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LETTER TO THE EDITOR

Electron–ion correlation effects in ion–atom single ionization

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Abstract. We study the effect of electron–ion correlation in single ionization processes of atoms by ion impact. We present a distorted wave model where the final state is represented by a correlated function solution of a non-separable three-body continuum Hamiltonian, that includes electron–ion correlation as coupling terms of the wave equation. A comparison of the electronic differential cross sections computed with this model with other theories and experimental data reveals that the influence of the electron–ion correlation is more significant for low energy emitted electrons.

Correlation is a well-known concept in a wide range of physical processes. It has been extensively discussed in many areas such as high energy, nuclear and solid state physics. In the atomic physics literature, correlation is generally associated with the long range Coulomb interaction between electrons. The ionization of atomic targets by collision with an electronic projectile is also a good example of such a kind of correlation. Here we deal with a more general definition of correlation, that can be understood as a lack of internal independence of the components of a system. This dynamical coupling between the motion of all particles plays a significant role in all many-body problems. The simplest system in which this correlation can be studied is the three-body Coulomb problem (3BCP). The ion–atom ionizing collisions can be modelled as a 3BCP with suitable effective charges and offers the opportunity to investigate the full continuum problem that describes the final state of the collision, as well as the bound-continuum state of the initial channel, where the ionic projectile interacts with the target atom. The details of these states can be revealed by the computation of cross sections of electronic emission in these collisions.

The theoretical models developed through the years to uncover the details of these processes can be classified according to the impact energy of the projectile. In the low-to-intermediate energy regime we found close-coupling methods based on a molecular approach [1,2], in the intermediate regime the classical trajectory Monte Carlo has been widely used [3], while in the intermediate-to-high energy regime, the distorted wave method can be applied [4, 5]. Furthermore, there are methods that can be applied throughout the complete range of impact energies [6–8]. The distorted wave method offers several advantages. First, it includes the correct asymptotic conditions of the wavefunctions due to the long-range behaviour of the Coulomb interaction between the particles [9]. In addition, the relative motion between the heavy partners can be treated under an impact parameter approximation and does not influence double differential cross sections (DDCSs) in the energy and scattering angle of the

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emitted electrons [10]. Then, in a three-body process such as the ionization of hydrogenic atoms, we can take into account only the electron–ion interactions. In this method, the cross sections are obtained from the transition matrix in prior form $T_{if} = \langle \Psi_f^+ | V_i | \Psi_i \rangle$ that can be calculated analytically for hydrogenic atoms. The state Ψ_i (Ψ_f^+) is a solution of the Schrödinger equation for the initial (final) channels. Different initial states have been proposed, ranging from the eikonal state (EIS) where the Coulomb interaction between the bound electron and the incoming projectile is treated only asymptotically [11], to the impulse approximation (IA), where the wavefunction is a convolution over the velocity distribution of the bound electron [12]. However, all these models share the same final state: a product of two solutions of the two-body Coulomb (2BC) problem, one for each of the relevant interactions: the electron–target and the electron–projectile pairs [13]. This state is commonly known as the C2 wavefunction and can be written as:

$$\Psi_{C2} = N_{C2} \Phi_{PW} \prod_{l=T,P} {}_1F_1 \left[\mathrm{i}a_l, 1, -\mathrm{i}k_l \xi_l \right]. \tag{1}$$

The Ψ_{C2} represents the independent motion of the electron in the field of two rapidly moving ions. The plane waves $\Phi_{PW} = \exp\left(\boldsymbol{K}_f \cdot \boldsymbol{R}_T + \boldsymbol{k}_T \cdot \boldsymbol{r}_T\right)$ describe the asymptotic motion of the particles without interaction, while the product of confluent hypergeometric functions fully introduces the long-range properties of the Coulomb potential. The function Ψ_{C2} has been written in the Jacobi pair $\{\boldsymbol{r}_T, \boldsymbol{R}_T\}$ as defined in [14], while the coordinates $\xi_j = r_j + \boldsymbol{k}_j \cdot \boldsymbol{r}_j$ with $j = T$ or P are the well-known parabolic coordinates for incoming boundary conditions [15]. The normalization factor N_{C2} in equation (1) is chosen in such a way that the wavefunction has asymptotic unitary flux.

