Nonperturbative B-spline R-matrix-with-pseudostates calculations for electron-impact ionization-excitation of helium to the n=3 states of He^{+}

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Non-perturbative B-spline R-matrix with pseudostates calculations for electron-impact ionization-excitation of helium to the n = 3 states of He^+

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We present fully-differential cross-section ratios for electron-impact ionization of helium without excitation and with simultaneous excitation of the residual ion. The results are obtained from a nonperturbative close-coupling formalism, with the resulting equations being solved by a B-spline R-matrix with pseudostates approach. Very encouraging agreement is obtained with directly measured cross-section ratios for ionization leaving the residual He^+ ion in either the $1s$ ground state or the $n = 3$ $(3s + 3p + 3d)$ excited states.

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I. INTRODUCTION

Electron-impact ionization with simultaneous excitation is a highly challenging problem, both experimentally and theoretically. Taking e − He as the prototype collision system, the challenges are due to the fact that all three electrons (the projectile and both target electrons) undergo significant changes in their respective quantum states. Hence the problem cannot be simplified further by effectively treating one of the target electrons as a "spectator" that remains in the $1s$ orbital.

In recent years, we have further developed a fully nonperturbative B-spline R-matrix (close-coupling) with pseudostates (BSRMPS) method and applied it to both direct ionization [1] and ionization with excitation problems [2, 3]. Despite the success achieved in employing a projection approach to obtain the cross sections from excitation amplitudes of the positive-energy discrete (i.e., finite-range) pseudostates, some skepticism remains about the validity of the approach. The method is indeed not exact [4], but it appears to provide a practical scheme that should systematically improve when the density of the pseudostate spectrum is increased.

To check this hypothesis, we further optimized our parallelized BSR codes. This allowed us to increase the number of pseudostates to 1,254 and thereby also consider ionization with excitation to the $n = 3$ states of He^+. Some details of the method and the specific calculation are provided in Sect. II, followed by the presentation and discussion of the results in Sect. III. Unless indicated otherwise, atomic units (a.u.) are used throughout this manuscript.

II. COMPUTATIONAL DETAILS

The spectroscopic bound states and continuum pseudostates were generated by the B-spline box-based close-coupling method [5]. The structure of the multichannel target expansion was chosen as

$$\Phi(n\ell, n'\ell', LS) = \mathcal{A} \sum_{i,j} a_{ij} \{\phi(n_i\ell_i)P(n_j\ell_j)\}^{L,S} + b\,\Phi(1s^2). \quad (1)$$

The functions $\phi(n\ell)$ are the hydrogen-like orbitals $1s, 2s, 2p, 3s, 3p, 3d$ for a nuclear charge $Z = 2$, while the $P(n_j\ell_j)$ are the "outer" orbitals that are expanded in the B-spline basis. We use a nonrelativistic model, with L and S denoting the total orbital and spin angular momenta, respectively. Furthermore, $\mathcal{A}$ is the antisymmetrization operator. A multi-configuration expansion for the $1s^2$ state was included as a correlation function.

The number of physical states that can be generated by this method depends on the radius a of the B-spline box. We chose $a = 25\,a_0$ (where $a_0 = 0.529 \times 10^{-10}$ m is the Bohr radius) and employed 74 B-splines of order 8 with a semi-exponential grid of knots. This yielded 29 physical and 1,226 target pseudostates that covered the energy region up to 100 eV with S, P, D, and F symmetries of both even and odd parity. The set of pseudostates contained the configurations $1sn'\ell'$, $2\ell n'\ell'$, and $3\ell n'\ell'$, with the latter two sets describing doubly excited autoionizing states as well as the ionization-excitation process. Note that the increase of the principal quantum of the "core" electron from $n = 2$ [2, 3] to $n = 3$ in the present work rapidly increased the number of states in the calculation, thereby limiting the highest pseudostate energy that we could handle.

