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Role of the Ground State in Electron-Atom Double Ionization

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Recently, absolute measurements have been reported for double ionization of helium by 5.6 keV electron-impact. At this high energy, one would think that the first Born approximation for the interaction of the projectile with the atom would be valid. However, on the basis of a lowest-order implementation of a Faddeev-type approach, Berakdar [Phys. Rev. Lett. **85**, 4036 (2000)] concluded that the approximation was *not* valid. Here we argue that (i) it is valid at this energy and (ii) the previous discrepancy between calculations in the first Born approximation and the overall magnitude of the measurements was due to a poor description of the ground state.

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Measurements of electron-impact double ionization of helium with fully determined kinematics present new challenges to atomic collision theorists. These so-called $(e, 3e)$ measurements are functions of the momenta of all three final-state electrons. The first *absolute* measurements of this kind were reported by Lahmam-Bennani *et al.* [1], based on the relative measurements of Taouil *et al.* [2]. The collision energy in the experiment was 5.6 keV and the momentum transfer was small, so one would expect a first Born approximation (FBA) for the interaction of the projectile with the atom to be reasonably accurate. FBA or similar results have been reported where the final double-continuum state of the atom was described by a product of three Coulomb waves [1] or a "convergent close-coupling" (CCC) wave function [3]. The CCC results had to be scaled up by a factor of 3.2 for comparison with the measurements and agreement in shape was relatively poor [3]. The "correlated four-body final-state," or C4FS, Coulomb-wave model showed better agreement in both shape and magnitude when very simplistic hydrogenic wave functions were used for the ground state; however, the overall magnitude was still off by nearly 50% [1]. Moreover, when a wave function ψ_{H} of the Hylleraas [4] type was used for the ground state, the C4FS magnitude became too large by a factor of 10 [1].

Agreement with the overall magnitude was eventually obtained by Berakdar [5], using an extension of the well-known Faddeev equations. These calculations, which used a product of two hydrogenic wave functions for the ground state [6], differed significantly for electron vs positron impact (by about a factor of 2). Since this should happen only if the higher-Born terms are strong, the conclusion was that the FBA is not valid at 5.6 keV.

Here we argue that the problem lies not with the FBA, but rather with a poor description of the ground state. What hydrogenic and Hylleraas wave functions have in common is that electron-electron correlation is treated imprecisely. Consequently, the cusp conditions of Kato [7] are generally not satisfied and the wave function will be inaccurate when the two electrons are close together. (Since the Coulomb potential $1/r$ is singular for $r \rightarrow 0$, the corresponding wave function must have a precise cusp at $r = 0$ such that the kinetic energy also becomes singular in a way which enables the total energy to remain constant.) This problem is resolved by using the Pluvinage wave function [8], which treats the electron-electron interaction to all orders of perturbation theory and satisfies Kato's cusp conditions exactly.

An exact evaluation of the first Born term for double ionization of helium requires the solution of the three-body problem for both the initial ground state and the final double-continuum state of the atom. These solutions, ψ_i and ψ_f^-, respectively, are necessarily orthogonal, i.e., $\langle \psi_f^- | \psi_i \rangle = 0$, since they are eigenfunctions of the same Hamiltonian (the minus sign in ψ_f^- denotes the solution with incoming scattered waves). In addition, since the ground state is a singlet spin state, both ψ_i and ψ_f^- must be spatially symmetric.

If approximations are made to either wave function, the orthogonality between ψ_f^- and ψ_i will generally be broken. Thus, when making approximations, some care must be taken to ensure that spurious contributions from the lack of orthogonality are small. A good way to test for false contributions is to impose orthogonality artificially by replacing ψ_f^- with $\psi_f^- - \langle \psi_f^- | \psi_i \rangle^* \psi_i$. Then, if (forced) orthogonalization leads to only small changes in the calculated cross sections, the original lack of orthogonality was not a serious problem.

