Convergent Close-Coupling Method: A "Complete Scattering Theory"?

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(Received 1 September 1995)

We demonstrate that a single convergent close-coupling calculation of 100 eV electron impact on the ground state of helium is able to provide accurate elastic and inelastic ($n \leq 3$ levels) differential cross sections, as well as singly, doubly, and triply differential ionization cross sections. This is a most promising step towards the development of a complete electron-atom scattering theory.

PACS numbers: 34.80.Bm, 34.80.Dp

Bederson [1] suggested that a goal for experimentalists is to perform "perfect scattering experiments," those which fully determine all aspects of a scattering process of interest. Such experiments have not only intrinsic value, but allow for the most detailed test of the scattering theory used to calculate the corresponding scattering amplitudes. Progress towards this goal has been substantial and continues to attract considerable attention (see Andersen and Bartschat [2], for example).

Similarly, but more generally, it has been our goal to provide a "complete scattering theory." Such a theory is not only able to describe individual perfect scattering experiments for a particular scattering process, but also the many possible scattering processes possible for a given incident projectile energy. In other words, a single calculation should yield accurate elastic, inelastic excitation, and ionization scattering amplitudes. Such a theory is possible only if the Schrödinger equation governing the motion of the interacting particles is solved accurately irrespective of the total energy in the system. In our view the convergent close-coupling (CCC) theory as introduced by Bray and Stelbovics [3] for the e-H scattering system is a candidate for such a description.

The CCC theory is based on the close-coupling (CC) formalism, with the coupled equations solved in momentum space as outlined by McCarthy and Stelbovics [4]. The coupled target states are obtained by diagonalizing the target Hamiltonian H_T in an orthogonal Laguerre (Sturmian) basis. The square integrability of the basis ensures that all of the resulting states Φ_n^N (with associated energies $\epsilon_n^N, n = 1, \ldots, N$) may be incorporated in the CC formalism. The negative-energy states provide a representation of the true discrete target spectrum ($\Phi_n^N \rightarrow \Phi_n, \epsilon_n^N \rightarrow \epsilon_n < 0$ as $N \rightarrow \infty$). The positive-energy states discretize the target continuum inducing a quadrature rule for the integration over the true target continuum (see Ref. [5] and references therein). Convergence, in say the cross sections, is studied by simply increasing the basis size as this yields a better description of the discrete spectrum and a more accurate quadrature rule for the integration over the true target continuum (see Fig. 1). It is the treatment of both the target discrete and continuum subspaces that allows the CCC method to be valid irrespective of the projectile energy or the scattering process of interest.

There is little argument regarding the validity of the CCC method for calculating the excitation cross sections at all energies. In this case the multichannel expansion has the correct boundary conditions (this is why the CC approaches have been historically successful with very few states at low energies), and, furthermore, expansion of the total wave function Ψ is performed using a complete basis. By unitarity it follows that the method is valid for the calculation of the total ionization cross section (see [6], for example). However, it is not so clear as to how to obtain detailed differential ionization cross sections. In particular, given a set of T-matrix elements arising upon solution of the CCC equations, how do we obtain the required ionization T matrix?