We then obtained the scattering amplitudes for excitation of all pseudostates using a fully parallelized version of the BSR complex [6] for electron collisions. Contributions from all symmetries for total orbital angular momenta $L_T \leq 25$ were included in the partial-wave expansion. The present model contained up to 3,027 scattering channels, leading to generalized eigenvalue problems with matrix dimension up to 200,000 in the B-spline basis.

Since it is the key step in our method, we now briefly repeat how the physical *ionization* cross sections are obtained from the *excitation* amplitudes for the pseudo-

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states [2, 3]. Recall that we are interested in the ionization amplitude

$$f(L_0M_0S_0M_{S_0}, \boldsymbol{k}_0\mu_0 \to L_fM_fS_fM_{S_f}, \boldsymbol{k}_1\mu_1, \boldsymbol{k}_2\mu_2) \quad (2)$$

for an initial target state with orbital angular momentum L_0 and spin S_0 (with projections M_0 and M_{S_0}, respectively) leading to a final ionic state with corresponding quantum numbers labeled by the subscript f, by an electron with initial linear momentum $\boldsymbol{k}_0$ and spin projection μ_0 resulting in two outgoing electrons described by $\boldsymbol{k}_1, \mu_1$ and $\boldsymbol{k}_2, \mu_2$. We obtain this *ionization* amplitude by projecting the *excitation* amplitudes for the pseudostates (superscript p)

$$\begin{aligned} &f^p(L_0M_0S_0M_{S_0}, \boldsymbol{k}_0\mu_0 \to LMSM_S, \boldsymbol{k}_1\mu_1) \\ &= \sqrt{\frac{\pi}{k_0k_1}} \sum_{l_0,l_1,L_T,S_T,\Pi_T,M_{L_T},M_{S_T}} i^{(l_0-l_1)}\sqrt{(2l_0+1)} \\ &\times(L_0M_0, l_00|L_TM_{L_T})(LM, l_1m_1|L_TM_{L_T}) \\ &\times(S_0M_{S_0}, \frac{1}{2}\mu_0|S_TM_{S_T})(SM_S, \frac{1}{2}\mu_1|S_TM_{S_T}) \\ &\times T_{l_0l_1}^{L_TS_T\Pi_T}(\alpha_0L_0S_0 \to \alpha LS)\, Y_{l_1m_1}(\theta_1, \varphi_1), \end{aligned} \quad (3)$$

to the true continuum functions for electron scattering from the residual ion, $\Psi^{k_2\mu_2(-)}_{L_fM_fS_fM_{S_f}}$, and summing over all energetically accessible pseudostates using the ansatz

$$\begin{aligned} f(L_0M_0S_0M_{S_0}, \boldsymbol{k}_0\mu_0 \;&\to\; L_fM_fS_fM_{S_f}, \boldsymbol{k}_1\mu_1, \boldsymbol{k}_2\mu_2) \\ &= \sum_p \langle \Psi^{\boldsymbol{k}_2\mu_2(-)}_{L_fM_fS_fM_{S_f}} | \Phi^p(nln'l', LS)\rangle f^p(L_0M_0S_0M_{S_0}, \boldsymbol{k}_0\mu_0 \to LMSM_S, \boldsymbol{k}_1\mu_1). \end{aligned} \quad (4)$$

In this multichannel generalization of Eq. (15) proposed by Bray and Fursa [7], $T_{l_0l_1}^{L_TS_T\Pi_T}(\alpha_0L_0S_0 \to \alpha_1L_1S_1)$ is an element of the T-matrix for a given L_T, total spin S_T, and parity Π_T of the collision system. Choosing the z-axis along the direction of the incident beam simplifies the formula to $m_0 = 0$ for the orbital angular momentum projection of the incident electron.

As seen from Eq. (4), the above procedure requires the overlap factors $\langle \Psi^{f,\boldsymbol{k}_2(-)}_{L_fM_fS_fM_{S_f}} | \Phi^p(nln'l', LS)\rangle$ between the true continuum states and the corresponding pseudostates. The continuum states, which describe electron scattering from the residual ion, are once again obtained using the R-matrix method, with the *same* close-coupling expansion that is employed for generating the bound pseudostates. This is a critical issue, since it allows for the preservation of the crucial channel information through the projection. For the present work, we used a six-state expansion coupling the $1s, 2s, 2p, 3s, 3p$, and $3d$ states of He^+.