In this work, we approximate ψ_f^- by the spatially symmetric part [9] of the well-known 3C wave function [10,11], which contains the product of three Coulombic distortion factors (one for each two-body Coulomb interaction). In the same spirit, we approximate ψ_i by a three-body product wave function ψ_{P} due to Pluvinage [8](the Pluvinage wave function is the doubly bound analog of the 3C wave function). Although our approximations to ψ_f^- and ψ_i are not orthogonal, they are constructed in a similar manner. Both treat all three two-body Coulomb

interactions exactly, albeit independently, and satisfy the cusp conditions of Kato [7] as any interparticle separation tends to zero. We call this approach "FBA(3C)" with a Pluvinage ground state.

We also performed FBA(3C) calculations with ψ_{H}, the same 3-parameter Hylleraas-type wave function that was used in the C4FS calculations [1]. We found that ψ_{H} leads to an overall magnitude that is about 50% larger than experiment (65% larger with orthogonalization), rather than a factor of 10 larger as was found in [1]. In our FBA(3C) calculations, the hydrogenic and Hylleraas-type wave functions gave very similar results in both shape and magnitude, and, as pointed out by Kheifets *et al.* [3], results from the C4FS effective-charge model should differ from FBA(3C) by only ~5% for the kinematics considered.

The present $(e, 3e)$ calculations are the first to use the Pluvinage wave function ψ_{P} for the ground state. We note that ψ_{P} has been used extensively by Crothers and coworkers [12] in *ion*-impact studies but has otherwise been ignored. Here we show that the use of ψ_{P} leads to much improved agreement with the overall magnitude of absolute $(e, 3e)$ measurements.

The fact that ψ_{P} leads to better agreement with experiment than ψ_{H} is not surprising. In 1935, Bartlett *et al.* [13] noticed that Hylleraas wave functions [4] lead to *infinite* errors in the local energy $(H\psi_i)/\psi_i$ (where H is the Hamiltonian for the atom) whenever the separation between any two particles vanishes. This is because Hylleraas wave functions do not diagonalize H in the (singular) Coulomb interactions. As discussed by Crothers and McCarroll [12], the Hamiltonian matrix for the Pluvinage wave function *is* diagonal in all Coulomb interactions and the remaining perturbations do not have singularities. Regarding the Hylleraas wave function near the Coulomb singularities, Bartlett *et al.* [13] wrote the following. *"The eigenfunction is thus probably very badly in error in the general neighborhood of any of these points."*

Atomic units are used in this work except where stated otherwise and unit vectors are denoted by a "hat,", i.e., $\hat{\mathbf{x}} = \mathbf{x}/x$, where $x = |\mathbf{x}|$. As usual, the mass of an electron is neglected relative to the mass of a proton.

The fully differential cross section (FDCS) for the $(e, 3e)$ reaction is given by [11]

$$\frac{d^8\sigma}{d\hat{\mathbf{k}}_1 d\hat{\mathbf{k}}_2 d\hat{\mathbf{k}}_3 d(k_2^2/2) d(k_3^2/2)} = (2\pi)^4 \frac{k_1 k_2 k_3}{k_0} |T_{fi}|^2. \quad (1)$$

Here $\mathbf{k}_0$, $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are the momenta of the incident, scattered, and two ejected electrons, respectively.

The exact transition-matrix element T_{fi} is given by

$$T_{fi} = \langle \Psi_f^- | V_i | \beta_i \rangle, \quad (2)$$

where Ψ_f^- is the exact scattering wave function developed from the final-state and

$$\beta_i = (2\pi)^{-3/2} e^{i\mathbf{k}_0 \cdot \mathbf{r}_1} \psi_i(\mathbf{r}_2, \mathbf{r}_3) \quad (3)$$

is the initial asymptotic state, where $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_3$ are the positions, relative to the atomic center, of the projectile and the two atomic electrons, respectively. The perturbation

$$V_i = -2/r_1 + 1/r_{12} + 1/r_{13} \quad (4)$$

is the interaction of the projectile with the target atom, where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$.

In the FBA, the projectile motion is treated with plane waves both initially and finally and exchange between the projectile and the atomic electrons is ignored. The integration over the projectile coordinate can then be easily performed analytically so that the transition amplitude in the FBA is given by

$$T_{fi}^{(\mathrm{FBA})} = (4\pi/q^2) \langle \psi_f^- | \tilde{V}_i | \psi_i \rangle. \quad (5)$$

Here

$$\tilde{V}_i = (-2 + e^{i\mathbf{q}\cdot\mathbf{r}_2} + e^{i\mathbf{q}\cdot\mathbf{r}_3})/(2\pi)^3, \quad (6)$$

where $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1$ is the momentum transferred from the projectile to the target atom.

