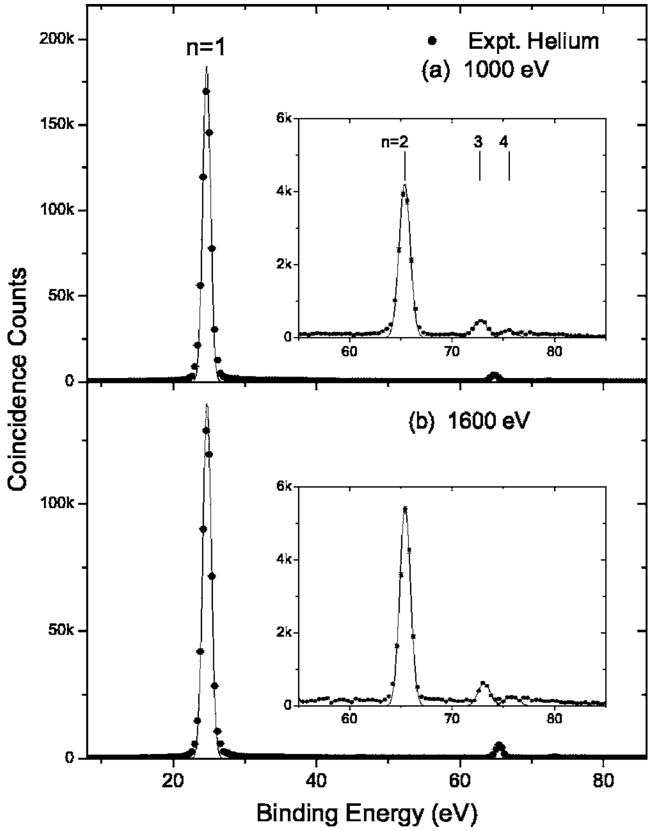

FIG. 1. Binding energy spectra of helium obtained at impact energies of (a) 1000 and (b) 1600 eV. The insets show an expanded view of the energy range of 55 to 85 eV.

Experimental momentum profiles obtained at impact energies of 1000 and 1600 eV of He 1s orbital is shown in Fig. 2, together with associated theoretical profiles. Theoretical momentum profiles were achieved by using the PWIA method. To compare with experiments, all the theoretical profiles were convoluted with the instrumental momentum resolution ($\Delta\theta = \pm 0.7°$, $\Delta\phi = \pm 1.9°$, $\Delta E = 1.2$ eV) according to the method in Ref. [13]. It is very clear from Fig. 2 that agreement between experiment and theory is satisfactory for the transition to the He 1s ground state at each used impact energy. The He 1s momentum profile exhibits no variations with impact energy except for slight changes due to the finite momentum resolution effects. The very slight discrepancies in the higher momentum region between experiment and PWIA theory are due to the distorted wave effects, where the PWIA regime is not valid [1]. Then the momentum profiles were zoomed in on the region above 1.5 a.u. momentum as shown in the insert view of Fig. 2. The dissimilarity of distorted wave effects in the above 2.0 a.u. momentum region can be observed at different impact energies, where the lower impact energy (1000 eV) suffer from a stronger influence of distorted wave effects than the higher impact energy (1600 eV). These observations indicate that the PWIA could provide a very good description of the primary ionization process of helium, and in the higher momentum region the distorted wave effects become smaller with the increase of impact energy.

Figure 3 shows experimental momentum profiles, obtained at impact energies of 1000 and 1600 eV for the transitions to the He$^+$ $n=2$ and 3 excited states. Also included in this figure are the PWIA calculations of Cook *et al.* [4], which were digitized from the literature. In contrast to the ground state transition, the He$^+$ $n=2$ and $n=3$ excited states experimental momentum profiles in Figs. 3(a) and 3(b) have somewhat difference from the PWIA calculations. Considering the momentum profile shape first, the PWIA calculations gives a good fit to the measured $n=2$ and $n=3$ cross section, in particular to the momentum region below 1 a.u. for the $n=2$ case. It is obvious that there are significant intensity difference for the He$^+$ $n=2$ momentum profiles between experiment and PWIA calculation, where the experimental intensity of $n=2$ is significant higher than the theoretical calculation in the region above 1 a.u. momentum. As for the $n=3$ momentum profiles, the theoretical profile at 1600 eV impact energy shows somewhat good fit to the experimental result in the lower momentum region. However, it provides lower intensity than the experiment measurements in the