The wavefunction Ψ_{C2} can be obtained by approximating the Schrödinger equation for the 3BCP in parabolic coordinates by a totally separable partial differential equation [15,16]. Then, the non-orthogonal kinetic energy that contains second-order cross derivatives in the spatial variables, is neglected on the basis that it falls as the inverse of the square for large interparticle distances [15], an order of magnitude faster than the Coulomb potential. In some sense, the Ψ_{C2} function includes a trivial correlation since the relative coordinates and momenta of the particles are linked [14]. This can be understood as a kinematic coupling, a property of all many-particle systems. However, the Ψ_{C2} state does not introduce any *dynamical* correlation, since the non-orthogonal kinetic energy is neglected at the wavefunction level and is considered as the perturbation in the calculation of the transition matrices. Moreover, since the Ψ_{C2} is a separable function, it implicitly includes the azimuthal symmetry of the 2BC functions, neglecting the influence of the third particle. Since the state is separable, the normalization factor holds the same property, that is, $N_{C2} = N(a_T)N(a_P)$, where $N(a_j) = \exp(\frac{\pi}{2}a_j)\Gamma(1 - \mathrm{i}a_j)$ are the 2BC normalization factors, $a_j = Z_j/k_j$ are the Sommerfeld parameters and Z_j are the charges of the heavy particles.

Recently, we have shown that it is possible to go one step beyond this separable approximation by the introduction of multiple variable hypergeometric functions. Some terms of the non-orthogonal kinetic energy that dynamically mix the relative motion between the charges can be incorporated in the equation based on physical foundations, even when they have a rather cumbersome expression in terms of parabolic coordinates [17]. Since all terms in the non-orthogonal energy involve mixed derivatives, the introduction of them in the differential equation leads to an intrinsic non-separability. The first solution of this kind can be written in terms of the two-variable hypergeometric function Φ_2 [18]:

$$\Psi_{\Phi_2}^+ = N_{\Phi_2} \Phi_{PW} \Phi_2 \left[\mathrm{i}a_T, \mathrm{i}a_P, 1, -\mathrm{i}k_T \xi_T, -\mathrm{i}k_P \xi_P \right]. \tag{2}$$

The Φ_2 function can be viewed as one of the possible generalizations of the ${}_1F_1$ hypergeometric function for two variables. The state given by equation (2) correctly behaves asymptotically

and introduces correlation in the wavefunction that modifies the behaviour of the function for all distances. In fact, the normalization factor to outgoing unitary asymptotic flux $N_{\Phi_2} = N(a_T + a_P)$ is a non-separable function depending on the sum of the Sommerfeld parameters of each relevant interaction. In addition, the $\Psi^+_{\Phi_2}$ functions behave correctly when both ions approach each other, that is $k_T \sim k_P$ and $r_T \sim r_P$, giving rise to a 2BC function of the electron and a particle with charge $Z_P + Z_T$. This important property is not fulfilled by the Ψ_{C2} function and makes the $\Psi^+_{\Phi_2}$ a suitable state not only for intermediate energy ion–atom collision processes but also for photo-double-ionization of molecular hydrogen where this kind of condition is relevant.

The function $\Psi^+_{\Phi_2}$ can be expanded as [19]:

$$\Psi^+_{\Phi_2} = N_{\Phi_2}\Phi_{PW}\sum_{m=0}^{\infty} a_m \mathcal{F}^m(\xi_T)\mathcal{F}^m(\xi_P) \tag{3}$$

$$a_m = (-1)^m \frac{(ia_T)_m\,(ia_P)_m}{m!\,(m)_m\,(1)_{2m}} \tag{4}$$

where $(\alpha)_m$ are Pochhammer symbols and

$$\mathcal{F}^m(\xi_j) = \left(ik_j\xi_j\right)^m\ {}_1F_1\left[ia_j + m, 1 + 2m, -ik_j\xi_j\right]$$

with $j = T$ or P for brevity. This series can be considered as an expansion of the function in terms of target-centered and projectile-centered two-body functions. It is clear that, besides the normalization factor, the lowest order ($m = 0$) of the series (3) *is* the Ψ_{C2} function and coupling is included in higher orders of the series expansion. The specific correlation introduced by the $\Psi^+_{\Phi_2}$ state determines the particular expression of the coefficients a_m. This leads us to propose a set of continuum correlated waves (CCW) given by:

$$\Psi^{CCW} = N_{CCW}\Phi_{PW}\sum_{m,n=0}^{\infty} A_{mn}\mathcal{F}^m(\xi_T)\mathcal{F}^n(\xi_P). \tag{5}$$