Finally, the fully differential cross section (FDCS) is given by

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega_1\mathrm{d}E_1\mathrm{d}\Omega_2\mathrm{d}E_2} = \frac{k_1k_2}{k_0}\left|f(L_0M_0S_0M_{S_0}, \boldsymbol{k}_0\mu_0 \to L_fM_fS_fM_{S_f}, \boldsymbol{k}_1\mu_1, \boldsymbol{k}_2\mu_2)\right|^2, \quad (5)$$

where E_i, Ω_i $(i = 1, 2)$ denote the energy and the solid angle element for detection of the two electrons.

III. RESULTS

Before we show results for ionization with excitation of the $n = 3$ ionic states, it seemed appropriate to investigate the stability of earlier predictions against significant

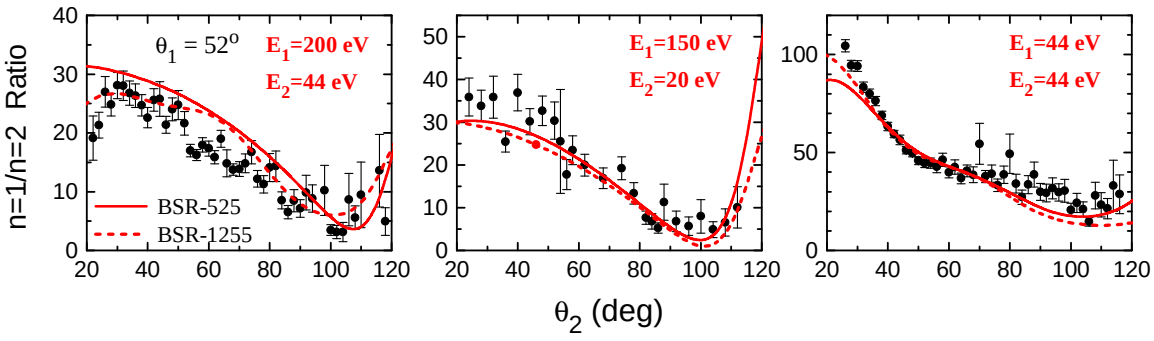

FIG. 1: (Color online) Cross-section ratio for electron impact ionization of helium, leaving the residual ion either in the $1s$ ground state or the $n = 2$ ($2s, 2p$) excited states. The two ejected electrons have fixed energies of 200 eV and 44 eV (left panel), 150 eV and 20 eV (center panel), or 44 eV both(right panel), hence requiring incident projectile energies of 269 eV, 195 eV, or 113 eV to leave He^+ in the ground state, and correspondingly 309 eV, 235 eV, or 153 eV to leave He^+ in $n = 2$. Results from the previous BSR-525 model (including ionic states up to $n = 2$) and the present BSR-1255 model are compared with the experimental data of Bellm *et al.* [8] for a detection angle of 52° for the faster of the two outgoing electrons.

changes of the model, in this particular case to more than doubling the number of states in the close-coupling expansion. Figure 1 shows earlier results [2] for ionization of $He(1s^2)^1S$ with simultaneous excitation of the residual ion to the $n = 2$ manifold. We chose the same three sets of energies for the two outgoing electrons that will be used below for ionization with excitation of $n = 3$. The results are for a detection angle of 52° for the faster of the two outgoing electrons, but the comparison between the previous predictions from a 525-state BSR model [2] and the present one with 1255 states is similar for other detection angles. Hence we conclude that both the numerics and the interpretation of the results are stable.

