

Theory of Electron Impact Ionization of Atoms

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Abstract. The theory of electron impact ionization of one- and two-electron atoms has advanced significantly in the past two years. This paper will summarize the progress that the members of our research center have contributed to.

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INTRODUCTION

We wish to report on our recent progress in several aspects of the electron-impact ionization from atoms. First we summarize the considerable developments in the formal aspects of the theory of ionization. Then we turn to a discussion of recent near-threshold numerical studies of electron impact from an atomic hydrogen target. This is followed by some examples of our recent work on electron impact ionization from the two-electron atoms, including helium and calcium.

FORMAL THEORY

The theoretical development we wish to report concerns the three body scattering wave function. We were able to derive the asymptotic form of the three-body scattering wave function [1, 2] and use it to establish integral representations for the scattering amplitude that are free of divergence problems characteristic of previous formulations.

It is well known that the ionization amplitude can be represented in terms of a trial integral which has a structure well suited for practical calculations. Here we show that it can be written in similar form without recourse to external trial quantities which is the requirement of a formally complete scattering theory. First we note that the total scattering wave Φ_i^+ developed from the initial two-fragment state $\Phi^{(i)}$ satisfies (in notation of Ref. [2])

$$(E - H)\Phi_i^{(\text{sc})+}(\mathbf{r}_1, \mathbf{r}_2) = \bar{V}_i\Phi^{(i)}(\mathbf{r}_1, \mathbf{r}_2), \quad (1)$$

where \bar{V}_i is the projectile-target interaction potential, and we separated Φ_i^+ according to $\Phi_i^+ = \Phi^{(i)} + \Phi_i^{(\text{sc})+}$. Therefore, for the ionization amplitude in prior form we can write that

$$T^{(\text{prior})}(\mathbf{k}_1, \mathbf{k}_2) \equiv \langle \Psi_f^- | \bar{V}_i | \Phi^{(i)} \rangle$$

$$\begin{aligned}
&= \langle \Psi_f^- | E - \vec{H} | \Phi_i^{(\text{sc})+} \rangle \\
&= \langle \Psi_f^- | \overleftarrow{H} - E | \Phi_i^{(\text{sc})+} \rangle + \langle \Psi_f^- | E - \vec{H} | \Phi_i^{(\text{sc})+} \rangle \\
&= \langle \Psi_f^- | \overleftarrow{H}_0 - \vec{H}_0 | \Phi_i^{(\text{sc})+} \rangle \equiv T^{(a)}(\mathbf{k}_1, \mathbf{k}_2), \tag{2}
\end{aligned}$$

where a left (right) arrow on the differential Hamiltonian operator indicates that it acts on the *bra* (*ket*) state. This is a new surface-integral form for the ionization amplitude.

Next we note that the total scattering wave Ψ_f^+ developed from the final three-fragment state $\Psi^{(f)-}$ satisfies

$$(E - H_0)\Psi_f^-(\mathbf{r}_1, \mathbf{r}_2) = V\Psi_f^-(\mathbf{r}_1, \mathbf{r}_2). \tag{3}$$

In addition, we note that

$$(E - H_0)\Phi^{(i)}(\mathbf{r}_1, \mathbf{r}_2) = V_i\Phi^{(i)}(\mathbf{r}_1, \mathbf{r}_2). \tag{4}$$

In the light of Eqs. (3) and (4) we get

$$\begin{aligned}
T^{(\text{prior})}(\mathbf{k}_1, \mathbf{k}_2) &\equiv \langle \Psi_f^- | V - V_i | \Phi^{(i)} \rangle \\
&= \langle \Psi_f^- | E - \overleftarrow{H}_0 - (E - \vec{H}_0) | \Phi^{(i)} \rangle \\
&= -\langle \Psi_f^- | \overleftarrow{H}_0 - \vec{H}_0 | \Phi^{(i)} \rangle \equiv T^{(b)}(\mathbf{k}_1, \mathbf{k}_2). \tag{5}
\end{aligned}$$

