Electron-Impact Ionization and Excitation of Helium to the $n = 1$–4 Ionic States

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We present high-precision $(e, 2e)$ measurements and calculations for the e-He four-body Coulomb breakup problem. Cross-section ratios for ionization and excitation of the first three excited states of He$^+$ relative to the ground state have been measured for incident energies between 112 and 319 eV. Comparing the data with predictions from a state-of-the-art hybrid distorted-wave + convergent R matrix with pseudostates (close coupling) approach shows that treating the projectile-target interaction at least to second order is crucial to obtain reasonable agreement between theory and experiment. Nevertheless, our benchmark studies reveal significant theoretical problems for the symmetric energy-sharing cases, thus indicating the need for further improvement.

The correlated dynamics of quantum few and many-body systems, mediated through the Coulomb potential, lie at the heart of atomic and molecular processes and chemical reactions in general. Thus, the accuracy with which we can describe this behavior bears direct relation to our ability to predict the outcomes of chemical reactions and carefully control chemical processes. Collisions of charged particles with isolated atoms provide a sensitive means to explore many-body behavior, as the momenta of the reaction participants and products can be determined in a coincidence experiment.

For atomic species, enormous progress has been made in recent years regarding the theoretical and computational treatment of electron-impact induced ionization. For the case of three interacting particles, the electron-hydrogen problem has provided an ideal testing ground for theory over many years. Nonperturbative approaches such as exterior complex scaling [1], time-dependent close coupling [2], convergent close coupling (CCC) [3], or a R matrix with pseudostates (RMPS) [4] are now able to predict the total, single-differential, double-differential, and triple-differential (TDCS) cross sections very accurately for this system, with the accuracy limit essentially set by the available computational resources and the algorithm stability. The CCC method was similarly successful in the description of coplanar He($1s^2$) ionization [5,6], provided the residual ion is left in He$^+$($1s$); i.e., one of the four electrons is effectively a "spectator."

In contrast, the full Coulomb four-body problem contains puzzling aspects, which are not properly described by current computational approaches. This was recently illustrated by work on ionization of helium by relatively fast electrons [7]. While the well-established first-order distorted-wave theory [8] and more elaborate "3DW" versions [9], which account for the correct asymptotic Coulomb boundary condition [10], are able to describe such processes very well for coplanar scattering geometries, where the momentum vectors of the incident projec-

tile and the two outgoing electrons all lie in the same plane, significant discrepancies between experiment and theory were found for "out-of-plane" scattering geometries. This result suggests that higher-order effects, not presently accounted for in many theories, are contributing substantially to the process. The latter experiments were performed as a follow-up on 100 MeV/amu C^{6+} heavy-particle impact ionization of helium [11]. Major discrepancies between the theoretical predictions and the experimental data raised serious questions regarding the ability of theory to describe such processes.

Further deficiencies in the current implementations of four-body theory become evident in the description of single ionization processes, when the electron in the residual ion is promoted to an excited state. Even for the coplanar geometry, significant discrepancies between theory and experiment can occur under such conditions, if the part of the projectile-target interaction responsible for ionization is accounted for only to first order [12]. Such experiments were performed by Dupré *et al.* [13] and later by Avaldi *et al.* [14] and Rouvellou *et al.* [15]. For their conditions of highly asymmetric energy sharing between the two outgoing electrons, hybrid methods based upon a perturbative treatment, up to second order, of the projectile-target interaction and an R matrix (close-coupling) approach for the ejected-electron-residual-ion interaction [16] were quite successful, especially when the latter part of the problem (e-He$^+$ collisions) was systematically driven to convergence [17]. Except for a pioneering study by Dogan and Crowe [18], the TDCS experiments mentioned above were concerned with ionization and excitation to the $n = 2$ residual ionic states. Because of the energy degeneracy of the He$^+$($2s$) and He$^+$($2p$) states, the measured TDCS was the sum of their respective contributions. However, by also detecting the emitted $2p \rightarrow 1s$ photon in a triple-coincidence $(e, 2e\gamma)$ measurement, Sakhelashvili *et al.* [19] were able to measure the $2p$ contribution individually. Comparison of their

results with several calculations once again showed that a higher-order theory (in the projectile-target interaction) is critical to describe the principal features of the observed angular dependence of the TDCS. This fact was further demonstrated by comparison of first- and second-order predictions with cross-normalized *relative* experimental data [19]. In some cases, first-order models were able to reproduce the angular dependence of the TDCS. However, different, *inconsistent* normalization factors had to be applied to the first-order results, while the second-order hybrid plus close-coupling method used in the present work was able to describe both the angular dependence and the relative magnitude of the data with consistent cross-normalization.

