Close-Coupling Approach to Coulomb Three-Body Problems

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The convergent close-coupling method is shown to overcome the remaining discrepancies with experiment of electron-hydrogen ionization. Consequently, this method is able to calculate accurately the Coulomb three-body problems which include electron collisions with hydrogen and helium (within the frozen core model), and helium double photoionization at all incident energies and kinematical or geometrical arrangements of the outgoing electrons.

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There has been much progress in the theoretical description of electron-atom collisions over the last decade or so, with the latest highlight being the numerical solution of the electron-impact ionization of hydrogen problem using the exterior complex scaling (ECS) technique by Rescigno *et al.* [1]. Until this time most of the progress had come from the many implementations of the close-coupling (CC) method, from the most widely used R-matrix approach [2], and its variants, through to our own momentum-space-based convergent close-coupling (CCC) method [3]. The CC techniques were developed in the early 1930s by Massey and Mohr [4] who gave a general formalism for treating the discrete atomic transitions, vital in numerous scientific and industrial applications. The key idea is to expand the total wave function using square-integrable states. Since the close-coupling equations yield stationary amplitudes upon variation in the expansion of the total wave function it is not surprising that they have been so successful in treating discrete transitions in widely varying collision systems. However, the usage of the square-integrable states suggests that the formalism will be inadequate for treating ionization.

Here we shall see that the extension of the CCC method to calculating ionization processes, as suggested by Bray and Fursa [5], also yields accurate ionization amplitudes solong as sufficient computational resources are utilized in their evaluation. Given the extensive literature on the complexity of the Coulomb three-body problem, see Rudge [6] and Peterkop [7] for example, the CCC approach is astonishingly simple, and has been severely criticized [8,9], but rebutted [10,11]. We also began doubting the method when application to ionization of hydrogen with equal-energy outgoing electrons proved less than satisfactory [12]. In addition, the complete presentation of the ECS results [13], which yielded unprecedented agreement with experiment, and correction of inconsistencies in the experiment at 17.6 eV [14], also made us question the formalism [5]. However, we shall show that the problem proved to be not with the formalism, but rather with limited computational resources.

The primary ingredient of the CCC method is the complete Laguerre basis

$$\xi_{kl}(r) = \left[\frac{\lambda_l (k-1)!}{(2l+1+k)!}\right]^{1/2} \times (\lambda_l r)^{l+1} \exp(-\lambda_l r/2) L_{k-1}^{2l+2}(\lambda_l r), \quad (1)$$

where the $L_{k-1}^{2l+2}(\lambda_l r)$ are the associated Laguerre polynomials, and k ranges from 1 to the basis size N_l. In the case of hydrogen, the target Hamiltonian $H_2 = K_2 + V_2$ is diagonalized in this basis yielding (pseudo) target states $|\phi_n^{(N)}\rangle$, with energies $\epsilon_n^{(N)}$, which for each $l \le l_{\max}$ satisfy

$$\langle \phi_m^{(N)} | H_2 | \phi_n^{(N)} \rangle = \delta_{mn} \epsilon_n^{(N)}, \quad (2)$$

where $N = \sum_{l=0}^{l_{\max}} N_l$. We use the (N) superscript to indicate the general dependence of the states on both the basis size N_l and the Laguerre exponential falloff parameter λ_l.

It is convenient to define the projection operator in the target space

$$I_2^{(N)} = \sum_{n=1}^{N} |\phi_n^{(N)}\rangle\langle\phi_n^{(N)}|, \quad (3)$$

with the understanding that the usage of the complete basis (1) ensures that

$$\lim_{N\to\infty} I_2^{(N)} = I_2, \quad (4)$$

the corresponding true identity operator. Then for the total Hamiltonian H acting to the left, energy E and spin S of the electron-hydrogen system we write the close-coupling approximation for the scattering amplitudes as [5]

$$\langle \Phi_f | H - E | \Psi_i^{(S+)} \rangle \approx \langle \Phi_f | I_2^{(N)} (H - E)[1 + (-1)^S P_r] I_2^{(N)} | \psi_i^{(S+)} \rangle, \quad (5)$$

