Discrepancy between theory and experiment in double ionization of helium by fast electrons

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We compute fully differential cross sections for double ionization of helium by electrons, within the high-impact-energy and low-momentum transfer regimes, using the generalized Sturmian functions approach. Our results are converged relative to the total angular momentum and variable domain size. The method shows very good agreement with convergent close coupling calculations performed by Kheifets *et al.* [J. Phys. B **32**, 5047 (1999)] for all ejection angles for the two electron emission energies considered in the experiments reported in that contribution. Both theoretical methods provide fully differential cross sections that require the same upscaling factors to compare with experimental data and are based on a first-order Born model for the projectile-target interaction. Since that reference was published, there were several theoretical efforts to account for the absolute scale of the experimental results, but agreement in the cross-section magnitude was not achieved even between theories. With the present contribution we conclude that the first-order Born model is now adequately solved, shifting the magnitude controversy towards either the experimental data and/or the addition of higher degrees of projectile-target interaction to the calculation.

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Ionization of atoms by electron impact is one of the key experiments in physics to explore the role of correlation and to analyze the features of the few-body quantum continuum. On one hand, ionization of hydrogen atoms has been the subject of many experimental studies, together with several very important theoretical developments along the years, for different impact energies and collision geometries, from basic models [1,2] to comprehensive fully numerically accurate calculations [3,4]. Pure single ionization of many electron atoms, however, still remains an important topic of research, due to the experimental difficulties of isolating this particular process from other emission channels, and an adequate theoretical description of the interaction between the ejected electron and the remaining bound partners in the residual target [5,6].

On the other hand, double ionization of helium by electron impact—usually noted as the $(e,3e)$ problem—has challenged both experimental and theoretical physicists in recent years [5]. The high-energy-impact regime used in the experiments results in sets of fully differential cross sections (FDCS) in the angles of the emitted electrons. There are experiments with conventional spectrometers in a coplanar geometry [7], with projectiles impinging at 5599 or 5587 eV, and ejection energies of 10 or 4 eV, respectively, for each electron. In this case, the momentum transfer $\mathbf{q}$ from the projectile to the nucleus is small, respectively, 0.24 and 0.22 a.u.. Also, there are kinematically complete experiments using the COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) apparatus [8,9], where the recoil momentum of the ion is also measured. Both methods complement each other in the understanding of the dynamics of the process.

There are many theoretical approaches applied to this process that can be divided into two different lines of work. The first ones are based on perturbation theory, where approximate initial and/or final wave functions of the collision are employed in the transition matrix calculation (see Refs. [10,11]). These methods usually make use of a Born series up to first or second order and are easier to compute from a numerical point of view. However, the FDCS obtained with them are remarkably different among themselves (both in shape and magnitude) and with the experimental data. Such differences have been puzzling, because these theories tend to agree with each other in simpler problems, such as the atomic single ionization by electron impact or the double photoionization of He [12,13]. This could indicate that second-order effects should be considered [14,15].

The other type of theoretical approach aims to solve the problem from first principles, making use of different basis sets to expand the solution accurately. Under this category one can find convergent close coupling (CCC) [7,16] and the J matrix [17–21]. The CCC approach gives good agreement in shape for all FDCS, but the magnitude is consistently lower than the experimental results by factors of 2.2 and 14 (10 and 4 eV cases, respectively). The J matrix approach gives overall agreement in shape with the experimental data, except for some ejection angles. Besides, it reproduces neither the absolute measurements nor the CCC theoretical results. One common ground of all these theoretical models is that they treat the description of the four-body problem as a three-body one. If these three-body theories were in agreement among themselves and all together in disagreement with the experimental data, one would conclude that the experimental data has flaws or that higher orders are required. Since even theories themselves do not agree with each other, it could be possible that some assumptions and/or numerical implementations are not adequate for all of them. If a new theory agrees any of the pre-existent calculations, one would be able to rule out the rest and perhaps question the experimental results.

The aim of this work is to shed some light into this long-standing controversy. Therefore, we propose a completely different theoretical approach based on generalized Sturmian

functions (GSF) [22,23], and compute the FDCS for $(e,3e)$ within this model.