Most of the presently available experimental TDCS data for ionization and excitation suffer from at least one, and often several, of the following drawbacks: (1) All the measured data are relative. (2) The signal-to-noise ratio is low; i.e., the statistical error bars alone are substantial. (3) The energy of the slower of the two outgoing electrons was often chosen between 5 and 10 eV. This is a most unfortunate choice, since resonance effects associated with the $n = 3$ and $n = 4$ doubly excited states of He make a straightforward comparison with theoretical predictions almost impossible [20] if the theory treats the e-He$^+$ problem via the generally desirable close-coupling approach.

The experimental work reported here is an attempt to address, and improve upon, most of the above problems, thereby providing highly accurate, ionic-state-specific and resonance-free data without normalization to theory. To achieve these goals, we applied our toroidal spectrometer to this problem. Its combination of high sensitivity and good energy resolution enabled us to measure relative cross sections for ionization and excitation leading up to the $n = 4$ states of He$^+$ (which account for only $\sim 0.1\%$ of the ionization cross section) for a variety of kinematical situations. By presenting the data as a *ratio* of cross sections for excitation of the $n \geq 2$ ionic states relative to the $n = 1$ "transition with a spectator" ionic state, the results can be directly compared to theory.

Figure 1 shows the scheme of our apparatus, the details of which can be found in Ref. [21]. An electron beam is created by illuminating a strained gallium arsenide photocathode by monochromatic laser light. The beam is transported at high energy through a differential pumping stage before entering the collision chamber in which it is focused and decelerated to the experimental collision energy E_0. An atomic-helium target beam, formed by effusion through a 1 mm internal diameter needle, intersects the electron beam orthogonally to define a localized interaction volume. The final-state $(e, 2e)$ electrons are momentum analyzed in one of two toroidal-sector electrostatic energy analyzers under coplanar scattering geometry. Each analyzer incorporates a pair of microchannel-plate electron multipliers followed by a crossed delay-line detector which determine the spatial and temporal electron-arrival coordinates (x_i, y_i, t_i) [22]. From these coordinates, pairs of
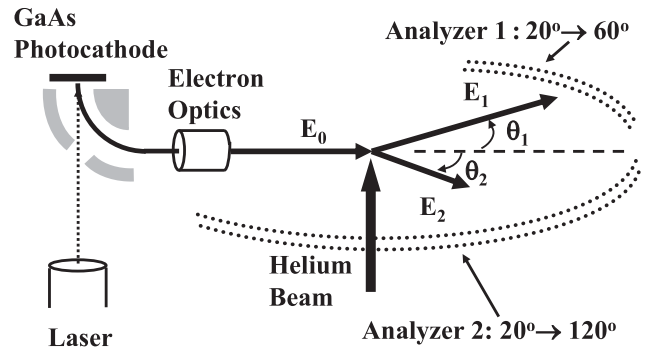

FIG. 1.   Scheme of the apparatus. See text for details.

electrons derived from common $(e, 2e)$ ionization events are identified by their correlated arrival times at the two separate detectors and their initial momenta $(\boldsymbol{p}_1, \boldsymbol{p}_2)$ deduced. One analyzer accepts electrons over the angular range $20° \leq \theta_1 \leq 60°$, while the other accepts electrons over the range $20° \leq \theta_2 \leq 120°$.

The present numerical calculations were performed using a hybrid distorted-wave + R matrix (close-coupling) method. Details of the approach were given by Bartschat and Burke [23]. The general idea is to treat the interaction of a "fast" electron with the target perturbatively, while the initial bound state and the e-He$^+$ half-collision of a "slow" ejected electron and the residual ion is treated via a close-coupling expansion. Major extensions of the method in recent years include the approximate treatment of the projectile-target interaction to second order [24] and the use of a convergent RMPS [17] expansion. Some of the approximations made in the evaluation of the second-order term were qualitatively supported by Chen and Madison [25]. As mentioned above, the method has been very successful for asymmetric energy-sharing conditions [17,19,20]. On the other hand, due to the fundamentally different treatment of the two final-state electrons, one would not expect it to be suitable for symmetric energy-sharing cases. Nevertheless, we decided to apply the DWBA-R matrix approach to these cases as well and thereby assess the size of the problem that will need to be fixed in the future. Consequently, we performed DWB1-RMPS and DWB2-RMPS calculations, in which the projectile-target interaction was treated to first (DWB1) or second (DWB2) order. For the asymmetric energy-sharing cases, the distortion potential for the projectile in both the initial and the final state was chosen as the static potential of the He$(1s^2)$ ground state. In contrast, for the symmetric-energy-sharing cases, the distortion potential in the final state was chosen as the ionic potential of the corresponding He$^+(n\ell)$ states.