For distinguishable electrons, the 3C wave function for a double-continuum state of helium is given by [11]

$$\psi_{3\mathrm{C}}^- = (2\pi)^{-3} e^{i(\mathbf{k}_2\cdot\mathbf{r}_2 + \mathbf{k}_3\cdot\mathbf{r}_3)} C(-2/k_2, \mathbf{k}_2, \mathbf{r}_2) \times C(-2/k_3, \mathbf{k}_3, \mathbf{r}_3) C(\mu/k_{23}, \mathbf{k}_{23}, \mathbf{r}_{23}). \quad (7)$$

Here $\mathbf{k}_{23} = \mu(\mathbf{k}_2 - \mathbf{k}_3)$, the reduced mass $\mu = 0.5$ a.u., and $C(\eta, \mathbf{k}, \mathbf{r}) = \Gamma(1 - i\eta) e^{-\pi\eta/2} {}_1F_1(i\eta, 1; -ikr - i\mathbf{k}\cdot\mathbf{r})$, where ${}_1F_1$ is the confluent hypergeometric function. Our approximation to ψ_f^- is the spatially symmetric part [9] of the above 3C wave function; thus exchange between the two atomic electrons is included in accordance with the Pauli principle.

The Pluvinage approximation for the ground state of helium is given by [8]

$$\psi_{\mathrm{P}}(\mathbf{r}_2, \mathbf{r}_3) = \phi(r_2)\phi(r_3)\chi(k, r_{23}). \quad (8)$$

Here $\phi(r) = \sqrt{8/\pi}\, e^{-2r}$ and

$$\chi(k, r) = N(k)\, e^{-ikr} {}_1F_1(1 - i\mu/k, 2, 2ikr), \quad (9)$$

where $k = 0.41$ a.u.(this value of k minimizes the ground-state energy) and $N(0.41) = 0.603\,37$.

$T_{fi}^{(\mathrm{FBA})}$ is evaluated using six-dimensional numerical (Gauss-Legendre) quadrature. We estimate that our numerical uncertainty for the FDCS is about 2%.

In the 5.6 keV experiment of Lahmam-Bennani *et al.* [1], both atomic electrons are ejected into the scattering plane (the plane containing $\mathbf{k}_0$ and $\mathbf{k}_1$) with the same energy (10 eV) and the scattering angle of the projectile is fixed at $\theta_1 = 0.45°$ ($|\mathbf{q}| \approx 0.24$ a.u., $\hat{\mathbf{q}} \approx 319°$). The scattering angles of the ejected electrons, θ_2 and θ_3, are

measured in the same sense as θ_1 relative to the incident beam direction.

In Fig. 1, we compare the complete set of the above absolute measurements with our FBA(3C) calculations. In the figure, θ_2 is fixed as indicated and the cross section is presented as a function of θ_3. There are 20 fixed-θ_2 cases in all.

We performed two different FBA(3C) calculations — one with ψ_P for the ground state and the other with ψ_H for the ground state. In both calculations, the final-state was orthogonalized to the initial-state. Calculations without orthogonalization (not shown) differed by about 10% from the corresponding calculations with orthogonalization for either choice of ground-state wave function. Thus spurious contributions from the lack of orthogonality are small for both ψ_P and ψ_H. Only the calculations with ψ_P are shown, since our results using ψ_H were simply 65% larger in overall magnitude, with no appreciable change in shape. The reason for the larger magnitude is that ψ_H has too large an amplitude in the region of space where the two electrons are close together, which increases the probability of double ionization.

When the Pluvinage wave function is used for the ground state (thick solid lines in Fig. 1), agreement with the absolute measurements [1] is quite good for all but four of the 20 cases ($\theta_2 = 83°$, 97°, 207°, and 221°). In these four cases, the measured FDCS is significantly smaller than theory for the larger theoretical peak. The C4FS calculations [1] exhibit a similar relative discrepancy with the shape of the measurements.