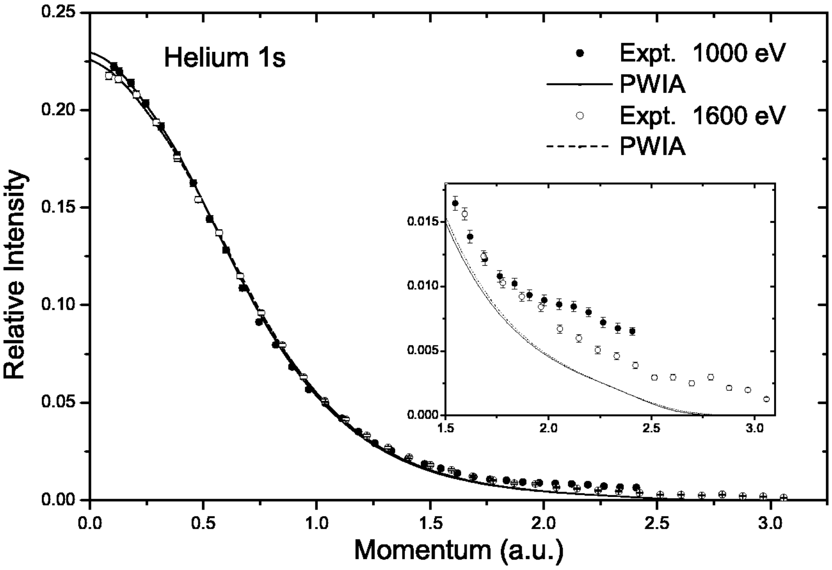

FIG. 2. Experimental momentum profiles of helium for the transition to the 1s ground ion state at impact energies of 1000 and 1600 eV compared with the PWIA calculations. The inset shows an expanded view in the momentum range of 1.5 to 3.1 a.u.

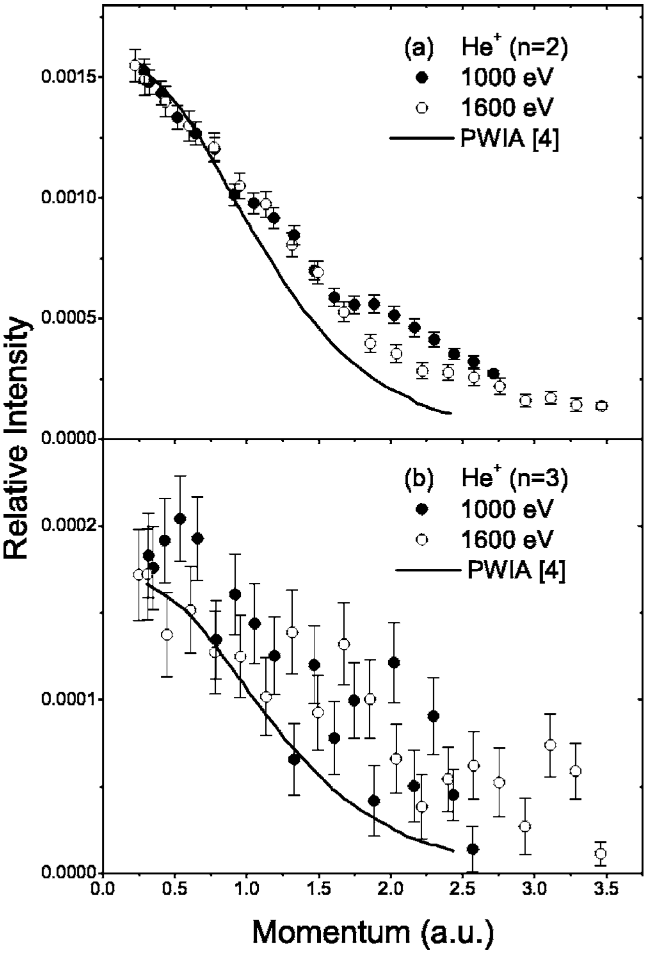

FIG. 3. Experimental momentum profiles of helium for the transition to the (a) $n=2$ and (b) $n=3$ excited ion states at impact energies of 1000 and 1600 eV. The solid lines are the PWIA calculations of Cook [4], which did not include resolution folding.