If we separate the unscattered and scattered parts of wave function Ψ_f^- according to $\Psi_f^- = \Psi^{(f)-} + \Psi_f^{(\text{sc})-}$ then we can also introduce

$$T^{(c)}(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi^{(f)-} | \overleftarrow{H}_0 - \vec{H}_0 | \Phi_i^+ \rangle, \tag{6}$$

$$T^{(d)}(\mathbf{k}_1, \mathbf{k}_2) = -\langle \Psi_f^{(\text{sc})-} | \overleftarrow{H}_0 - \vec{H}_0 | \Phi_i^+ \rangle. \tag{7}$$

Forms $T^{(a)}$, $T^{(b)}$, $T^{(c)}$ and $T^{(d)}$ are convenient for numerical calculations as the result depends only on the asymptotic behaviour of the scattered wave functions. The asymptotic forms of Φ_i^+ and Ψ_f^- have been given in [2, 3].

We emphasize the importance of the new form of the ionization amplitude $T^{(c)}$ given by Eq. (6), from the point of view of the general scattering theory. Eq. (6) leads to a well-defined conventional volume-integral form of the ionization amplitude in terms of the total three-body scattering wave function Φ_i^+ , being developed from the initial two-fragment channel $\Phi^{(i)}$. In the stationary-state scattering theory the *post* form of the breakup amplitude is defined by

$$T^{(\text{post})}(\mathbf{k}_1, \mathbf{k}_2) = \langle \mathbf{k}_1, \mathbf{k}_2 | V | \Phi_i^+ \rangle, \tag{8}$$

where $\langle \mathbf{r}_1, \mathbf{r}_2 | \mathbf{k}_1, \mathbf{k}_2 \rangle = e^{i\mathbf{k}_1 \cdot \mathbf{r}_1 + i\mathbf{k}_2 \cdot \mathbf{r}_2}$ is the undistorted three-body plane wave. However, this form is valid only when interaction between particles is short-ranged. The commonly accepted stationary theory of scattering fails to define the same for long-range

interactions. At the same time from the c -form of the ionization amplitude we get

$$T^{(c)}(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi^{(f)-} | \overleftarrow{H}_0 + V - E - \overrightarrow{H}_0 - V + E | \Phi_i^+ \rangle \quad (9)$$

$$= \langle \Psi^{(f)-} | \overleftarrow{H}_0 + V - E | \Phi_i^+ \rangle. \quad (10)$$

This allows us to introduce

$$T^{(\text{post})}(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi^{(f)-} | \overleftarrow{H} - E | \Phi_i^+ \rangle. \quad (11)$$

Eq. (11) takes the form of Eq. (8) when the full interaction is short-ranged. Thus, Eq. (11) extends the definition of the *post*-form of the breakup amplitude to long-range potentials including the Coulomb interaction.

e -H NUMERICAL SOLUTION

We have recently made important progress in the direct numerical solution of electron-hydrogen ionizing collisions. Our method, propagating exterior complex scaling (PECS), combines exterior complex scaling [4] with a highly efficient numerical technique to obtain solutions to the full time-independent Schrödinger equation for e -H collisions in coordinate space [5]. This method was used to calculate accurate total and differential ionization cross sections at energies to within 0.01 a.u. of threshold [6], which provided convincing fully-quantal *ab initio* support for the Wannier [7] and related ionization threshold laws.