In general, the coefficients A_{mn} can be obtained by constraining the function Ψ^{CCW} to satisfy the Schrödinger equation and other physical properties such as Kato cusp conditions or asymptotic restrictions [20]. Then, the series (5) breaks the artificial azimuthal symmetry of the independent particle approach considered in the C2 state. Furthermore, it is not necessary to include all the terms in the series, since a finite sum of these kind of terms always leads to a well-behaved function in the asymptotic regions. This could be useful for introducing a coupled description of the initial channel in any ion–atom collision [21].

From a computational point of view, the most important feature of the state (5) is that the transition matrices for single ionization within a distorted wave framework can be obtained analytically. They can also be represented by a series expansion that has an excellent numerical convergence [22, 23]. The final expressions for the transition matrices involve the calculation of a two-variable non-confluent hypergeometric F_1 of Appell and Kampé de Feriet [24]. Using some of the analytic continuation studied in detail by Le Vavasseur [25] and Olsson [26] a long time ago, we developed a code to perform an efficient numerical evaluation of F_1. The numerical computation of DDCS can be performed with reasonable computing time on a desktop computer. We have chosen an eikonal initial state because this simultaneously fulfills the asympotic Coulomb conditions and a proper normalization during the collision [11]:

$$\Psi_i^{EIS} \propto \psi_i(\boldsymbol{r}_T)\exp\left[-i\frac{Z_P}{v_i}\ln\left(k_i r_i - \boldsymbol{k}_i\cdot\boldsymbol{r}_i\right)\right].$$

The bounded electron in the target is described by the state $\psi_i(\boldsymbol{r}_T)$. We restrict our calculations to fundamental states of hydrogenic atoms. This enables us to compute cross sections in collisions with helium by using a Roothan–Hartree–Fock $5z$ state [27].

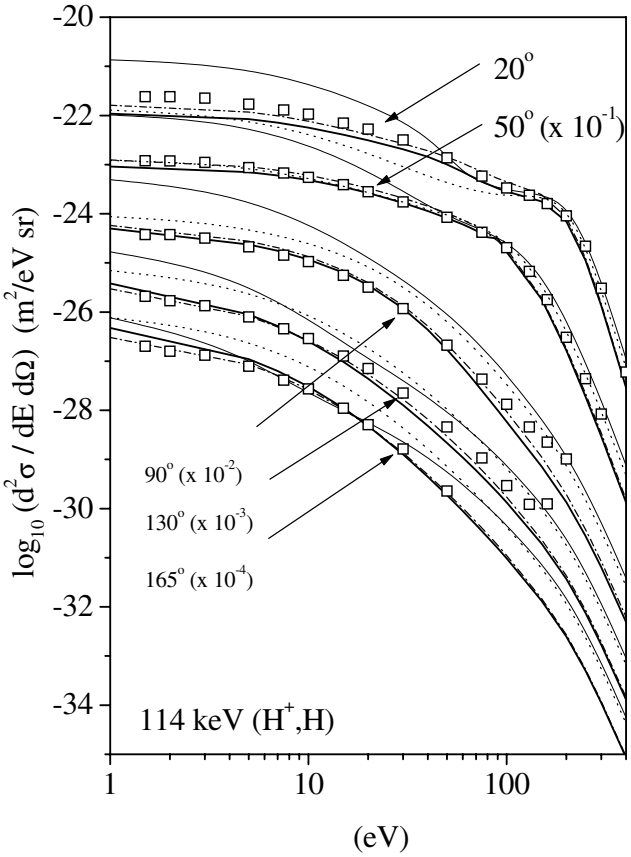

Figure 1. DDCS for the collision 114 keV $H^+ \rightarrow H$. Heavy curve, CCWEIS1 model; thin curve, Born–Φ_2 approximation; dotted curve, first Born approximation; dash–dotted curve, CDWEIS model. Squares represent the experimental data obtained by Kerby *et al* [28].