where the required symmetry is imposed with the usage of the space exchange operator P_r, and $\langle \Phi_f |$ is an eigenstate of the asymptotic Hamiltonian. Since $I_2^{(N)}$ ensures

that the target-space electron is always bounded the asymptotic Hamiltonian K is taken to be the same for both discrete excitation and ionization, namely $K = K_1 + H_2$. Then for discrete transitions $\langle\Phi_f| = \langle \boldsymbol{k}_f \phi_f|$, where ϕ_f is a discrete eigenstate and $\boldsymbol{k}_f$ a plane wave. For ionizing collisions $\langle\Phi_f| = \langle \boldsymbol{k}_f \boldsymbol{q}_f^{(-)}|$, where $\boldsymbol{q}_f^{(-)}$ is a Coulomb wave of energy $q_f^2/2$, and the amplitude (5) for some initial state i may be written as

$$f^{(NS)}(\boldsymbol{k}_f, \boldsymbol{q}_f) = \langle \boldsymbol{q}_f^{(-)}|\phi_f^{(N)}\rangle\langle \boldsymbol{k}_f \phi_f^{(N)}|T^{(NS)}|\phi_i^{(N)}\boldsymbol{k}_i\rangle, \quad (6)$$

where $q_f^2/2 = \epsilon_f^{(N)}$, and we used the fact that $\langle \boldsymbol{q}_f^{(-)}|\phi_n^{(N)}\rangle = \delta_{fn}\langle \boldsymbol{q}_f^{(-)}|\phi_f^{(N)}\rangle$. The excitation amplitudes $T^{(NS)}$ are found by solving the close-coupling equations in momentum space [3]

$$\langle \boldsymbol{k}_f \phi_f^{(N)}|T^{(NS)}|\phi_i^{(N)}\boldsymbol{k}_i\rangle = \langle \boldsymbol{k}_f \phi_f^{(N)}|V^{(NS)}|\phi_i^{(N)}\boldsymbol{k}_i\rangle + \sum_{n=1}^{N}\int d^3k \frac{\langle \boldsymbol{k}_f \phi_f^{(N)}|V^{(NS)}|\phi_n^{(N)}\boldsymbol{k}\rangle\langle \boldsymbol{k}\phi_n^{(N)}|T^{(NS)}|\phi_i^{(N)}\boldsymbol{k}_i\rangle}{E + i0 - \epsilon_n^{(N)} - k^2/2}. \quad (7)$$

The sum above effectively approximates a sum over the true negative-energy eigenstates and an integral over the true target continuum, with the energy integration ending at E on the energy shell.

The formulation (6) is extraordinarily simple and some immediate problems are apparent. For any $q_f < k_f$ there are two independent amplitudes $f^{(NS)}(\boldsymbol{k}_f, \boldsymbol{q}_f)$ and $f^{(NS)}(\boldsymbol{q}_f, \boldsymbol{k}_f)$ that arise from excitation of pseudostates of energy $\epsilon_f^{(N)} = q_f^2/2$ and $\epsilon_{f'}^{(N)} = k_f^2/2$, respectively. Superficially, there appears to be a double counting problem, yet within a unitary formalism. Following a numerical study a resolution was found by suggesting that the scattering amplitudes in (7) form a step-function with

$$\lim_{N\to\infty}\langle \boldsymbol{k}_f \phi_f^{(N)}|T^{(NS)}|\phi_i^{(N)}\boldsymbol{k}_i\rangle = 0 \quad \text{for } k_f^2/2 < \epsilon_f^{(N)}, \quad (8)$$

and hence the on-shell target-space energy integration in (7) effectively ending at $E/2$ [15]. Further analysis of the problem by Stelbovics [16] suggested that the close-coupling expansion behaves like a Fourier expansion of a step-function with convergence of the amplitudes at the step to half the step height, thereby explaining the observed numerical behavior. The most convincing example of this was recently demonstrated by Scott *et al.* [17], who showed how the step-function is approached with increasing size of the calculations, and that the average over the diminishing oscillations yields physically accurate results.