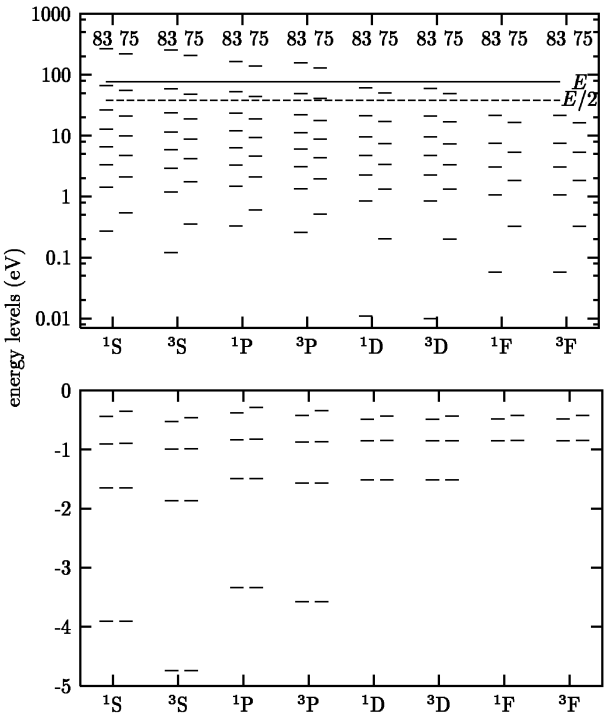

FIG. 1. One-electron excited-state energy levels arising from the 83- and 75-state CCC calculations. The total energy of the e-He system for 100 eV incident electrons is denoted by E.

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In the CCC formalism the total wave function is expanded in an explicitly antisymmetric set of N square-integrable states Φ_n^N via

$$|\Psi_i^{(+)}\rangle \approx |\Psi_i^{N(+)}\rangle = \mathcal{A} \sum_{n=1}^{N} |\Phi_n^N f_{ni}^{N(+)}\rangle, \qquad (1)$$

where $\mathcal{A}$ is the antisymmetrization (space and spin) operator, and $f_{ni}^{N(+)}$ are the unknown one-electron projectile-space functions with outgoing spherical-wave boundary conditions determined upon solution of the coupled equations. We rely on the completeness of our basis so that equality may be achieved in (1) as $N \to \infty$. However, for any finite N it is clear that such an expansion does not allow for two (or more) electrons to be at infinity. In other words, arbitrary three-body boundary conditions cannot be satisfied by (1) for any finite N. The expansion (1) may be viewed as a "shielding" approximation in that, irrespective of the energy distribution of the two (or more) outgoing electrons, only one electron is allowed to be at infinity, and moves in the field of an asymptotically neutral target. This problem does not invalidate the CCC formalism, but indicates that certain kinematical regions may require prohibitively large N for computational purposes. On the other hand, the process which dominates the total ionization cross section, where the "ejected" electron has energy much smaller than the "scattered" electron, is likely to be well modeled by the expansion (1) with relatively small N.

The above considerations make it simple for us to move from the consideration of excitation T-matrix elements to those corresponding to ionization within the CCC framework. We take the view that the calculation of ionization processes is essentially the same as that for excitation processes. We form the ionization T-matrix elements from those corresponding to the excitation of states Φ_n^N with $\epsilon_n^N > 0$, which arise upon solution of the coupled equations simultaneously with those corresponding to excitation. As the states Φ_n^N are normalized to unity we restore the continuum normalization and boundary conditions by multiplying these T-matrix elements by the overlap $\langle \boldsymbol{q}_f^{(-)} | \Phi_f^N \rangle$, where the continuum wave $\boldsymbol{q}_f^{(-)}$ is an eigenstate of H_T and has the energy $q^2/2 = \epsilon_f^N$. This is much the same as was done by Bray *et al.* [7] in the case of e-H ionization, which followed the work of Curran and Walters [8].

Thus, upon solution of the CCC equations we obtain an N-state approximation

$$\langle \boldsymbol{k}_f \Phi_f^N | T^N | \Phi_i \boldsymbol{k}_i \rangle = \langle \boldsymbol{k}_f \Phi_f^N | V | \Psi_i^{N(+)} \rangle \qquad (2)$$

for the excitation T matrix ($\epsilon_f^N \approx \epsilon_f < 0$), and use

$$\langle \boldsymbol{k}_f \boldsymbol{q}_f^{(-)} | T^N | \Phi_i \boldsymbol{k}_i \rangle = \langle \boldsymbol{q}_f^{(-)} | \Phi_f^N \rangle \langle \boldsymbol{k}_f \Phi_f^N | V | \Psi_i^{N(+)} \rangle \qquad (3)$$