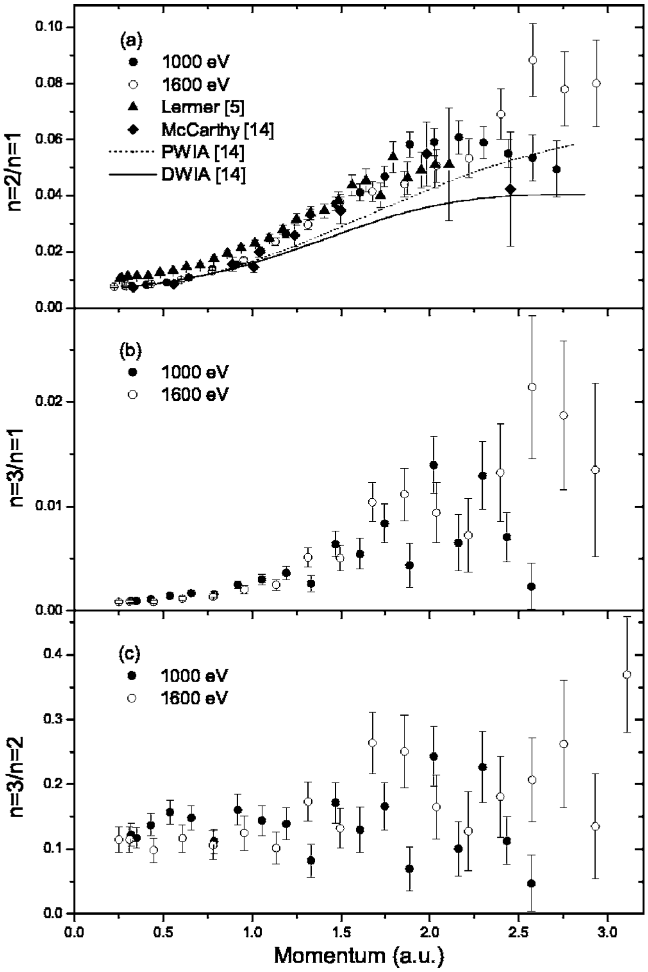

FIG. 4. Cross section ratios of helium (a) $n=2/n=1$, (b) $n=3/n=1$, and (c) $n=3/n=2$. The experimental data of Lermer [5] and McCarthy [14] and the PWIA and DWIA calculations [14] are also included for the $n=2/n=1$ cross section ratio.

higher momentum region. And the 1000 eV result displays higher intensity in general than the PWIA calculation.

C. Cross section ratios

It is interesting to see how the $(e,2e)$ cross sections for the transitions to the He$^+$ $n=1$, 2, and 3 states compares with one another as a function of momentum p. The cross sections of $n=1$, 2, and 3 were measured at the same out of plane azimuthal angles ϕ, and not at the same values of electron momentum p, since the different binding energies give slightly different values for p with the same values of ϕ as described by Eq. (3). Therefore interpolated $n=1$ cross sections were used in driving $n=2$ to $n=1$ and $n=3$ to $n=1$ cross section ratios, and $n=2$ cross sections were interpolated in driving $n=3$ to $n=2$ cross section ratio, as shown in Fig. 4. Figure 4(a) also includes the experimental cross section ratio of $n=2$ to $n=1$ of Lermer *et al.* [5] and the experimental result and theoretical calculations of McCarthy *et al.* [14], which were digitized from their literature. Comparing the present and previous experimental results with PWIA and DWIA calculations indicates that experimental cross section ratios agree reasonably well for shape with the theoretical calculations, and the PWIA and DWIA calculations can provide good description of present experimental results in the momentum region below 1.0 a.u. However, these calculations underestimate the measured $n=2/n=1$ cross section ratio intensity above ~1 a.u. Surprisingly, these calculations show that the cross section ratio $\sigma(n=2)/\sigma(n=1)$ calculated within the DWIA is smaller than the PWIA ratio at the higher momentum region. Since the DWIA using static exchange potentials is adequate to describe $(e,2e)$ on argon and xenon [1] there must be an aspect of the helium case for which the distorted wave description of the initial or final state is less adequate than for larger atoms. A possible source is the fact that coupling between the degenerate $2s$ and $2p$ states of He$^+$ is ignored in these calculations [14], or some other effects that these theoretical treatments have not given full consideration. The present experimental data urges the need of further refinements to the calculation. Moreover, significant intensity dissimilarity between 1000 and 1600 eV impact energies was observed in the higher momentum region of these $n=2/n=1$ cross section ratios. It indicates that the measurements are competent to evaluate the validity of theoretical methods at different impact energies.

The measured $n=3/n=1$ and $n=3/n=2$ cross section ratios at 1000 and 1600 eV impact energies are shown in Figs. 4(b) and 4(c), respectively. In the form of $n=3/n=1$ ratios the lower momentum region are identical in the measurements of 1000 and 1600 eV impact energies. However, there is also some dissimilarity in the higher momentum region between these two impact energies results. The shape of $n=3/n=1$ cross section ratios are similar to that of $n=2/n=1$ ratios. As for the $n=3/n=2$ ratios there are some slight dissimilarity in the lower momentum region between the measurements of 1000 and 1600 eV impact energies. Since the experimental cross section in the higher momentum region (also larger ϕ) for the transition to the $n=3$ state is very small, and we could give the semiquantitative assessment of the $n=3/n=2$ ratio in the higher momentum region.

IV. SUMMARY

The EMS measurements were carried out for the ionization excitation of helium for the transition to the ground $n=1$ state and the $n=2$ and 3 excited states at impact energies of 1000 and 1600 eV using a high-resolution and high-sensitivity spectrometer we developed. The comparison of experimental and theoretical results of He $1s$ state indicates that the PWIA could provide a very good description of the binary $(e,2e)$ reaction for the primary ionization process of helium at different impact energies. The impact energy dependence of the He^+ $n=2$ and 3 excited final ion states momentum profiles were examined for the first time and the disagreements between experiment and theory confirmed the invalidity of PWIA description of ionization-excitation processes. Moreover, the cross section ratios of He^+ $n=2/n=1$, $n=3/n=1$, and $n=3/n=2$ were also reported as a function of momentum p at 1000 and 1600 eV impact energies. Discrepancies at higher momenta between experimental measurements and DWIA calculation are especially arrestive, which indicates that the DWIA using static exchange potentials is less adequate to describe $(e,2e)$ on the transition to the He^+ excited states. A possibility could be the fact that coupling between the degenerate $2s$ and $2p$ states of He^+ is ignored in calculation, or some other effects that these theoretical treatments have not given full consideration. Further refinements to the calculation could be justified by using this kind of experimental data.

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