Though the $f^{(NS)}(\boldsymbol{q}_f, \boldsymbol{k}_f)$ do not satisfy the expected symmetry condition $f^{(S)}(\boldsymbol{q}_f, \boldsymbol{k}_f) = (-1)^S f^{(S)}(\boldsymbol{k}_f, \boldsymbol{q}_f)$ generally, they do so for $q_k = k_f$ [12]. The doubling at $q_f = k_f$, and satisfying the required symmetry is achieved by writing the final CCC amplitude $F^{(NS)}$ for comparison with experiment as

$$F^{(NS)}(\boldsymbol{q}_f, \boldsymbol{k}_f) = f^{(NS)}(\boldsymbol{q}_f, \boldsymbol{k}_f) + (-1)^S f^{(NS)}(\boldsymbol{k}_f, \boldsymbol{q}_f), \quad (9)$$

with the understanding that for $q_f \neq k_f$ one of the two terms on the right-hand side above is negligible for sufficiently large N.

This should have been the end of the story, particularly since it confirmed the utility of the CCC method for electron-impact ionization of helium as explained by Bray, Fursa, and Stelbovics [18]. Similarly for the case of helium double photoionization, see Kheifets and Bray [19], and references therein. However, not so in the case of e-H ionization with equal-energy outgoing electrons [12], thus bringing into question the entire formalism.

It turns out that there were two problems with the earlier calculations [12]. The primary problem was that the then available computational resources did not allow calculations with large-enough basis sizes N_l. This resulted in having to vary the λ_l to ensure a pseudostate energy at exactly the value of interest in the experiment. Since the CCC amplitudes (6) are available only at the energies of the pseudostates we had to avoid interpolation of complex numbers across sparsely spaced energies. It is the systematic variation of the λ_l, as N_l were varied, that resulted in the apparent convergence to the wrong result.

Here we consider the key test case of 17.6 eV e-H ionization of atomic hydrogen with 2 eV outgoing electrons. The initial measurements of this case have been recently revised [14], and are now much more internally consistent than was the case earlier [12].

The CCC calculations depend on the parameters $l_{\max}$, N_l, and λ_l. The earlier 17.6 eV calculation [12] had $l_{\max} = 5$, $N_l = 20 - l$, and $\lambda_l \approx 0.8$, which was varied to ensure one pseudostate energy was exactly 2 eV for each l. The results presented here are from a CCC calculation which has $l_{\max} = 5$, $N_l = 50 - l$, and $\lambda_l = 2$. The usage of the much larger N_l and λ_l ensures that the continuumlike pseudostates go out considerably further than in the earlier calculation. It is more important to have accurate oscillations in the continuumlike pseudostates, which come from the larger N_l, than attempting to extend the states by taking a smaller λ_l. The much denser discretization allows to take a single value for λ_l and then interpolate amongst the complex amplitudes to obtain the result at 2 eV. The combined usage of $N_l = N_0 - l$ and $\lambda_l = \lambda$ ensures a commensurate treatment of the target-space l, and leads to the most efficient rate of convergence.

We begin with the largest cross sections measured, which are for the fixed-θ_A geometries. These are presented in Fig. 1 together with the ECS and CCC theories. The experimental uncertainty in the absolute value determination is $\pm 40\%$ [20], and so the uniform multiplication of experiment by 0.7, for best visual fit to the theories, is

