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Modelling of three-body effects in a double continuum

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Abstract: Theoretical calculations of double ionisation by electron or photon impact require a final state wavefunction which takes account both of the Coulomb repulsion between the pair of free electrons and of their interaction with the residual ionic core. It is desirable that this should be separable so as to facilitate the introduction of electron-pair correlations in the initial state wavefunction for the collision system. We propose a method for calculating coupled classical trajectories for the free electrons and deducing from these potentials from which their quantum mechanical wavefunctions may be obtained. Test calculations are reported for electron impact single ionisation of hydrogen.

1. INTRODUCTION

Two major problems are encountered when making calculations for $(e,2e)$ processes. One is how to take account of the Coulomb repulsion between a pair of final state free electrons. This also exists in the case of double ionisation by photon impact $(\gamma,2e)$ or by high energy electron impact $(e,3e)$. It is resolved by explicit introduction of a Coulomb correlation factor in the wavefunction of Brauner, Briggs and Klar [1] and in subsequent developments by Altick and collaborators [2]. The other difficulty is that of coupling between competing reaction channels, which also results from Coulomb forces between electron pairs. This must obviously be taken into account when obtaining the final state of the target system in the cases of ionisation-with-excitation and of autoionisation. A similar problem exists for the complete collision wavefunction describing the projectile and the target system together. Curran and Walters [3] deal with it by using close-coupling wavefunctions, describing the elastic and two of the excitation channels exactly and other channels through the use of pseudostates. Joachain and collaborators [4] employ Born 2 or Eikonal Born series models including some channels explicitly and compensating for the rest by a closure approximation. Distorted wave Born models [5] only take account of the elastic channel. Because the Brauner et al wavefunction is an asymptotic solution of the full three-body Coulomb Hamiltonian it implicitly contains some coupling to elastic and excitation channels. Pseudostates or closure can introduce

approximate coupling to double continuum channels. All these models can give good results in appropriate circumstances, but none fully includes the physics of the problem.

The Brauner et al wavefunction, although a considerable advance, is strictly correct only in the asymptotic region. It provides a good target final state for double ionisation of helium by photons [6] or electrons [7]. But its extension to more complex targets is delicate. The problem of introducing the interaction of two free electrons with a many-electron ionic core can in principle be solved by replacing the Coulomb waves for the individual electrons by wavefunctions calculated in the static field of the core. But the Coulomb correlation term (involving the relative directions of the two free electrons) is not, without further approximation, easily expressed in partial wave form. Nevertheless, it is inevitable that the target initial state and the ionic core be described by multiconfiguration Hartree-Fock type wavefunctions. Formulation of the collision problem, without the use of partial wave expansions and the support of Racah algebra, then becomes arduous.

In high energy electron impact, both for double ionisation and for ionisation-with-excitation, the target initial and final states must be described by wavefunctions taking proper account of electron-pair correlations. Plane waves may be used to describe the projectile. At lower energies it is certainly necessary to take account of the coupling between competing reaction channels. A possible approach would be to combine the representation of the complete collisional system by a close-coupling wavefunction and an approximate description of the final state, including electron-pair correlations for the target system. This is envisagable only if the final state wavefunction is simple enough. In previous work [8] we proposed a parametric potential method for treating coupled channels based on different excited states of the residual ion. Here we examine a classical trajectory method for dealing with the double continuum.

