Nonperturbative Treatment of Ionization with Excitation of Helium by Electron Impact

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We present cross sections for electron-impact ionization and simultaneous ionization plus excitation of helium by electron impact. The results are obtained from a fully nonperturbative close-coupling formalism using our B-spline R-matrix approach. A large number of pseudostates in the expansion of the wave function represent the coupling to the ionization continuum. We obtain excellent agreement with the directly measured experimental cross section ratios (Bellm *et al.*, Phys. Rev. A **75**, 042704 (2007)) for ionization leaving the residual He^+ ion in either the $1s$ ground state or the $n = 2$ ($2s + 2p$) excited states.

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It has been over a decade since one of the most fundamental problems in atomic collision physics, the ionization of atomic hydrogen by electron impact, a so-called $(e, 2e)$ process, was solved in a fully nonperturbative way by Rescigno *et al.* [1] using the "exterior complex scaling" (ECS) method. The very same problem was also solved, to comparable accuracy, by the time-dependent close-coupling (TDCC) [2] and the time-independent convergent close-coupling (CCC) [3,4] approaches. The major difficulty in the treatment, namely, the complicated boundary conditions due to three free charged particles in the final state, is effectively avoided in both the ECS and TDCC formulations. In the CCC method, one first solves an excitation problem with simpler boundary conditions for the discrete (bound) pseudostates representing the ionization continuum. In a second step, the corresponding transition matrices are transformed to approximate the ionization process. Many of the lessons learned in describing the $(e, 2e)$ process for atomic hydrogen have since been used also in the full breakup (double ionization) of the He atom by photon impact, either in the steady-state [4,5] or the short-pulse regime [6,7].

The natural extension of the three-body Coulomb problem to a four-body case is electron impact of helium, in which four charged particles (three electrons and the nucleus) are involved. In this case, the situation is much more complicated than in atomic hydrogen, due to the significant electron-electron correlations already in the initial bound state. Nevertheless, provided one electron remains in the ground $(1s)^2S$ state of the residual He^+ ion, this electron may, to a high degree of accuracy, be treated as a spectator. With this simplification, the CCC method has been very successful in describing the corresponding $(e, 2e)$ processes as well [8,9]. In fact, the CCC predictions for the triple-differential cross section (TDCS) for ionization without excitation [10] were believed to be sufficiently accurate that they were used [11]—*instead* of measured data—to generate absolute cross sections for ionization with excitation to the $n = 2$ and $n = 3$ states of He^+ from the experimentally determined cross section ratios [12,13] for ionization with and without excitation.

To our knowledge, there has been no fully nonperturbative theory available to date to treat in detail the highly correlated ionization plus excitation process in helium. This is the basic four-body Coulomb problem, even though only two electrons are free in the final state while the third one remains bound to the He^{2+} nucleus. Interestingly, not even the total cross section (integrated over the detection angles of both final-state continuum electrons and the energy loss of the projectile) is currently known to a satisfactory degree of accuracy (see [14] and references therein for a detailed discussion). Although Pindzola *et al.* [15] published total cross sections for three energies from a TDCC model, the work was not pursued to angle-differential observables. No ECS or CCC predictions of this process have been published either.

A partially successful theory for the ionization-excitation process has been a hybrid approach, in which the interaction of a "fast" projectile electron with the target is described by a first-order or second-order distorted-wave approach, while the initial bound state and the scattering of a "slow" ejected electron from the residual ion is treated by a convergent R matrix with pseudostates (RMPS) expansion. These DWB1-RMPS [16] and DWB2-RMPS [17] models were formulated for highly asymmetric kinematics and small energy losses compared to the incident energy. The method proved indeed applicable to such situations (see [18] as an example), but it failed when these conditions were no longer fulfilled [11,13]. A four-body distorted-wave method [19] used asymptotically accurate wave functions, but it suffered from serious shortcomings in the region where the collision most likely occurs and, consequently, was unable to reproduce the experimental data [11].

