Close-coupling approach to ionization processes

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Abstract. We briefly review recent progress in the field of electron-impact ionization of light atoms concentrating on those theories which attempt to fully solve the underlying scattering problem. Comparison between competing theories and experiment shows up some unexpected discrepancies.

INTRODUCTION

The continued growth in computational power has allowed the emergence of highly computationally intensive techniques for solving ionization problems. The most spectacular example is the work of Rescigno et al. [1] who claimed to have solved the electron-hydrogen ionization problem. They utilise the exterior complex scaling (ECS) method which requires a two-dimensional direct numerical integration out to large distances. Careful usage of the transition of the coordinates from real to complex numbers enables the evaluation of the total wavefunction of the system, without recourse to three-body boundary conditions. Having a numerical wavefunction then allowed the extraction of the scattering information, first via a flux method [2], and then more accurately utilising amplitude formulations [3]. The resulting cross sections are in best overall agreement with available e-H experiments to date.

Another example of substantial progress made possible by modern computational resources is the development of time-dependent techniques [4, 5]. Application to double photoionization of helium [6], a near equivalent of electron-impact ionization of He^+, has shown excellent agreement with experiment [7], as well as other computer-intensive approaches including the hyperspherical R-matrix method [8] and the convergent close-coupling (CCC) theory [7].

It is the latter approach that has been pursued by the present authors. Though initially the close-coupling method [9] was designed for elastic scattering and discrete excitation, a simple extension to ionizing processes is possible [10]. In this paper we consider application of the CCC method to low energy e-H ionization with equal-energy outgoing electrons. We compare with experiment and the ECS theory.

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CCC THEORY

The details of the CCC approach to ionization have been given by Bray and Fursa [10], and subsequently, following the work of Stelbovics [11], slightly modified for the case of equal-energy outgoing electrons [12]. Briefly, we first obtain N square-integrable target states by diagonalising the target Hamiltonian using a Laguerre basis

$$\langle\phi_f^N|H_{\rm T}|\phi_i^N\rangle = \varepsilon_f^N\delta_{fi}. \quad (1)$$

The idea relies on

$$\lim_{N\to\infty}\sum_{n=1}^{N}|\phi_n^N\rangle\langle\phi_n^N| = \sum\!\!\!\!\!\!\int_n |\phi_n\rangle\langle\phi_n| = I. \quad (2)$$

The states are used to expand the total electron-atom wavefunction

$$|\Psi_i^{(+)}\rangle = \mathcal{A}|\psi_i^{(+)}\rangle \approx \mathcal{A}\sum_{n=1}^{N}|\phi_n^N\rangle\langle\phi_n^N|\psi_i^{(+)}\rangle. \quad (3)$$

Close-coupling equations are formed in momentum space for the T matrix at a total energy $E = \varepsilon_i^N + k_i^2/2 = \varepsilon_f^N + k_f^2/2$

$$\begin{aligned}\langle\mathbf{k}_f\phi_f^N|T|\phi_i^N\mathbf{k}_i\rangle &= \langle\mathbf{k}_f\phi_f^N|V|\phi_i^N\mathbf{k}_i\rangle \\ &+ \sum_{n=1}^{N}\int d^3k\frac{\langle\mathbf{k}_f\phi_f^N|V|\phi_n^N\mathbf{k}\rangle\langle\mathbf{k}\phi_n^N|T|\phi_i^N\mathbf{k}_i\rangle}{E+i0-\varepsilon_n^N-k^2/2}.\end{aligned} \quad (4)$$

Upon solution of (4) the discrete amplitudes show step-function behaviour [13]

$$\lim_{N\to\infty}\langle\mathbf{k}_f\phi_f^N|T|\phi_i^N\mathbf{k}_i\rangle = 0, \text{ for } k_f^2/2 < \varepsilon_f^N. \quad (5)$$

The ionization, or (e,2e) amplitude is defined by

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

where $\mathbf{q}_f^{(-)}$ is a continuum eigenstate of H_T with energy $q_f^2/2 = \varepsilon_f^N \leq k_f^2/2$. Following the work of Stelbovics [11] it follows that solving (4) is like taking a finite Fourier expansion of a step-function. Accordingly, at the step the amplitudes converge to half the required values. Hence for $q_f^2/2 = k_f^2/2 = E/2$ we use $2f^N(\mathbf{k}_f,\mathbf{q}_f,\mathbf{k}_i)$.