2. COULOMB CORRELATIONS IN THE DOUBLE CONTINUUM

As an alternative to introducing an explicit coulomb correlation factor, we wish to develop a model in which the wavefunction for the double continuum is separable. Each electron is to be described by a distorted wave, calculated using a potential obtained from classical trajectories and specific to particular kinematics (energies and relative angle). Such an approach was adopted by Jones et al [9] and by Robaux et al [10], using constant speed linear trajectories; as compared to a standard Distorted Wave Born (D.W.B.A.) model this implies modifying the potentials used for the calculation of the final state free electron wavefunctions. For e^- - He collisions this implies adding a term with λ/r behaviour to standard distortion potentials; such a procedure gives fairly good cross sections. But for e^- - H collisions it comes down to using a product of coulomb waves with an effective charge prescription similar to that of Schultz [11] and the results are unsatisfactory. Very recently, Jones et al [12] have developed a D.W.B.A. model in which a correlation factor, related to the Brauner-Briggs-Klar (B.B.K.) wavefunction, is introduced in the final state. This apparently gives excellent results for e^- - H collisions. Whelan et al [13] have applied a correlation correction to a standard D.W.B.A. model and get very good shape comparison with the experiments of Rösel et al [14] for e^- - He collisions. We note that the B.B.K. theory can have problems for angular distributions at low incident electron energy in e^- - H collisions under coplanar symmetric energy-sharing conditions [15] and neither the B.B.K. nor the D.W.B.A

theories give a reliable absolute value for cross sections below about 100 eV. The success of the theories of Jones et al and of Whelan et al results from the fact that they combine the B.B.K. and D.W.B.A. approaches. The model described in the present paper is an alternative approach to theirs, with the same object.

We have developed methods for the calculation of coupled classical trajectories, taking account of their curvature and of the change of energy due to the coulomb repulsive force. The system of two electrons moving in the field of a nucleus can become chaotic if their trajectories are taken out to infinite distance [16]. Thus we integrate to a finite but very large radius (at least $8000 a_0$). Furthermore, the trajectories have closest approach distances related to the energetics in a particular kinematical situation. This forces us to make an analytical continuation of our perturbation potentials down to zero radius, avoiding singular behaviour at the origin. This is one of the difficulties in our method, but is essentially a numerical one because the perturbation is insignificant compared to the nuclear attraction at small radius. A much more serious problem is the conceptual one related to the wavelength associated with the energy of the colliding electron. For slow electrons this is very much larger than the dimensions of the target, so one could naively suppose that a classical treatment has no sense. However, when the colliding electron approaches the nucleus, it is accelerated by falling into a deep attractive potential well and so its wavelength decreases. The adiabatic approximation used to derive polarisation potentials fails at small radius (when the colliding electron penetrates well inside the target electron cloud) precisely because of this acceleration.

Consider the general problem of two free electrons interacting with each other in the field of an ion, represented by the central potential V . Their coordinates are $\mathbf{r}_1$ and $\mathbf{r}_2$ and their separation is $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1$. They are detected at angles θ_a and $-\theta_b$, with energies E_a and E_b respectively. We consider their motion to be confined to the plane defined by the ion and the detectors, so $\mathbf{r}_i = \{r_i, \theta_i\}$ and $\mathbf{r} = \{r, \theta\}$ with $\theta = \theta_2 - \theta_1$. We calculate their classical trajectories as a function of time, taking into account the effect of their mutual repulsion on their individual energies $E_i(t)$ and angular momenta $l_i(t)$. For electron 1 we have:

$$T_1(t) = \frac{m}{2} \left[\left(\frac{dr_1}{dt} \right)^2 + r_1^2 \left(\frac{d\theta_1}{dt} \right)^2 \right] = E_1(r_1) - V(r_1) \quad (1)$$

$$l_1(t) = m r_1^2 \left(\frac{d\theta_1}{dt} \right) = l_1(0) - \int_0^t \frac{r_1(t') r_2(t') \sin \theta(t')}{r^3(t')} dt' \quad (2)$$

$$E_1(t) = E_1(0) + \int_0^t \frac{1}{r^3(t')} \left\{ [r_1(t') - r_2(t') \cos \theta(t')] \frac{dr_1}{dt'} - r_1(t') r_2(t') \sin \theta(t') \frac{d\theta_1}{dt'} \right\} dt' \quad (3)$$

A similar set of equations for electron 2 are obtained by permuting the indexes 1 and 2 and changing the sign before $\sin \theta$. The factors in the integrals come from the radial and angular components of the repulsive force. The $T_i(t)$ are the kinetic energies on the trajectories, so the effective potential for the motion of electron i is simply $V_i(t) = E_i(\infty) - T_i(t)$. This gives us the potential for quantum mechanical calculation of its wavefunction.