Two experiments were performed, one for asymmetric- and one for symmetric-energy sharing between the two final-state electrons. The average scattered or ejected-electron energies E_1 and E_2 were 200 and 44 eV, respectively, for the asymmetric-energy-sharing case and 44 and

44 eV for the symmetric case. The required values for the primary energy E_0 in the case of ionization to $He^+(1s)$ are then 268.6 and 112.6 eV, respectively. To maintain the same average scattered or ejected-electron energies for ionization and excitation, these values need to be increased by 40.8 ($n = 2$), 48.4 ($n = 3$), and 51.0 eV ($n = 4$). For the asymmetric experiment, coincident events were accepted over a 16 eV energy window in each analyzer, i.e., $192 \text{ eV} \leq E_1 \leq 208 \text{ eV}$, $36 \text{ eV} \leq E_2 \leq 52 \text{ eV}$, while a better energy resolution was achieved for the symmetric case with reduced 8 eV energy windows, i.e., $40 \text{ eV} \leq E_1 \leq 48 \text{ eV}$, $40 \text{ eV} \leq E_2 \leq 48 \text{ eV}$. Within each window, electron energies were determined to within 0.65 (asymmetric kinematics) and 0.40 eV (symmetric kinematics). In combination with an energy spread of 0.30 eV for the primary beam, $(e, 2e)$ binding-energy resolutions of 1.0 and 0.65 eV, respectively, were achieved for the asymmetric and symmetric experiments. To reduce the data collection time for the symmetric-energy-sharing experiment, the primary beam energy was maintained at the $n = 3$ value of 161.0 eV when performing the $n = 4$ measurement, resulting in a reduced average energy of 42.7 eV for the two final-state electrons in that case. To ensure correct cross-normalization, the beam focusing optics was adjusted with the incident energy to maintain a constant interaction-volume geometry. Additionally, the beam current at each incident energy was recorded to enable energy-dependent changes in the primary-electron beam current to be corrected for in software. For both experiments, E_0 was scanned to average over long-term instrumental drifts.

Figure 2 shows our data for the asymmetric energy-sharing case, where the faster electron is detected at $\theta_1 = 32°$. The theoretical results are for the average energies listed, and we checked that convolution with the experimental energy windows would change the numbers by less than 10%. Interestingly, the DWB1-RMPS approach, which accounts only for the projectile-target interaction to first order, describes the angular dependence of the TDCS ratios $He^+(1s)/He^+(n = 2)$ quite well, except for a severe cross-normalization problem. Since the results were multiplied by 0.2 in the figure, the DWB1-RMPS ratio is approximately 3–5 times too large compared to experiment. Looking at the individual theoretical TDCS results, this factor can be traced back to problems with the predicted magnitude of the TDCS for simultaneous ionization and excitation to $n = 2$ and $n = 3$, respectively. These cross sections are predicted significantly too small by the first-order method. The DWB2-RMPS results, on the other hand, are much more consistent in predicting the ratio of the absolute sizes correctly, although the agreement between experiment and theory is certainly not perfect.

Similar qualitative statements can be made for the results exhibited in Fig. 3 for the symmetric energy-sharing case, again with one electron detected at $\theta_1 = 32°$. Quantitatively, however, the DBW1-RMPS model is now wrong by about an order of magnitude for many angles, and the angular dependence itself is not predicted correctly
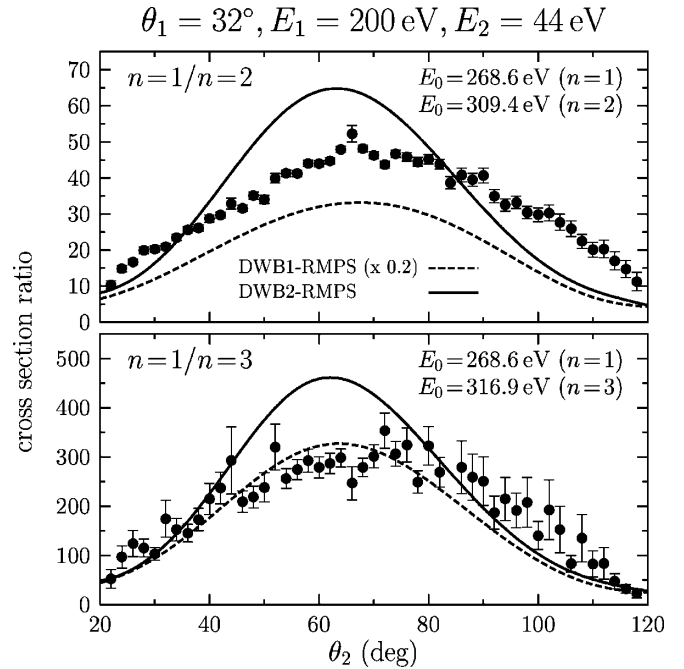