COMPARISON OF THEORY AND EXPERIMENT

With such an approach the CCC theory yields absolute agreement with all e-He ionization measurements where the two outgoing electrons share the excess energy E equally [see 14, and references therein]. Surprisingly, the situation for the simpler atomic hydrogen target is less clear.

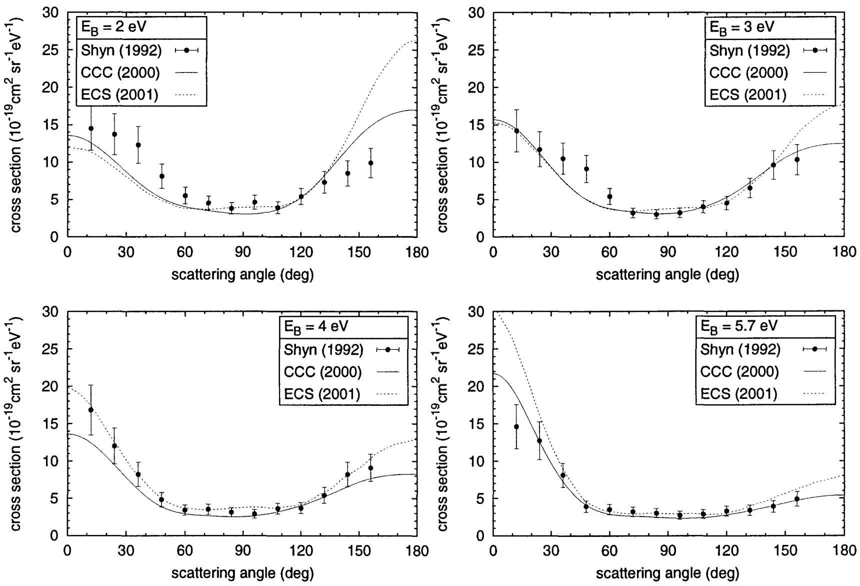

FIGURE 1. Doubly differential cross sections for 25 eV e-H ionization. The experiment is due to Shyn [15], the ECS theory is due to Isaacs et al. [16] and the CCC calculations are due to Bray [17].

We begin by looking at e-H ionization by 25 eV electrons. Here we have absolute doubly differential cross sections (DDCS) measured by Shyn [15]. These describe the angular distribution of the electron ejected with energy E_B. In Fig. 1 we present the experiment and the ECS [16] and CCC [17] theories. We see generally good agreement between the two theories and experiment. The biggest discrepancies between the theories are for the small and large scattering angles, which contribute least to the singly differential, and hence the total ionization, cross section owing to the $\sin(\theta)$ factor in the integration. Of particular interest is the case of $E_A = E_B = 5.7$ eV, for which relative triply differential cross section (TDCS) data are available [18].

The comparison of the 25 eV equal-energy-sharing TDCS is given in Fig. 2. The data are only available for the fixed separation angle θ_{AB} of the two outgoing electrons. For brevity of presentation we take the two smallest available θ_{AB} and two largest. As the data are relative we are free to move it collectively up and down by a single factor. We choose to normalise the experiment at the small θ_{AB}, where ECS and CCC are in good agreement. In doing so we see that at the larger θ_{AB} ECS agrees well with experiment, but the CCC theory is substantially too low. The fact that CCC is much lower than ECS is related to the discrepancy at the small and large scattering angles of the corresponding DDCS of Fig. 1. We are at a loss to explain why CCC would agree with ECS for the smallest θ_{AB}, which yield the smallest cross sections, yet be so different for the largest θ_{AB} with the largest cross sections. Apparently, upon integration over the solid angle $d\Omega_A$ the resultant DDCS only disagrees at the extreme angles.