Given its success for electron impact ionization of atomic hydrogen and ionization without excitation of helium, the close-coupling expansion has remained a promising candidate to tackle the ionization with excitation process. In this Letter, we formulate the problem in the framework of the B-spline R-matrix (BSR) approach [20]. This method has enjoyed significant success in the treatment of low-energy, near-threshold electron collisions with

complex atoms. Since a comprehensive list of publications is impossible, we refer to [21] as a recent example and to [22–24] for applications to e-He collisions.

The key point of the method is the use of B splines as a universal and effectively complete basis to describe the projectile electron in the close-coupling expansion of the collision system. A distinctive feature of our BSR implementation is the possibility to employ individually optimized, and hence "nonorthogonal" orbitals to describe the target states, and we do not restrict the projectile orbitals to be orthogonal to the target orbitals either. Although the lack of these restrictions makes the setting up of the Hamiltonian matrix significantly more complicated than in the standard R-matrix approach [25], the flexibility of the method has proven to be a critical advantage on many occasions.

In recent calculations [21–24], we already introduced a few pseudostates in the BSR close-coupling expansion. The principal purpose of these states, however, was to further improve the target description and to represent the contribution from the ionization continuum to such physically important effects as the polarization of the target by the projectile. Here we extended this approach substantially by including a large number of pseudostates for a rigorous treatment of the target continuum. As a result, we can now consider ionization processes as well.

Specifically, we are interested in the ionization of an atom by electron impact, schematically written as

$$e_0(\boldsymbol{k}_0, \mu_0) + A(L_0, M_0; S_0, M_{S_0}) \rightarrow e_1(\boldsymbol{k}_1, \mu_1) + e_2(\boldsymbol{k}_2, \mu_2) + A^+(L_f, M_f; S_f, M_{S_f}), \quad (1)$$

where the $\boldsymbol{k}_i$ are the linear momenta of the incident, scattered, and ejected electrons, respectively, and the μ_i are their spin projections. Furthermore, L_0, S_0 and L_f, S_f are the orbital and spin angular momenta of the initial $(N+1)$-electron atom and the residual N-electron ion, with the corresponding magnetic quantum numbers M_0, M_{S_0} and M_f, M_{S_f}. Since we are using a nonrelativistic model, we can handle all spins and spin components by Clebsch-Gordan coefficients. To simplify the notation, we will omit these quantum numbers below.

For a complete description of this process, we need the ionization amplitude

$$f(L_0, M_0, S_0; \boldsymbol{k}_0 \rightarrow L_f, M_f, S_f; \boldsymbol{k}_1, \boldsymbol{k}_2). \quad (2)$$

In the continuum pseudostate approach [3], one begins by replacing the true continuum orbitals of the ejected electron by a square-integrable representation, usually obtained by diagonalizing the target Hamiltonian in an appropriate basis. The total, angle-integrated ionization cross section can then be obtained from the excitation cross sections of the pseudostates. To obtain the more detailed angle-differential cross sections, however, one needs to first project the discrete pseudostate functions to the true continuum functions at the proper ejected electron energy and construct the ionization amplitude (2).

In our method, the atomic wave function describing the $(N+1)$-electron system is expanded in terms of products of the N-electron ionic states and radial functions for the outer electron. For He ($N = 1$) specifically, we use

$$\Phi(LS) = \mathcal{A} \sum_{i,j} \{\varphi(n_i l_i) P(n_j l_j)\}^{LS} + \phi(1s^2). \quad (3)$$

The operator $\mathcal{A}$ denotes the proper antisymmetrization. It implies that the target function $\varphi(n_i l_i)$ is coupled to the outer electron represented by $P(n_j l_j)$ through the usual angular momentum rules for total orbital angular momentum L and spin S. In our case the functions $\varphi(n_i l_i)$ are the hydrogenic orbitals $1s$, $2s$, and $2p$ for He^+.