$\theta_1 = 32°, E_1 = 200 \text{ eV}, E_2 = 44 \text{ eV}$

FIG. 2. TDCS for ionization of $He(1s^2)$ leading to $He^+(1s)$ divided by the corresponding TDCS for leaving the He^+ ion in the $n = 2$ and $n = 3$ states. The final-state electrons have average energies of 200 and 44 eV in all cases, while the primary energy E_0 was set to 268.6 eV for $n = 1$, 309.4 eV for $n = 2$, and 316.9 eV for $n = 3$, respectively. The 200 eV electron is detected at $\theta_1 = 32°$, while the detection angle θ_2 of the 44 eV electron is varied (see Fig. 1). The DWB1-RMPS (dashed line) and DWB2-RMPS (solid line) models are described in the text. Note that the DWB1-RMPS results were multiplied by 0.2.

either. The DWB2-RMPS model does much better for $n = 2$ and $n = 3$, although discrepancies of about a factor of 2 exist for many individual angles, and the maximum predicted in the ratios for $\theta_2 \approx 50°$ is almost flat in the experimental data. Even more significant problems exist for the $n = 4$ case, which is very challenging to both experiment and theory due to the small TDCS values.

In conclusion, we have presented highly accurate experimental benchmark data for simultaneous ionization and excitation of $He(1s^2)$, leading to He^+ in the $n = 1$–4 states. Special emphasis was placed on a good signal-to-noise ratio, the elimination of resonance effects, and a consistent, purely experimental cross-normalization. The data were compared to predictions from a state-of-the-art hybrid distorted-wave + convergent R matrix with pseudostates approach. For the asymmetric energy-sharing case, the agreement between experiment and theory is satisfactory, indicating that the theoretical approach contains the principal ingredients to predict the outcome of such collision processes. We emphasize again that accounting for the projectile-target interaction to at least second order is absolutely critical for the correct cross-normalization of the data. Significant theoretical problems, however, remain for the symmetric energy-sharing cases.
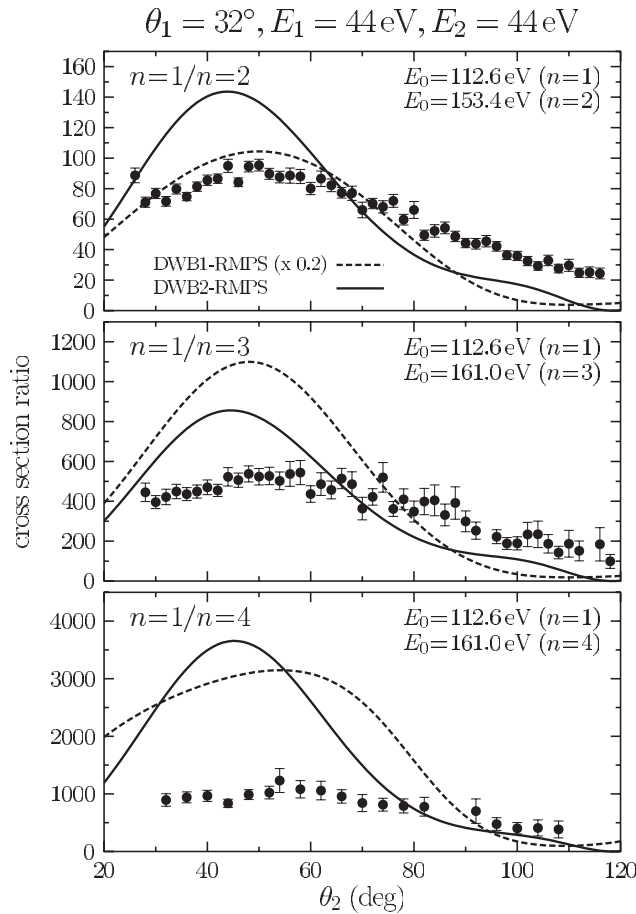
FIG. 3. TDCS for ionization of He($1s^2$) leading to He$^+$($1s$) divided by the corresponding TDCS for leaving the He$^+$ ion in the $n = 2$, $n = 3$, and $n = 4$ states. Both outgoing electrons have an average energy of 44 eV for $n = 1, 2, 3$ (42.7 eV for $n = 4$), while the primary energy E_0 was chosen as 112.6 eV for $n = 1$, 153.4 eV for $n = 2$, and 161.0 eV for $n = 3, 4$, respectively. One of the final-state electrons is detected at $\theta_1 = 32°$, while the detection angle θ_2 of the other electron is varied (see Fig. 1). The theoretical models are as in Fig. 2.

Further improvements of the present model, or any new approach, should include a proper treatment of both exchange effects and the three-body Coulomb boundary condition for the residual ion and the two outgoing electrons. Work in this direction is currently in progress. We hope that the present data will stimulate further experimental and theoretical interest in this highly correlated fundamental four-body Coulomb problem.

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