Figures 2-4 depict the present results for the ionization with excitation to the $n = 3$ manifold of He^+. For all three cases, two with different asymmetric energy sharings and the third for the same energy of both outgoing electrons, the agreement between the BSR-1255 predictions and experiment is not perfect but certainly very encouraging. For comparison, we also show hybrid DW2-RMPS results, which were obtained by describing the projectile-target interaction by a second-order distorted-wave (DW2) approach, while the ejected-electron–residual-ion interaction as well as the initial bound state were treated by a convergent R-matrix (close-coupling) with pseudostates (RMPS) expansion [9, 10]. Such an unequal treatment of the two electrons can be appropriate if they are essentially distinguishable, i.e., for sufficiently high incident projectile energies and highly asymmetric energy sharing between the two free electrons in the final state. In such cases, neglecting exchange and also channel coupling for the fast electron may be a good approximation [11]. We also emphasize that the hybrid model does not account for post-collision interaction, while the BSR model includes the effect within the R-matrix box.

It should be noted that both the experimental and theoretical results for the cross-section ratios can be transformed into absolute cross sections for ionization with excitation by normalizing to the well-known results for ionization without excitation [12], where the agreement between experiment and several theories, most notably convergent close-coupling (CCC) [7, 13], time-dependent close-coupling (TDCC) [14, 15], and BSRMPS has been shown to be excellent on a number of occasions [1, 16].

We finish this paper with a comparison between experiment and theory for the angle-integrated cross section for ionization of $He(1s^2)^1S$ with simultaneous excitation of the residual ion to the $He^+(3p)$ state. The theoretical cross section was obtained for a number of incident projectile energies by numerically integrating the FDCS over a suitable grid of angles and energy sharings for the two outgoing electrons. In contrast to ionization with excitation to the $2p$ state [3], the much simpler alternative procedure of decomposing each pseudostate into the various $n_i\ell_i n_j\ell_j$ components and obtaining the $3p$ results by weighting the contribution from excitation of each pseudostate by its admixture from the $3pn_j\ell_j$ configuration turned out to be less suitable in this case.

The results are shown in Fig. 5. The energy dependence of the cross section can be determined experimentally by observing the radiation emitted in the subsequent optical decay to $He^+(1s)$. As discussed in detail for the corresponding $He^+(2p)$ case [3], the absolute value of the cross section was the subject of a long-standing debate due to the difficulties associated with the required experimental normalization procedure. For the $2p$ case, our recent calculations [3] strongly supported the early normalizations suggested by Bloemen *et al.* [17] and Forand *et al.* [18] over a later attempt by Merabet *et al.* [22]. Furthermore, we found that hybrid models as well as fully perturbative calculations underestimated the cross section significantly.

In contrast to what one might expect from the above discussion, Fig. 5 shows good agreement between two experimental datasets [20, 21], the present BSR-1255 pre-