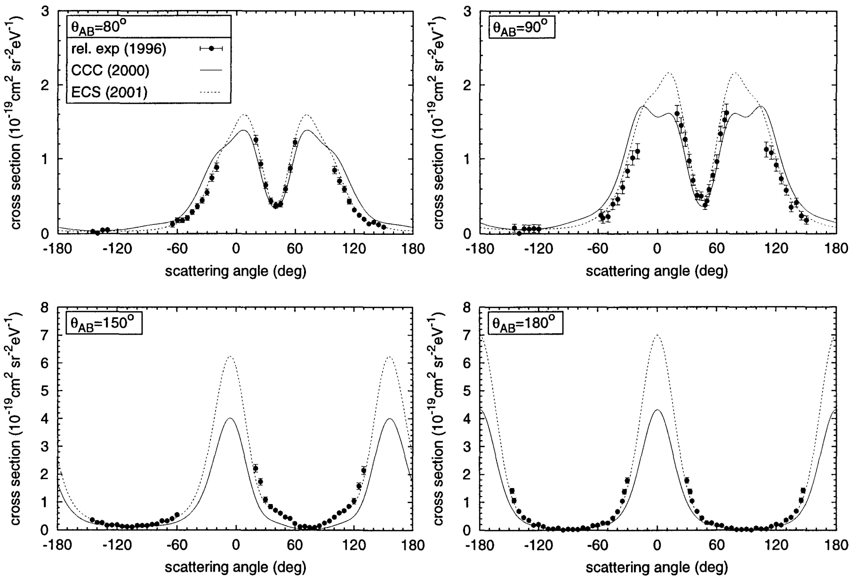

FIGURE 2. Triply differential cross sections for 25 eV e-H ionization with equal-energy outgoing electrons. The experiment is due to Röder et al. [18], the ECS theory is due to Baertschy et al. [3] and the CCC calculations are due to Bray [12].

We next turn our attention to the two energies where absolute TDCS data exist, that of 15.6 and 17.6 eV. In both cases the outgoing electrons have equal energy, $E_A = E_B = 1$ eV and $E_A = E_B = 2$ eV, respectively.

The 15.6 eV data are presented in Fig. 3. The experiment [19] is absolute, but has uncertainty of $\pm 35\%$ in the overall normalisation. However, internormalisation is claimed to be accurate to within 10%. At this energy there are not only fixed θ_{AB} data, but also fixed θ_A data. The latter are particularly important because the cross sections are often large, and also, they allow for internal consistency checks. Whenever the two sets of data intersect they must have a common point, as is generally the case with the 15.6 eV data [12].

From the figure we see remarkable agreement between the two theories and experiment so long as uniform scaling factors are applied. Experiment is a factor of two larger than the ECS theory, and a factor of three larger than the CCC theory. It is particularly surprising that the two theories disagree with each other only in the overall magnitude, with 3 CCC$\approx$ 2 ECS.

A similar situation occurs at 17.6 eV, presented in Fig. 4. Though at this energy the experimental data show some internal inconsistency [12] this is likely to affect the smallest cross sections measured. The geometries presented are similar to those of Fig. 3, and once more show excellent agreement between theory and experiment except for overall normalisation factors. Once again experiment is a factor of two greater than the ECS theory and a factor of three than the CCC theory.