for the ionization T matrix ($\epsilon_f^N > 0$). Here the projectile is denoted by a plane wave $\boldsymbol{k}_f$ with corresponding energy $k_f^2/2$. Convergence is tested by increasing N and it is necessary to establish that convergence in both (2) and (3) is indeed observed, particularly so in the latter case as $\langle \boldsymbol{q}_f^{(-)} | \Phi_f^N \rangle \to \infty$ as $N \to \infty$. If convergence is observed, it is equally important that convergence be to the correct result. In the case of excitation this has already been extensively studied (see [5,9]). However, in the case of ionization we immediately run into a problem. As $N \to \infty$ completeness of our basis should result in $|\Psi_i^{N(+)}\rangle \to |\Psi_i^{(+)}\rangle$ and $\langle \boldsymbol{q}_f^{(-)} | \Phi_f^N \rangle \langle \Phi_f^N | \to \langle \boldsymbol{q}_f^{(-)}|$ [10]. This implies

$$\langle \boldsymbol{q}_f^{(-)} | \Phi_f^N \rangle \langle \boldsymbol{k}_k \Phi_f^N | V | \Psi_i^{N(+)} \rangle \to \langle \boldsymbol{k}_f \boldsymbol{q}_f^{(-)} | V | \Psi_i^{(+)} \rangle, \quad (4)$$

which should have a divergent phase factor, irrespective of the kinematics [see Eq. (2.51) of Rudge [11] for $z_1 = 0, z_2 = 1$], if $|\Psi_i^{(+)}\rangle$ satisfies the correct three-body boundary conditions. As we do observe convergence in (3), particularly in the asymmetric kinematics region, we suspect that the limiting procedure provided by simply increasing N does not lead to equality in (4). We use experiment to check that convergence is to the correct result.

In the CCC formalism the T-matrix elements $\langle \boldsymbol{k}_m \Phi_m^N | T^N | \Phi_i \boldsymbol{k}_i \rangle$ and $\langle \boldsymbol{k}_n \Phi_n^N | T^N | \Phi_i \boldsymbol{k}_i \rangle$, where $k_m^2/2 = \epsilon_n^N$ and $k_n^2/2 = \epsilon_m^N$, correspond to two distinct theoretical processes which are observed simultaneously in the experiment. For this reason we sum the cross sections for both of these processes. Each of these T-matrix elements may be written as a coherent sum of direct and exchange amplitudes, but in the case where $\epsilon_m^N \ll k_m^2/2$ the former T matrix is much larger than the latter. Summing the probabilities allows us to stay consistent with the definition of the total and the singly differential ionization cross sections [10].

The aim of this Letter is to demonstrate the ability of the CCC theory to simultaneously describe accurately elastic, excitation, and detailed ionization processes at a single projectile energy. For this to be possible we require the existence of the corresponding experimental data at a single energy. Thanks to the recent measurements of triply differential cross sections by Röder, Jung, and Ehrhardt [12] for 100 eV electron impact ionization of helium we now have such a set at this projectile energy.

The CCC theory for obtaining the T-matrix elements for electron-impact excitation of helium have been given by Fursa and Bray [13]. The only addition necessary to that work is to calculate the above-stated overlaps, which is done after evaluation of the continuum waves $\boldsymbol{q}_f^{(-)}$ (separately for singlet and triplet symmetries) in the same frozen-core approximation as used for generating the states Φ_n^N. In testing convergence we write $N = \sum_{l=0}^{l_{\max}} N_l$ and so need to demonstrate convergence separately with increasing $l_{\max}$ and N_l. For this purpose we shall present the results of the 69-, 75-, and 83-state calculations. The one-electron energy levels for the latter two calculations ($l_{\max} = 3$) are given in Fig. 1, with the 69-state energy

levels being the same as those for the 83-state calculation except for the absence of the F states, i.e., $l_{\max} = 2$. Thus comparison of the 83- and 75-state calculations shows the variation in the results by varying N_l while keeping $l_{\max} = 3$, and comparison of the 83- and 69-state calculations shows the effect of varying $l_{\max}$.