In our approach, the radial functions

$$P(n_j, l_j) = \sum_k b_{kj} B_k(r) \quad (4)$$

are expanded in a B-spline basis. The vectors of unknown expansion coefficients $\boldsymbol{b}_j$ for each orbital are found by diagonalizing the target Hamiltonian matrix inside a box of radius a. These functions are forced to vanish at the edge of the box. Note that the nonorthogonal orbital technique implemented in our BSR code allows us to use an independently optimized multiconfiguration expansion for the initial $1s^2$ state. We obtained an energy of -2.90175 a.u. for this state.

As mentioned above, an important property of B splines is that they form an effectively complete basis over the interval spanned by the knot sequence. The number of physical states that we can generate depends on the radius of the box. Along with the physical states, the scheme provides a set of pseudostates that represent the Rydberg spectrum and the continuum. We chose $a = 15a_0$ (where $a_0 = 0.529 \times 10^{-10}$ m is the Bohr radius) and used 44 B splines of order 8 on a semiexponential grid of knots. This resulted in 525 physical and pseudo target states that covered the energy region up to 300 eV with S, P, D, and F symmetries. The set of pseudostates contained the configurations $1sn_1l_1$, $2sn_2l_2$, and $2pn_3l_3$, with the latter two sets describing doubly excited autoionizing states and the ionization-excitation process.

We then obtained the scattering amplitudes for *excitation* of all pseudostates using our BSR complex [20] for electron collisions. Contributions from all $(N+2)$-electron symmetries with total orbital angular momentum ≤ 25 were included in the partial-wave expansion. The present model contained up to 1303 scattering channels, leading to generalized eigenvalue problems with matrix dimension up to 80 000 in the B-spline basis. The corresponding solutions were obtained with a newly developed parallelized version of the BSR complex.

The last, and most crucial step in the process, is the generation of the ionization amplitudes (2). This is done by summing up the amplitudes for excitation of all the energetically accessible pseudostates (index p in Eq. (5)

below), with the expansion factors given by the overlap of the pseudostates and the true continuum function. Specifically, we calculate

$$f(L_0, M_0, S_0; \boldsymbol{k}_0 \rightarrow L_f, M_f, S_f; \boldsymbol{k}_1, \boldsymbol{k}_2) = \sum_p \langle \Psi_f^{\boldsymbol{k}_2^-} | \Phi(L_p S_p) \rangle f(L_0, M_0, S_0; \boldsymbol{k}_0 \rightarrow L_p, M_p, S_p; \boldsymbol{k}_{1p}). \quad (5)$$

Here the function $\Psi_f^{\boldsymbol{k}_2^-}$ is the close-coupling solution of the e-He^+ collision problem with the boundary condition of incoming waves in all channels and an outgoing wave in the channels represented by the ionic state with quantum numbers L_f, M_f, and S_f, respectively.

Note that $\Psi_f^{\boldsymbol{k}_2^-}$ and $\Phi(L_p S_p)$ have *different* energies for the continuum electron represented by $\boldsymbol{k}_2$ and the electron in the pseudostate. Because of energy conservation, excitation of $\Phi(L_p S_p)$ leads to $\boldsymbol{k}_{1p} \neq \boldsymbol{k}_1$ for the projectile. While interpolating the transition-matrix elements works well for the single-channel case [3], our direct projection method is necessary to maintain the crucial channel information in multichannel situations. This makes Eq. (5), which is the generalization of Eq. (15) of [8] for multichannel cases, a suitable approximation for the true ionization amplitude. More details will be given in a separate publication.