It is obviously impossible to define initial conditions for $r_i(0)$, $l_i(0)$ and $E_i(0)$ in a non-arbitrary way. However, the derivation in quantum mechanics of scattering amplitudes, for single ionisation by Peterkop [17] and by Rudge and Seaton [18], and for double ionisation by Tweed [19], imply an asymptotic condition on the momenta k_a , k_b and $k = k_b - k_a$:

$$r_1 = \tau k_a$$

$$r_2 = \tau k_b$$

$$r = \tau k$$

where τ is a constant. Imposing this condition in the calculation of our classical trajectories, we obtain from equation (2):

$$l_1(t) \underset{t \rightarrow \infty}{\sim} - \frac{k_a k_b \sin(\theta_a + \theta_b)}{v k^2} \ln(r) + \text{constant} \quad (4)$$

At sufficiently large radius and time, $r_1 d\theta_1/dt$ becomes small in comparison with dr_1/dt . So from equation (3) we obtain:

$$E_1(t) \underset{t \rightarrow \infty}{\sim} - \frac{k_a - k_b \cos(\theta_a + \theta_b)}{k} r_1^{-1} + \text{constant} \quad (5)$$

Equation (4) corresponds to the result obtained by Gaillitis [20]: the Coulomb repulsion between electrons causes their individual angular momenta to increase logarithmically with radius, but the total angular momentum of the electron pair is conserved and is finite. The choice of constant is arbitrary; for the results presented here it was taken to be zero. In equation (5) the constant is E_a , so as to get the required energy at the detector, assuming this is at sufficiently large radius for the interaction potentials to be negligible.

We use the asymptotic forms from equations (4) and (5) to provide initial estimates for l_i and E_i . These are injected into equations (1) and (2), which are then solved simultaneously for r_i and θ_i by numerical integration. The initial values of θ_1 and θ_2 are adjusted so as to get the required asymptotic angles, θ_a and $-\theta_b$ respectively. Those of r_1 and r_2 are essentially closest approach distances imposed by the repulsive force between the electron pair becoming too strong. The l_i and E_i are then recalculated from the trajectories, with initial values chosen so as to get the required asymptotic result. The procedure is then continued iteratively; convergence is rapid.

From these trajectory calculations we obtain the $V_i(t)$ and $r_i(t)$ on the same time grid. Thus we can express V_i as a function of r_i on any radial grid by means of cubic spline interpolation. We finally obtain a potential $V_a(r_1)$ for the electron which is detected at angle θ_a with energy E_a and a potential $V_b(r_2)$ for the electron which is detected at angle $-\theta_b$ with energy E_b .

3. ELECTRON-IMPACT SINGLE IONISATION OF HYDROGEN

We have used electron impact single ionisation of hydrogen as a test case. The triple differential cross section is given by:

$$\frac{d^3 \sigma}{d\Omega_a d\Omega_b dE} = (2\pi)^4 \frac{k_a k_b}{k_0} \frac{1}{4} \sum_S (2S + 1) |f_s(k_a, k_b)|^2 \quad (6)$$

with

$$f_s(\mathbf{k}_a, \mathbf{k}_b) = \int d\mathbf{r}_1 \int d\mathbf{r}_2 \Psi_s^*(\mathbf{r}_1, \mathbf{r}_2) \{1/r_1 - 1/r_2 - V_s(\mathbf{r}_2)\} \\ \times [\Phi_a(\mathbf{r}_1) \Phi_b(\mathbf{r}_2) + (-1)^S \Phi_a(\mathbf{r}_2) \Phi_b(\mathbf{r}_1)] \quad (7)$$

where Φ_a and Φ_b are distorted waves calculated using potentials V_a and V_b corresponding to the particular final momenta $\mathbf{k}_a$ and $\mathbf{k}_b$. Ψ_s describes the collisional system for total spin S , with an incident electron of momentum $\mathbf{k}_0$. It is obtained by the distorted wave polarised orbital method of McDowell et al [21] and takes into account, through the distortion potential V_s , of elastic scattering of the colliding electron. For the sake of comparison, we also make calculations using the same initial state Ψ_s but with Coulomb waves for Φ_a and Φ_b . These correspond essentially a standard distorted wave Born model. We refer to them as "DWB" in figure legends whereas the calculations with classical potentials are referred to as "DWB+PCI".