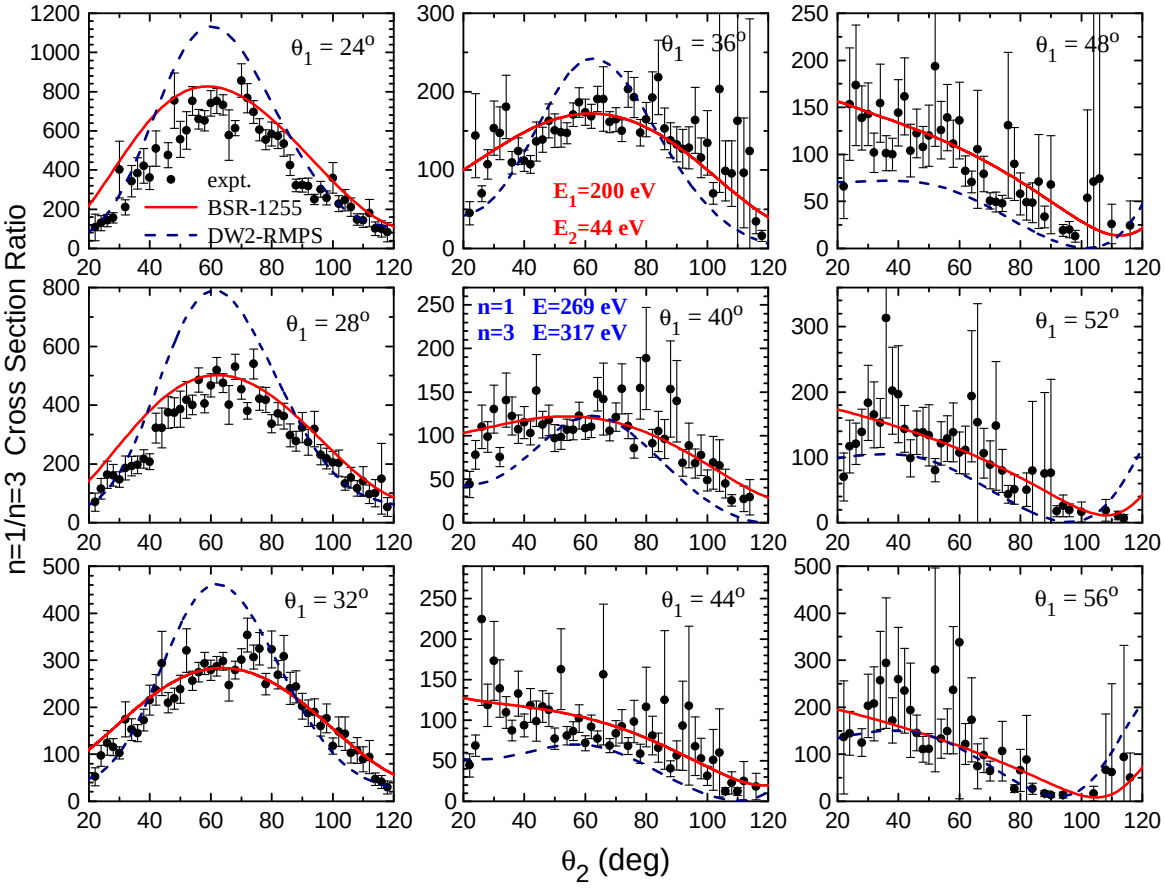

FIG. 2: (Color online) Cross-section ratio for electron impact ionization of helium, leaving the residual ion either in the $1s$ ground state or the $n = 3$ $(3s, 3p, 3d)$ excited states. The two ejected electrons have fixed energies of 200 eV and 44 eV, respectively, hence requiring incident projectile energies of 269 eV (to leave He^+ in the ground state) or 317 eV (to leave He^+ in $n = 3$). The experimental data and the predictions from the DW2-RMPS hybrid theory are from Bellm *et al.* [8].

dictions, and the hybrid DW-RM6 results of Raeker *et al.* [19], which were obtained with a first-order distorted-wave description of the projectile and a six-state close-coupling model for the e−He^+ collision process involving the ejected electron. Since the model did not do very well for ionization with excitation to $n = 2$ [3], we believe that the good performance of the DW-RM6 approach is fortuitous in the present case. This example shows that care should be taken when evaluating the quality and reliability of a theoretical approach based on a very small set of data.

IV. SUMMARY

We have presented a detailed comparison of results for ionization of helium in its ground state with simultaneous excitation to the $n = 3$ manifold of the residual He^+ ion. The good agreement between the experimental data and the theoretical predictions lends further support to the method used to extract the ionization-excitation cross sections via a two-step process of first calculating the excitation of positive-energy discrete pseudostates before mapping the results to the true continuum states by weighting the contributions from various scattering amplitudes with appropriate overlap integrals.