Berakdar [5,6] has presented a Faddeev-type approach where, in lowest order, the four-body final state is a sum of four 3C states — each corresponding to switching off all interactions for one of the four particles. One of Berakdar's resulting four amplitudes corresponds to FBA(3C) with a hydrogenic ground state. Using this amplitude alone would overestimate the overall magnitude by about 50%. Thus, in Berakdar's calculations (thin solid lines in Fig. 1), agreement with the overall magnitude is obtained as a result of interference between an FBA amplitude and three other three-body amplitudes, all of which use a hydrogenic ground state. Such interference is common in lowest-order implementations of the Faddeev method and is known

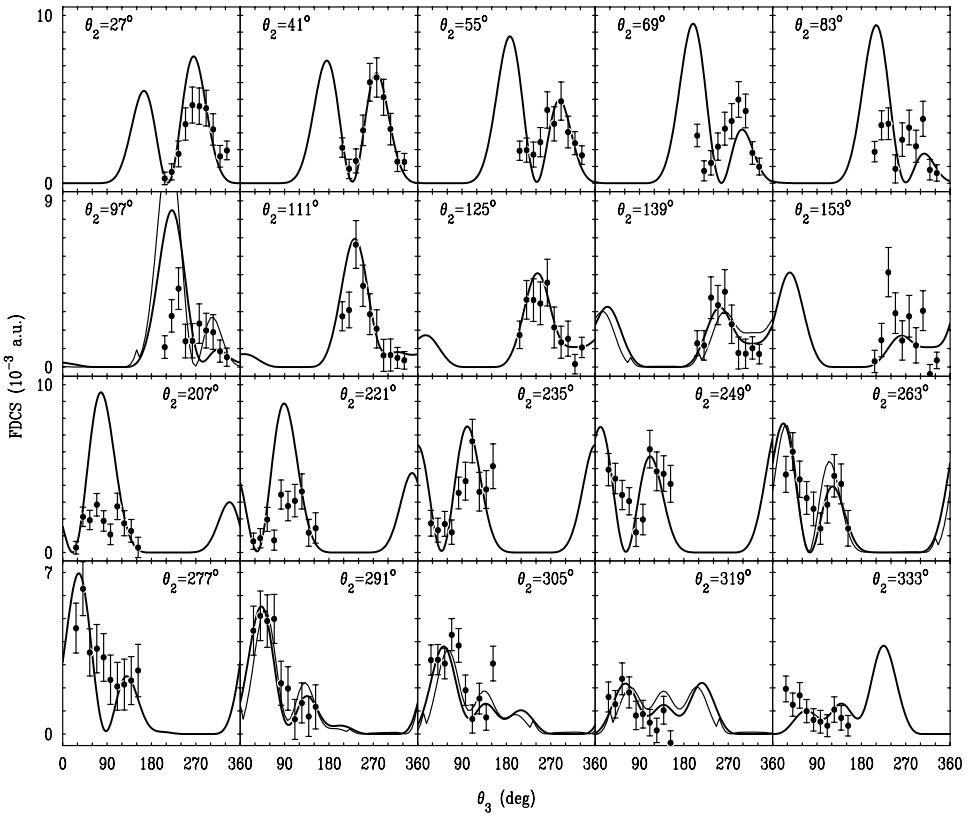

FIG. 1. Fully differential cross section (FDCS) in the scattering plane for electron-impact double ionization of the ground state of helium. The collision energy is $E_0 = 5599$ eV and the energies of the ejected electrons are given by $E_2 = E_3 = 10$ eV. The incident electron is scattered by an angle $\theta_1 = 0.45°$ and the scattering angles for the ejected electrons are θ_2 and θ_3. The present FBA(3C) calculations with a Pluvinage ground state are shown as the thick solid lines. The absolute measurements (solid circles with error bars) are from Lahmam-Bennani *et al.* [1]. Also shown, where available ($\theta_2 = 97°$, 139°, 263°, 291°, 305°, and 319°), are calculations from Berakdar [5] using a Faddeev-type approach (thin solid lines).

