Double ionization of two-electron systems

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Abstract. We address various issues related to the double ionization by electron impact of two-electron systems. The emphasis will be put on the theoretical description of high incident energy \((e, 3e)\) processes, for which the first Born approximation should be suitable. In the case of helium, absolute experimental data for fivefold differential cross sections are available in coplanar geometry. We will review and discuss the divergencies existing between the results obtained with different theoretical models, and those appearing when compared to the experiments in particular with respect to the absolute scale. We will then discuss some results obtained in a recently proposed out of plane geometry.

1. Introduction
The study of the double ionization of atoms by electron impact allows one to gain information on correlated systems. In a kinematically complete \((e, 3e)\) experiment, the particles are detected in coincidence and a fivefold differential cross sections (FDCS) is deduced. In the case of two-electron atomic targets, like helium, one deals in the final state with a pure four-body Coulomb problem which, for high incident energies, can be reduced to a three-body problem from a theoretical point of view. However, even in this case, no exact wavefunction is known for either the scattering or the bound states. Hence, approximations are made, and \((e, 3e)\) cross sections obtained with different theoretical descriptions of the initial and final states are generally not in agreement with each other. Moreover, when compared with high energy absolute experimental data on helium [1, 2], a rather confusing picture emerges. This has been the subject of many recent studies (see, e.g., [3, 4] and references therein), and will be further discussed in Section 2. We shall show how several models disagree with each other and with the experimental data: a particular attention will be put on the absolute scale of cross sections. In Section 3, we shall make some comments on the results obtained in a new out-of-plane geometry which was suggested as a tool to explore second Born effects. This contribution will end with some concluding remarks.

2. Coplanar geometry
Absolute \((e, 3e)\) FDCS have been measured by Lahmam-Bennani et al. for high incident energy \((E_i = 5599 \text{ eV})\) and ejected energies of (A) \(E_1 = E_2 = 10 \text{ eV} [2]\) or (B) \(E_1 = E_2 = 4 \text{ eV} [1]\). In these coplanar experiments, the scattering angle was set at \(\theta_0 = 0.45^0\), so that these conditions correspond to small momentum transfer \(q\) (\(q = 0.24\) and \(q = 0.22\) a.u., respectively) and thus to the dipolar regime. The absolute data allow one to make a detailed study – both in shape
and magnitude – of the double ionization of helium. Cross sections were reported as a function of the angle of one of the ejected electrons \( \theta_1 \), for 20 (A) and 16 (B) fixed values of \( \theta_1 \).

The fully differential \((e,3e)\) cross section for the ejection of two electrons from a two-electron atom, is given theoretically within the first Born approximation (FBA) by

\[
\frac{d^5\sigma}{d\Omega_0d\Omega_1d\Omega_2dE_1dE_2} = (2\pi)^4 \frac{k_0k_1k_2}{k_i} |T_{fi}(k_0, k_1, k_2)|^2,
\]

where \( k_i \) and \( k_0 \) are the momenta of the incoming and outgoing projectile which define the scattering plane; \( k_1 \) and \( k_2 \) are the momenta of the ejected electrons after the collision (energy \( E_1, E_2 \)); \( d\Omega_0, d\Omega_1 \) and \( d\Omega_2 \) denote, respectively, the solid angle elements for the scattered and the two ejected electrons. The transition matrix \( T_{fi} \) depends on the choice of the initial double bound state \( \Psi_i \) and the final double continuum \( \Psi_f \); these wavefunctions which describe, respectively, the two electrons belonging to the target before and after their ejection, are not known exactly.

On the theoretical side, there have several published calculations which attempted to reproduce these experimental data: the Convergent Close Coupling (CCC) approach [1, 5]; the J–matrix method [6, 7]; a wave–packet evolution approach [8, 9]; the Green function expansion [10]; the distorted wave approaches with the "pure" 3C model [11, 12, 13, 14, 15, 16, 3, 4] or variants with effective charges [2, 17, 18] (it is worth underlying that all the 3C results, published by several groups and obtained with independent numerical codes, are consistent with each other); and the 6C model [11] (the latter takes into account the pairwise final state interactions between all four particles; as in the considered kinematical conditions the 6C model results do not differ much from those of the 3C [11], we shall no longer discuss it). Since the choice of effective charges is arbitrary, and the improvement with respect to the pure 3C model is highly dependent on the kinematical conditions, we shall not consider here any of its variants.

Since the experimental energy of the incoming projectile is high and the momentum transfer is small, the comparison between the theoretical calculations and the measured data can be performed within the frame of the FBA in the interaction of the projectile with the target atom. Indeed, explicit second Born calculations showed that little difference (about 10%) is observed with either the CCC approach [19] or the 3C model [20, 21]. The validity of the FBA, is further supported by the comparison of 3C calculations for electron and positron impact [11] (in contrast, differences between the two projectiles were observed in a Green function expansion combined with a very simple \( \Psi_i \) wavefunction [10], suggesting that the Born limit is not reached).