Cross sections are plotted as a function of θ_b for incident energy $E_0 = 54.4$ eV in figure 1, for $E_b = 5$ eV and $\theta_a = 4^\circ$. We compare the results of our two calculations to the experimental data of Schlemmer et al [22] as normalised by Brauner et al [15]. The classical potentials give a good result, seemingly overestimating slightly the magnitude of the binary lobe.

Figure 1: T.D.C.S in $a_0^2/\text{sr}^2/2Ry$ for $E_0 = 54.4$ eV, $E_b = 5$ eV and $\theta_a = 4^\circ$

In figure 2 we compare our two calculations to those of Curran and Walters [3]. The experimental points are as in figure 1, but now normalised to the latter calculation. This gives a different picture: our classical potential calculations give a good description of the binary lobe but a poor description of the recoil lobe. A similar comparison is made in figure 3 for the same energies and $\theta_a = 10^\circ$. Since the calculations of Curran and Walters take account of the coupling between excitation channels they correctly describe the recoil lobe, whereas our Ψ_s is too simple an approximation. However, as they have no post-collision interaction in their final state, we do better than them for the binary lobe.

Figure 3: T.D.C.S in $a_0^2/\text{sr}^2/2Ry$ for $E_0 = 54.4$ eV, $E_b = 5$ eV and $\theta_a = 10^\circ$

It is also interesting to consider the case of an incident energy of 27.2 eV with $E_a = E_b = 6.8$ eV. We compare our two calculations to the data of Brauner et al [15], plotting the cross section as a function of $\theta_a + \theta_b$, for $\theta_a = 15^\circ$ in figure 4 and for $\theta_a = 30^\circ$ in figure 5. Our results are acceptable except for the fact that the cross section does not drop to zero for a relative angle of 0° . This is not surprising when ϕ_a and ϕ_b are Coulomb waves, but the introduction of a post-collision interaction should prevent two electrons of the same energy from having the same direction. We are able to reproduce the double peak form of the experiment because our initial state Ψ_0 gives a good description of the scattering of the projectile by the nucleus. However, the peaks are not in the right place. Our classical potentials are not giving a strong enough effect at small relative angle.

The problem is the choice of a zero constant in the asymptotic condition for the angular momentum (equation 4). As a result, the angular momenta at small radius are not large enough to permit a rapid separation of the electrons and the trajectories have to be started too far out. We are now testing a computer code using inward integration of the coupled classical equations of motion in Cartesian coordinates $\{x_i, y_i\}$. Boundary conditions on derivatives dx_i/dt and

dy_i/dt at large time are affected only slightly by the $\ln(r)$ factor in equation 4, and negligibly by the constant.

Figure 4: T.D.C.S in $a_0^2/\text{sr}^2/2Ry$ for $E_0 = 27.2$ eV, $E_a = E_b = 6.8$ eV and $\theta_a = 15^\circ$ as a function of $\theta_a + \theta_b$.

Figure 5: As figure 4 for $\theta_a = 30^\circ$; experimental data $\times 2$.

In figure 6 we show some typical results of these computations for the case of two free electrons of energy 6.8 eV. We plot the r -multiplied correction to the central residual ion potential: $r_1 (V_a(r_1) - V(r_1))$. It may be seen that as the angle between the directions of the electrons decreases, the potential becomes increasingly important at small to medium radius. This is precisely what we wish to obtain,

Figure 6: Dependence on relative angle of the classical correction potentials, for two electrons of energy 6.8 eV

since the high peak in the potential at small radius will push the wavefunctions of the final state free electrons out to larger radius. This will reduce their overlap with the wavefunction of the bound target electron in the initial state and so give smaller radial integrals. The larger is the peak in the potential, the smaller will be the cross section. So at very small relative angles of the electrons we should obtain a near-zero cross section.