In Fig. 2 we present the differential cross sections for excitation of $n \leq 3$ states by 100 eV electrons incident on the ground state of helium. We find excellent convergence and essentially quantitative agreement with the measurements of various groups. This is a substantial achievement in itself as no other available theory is able to do so well [13].

The least detailed ionization process, the total ionization cross section, may be obtained using the optical theorem, or by simply summing the integrated cross sections corresponding to the excitation of states Φ_n^N with $\epsilon_n^N > 0$ [6]. The 69-, 75-, and 83-state calculations yield (units 10^{-17} cm^2) 3.53, 3.56, and 3.56, respectively, which compares favorably with 3.63 ± 0.2, the experimental estimate [14]. This result is what should be obtained when the singly differential ionization cross section (SDCS) is integrated from zero to $E/2$. The details of how we obtain the SDCS in the CCC formalism may be found in Ref. [10]. Our results are in good agreement with experiment [15] (see Fig. 3). This is particularly encouraging at the midpoint where one of the electrons is treated as a plane wave expanded using 40 partial waves, and the other is described by Coulomb-like states expanded using only 3 or 4 partial waves. Here, as was noted earlier [10], we do not have convergence in the l-dependent contributions in the vicinity of $e = E/2$, yet unitarity ensures convergence in the summed result.

Next, we move on to the more detailed doubly differential cross sections which give the angular distribution for an outgoing electron of particular energy. The integral over the angular distribution gives the SDCS. These are presented in Fig. 4. We find convergence and agreement with experiment to be surprisingly good. The fact that increasing $l_{\max}$ from 2 to 3 has a small effect (difference between the 83- and 69-state results) even when both electrons have energy >20 eV is due to the fact that these cross sections are dominated by the ejection of the target electron into the ^1P continuum wave. Such cross sections are well described by the theory as convergence in this wave is readily obtained by the inclusion of $^{1,3}D$ and $^{1,3}F$ states in the CCC formalism.

Finally, we turn to the most detailed ionization information, the triply differential cross sections (TDCS). These describe the angular behavior of one of the outgoing electrons for a given energy and position of the other. In Fig. 5 we present the calculated TDCS for the coplanar asymmetric geometry, where the angular distribution θ_B is for the slow electron $E_B = 4$ eV with the corresponding fast electron being observed at the four angles $\theta_A = -20°, -25°, -30°$, and $-150°$. The measurements, due Röder, Jung, and Ehrhardt [12], are internormalized so that a single experimentally estimated normalization point at $(\theta_A, \theta_B) = (-20, 75)$ fixes the scale for all four figures. We see that the CCC theory obtains essentially quantitative agreement with experiment. Such excellent agreement with experiment poses the question as to whether the CCC theory will be able to obtain correct TDCS irrespective of the choice of the kinematics and

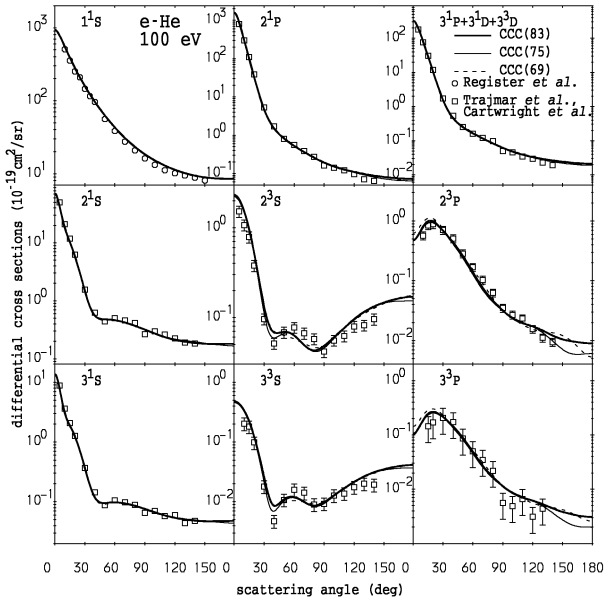

FIG. 2. Differential cross sections for e-He scattering at a projectile energy of 100 eV. The measurements are from Register, Trajmar, and Srivastava [16], Cartwright *et al.* [17], and Trajmar *et al.* [18].