The TDCS for He can be written as

$$\frac{d\sigma_f}{dE_{1(2)} d\Omega_1 d\Omega_2} = \frac{k_1 k_2}{k_0} \sum_{M_f} |f(M_f; \boldsymbol{k}_1, \boldsymbol{k}_2)|^2, \quad (6)$$

where we have simplified the notation of the amplitude (5) by omitting $\boldsymbol{k}_0$ as well as the quantum numbers $L_0 = M_0 = S_0 = 0$ and $S_f = 1/2$. We emphasize that the primary and the ejected electron are not treated symmetrically in the pseudostate approach. For comparison with experiment, therefore, we should also consider the ionization process, in which the primary electron has the final momentum $\boldsymbol{k}_2$ while the ejected electron has momentum $\boldsymbol{k}_1$. Since the two processes cannot be distinguished, the corresponding amplitudes should be added coherently. For details, including spin-related factors, see [9].

As a first test of our method, Fig. 1 exhibits the TDCS for electron impact ionization of helium in its $(1s^2)^1S$ ground state with the residual ion left in the $He^+(1s)^2S$ state. The primary energy E_0 is 112.6 eV and the two final-state electrons both have energies of 44.0 eV. Comparing the present BSR results with the predictions from the CCC and the hybrid DWB2-RMPS approaches [11], we find overall good agreement between CCC and BSR regarding the shape of the TDCS for all four scattering angles for which results are available. The discrepancies between the CCC and BSR absolute values (up to 30%) are most likely due to the different descriptions of the initial bound state. We use a multicore multiconfiguration expansion while the CCC calculations were performed in the frozen-core approximation with one electron fixed in the He^+ $1s$ orbital.

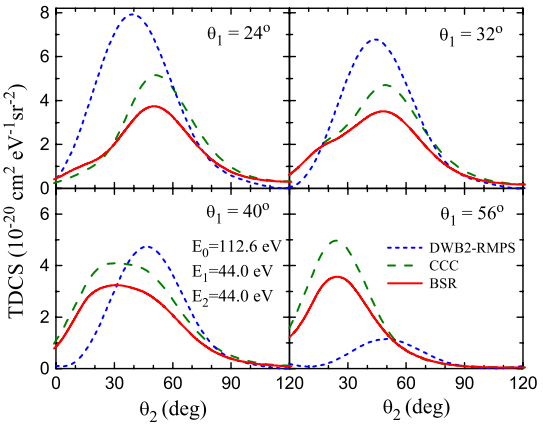

FIG. 1 (color online). TDCS for electron impact ionization of helium in its $(1s^2)^1S$ ground with the residual ion left in the $He^+(1s)^2S$ state. The primary energy E_0 is 112.6 eV and both final-state electrons have energies of 44.0 eV.