4. CONCLUSIONS

Our present method shows promise but needs improvement, especially when the energies of the two free electrons are both small. The problem comes from the correct definition of asymptotic boundary conditions, and in current work we are systematically extending the final time and radii out until we can be sure of the uniqueness of the classical solutions. Use of Cartesian coordinates simplifies the numerical aspects and makes it possible to bring the trajectories in to a very small radius. So we should be able to introduce the Coulomb repulsion between electrons at short distance. Extension to many-electron systems is simple. There remains the problem of short range correlations. This affects somewhat the potentials which we derive from our classical trajectories, but it seems probable that the real problem is with the wavefunction of the colliding electron in the initial state. We are currently introducing potentials obtained by the method of Perdew and Zunger [23] and Bachelet et al [24].

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REFERENCES

- [1] Brauner M. Briggs J.S. and Klar H., *J. Phys. B* **22** (1989) 2265
- [2] Franz A. and Altick P.L., *J. Phys. B* **25** (1992) 1577
Ming Tong and Altick P.L., *J. Phys. B* **25** (1992) 741
- [3] Curran E.P. and Walters H.R.J., *J. Phys. B* **20** (1987) 337
- [4] Byron F.W. Jr., Joachain C.J. and Piraux B., *J. Phys. B* **13** (1980) L673
Potvliege R.M., Vucic S. and Joachain C.J., *J. Phys. B* **20** (1987) 4883
- [5] McCarthy I.E., *Z. Phys. D* **4** (1992) 287
- [6] Maulbetsch F. and Briggs J.S., *J. Phys. B* **26** (1993) 1679
- [7] Joulakien B., Dal Cappello C. and Brauner M., *J. Phys. B* **25** (1992) 2863
- [8] Robaux O., Tweed R.J. and Langlois J., *J. Phys. B* **24** (1991) 4567
- [9] Jones S., Madison D.H. and Srivastava K.M., *J. Phys. B* **25** (1992) 1899
- [10] Robaux O., Tweed R.J. and Langlois J., *Z. Phys. D* **23** (1992) 331
- [11] Schulz M., *J. Phys. B* **6** (1973) 2580
- [12] Jones S., Madison D.H., Franz A. and Altick P.L., *Phys. Rev. A* **48** (1993) R22
- [13] Whelan C.T., Allan R.J., Walters H.R.J. and Zhang X. in "(e,2e) & related processes" ed. C.T. Whelan, H.R.J. Walters, A. Lahmam-Bennani and H. Ehrhardt (Kluwer Academic Publishers, 1993)
- [14] Rösel T., Dupré C., Roder J., Duguet A., Jung K., Lahmam-Bennani A. and Ehrhardt H., *J. Phys. B* **24** (1991) 3059
- [15] Brauner M., Briggs J.S., Klar H., Broad T., Rösel T., Jung K. and Ehrhardt H., *J. Phys. B* **24** (1991) 657
- [16] Yan Gu and Jian-Min Yuan, *Phys. Rev. A* **47** (1993) R2442
- [17] Peterkop R., *Opt. Spectros.* **13** (1962) 87
- [18] Rudge M.R.H. and Seaton M.J., *Proc. R. Soc A* **283** (1965) 262
- [19] Tweed R.J., *J. Phys. B* **6** (1973) 259
- [20] Gailitis M., *J. Phys. B* **23** (1990) 85
- [21] McDowell M.R.C., Morgan L. and Myerscough V.P., *Comput. Phys. Commun.* **7** (1974) 38
- [22] Schlemmer P., Rösel T., Jung K. and Ehrhardt H., *J. Phys. B* **22** (1989) 2179
- [23] Perdew J.P. and Zunger A. *Phys. Rev. B* **23** (1981) 5048
- [24] Bachelet G.B., Hamann D.R. and Schlüter M. *Phys. Rev. B* **26** (1982) 4199