The $(e,3e)$ problem formally involves four bodies, three electrons and the nucleus. Since the center of mass of the system lies on the nucleus, the problem can be described by three coordinates $\{\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3\}$ of the electrons relative to the helium core, totaling nine real variables. We consider that electron 1 is the projectile with initial momentum $\mathbf{k}_i$ that leaves the collision region with final momentum $\mathbf{k}_f$, while electrons 2 and 3 are bound in the initial state and ejected by the collision with electron 1. The total energy of the system is $E = E_0 + |\mathbf{k_i}|^2/2 = E_a + |\mathbf{k_f}|^2/2$. The bound energy of the He atom is represented by E_0, while E_a is the energy of the ejected electrons. The accurate solution of the corresponding four-body Schrödinger equation remains a formidable task from a numerical point of view. However, in the high-energy regime it is possible to reduce this problem to a three-body one, by keeping the first order of a series expansion of the exact solution. In a recent work, we have shown that the three-body equivalent problem for the $(e,3e)$ ionization can be described by the time-independent Schrödinger equation [24–26]:

$$[E_a - h_{He}]\Phi^+_{sc}(\mathbf{r}_2,\mathbf{r}_3) = \frac{4\pi}{q^2}\frac{1}{(2\pi)^3}(-2 + e^{i\mathbf{q}\cdot\mathbf{r}_2} + e^{i\mathbf{q}\cdot\mathbf{r}_3})\Phi_i(\mathbf{r}_2,\mathbf{r}_3). \quad (1)$$

Here the wave function $\Phi^+_{sc}(\mathbf{r}_2,\mathbf{r}_3)$ dictates the ionized electron dynamics, while $\Phi_i(\mathbf{r}_2,\mathbf{r}_3)$ is the $1s^2$ state of He, with Hamiltonian h_{He}.

The GSF method makes use of orthogonal one-electron radial basis sets. Each element of this basis is the solution of the Sturmian equation

$$\left[-\frac{1}{2}\frac{d^2}{dr^2} + \frac{l(l+1)}{2r^2} + \mathcal{U}(r) - E_s\right]S^{\pm}_{nl}(r) = -\beta_{nl}\mathcal{V}(r)S^{\pm}_{nl}(r), \quad (2)$$

where $\mathcal{U}(r)$ and $\mathcal{V}(r)$ are real functions of the variable r, called *generating* and *auxiliary* potentials, respectively. The main difference between this equation and the traditional Schrödinger one is that the energy E_s is a fixed parameter and one searches the eigenvalues β_{nl} [27]. The magnitude l is the single-particle angular momentum. An important feature of the GSF method is that the solutions of Eq. (2) are chosen with outgoing (+) or incoming (−) asymptotic condition, if one is interested in a continuum state [28], or as exponentially decaying functions if one is looking for bound states [29]. Therefore, one can expand the scattering wave as

$$\Phi^+_{sc}(\mathbf{r}_2,\mathbf{r}_3) = \sum_{L,M}\sum_{l_2,l_3}\sum_{n_2,n_3}\phi_\nu\Theta_\nu(\mathbf{r}_2,\mathbf{r}_3), \quad (3)$$

with

$$\Theta_\nu(\mathbf{r}_2,\mathbf{r}_3) = \mathcal{Y}^{LM}_{l_2l_3}(\hat{\mathbf{r}}_2,\hat{\mathbf{r}}_3)\frac{S^+_{n_2l_2}(r_2)}{r_2}\frac{S^+_{n_3l_3}(r_3)}{r_3}, \quad (4)$$

where the $\mathcal{Y}^{LM}_{l_2l_3}(\hat{\mathbf{r}}_2,\hat{\mathbf{r}}_3)$ are coupled spherical harmonics, while the GSFs $S^+_{n_il_i}(r_i)$ $(i = 2,3)$ have purely outgoing asymptotic behavior. The initial state $\Phi_i(\mathbf{r}_2,\mathbf{r}_3)$ in Eq. (1) can be computed in terms of bound GSFs, as shown in Ref. [30]. Projection onto the final-state basis $\Theta_{\nu'}(\mathbf{r}_2,\mathbf{r}_3)$ leads to a large, dense linear system on the unknowns ϕ_ν. The label ν denotes the set of indices $\{L,M,l_2,l_3,n_2,n_3\}$.