In the present contribution, we shall restrict the analysis to FBA results. Even within this approximation, the FDCSs obtained with different theoretical descriptions of the initial and final states are not in agreement with each other, and yield a rather confusing picture which is the subject of a lively debate amongst theorists.

Consider first the set, named (A) hereafter, of experimental data for two electrons ejected at \( E_1 = E_2 = 10 \) eV. Based on the published material, we may state that practically all theoretical models manage to reproduce overall the FDCS shapes; some minor details of the 20 situations may differ from one approach to the other. On the other hand, large differences appear as the magnitude is concerned and will be now investigated. To facilitate the task, we introduce the following experimental to theoretical FDCS ratio

\[
R_{\theta_1}(E_1, E_2) = \frac{FDCS[\text{Experimental}]}{FDCS[\text{Theoretical}]}
\]

evaluated at one of the cross sections maxima (see choice below), for a given set of energies \( (E_1, E_2) \) and geometrical configuration \( (\theta_1) \). Ratios found for several models are presented in Table 1 with the intention of providing an overview of the magnitudes (different, but relatively similar, ratios can be deduced for other angles \( \theta_1 \)).
Let us start with those theoretical approaches which are fundamentally "numerical", and which should – within their convergence limits – be "exact". To compare with the measured data, all the FDCS obtained with the CCC approach were rescaled – at the theoretical maximum at $\theta_2 = 263^\circ$ for $\theta_1 = 41^\circ$ – by a factor 3.2 [1], hence $R_{41}(10, 10) = 3.2$. This important magnitude disagreement surprised Kheifets and Bray [5], as the same model gave excellent gauge agreement for $(\gamma, 2e)$ processes with two electrons escaping with 10 eV each. Recently, a purely numerical calculation based on a wave-packet evolution approach [8] yielded $(e, 3e)$ cross sections which are close, in both shape and magnitude, to those found with the CCC approach. However, a magnitude difference of about 20% is observed [9], hence $R_{41}(10, 10) = 2.6$. While these two independent "numerical" approaches support each other, and agree also when applied to $(\gamma, 2e)$ processes, they do not reproduce the absolute scale of the experimental $(e, 3e)$ data. It is worth mentioning that these two calculations were performed using different type of initial states: the CCC used a 20-parameters Hylleraas helium function while the second [8] derived its own wavefunction using a relaxation method. Calculations with the J-matrix approach to Faddeev-Merkuriev differential equations [6, 7] yield a reasonable agreement in $(e, 3e)$ cross sections magnitude, but to a lesser extent in their shapes. It should be noted that previous published calculations [6], where the pseudostates method was employed, showed important magnitude disagreements with experimental data, similarly to the CCC approach. The latest, presumably more converged, calculations [22] give a ratio $R_{41}(10, 10) = 1.2$.

Consider now more "analytical" descriptions of the double continuum. These include the Green’s function expansion [10] which corresponds to the lowest order iteration of an incremental approach to the four-body Coulomb problem. In combination with an independent particle description of the helium bound state, the published results indicate that FDCS magnitudes are reproduced for five $\theta_1$ values, i.e. $R_{\theta_1}(10, 10) \simeq 1$; the cross section for $\theta_1 = 41^\circ$ was not given. The combination of the "pure" 3C double continuum with different bound wavefunctions yields results which depend on the level of correlation included in the initial state. When highly correlated ("advanced") $\Psi_i$ are used for helium [12, 13, 21, 14], a disagreement in magnitude is found, $R_{41}(10, 10) \simeq 0.5$. On the other hand, overall agreement in magnitude is found [11, 12, 15, 3] when using "simple" $\Psi_i$, like that proposed by Pluvinage (which solves or "diagonalizes" the three two-body Coulomb potentials) or others with a similar analytical structure; at $\theta_1 = 41^\circ$ we have $R_{41}(10, 10) = 0.9$ i.e. a theoretical cross section about 1.8 times smaller than with advanced $\Psi_i$.

In summary, when compared to the "numerical" models, the 3C model and the Green’s function expansion, combined with simple helium wavefunctions, lead to the best level of agreement, on the absolute scale, with the experimental set (A). The agreement of the 3C model, however, is fortuitous [3] as will be briefly explained below.

**Table 1.** Ratio $R_{\theta_1}(E_1, E_2)$ defined by (2) obtained with different theoretical models.

|                | $R_{41}(10, 10)$ | $R_{45}(4, 4)$ |
|----------------|------------------|----------------|
| CCC [1]        | 3.2              | 14             |
| Time Dependent [8, 9] | 2.6            | 8.5            |
| J-matrix [6, 7, 22]  | 1.2              | 7              |
| 3C (advanced $\Psi_i$) [Present] and [12, 4] | 0.5              | 30             |
| 3C (simple $\Psi_i$) [Present] and [11, 12] | 0.9              | 46             |

Let us now turn to the second set of data, named (B) hereafter, at $E_1 = E_2 = 4$ eV ejected energies. Contrary to the $E_1 = E_2 = 10$ eV case, the theoretical models do not reproduce so
well the shapes of the 16 measured FDCS. Moreover, large magnitude differences appear again, and yield a picture which differs with the one described above.