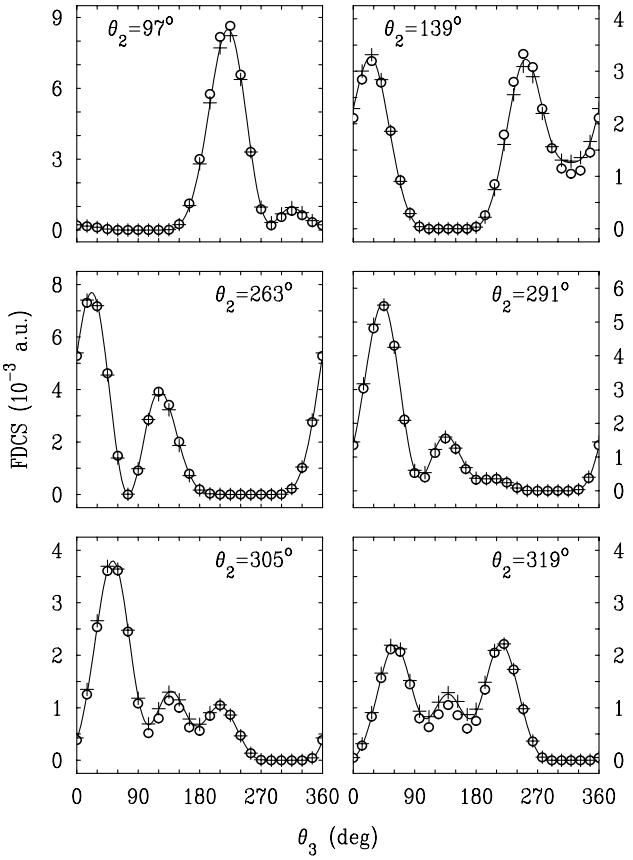

FIG. 2. Comparison of FBA(3C) calculations (solid lines) with 6C calculations for electron (open circles) and positron (plus signs) impact for the kinematics of Fig. 1.

to produce unphysical results in the case of single ionization [14,15].

To explore this issue further, we have implemented a genuine four-body method where the final-state wave function for the whole system (projectile plus target) is approximated by a "6C" wave function that treats all six two-body Coulomb interactions between the four particles exactly, albeit independently (and necessitates a nine-dimensional numerical quadrature to compute T_{fi}). This four-body product wave function (6C) is superior to the sum of four three-body (3C) wave functions used by Berakdar [5], for the reasons discussed by Briggs [14]. In Fig. 2, our 6C calculations (numerically accurate to 5%) for electron and positron impact are compared with the FBA (the Pluvinage wave function is used for the ground state). These calculations clearly support the validity of the FBA — the 6C results closely follow the FBA curves and the difference between electron and positron impact is an order of magnitude smaller than predicted by Berakdar [5].

Our FBA(3C) model is reasonable only for very fast projectiles, since the projectile-target interaction is treated only to first order. In addition, our model will not be valid if the sum of the energies of the two ejected electrons is very low, since their motion is treated using 3C perturbation theory. Indeed, it may seem surprising that our theory is reasonably accurate even for 10-eV ejected electrons. A recent $(e, 2e)$ study [16] showed that the 3C cross section has errors of $\pm 50\%$ for 20-eV outgoing electrons (54-eV collision energy, atomic hydrogen target). The reason why 3C is accurate for 10-eV ejected electrons here, but not for 20-eV outgoing electrons in the case of $(e, 2e)$, is because initial-state projectile-target interactions beyond first order (which are not included in the 3C approximation) are negligible in the former case of a 5599-eV projectile but important in the latter case of a 54-eV projectile.

In conclusion, we have presented a simple three-body model for electron-impact double ionization of helium. Like many of the previous FBA studies, we used the 3C wave function to describe the final double-continuum state of the atom. Unlike all previous $(e, 3e)$ studies, however, we used the Pluvinage wave function [8] for the ground state. Consequently, electron-electron correlation is treated precisely (and the cusp conditions of Kato are satisfied exactly) by both our initial and final target wave functions and we found that this is crucial for reproducing the absolute measurements.

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