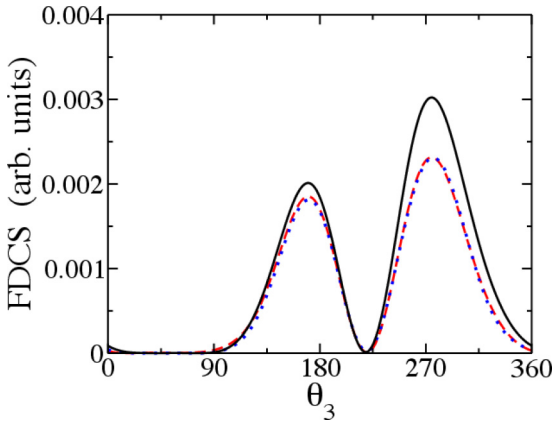

FIG. 1. (Color online) Example of convergence of FDCS for different values of the numerical asymptotic limit. FDCS is computed for the 10 + 10 eV energy sharing and an ejection angle of $\theta_2 = 41^\circ$ in arbitrary units, without renormalization factors. The solid black line was computed at 28 a.u., dashed red (gray) line at 48 a.u., and the dotted blue (gray) line at 68 a.u.

In the present work, we apply the GSF strategy to the $(e,3e)$ collisional configuration of Ref. [7], where absolute FDCSs in the coplanar geometry have been obtained for equal emission energies, of 4 and 10 eV. The energy parameter E_s for each Sturmian was set equal to the total available energy for the ejected electrons. For each total angular momentum L wave, five sets of values (l_2,l_3) were considered. To test the convergence respect to total angular momentum L, we took the mean value of each partial wave's coefficients ϕ_ν to compare the contribution to the cross section. We choose to compute five partial waves with $L = 0,1,2,3$, and 4. The $|\phi_\nu|$ mean value peaks at $L = 1$, drops one order of magnitude for $L = 2$, and drops two additional orders of magnitude for $L = 3$ and 4. Hence, the waves with L beyond 2 contribute negligibly to the FDCS, at least within this $(E_a,\mathbf{q})$ regime. This is further supported by the results of Knyr *et al.* [21], where they required only up to $L = 2$.

The transition matrix $T_{\tilde{\mathbf{k}}_2,\tilde{\mathbf{k}}_3}$ in terms of the momenta of the ionized electrons is computed from the asymptotic part of the wave function [24]. For practical purposes, the transition matrix is obtained at different fixed hyperradii $\rho^2 = r_2^2 + r_3^2$, until convergence is achieved. The differential cross section is computed as

$$\frac{d^5\sigma}{d\Omega_2 d\Omega_3 d\Omega_f dE_2 dE_3} = (2\pi)^4\frac{k_f k_2 k_3}{k_i}|T_{\tilde{\mathbf{k}}_2,\tilde{\mathbf{k}}_3}|^2. \quad (5)$$

In Fig. 1 we show the FDCS for $\theta_2 = 41^\circ$ computed for three different domains. It is clear that convergence is achieved for ≈50 a.u., since that FDCS varies little from the one obtained at ≈70 a.u. This behavior replicates for all ejection geometries.

In the next figures we show our calculated fully differential cross sections with the GSF theory for two ejection angles: $\theta_2 = 135^\circ$ and $\theta_2 = 315^\circ$ for 4 + 4 eV configuration (Fig. 2), and $\theta_2 = 55^\circ$ and $\theta_2 = 291^\circ$ for 10 + 10 eV emission (Fig. 3). We compare the GSF results with those of the CCC and J matrix theories. Our calculations present a remarkable agree-