Our TICS results, shown in Fig. 1a, are scaled by $E^{1.127}$ to give emphasis to the low-energy results and highlight the improvement in precision over older convergent close-coupling (CCC) and R-matrix with pseudo states (RMPS) calculations. The estimated

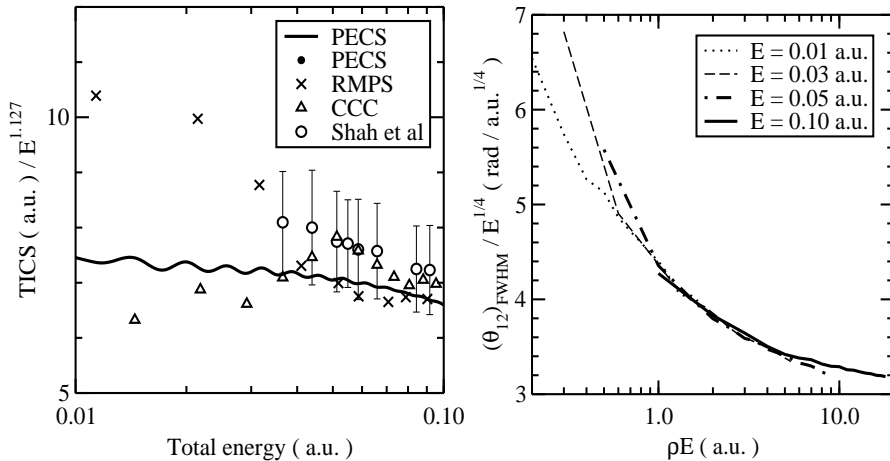


FIGURE 1. PECS calculations for e -H ionizing collisions near threshold. (a) Total ionization cross section (TICS) scaled by $E^{1.127}$, and (b) Full-width-half-maximum (FWHM) of the angle between ejected electrons scaled by $E^{1/4}$. CCC and RMPS calculations from [8] and measurements from [9].

standard errors of our calculations range from $\pm 3\%$ at 0.01 a.u. to $\pm 1\%$ at 0.10 a.u.. We applied non-linear curve-fitting techniques to these results and calculated the threshold power law to be $\sigma \propto E^{1.122 \pm 0.015}$, and the $L=0$ triplet power law to be $\sigma \propto E^{3.36 \pm 0.02}$, in accord with Wannier theory. Analysis of the single-differential cross sections showed a deviation of approximately 4% from energy-independence of the outgoing electrons, and we estimated the spin-asymmetry to approach the limiting value $A_s = 0.54 \pm 0.01$, as threshold is approached.

Wannier theory predicts that the full-width-half-maximum of the angular separation of the outgoing electrons is given by $(\theta_{12})_{\text{FWHM}} = \alpha E^{1/4}$, but there is disagreement on the value of α (see [6] and references therein). An important outcome from our near-threshold calculations was that we were able to establish that $\alpha = 3.0 \pm 0.2$ (in atomic units), which is in the middle of the range of values predicted by semi-classical methods (2.66 to 3.55). Figure 1b shows our results for $(\theta_{12})_{\text{FWHM}}$ as a function of ρE , where ρ is the hyperradius where the cross sections are calculated, and scaled by $E^{1/4}$. The results have not fully converged with respect to increasing ρE , but give convincing evidence of a $E^{1/4}$ relationship, and an estimate for α was made from an asymptotic extrapolation. For the energies considered here, we estimate that full convergence of α would require a calculation grid extending to $\rho \approx 2000 a_0$, well beyond the 180 a_0 used for these calculations, and well beyond our present computational resources.

TWO-ELECTRON TARGETS

Two electron-atoms present a more challenging vista for theorists. As we have indicated, the atomic three-body problem is largely solved numerically. The treatment of the four-body problem requires further simplifying assumptions to reduce it to a form suitable for practical solution. In this section we illustrate how the CCC method can be applied to the problem of electron impact ionization of two-electron atoms. We will consider the case of equal-energy sharing for the ionizing collisions as the CCC theory for this case is has received detailed analysis and is now well understood. We will contrast recent studies for Helium where there are numerous investigations with Calcium which has been far less studied.