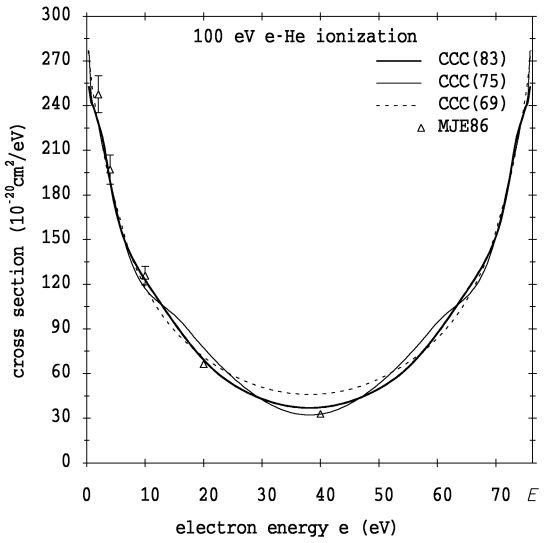

FIG. 3. The singly differential cross section for 100 eV electron impact ionization of the ground state of helium. The energy of an outgoing electron e ranges from 0 to the total energy E. The measurements, denoted by MJE86, are from Ref. [15].

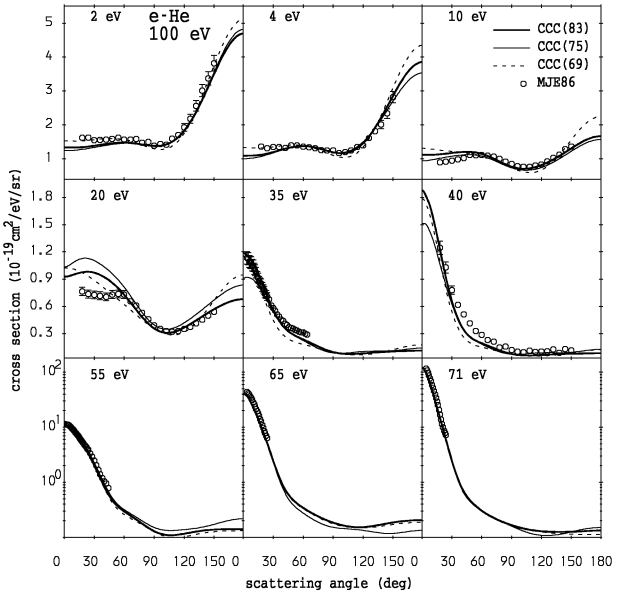

FIG. 4. The doubly differential cross sections for 100 eV electron impact ionization of the ground state of helium for various indicated energies of an outgoing electron. The measurements, denoted by MJE86, are from Ref. [15].

geometry. We are unable to answer this question at this stage. For example, preliminary calculations have reproduced the shape of the symmetric coplanar TDCS in a number of cases, but appear to be too low in magnitude.