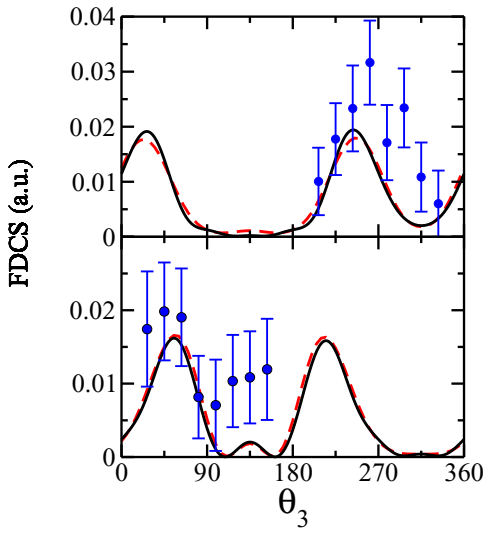

FIG. 2. (Color online) Two examples of FDCS compared with the experimental and theoretical data for $4+4$ eV ejected energies. Emission angles are $\theta_2 = 135^\circ$ (top) and $\theta_2 = 315^\circ$ (bottom). Solid black line, present GSF model; dashed red (gray) line, CCC; and circular blue (gray) symbols and bars, experimental data from Ref. [7].

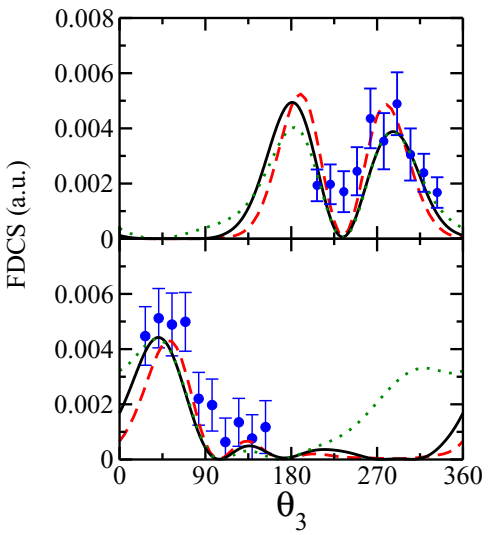

FIG. 3. (Color online) Same as Fig. 2, for $10+10$ eV ejected energies. Emission angles are $\theta_2 = 55^\circ$ (top) and $\theta_2 = 291^\circ$ (bottom). Solid black line, present GSF model; dashed red (gray) line, CCC; green (gray) solid line with symbols, J matrix ([31], based on the methods from Ref. [21]); and circular blue (gray) symbols and bars, experimental data from Ref. [7].

ment with the CCC approach, in both shape and magnitude, for $4+4$ eV electrons. A good agreement is obtained for the $10+10$ eV emission, although the interelectronic correlation described by the separation of the main peaks is somewhat different. We also note that the J matrix predicts a peak at $\theta_2,\theta_3 = 291^\circ$ (collinear emission), that does not show up in our calculations or in the CCC's (see Fig. 2). Disagreement in magnitude with the absolute experimental data available is observed for all geometries. Factors of 2.2 and 14 were used to scale up the theoretical GSF and CCC results, while the J matrix required factors of 1.2 and 7 [6]. Another striking difference with experimental result can be seen in the sharp zero of the theoretical cross section for $\theta_2 = 55^\circ$ and $\theta_3 = 220^\circ$ at $10+10$ eV, where the experiment exhibits a null value. This behavior can also found in other geometries.

Although differences in magnitude still remain when theory is compared with the experiment, we can state that now there are two completely different theoretical approaches agreeing well with each other, for a wide range of ejection angles. The agreement between the theories is the best reported up to this moment, and it applies to the two emission energies of the Orsay experiment. One possible solution to the puzzle is that the assumption that the process can be modeled as a three-body system within a first-order Born style expansion is not valid, and some of the features of the real four-body collision are left out. There is evidence that including second-order Born terms in the calculations does not lead the theoretical results to agree with experimental data magnitude [32]. Therefore, it is possible that the experimental data present some systematic error that results in the uniform overestimation of the cross sections along all geometries. However, for that suggestion to be verified, good agreement in second-order calculations should be attained by at least two independent schemes, as we have now presented for the first order. We believe that our work has helped to advance the controversy away from the first-order stage.

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