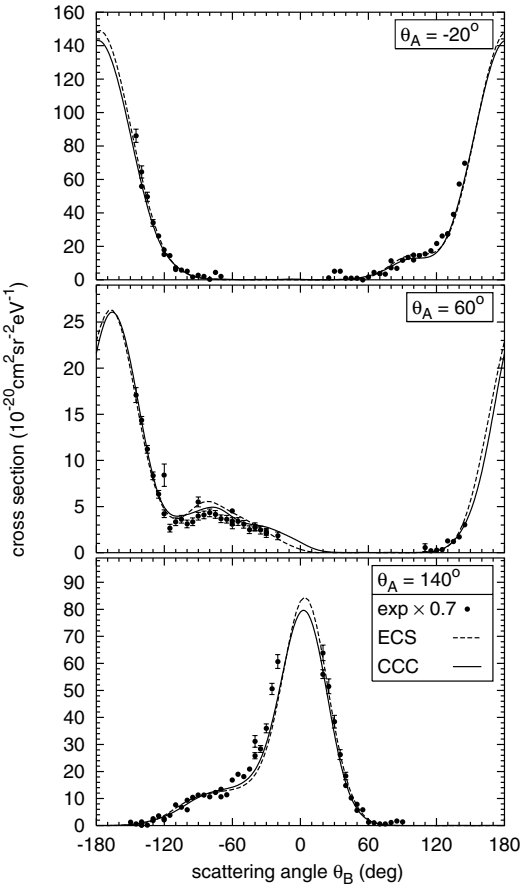

FIG. 1. Coplanar triply differential cross sections, in the specified fixed-θ_A geometries, for 17.6 eV electron-impact ionization of atomic hydrogen with 2 eV outgoing electrons. The experimental data, with ±40% uncertainty in overall absolute value, are the full set presented in Ref. [14] multiplied uniformly by 0.7 for best visual comparison to the theory. The ECS calculation is from Ref. [13]. The CCC calculation is outlined in the text.

well within this uncertainty. We see excellent agreement between experiment and the two theories. The theories and the experiment are barely distinguishable, a much improved result on the comparison presented earlier [12].

The so-called fixed-θ_{AB} geometries keep the angle between the two detectors constant. They succinctly capture the underlying physical processes, and are presented in Fig. 2. Going down from the $\theta_{AB} = 90°$ to $\theta_{AB} = 180°$ the electrons are detected further and further apart, and consequently the cross sections generally increase. Agreement between the two theories and experiment is again excellent.

The last geometry for which data are available is the so-called doubly symmetric geometry (Fig. 3), where the two electrons of same energy go out on the opposite sides of the incident beam, i.e., $\theta_A = -\theta_B$. Once more we see generally good agreement between theories and experiment, except at the very forward angles. Here the present calculation shows its limitations since near-zero cross