To analyse the changes in cross sections introduced by the CCW approach, we present the CCWEIS1 model that includes the first correlated order of the $\Psi^+_{\Phi_2}$ function and choose the normalization factor according to this prescription. In figure 1 we show the DDCS for a variety of angles of the emitted electron for the 114 keV $H^+ \rightarrow H$ collision. Experimental data have been reported by Kerby *et al* [28]. We observe that the CCWEIS1 model has a good agreement with the experiment for all emission energies. We note that this model introduces an improvement over the previous Born–Φ_2 theory [23, 29] that includes an undistorted initial channel. The CCWEIS1 theory gives a slightly lower contribution than the non-correlated CDWEIS model. Both theories underestimate the data for small emission angles, which is characteristic of all models with eikonal initial states, since they do not properly describe the electron–projectile interaction for small distances in the initial state [32].

We can now turn our attention to the zero degree emission DDCS. Up until now, data on $H^+ \rightarrow H$ collisions in the forward direction have not been available, and hence we should move to the helium target. In figure 2 we show the DDCS for a 1.5 MeV $H^+ \rightarrow He$ collision. Here we have modelled the final interaction with the target by an effective charge $Z^*_T = 1.3549$ that leads to the correct energy of the bound state. Experimental data have been measured by

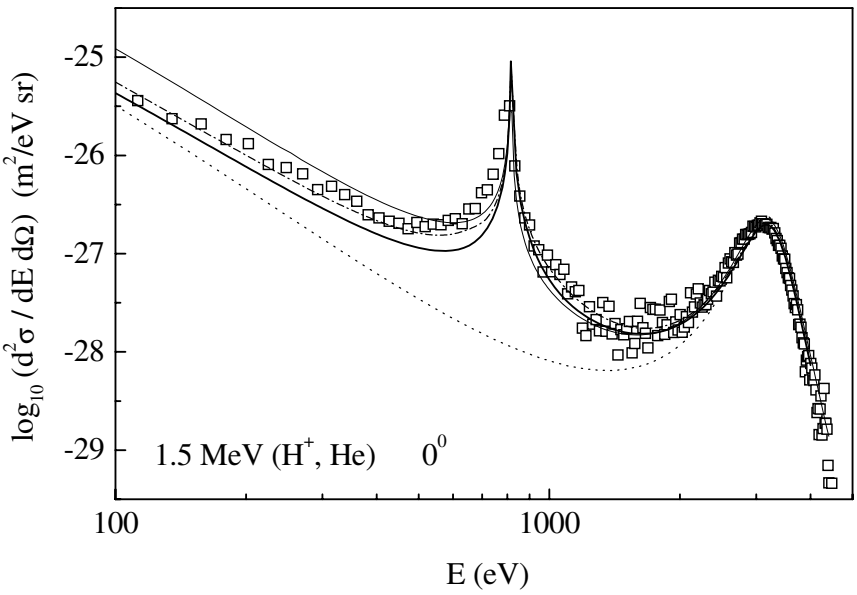

Figure 2. DDCS for zero degree emission for the process 1.5 MeV $H^+ \rightarrow$ He. The notation is the same as in figure 1. Experimental data from Lee *et al* [33].

Lee *et al* [33]. Three structures appear in this DDCS: the soft electron (SE) peak (electrons emitted with low velocities relative to the target), the electron capture to the continuum (ECC) cusp built by electrons with small velocity relative to the projectile and the binary peak, where electrons would lie after a binary collision with the incoming ion. This binary encounter peak (around $E \sim 3$ keV in figure 2) is mainly driven by a head-on collision between the projectile and the electron and is well described by simple two-body theories [33]. Since both correlated and uncorrelated distorted wave models exactly account for this collision, they give a very good agreement with the experiment. Turning our attention to the ECC peak (at $E = E_{\mathrm{ECC}} = 817$ eV in figure 2), we can see that the agreement is good in the high wing ($E > E_{\mathrm{ECC}}$) of the cusp, while in the low one the CCWEIS1 slightly underestimates the experimental data. We should note that the Born–Φ_2 exaggerates the DDCS for SE electrons by an order of magnitude. It is clear that the main differences between correlated and uncorrelated models arise in the SE region. To analyse this fact in detail, we have computed DDCS for small emitted energies in a 2.5 MeV amu^{-1} C^{6+} collision with helium that has been recently measured by Tribedi *et al* [34]. In figure 3 we plot the DDCS as a function of the emission angle. For the lowest electron energy measured (1 eV), the correlation modifies the behaviour in the small and large angle regions with respect to the CDWEIS model. For larger energies, the DDCS is bigger for large emission angles while the behaviour for forward angles remains similar in both theories. These soft collisions occur for large impact parameters, i.e. a large distance between the electron and the projectile. Then, we can conclude that even for these distances correlation is still relevant and cannot be neglected. This effect can be traced in the correlated normalization factor $N(a_T + a_P)$ of the final wavefunction. The changes introduced by the correlated state are in the right direction, but there still exist some quantitative differences due to the approximate way in which the electron–ion correlation has been considered. We would like to emphasize that this enhancement is due to the coupling of the motion of the electron