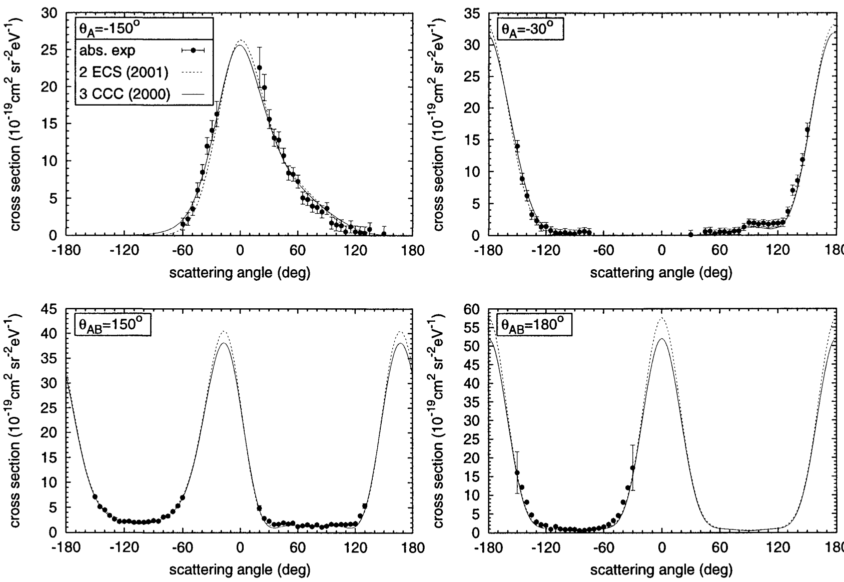

FIGURE 3. Triply differential cross sections for 15.6 eV e-H ionization with equal-energy (1 eV) outgoing electrons. The experiment is due to Röder et al. [19] and references therein, the ECS theory is due to Baertschy et al. [3] and the CCC calculations are due to Bray [12].

CONCLUSIONS

There has been much progress in the last few years in the ability of theory to reproduce measurements of electron-impact ionization fully differential cross sections. In the process some astonishing and unexpected discrepancies between competing theories have been found. The ECS theory yields the most accurate e-H angular distributions at the higher energies suggesting that something is going wrong with the CCC theory for these cases. At the lower energies both theories yield comparable angular distributions which, however, are around a factor of 2/3 apart in overall magnitude. Nevertheless, both theories yield accurate total ionization cross sections.

While we are presently investigating the CCC implementation at the higher energies, we would be grateful for the application of the time-dependent close-coupling (TDCC) theory [4] to this problem. Most importantly, new accurate absolute experimental observations would be very welcome. In particular, measurements of absolute double differential cross sections at the lower energies would be helpful in establishing the required magnitudes.

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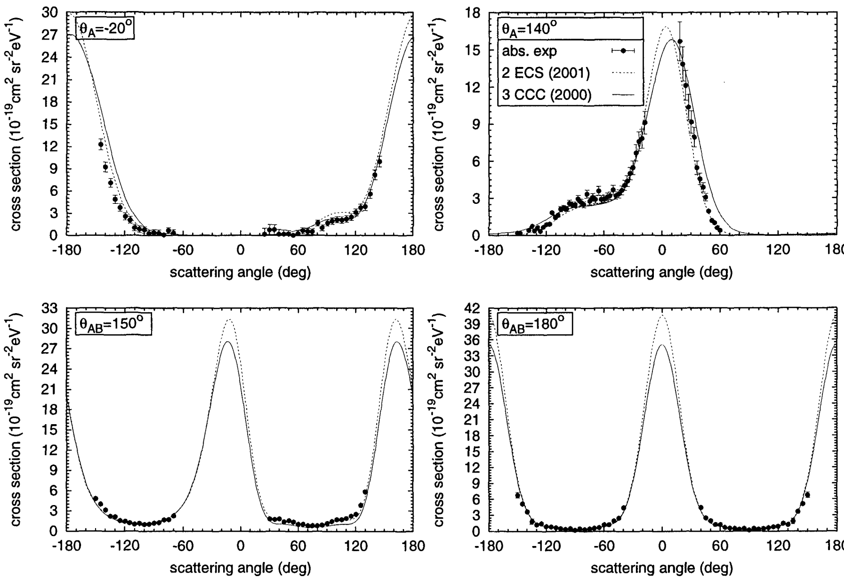

FIGURE 4. Triply differential cross sections for 17.6 eV e-H ionization with equal-energy (2 eV) outgoing electrons. The experiment is due to Röder et al. [19] and references therein, the ECS theory is due to Baertschy et al. [3] and the CCC calculations are due to Bray [12].

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