Helium

Helium is the ideal target for the experimentalists with by far the most data available than for any other target. What is particularly helpful is that much of these data are on the absolute scale allowing for quantitative comparison with theory. From the theoretical perspective while helium is not as ideal as atomic hydrogen, it turns out that by far the most dominant transitions involve the excitation of only one electron. This allows the treatment of the e-He problem as predominantly a three-body problem. The details of the CCC theory for e-He ionization have been given some time ago [10, 11]. The case of calculating electron-helium ionization with equal-energy outgoing electrons has been studied in great detail very recently [12]. Here, in Fig. 2, we show a representative ex-

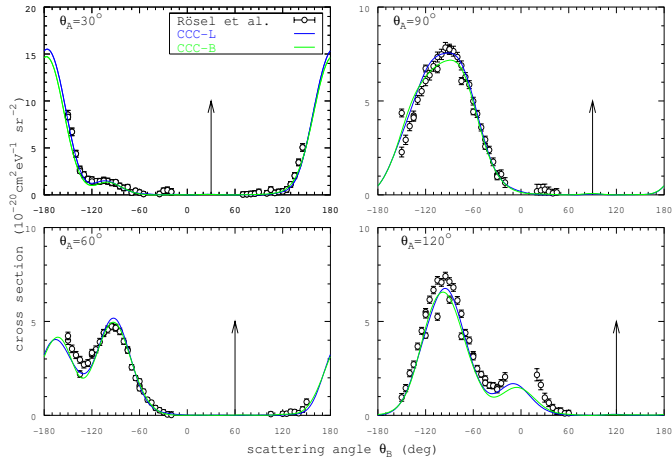


FIGURE 2. 26.6 eV electron-impact ionization of He with two 1 eV outgoing electrons. The theory and experiment are from Refs. [12] and [13], respectively.

ample of the excellent quantitative agreement between the CCC theory and experiment.

Calcium

We treat the calcium target as a two-electron atom with an inert Hartree-Fock core [15]. This way the theory developed for ionization of helium is equally applicable to

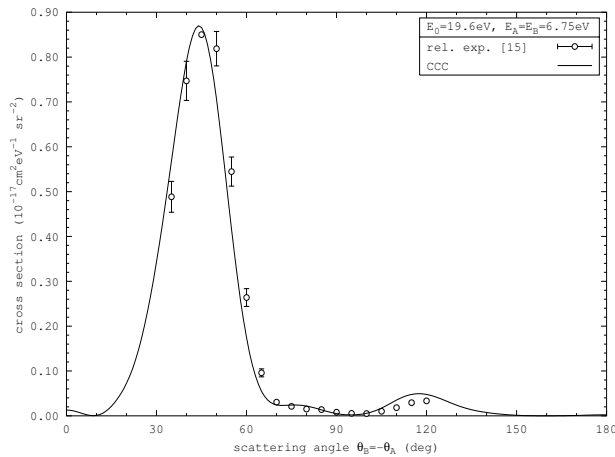


FIGURE 3. 19.6 eV e-Ca ionisation with two 6.75 eV outgoing electrons. The relative measurements of Murray and Cvejanovic [14] have been normalised to the present CCC calculation.

calcium. In Fig. 3 we give the results of a previously unpublished CCC calculation and compare with existing measurements [14]. While agreement looks excellent for the case considered, as the threshold is approached substantial discrepancies arise. This is currently under investigation.

CONCLUDING REMARKS

The review has presented snapshots of our most recent work. The progress in our understanding of the theory that has led to the closing of some formal long-standing problems is satisfying. Our next work will be to fully implement the new approach as a viable calculational framework. Similarly for electron-hydrogen scattering, following on from the earlier successes of CCC and ECS methods the PECS calculations have been able to reveal details of the near threshold ionization behaviour and to confirm the predictions of the semiclassical models. Our continuing focus will be now to model the two-electron systems in detail. While complete solution of the full Schrödinger equation for a three-electron system is still not possible, it should be within the next few years. One line of our research will be to extend the PECS method to this system. From a practical point of view the CCC formulation of ionization is well advanced and it now remains to investigate more fully application to quasi two-electron targets.

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