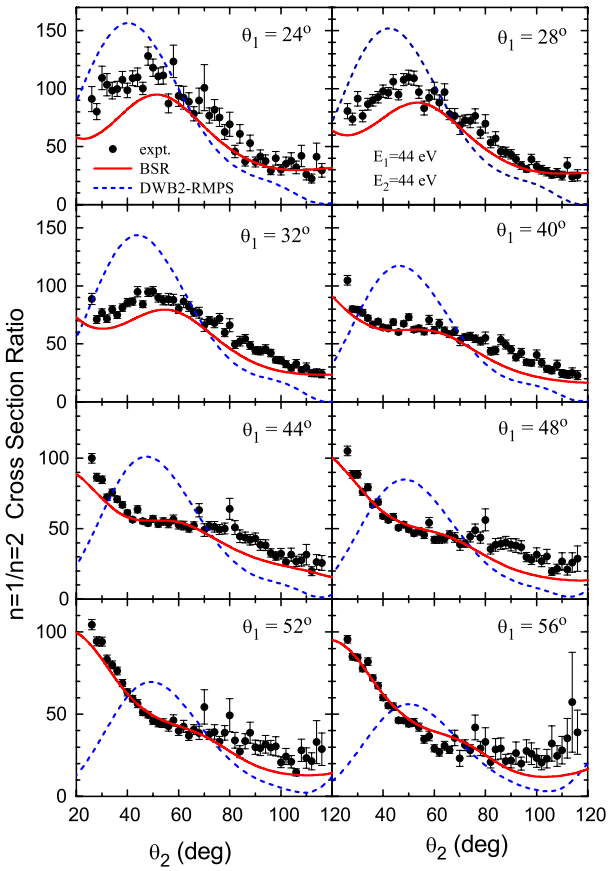

FIG. 2 (color online). TDCS ratio for electron impact ionization of helium in its $(1s^2)^1S$ ground with the residual ion left in either the $He^+(1s)^2S$ state or the excited He^+ $(n = 2)$ states. The primary energy E_0 is either 112.6 eV $(n = 1)$ or 153.4 eV $(n = 2)$ and both final-state electrons have energies of 44.0 eV. The experimental data of Bellm *et al.* [13] are compared with the present BSR results and with predictions from the hybrid DWB2-RMPS approach.

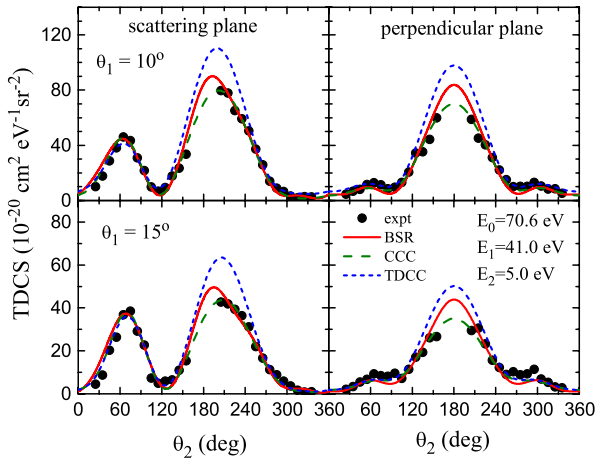

FIG. 3 (color online). TDCS for electron impact ionization of helium in its $(1s^2)^1S$ ground with the residual ion left in the $He^+(1s)^2S$ state. The primary energy E_0 is 70.6 eV, and the final-state electrons have energies of 41.0 eV and 5.0 eV. The experimental data for the coplanar and perpendicular geometries, as well as the CCC and TDCC predictions, are excerpts from Fig. 2 of [26].

The DWB2-RMPS approach shows significant problems for the symmetric energy sharing case in general, but particularly for the largest detection angle of the reference electron.

The most significant finding of the present work is presented in Fig. 2, where we compare the BSR and DWB2-RMPS predictions with the directly measured experimental cross section ratios [13] for ionization without excitation (leaving the electron in He^+ in the $1s$ state) and ionization with excitation to He^+ $(2s + 2p)$. The agreement between the BSR results and the experimental data is excellent at all angles of the reference electron between 24° and 56°, and all detection angles of the other electron between 25° and 115°. As expected, the hybrid approach is inappropriate for large angles of the fixed electron.

We have presented an entirely nonperturbative treatment of the Coulomb four-body problem, as it appears in ionization and particularly ionization with excitation of helium by electron impact. Our formulation is based on the ideas of treating such processes within the framework of the close-coupling expansion. After mapping the amplitudes for ionization of doubly excited pseudostates to the ionization continuum with only one of the two electrons remaining bound in the residual ion, we obtained excellent agreement between our numerical results and directly measured cross section ratios.

We plan to perform a number of additional calculations for the $(e, 2e)$ process in helium, especially extending our initial tests to asymmetric kinematics. As shown in Fig. 3, our first results are very encouraging and show similarly good agreement with the very recent absolute experimental data as the CCC predictions [26]. Where differences remain, the available data do not allow for an unambiguous judgment of which theory might be closer in the gaps where measurements are missing.

Most importantly, however, we are already in a position to move towards more complex targets, particularly Ne and Ar. Many experimental data exist for these systems. While (partially) perturbative theories have had some success and the CCC method has been used for the ionization of s electrons in a frozen-core model, theory has overall been unable to systematically reproduce these data to an acceptable degree of accuracy (see, for example, [27]). We expect to change this situation in the near future.

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