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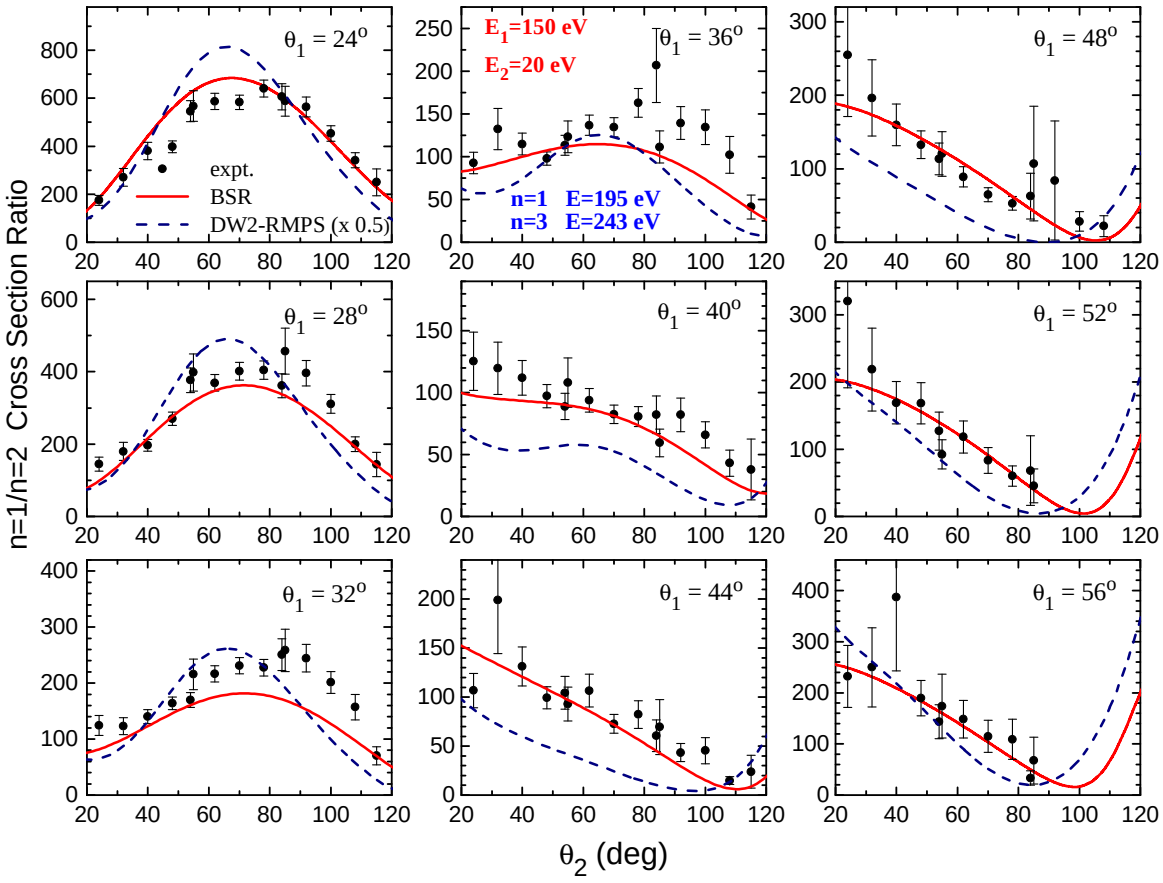

FIG. 3: (Color online) Same as Fig. 2, except that the two ejected electrons have energies of 150 eV and 20 eV, respectively, hence requiring incident projectile energies of 195 eV or 243 eV. Also shown are the hybrid calculations (multiplied by 0.5 to improve visibility) given in Bellm *et al.* [8].