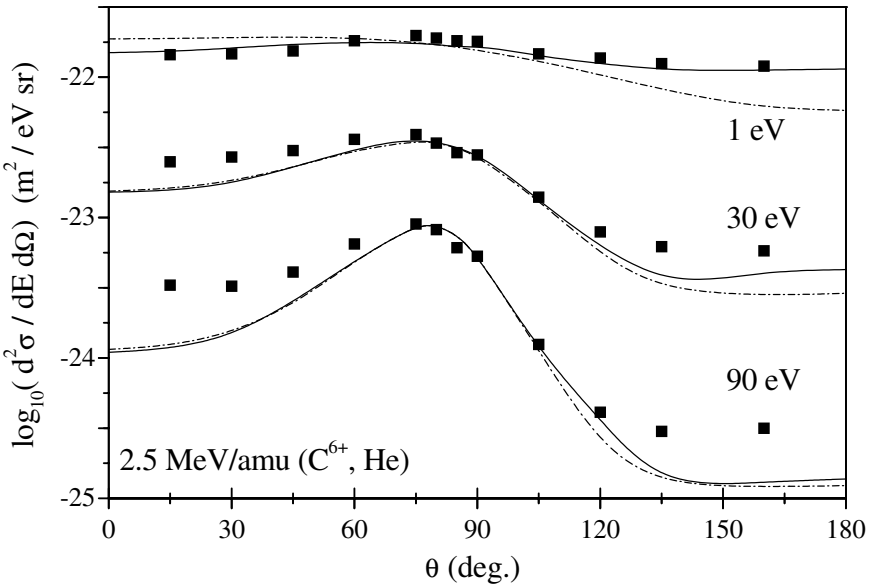

Figure 3. Low energy spectra for the 2.5 MeV amu^{-1} C^{6+} → He collision. Full curve, CCWEIS1 model; dash–dotted curve, CDWEIS model. Experimental data were measured by Tribedi *et al* [34].

relative to both target and projectile in the final state. This is different from a similar effect obtained when the role of passive electrons is taken into account in the target wavefunction, in the CDWEIS approach [30–32]. In the last case, the electron–ion correlation is treated only in a perturbative way, while the screening of the passive electrons is represented by a suitable model potential within the independent particle approximation. However, it is clear from figure 3 that electron–ion correlation is very important in this region and should not be neglected in any case, even in a simple Coulomb effective charge final state model used in this work.

In summary, we have shown that the electron–ion correlation plays an important role in single ionization of atoms by ion impact at high energies. A comparison with our previous Born-like models shows that the CCWEIS1 model improves the agreement with experimental results. The main differences with separable models, such as CDWEIS, can be found in the soft electron region, where the CCWEIS1 model enhances the emission for backward angles and slightly lowers the forward DDCS. It has been usual to neglect the electron–ion correlation on the basis that it is less significant than the long range behaviour of the Coulomb potential. However, the changes in DDCS are significant for low-energy electrons where the intermediate and asymptotic wavefunctions are dominant. Then, a detailed description of the ionization process should properly include the electron–ion correlation in a non-perturbative way, even for asymptotic distances. Finally, we note that the role of inactive electrons has been taken into account only through effective charges. The introduction of Hartree–Fock solutions for the description of many-electron targets or projectiles would improve the quantitative agreement with the experimental data. Hence, our future work will be devoted to analysing the whole non-orthogonal kinetic energy, searching for more exact correlated wavefunctions.

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