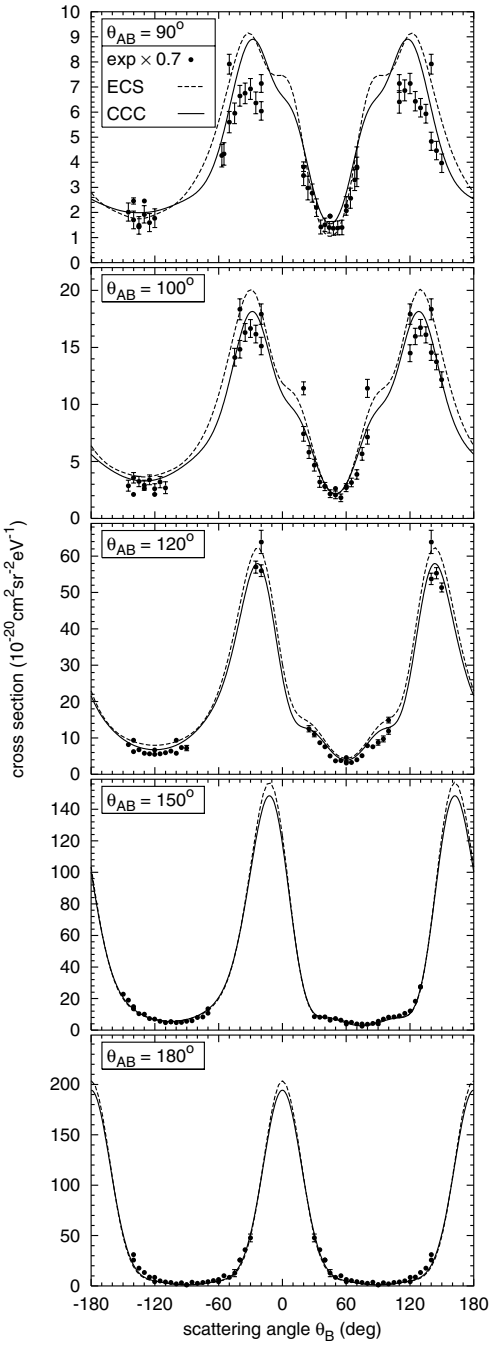

FIG. 2. Same as for Fig. 1, but fot the fixed-θ_{AB} geometries.

sections are expected. The problem comes from inaccuracies associated with the complex amplitude interpolation. Here we require large complex numbers to cancel each other to yield near-zero cross sections.

The CCC results presented here confirm that the evaluation of the ionization amplitudes from the simple Eq. (6) after solution of (7) does result in accurate amplitudes so long as N is sufficiently large. Such calculations are now practical owing to the ever-growing computational power. The CCC calculations have also been performed at the other incident energies of 15.6, through to 30 eV where ECS and experimental data are available. Similarly

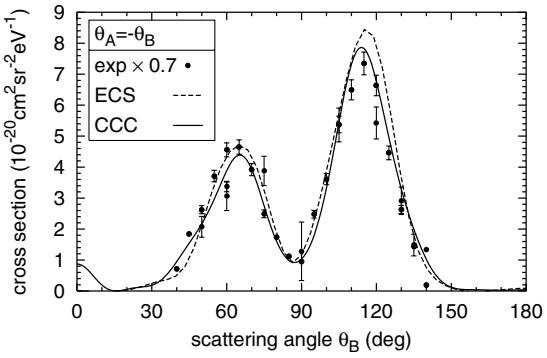

FIG. 3. Same as for Fig. 1, except for the $\theta_A = -\theta_B$ geometry.

excellent agreement has been found at all energies (see [21] for the 25 eV case), and will be presented subsequently together with details of convergence studies. Here we just mention that the same value of $\lambda_l = 2$ was able to be used with $N_l = 65 - l$ being necessary for satisfactory convergence at the lowest energy through to $N_l = 20 - l$ at 30 eV.

The recent calculations also explain why the CCC method worked so well in solving the closely related problems of double photoionization and electron-impact ionization of helium. In both these cases the CCC calculations had to obtain an accurate representation of the He^+ 1s orbital, and hence had λ_l typically around twice the value used for hydrogen previously. This allowed denser discretization of the continuum and hence smaller variation in the λ_l. The shorter range of the interactions also contributed substantially to the ease of the numerical solution.

While we have concentrated on the most computationally difficult case where the two outgoing electrons have the same energy, the generally more probable case of ionization with unequal energy electrons requires some discussion. In these cases, taking $q_f < k_f$ in (6), we find that widely varying calculations yield nearly identical results so long as account is taken of how the underlying Fourier expansion is behaving. Since we know *ab initio* the size of the step, the integral, and that the oscillations are about the true values, accurate amplitudes are able to be obtained from relatively small calculations. We have compared the doubly differential cross sections arising from the present calculations with those of the ECS theory in steps of 0.25 eV, and found excellent agreement in shape and magnitude. Alternatively, increasing N yields diminishing oscillations in the calculated amplitudes, and may be used to check the results from smaller calculations, as most recently confirmed by Scott *et al.* [17].

In summary, the present results now give us a complete picture of how the close-coupling method works. The promise of CCC being a "complete scattering theory," one which is able to describe all possible electron-atom collision processes of practical interest [22], has now been realized. We acknowledge the great contribution to the field made by the development of the ECS theory for the *e-H* collision system [1]. Our strength is the ready generalization to targets more complicated than hydrogen, with success in helium double photoionization and helium one-electron excitation and ionization already established. We now suggest that all these Coulomb three-body problems have been effectively solved.

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