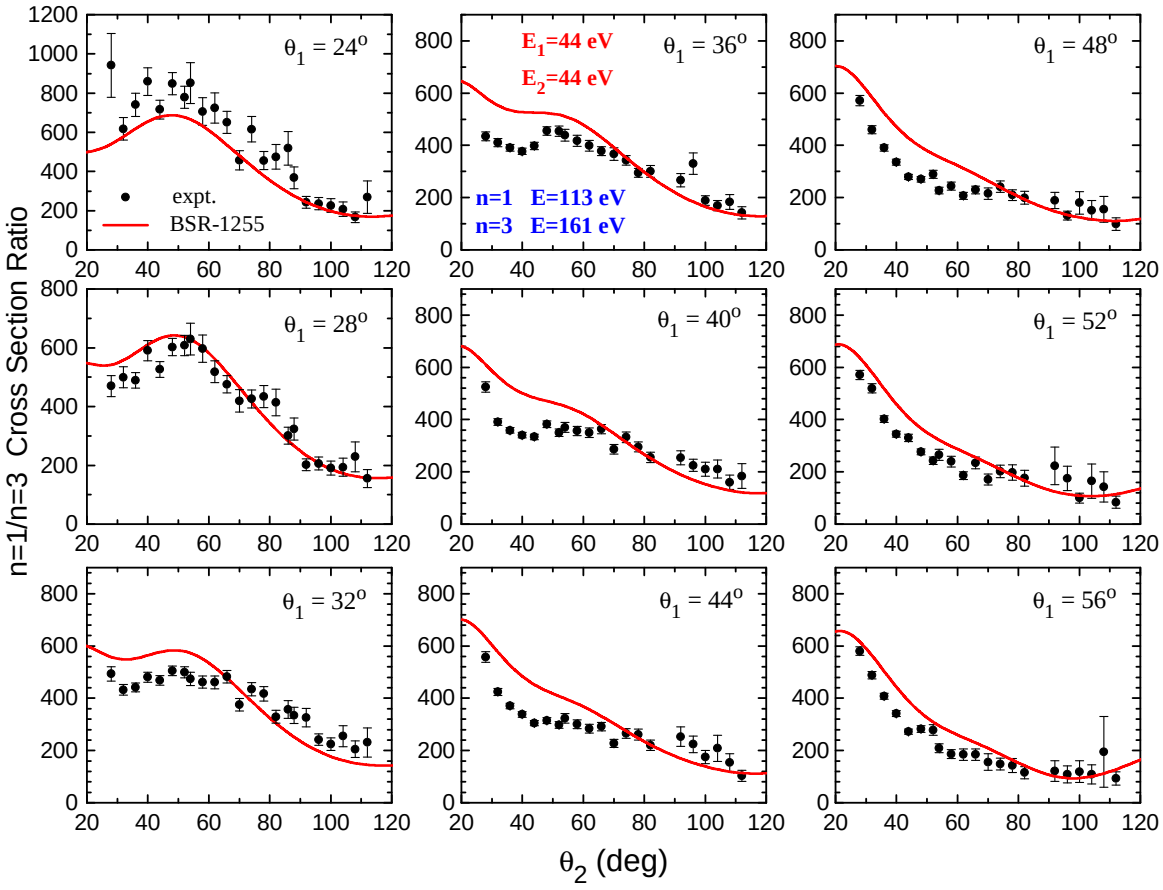

FIG. 4: (Color online) Same as Fig. 2, except that the two ejected electrons have both an energy of 44 eV, hence requiring incident projectile energies of 113 eV or 161 eV. No hybrid results are given, since the asymmetric treatment of the two electrons is inappropriate.

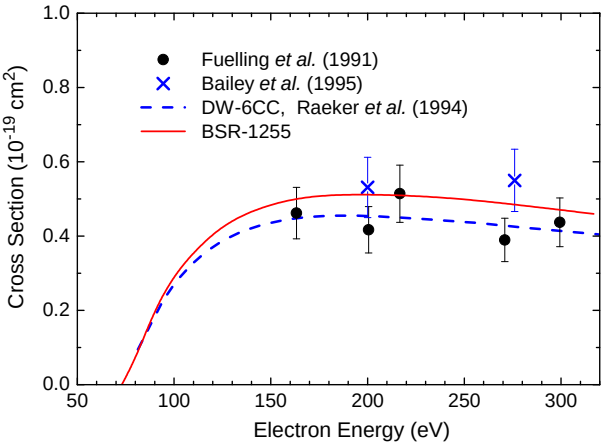

FIG. 5: (Color online) Angle-integrated cross sections for electron impact ionization of helium in its ground state with simultaneous excitation to the $3p$ final state of the residual He^+ ion. Also shown are the hybrid calculations by Raeker *et al.* [19] and the absolute experimental data are of Fuelling *et al.* [20] and Bailey *et al.* [21].

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