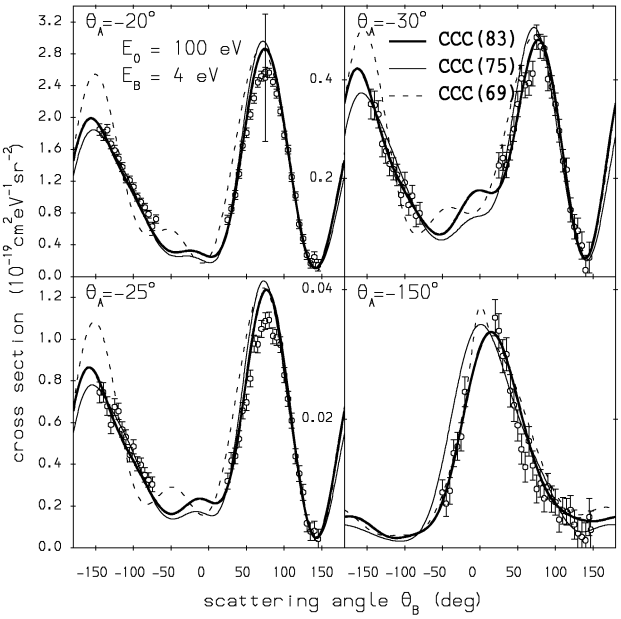

FIG. 5. The triply differential cross sections for 100 eV electron impact ionization of the ground state of helium. The measurements are from Röder, Jung, and Ehrhardt [12].

In conclusion, we have seen that a single CCC calculation may readily obtain accurate cross sections for discrete excitation, as well singly, doubly, and triply differential ionization. For this reason we believe the CCC theory goes some way to being a complete scattering theory. It now remains to extensively test the CCC approach for calculating differential ionization processes at lower impact energies with the vast array of the available experimental TDCS at various geometries and kinematic combinations.

We are indebted to Jochen Röder for performing the TDCS measurements at our request. We thank Joe Macek for bringing to our attention the problem of the divergent phase factor arising in Eq. (4). Stimulating discussions with Ian McCarthy and Colston Chandler are gratefully appreciated. Support of the Australian Research Council and the Flinders University of South Australia is acknowledged. Research sponsored in part by the Phillips Laboratory, Air Force Materiel Command, USAF, under cooperative agreement, No. F29601-93-2-0001.

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[1] B. Bederson, Comments At. Mol. Phys. **1**, 41 (1969).
[2] N. Andersen and K. Bartschat, Comments At. Mol. Phys. **29**, 157 (1993).
[3] I. Bray and A. T. Stelbovics, Phys. Rev. A **46**, 6995 (1992).
[4] I. E. McCarthy and A. T. Stelbovics, Phys. Rev. A **28**, 2693 (1983).
[5] I. Bray and A. T. Stelbovics, Comput. Phys. Commun. **85**, 1 (1995).
[6] I. Bray and A. T. Stelbovics, Phys. Rev. Lett. **70**, 746 (1993).
[7] I. Bray, D. A. Konovalov, I. E. McCarthy, and A. T. Stelbovics, Phys. Rev. A **50**, R2818 (1994).
[8] E. P. Curran and H. R. J. Walters, J. Phys. B **20**, 337 (1987).
[9] I. Bray and A. T. Stelbovics, Phys. Rev. Lett. **69**, 53 (1992).
[10] I. Bray and D. V. Fursa, J. Phys. B **28**, L435 (1995).
[11] M. R. H. Rudge, Rev. Mod. Phys. **40**, 564 (1968).
[12] J. Röder, K. Jung, and H. Ehrhardt (private communication).
[13] D. V. Fursa and I. Bray, Phys. Rev. A **52**, 1279 (1995).
[14] R. G. Montague, M. F. A. Harrison, and A. C. H. Smith, J. Phys. B **17**, 3295 (1984).
[15] R. Müller-Fiedler, K. Jung, and H. Ehrhardt, J. Phys. B **19**, 1211 (1986).
[16] D. F. Register, S. Trajmar, and S. K. Srivastava, Phys. Rev. A **21**, 1134 (1980).
[17] D. C. Cartwright, G. Csanak, S. Trajmar, and D. F. Register, Phys. Rev. A **45**, 1602 (1992).
[18] S. Trajmar, D. F. Register, D. C. Cartwright, and G. Csanak, J. Phys. B **25**, 4889 (1992).