The CCC results were rescaled – at the experimental maximum at $\theta_2 = 279^0$ for $\theta_1 = 45^0$ – by a factor 14 [1], hence $R_{45}(4, 4) = 14$. At the same angle, the wave–packet evolution approach [9] reproduce again the CCC shapes but with $R_{45}(4, 4) = 8.5$. The agreement (within 20%) observed for the (A) set does not hold for the (B) case, since the magnitude differs by up to 50%. On the other hand, the J-matrix method gives a similar FDCS shape but with $R_{45}(4, 4) = 7$ [22]. In summary, the three "numerical" approaches, which in principle are exact solutions for the three-body problem, do not give the same conclusions as in the (A) case: they disagree on the absolute scale not only with the experimental data, but also between each other (moreover the relative ratios are different for sets (A) and (B)).

Turning to "analytical" descriptions, some results with the Green’s function expansion were presented in [10] but with an unspecified scaling factor. As the 3C double electron continuum model is expected to be very poor for ejected energies as low as 4 eV, FDCS have not been published previously. Indeed, very important magnitude disagreements are found: $R_{45}(4, 4) = 30$ with advanced helium $\Psi_i$ and $R_{45}(4, 4) = 46$ with simple $\Psi_i$ (present calculations). Note that, again, a ratio of about 1.5 is observed between these two results: while this ratio played in favor in case (A), here it implies a worse disagreement with the measurements. Hence, the 3C magnitude agreement obtained in case (A) has to be considered as fortuitous. This conclusion could be reached after a series of investigations [12, 16, 21, 3, 4] which will now be summarized.

Let us recall that the 3C model consists in describing the double continuum by a symmetrized and orthogonalized version of the pure C3 (or BBK) function [23]. While this function diagonalizes all three two-body Coulomb interactions and has the correct asymptotic behavior when all interparticle distances are large, it describes poorly the behavior at intermediate distances or when one particle is far away from the other two. Previous applications to $(e, 2e)$, $(\gamma, 2e)$ or $(e, 3e)$ processes have shown that the 3C model is able to reproduce reasonably well most of the shapes of the observed differential cross sections, but rather badly their magnitude. Curiously, when combined with simple $\Psi_i$, the 3C model leads to a very good agreement, on the absolute scale, for the $(e, 3e)$ experimental data at $E_1 = E_2 = 10$ eV.

The particular choice used to describe the helium initial state $\Psi_i$ has been thoroughly studied in combination with the 3C double continuum. In [11], the theory-experiment magnitude agreement was directly related to the quality of Pluvinage’s wavefunction, in particular because it diagonalizes the Hamiltonian in all three Coulomb interactions. In [12, 16], it was shown that this property is not the deciding factor. The agreement was otherwise attributed [13] to the fact that the Pluvinage’s wavefunction treat the interactions in the initial and final channels in a ”balanced” way (on equal footing). In order to check this, we have studied [3] the interplay of the 3C final state with several initial bound state descriptions, not only for $(e, 3e)$ but also for $(\gamma, 2e)$ processes. The approximate initial (Pluvinage) and final (3C) wavefunctions appear to be "balanced" for $(e, 3e)$ – but not for $(\gamma, 2e)$ – processes since large $(\gamma, 2e)$ gauge discrepancies are observed. Moreover, Pluvinage wavefunction is not the doubly bound analog of the C3 [24].

As the behavior of $\Psi_i$ near the two-body coalescence points is concerned (Kato cusp conditions [25]), it has been demonstrated, unambiguously, that it has no influence in the experimental $(e, 3e)$ conditions under scrutiny, with helium wavefunctions of both moderate (with several asymptotic behaviors) [12, 3] and very good quality [14, 4].

In view of the present analysis no theoretical model, whether of "numerical" or "analytical" nature, is able to reproduce the absolute scale of all presently available $(e, 3e)$ measurements [1, 2]. Moreover, no agreement is found between numerically exact approaches like the CCC, wave-packet evolution and J-matrix, placing doubts about the approaches themselves since, within the FBA, agreement between all of them is expected. On the more "analytical" side, the 3C model is able to reproduce reasonably the FDCS shapes. However, whatever the chosen
initial state trial wavefunction – diagonal or not, and satisfying or not Kato cusp conditions – the 3C model is not suitable to describe the absolute scale of the considered \((e, 3e)\) processes. The magnitude agreement obtained for set (A) with simple functions should be considered as fortuitous; the combination of simple functions can accidentally yield good absolute results by a lucky compensation of deficiencies.

In spite of these considerations on the absolute scale, we have calculated [4], for the first time, FDCSs for helium-like ions within the 3C model in combination with highly correlated initial states which, by construction, satisfy exactly two-body cusp conditions. No experimental data exist, but under similar kinematical and geometrical conditions, the \(H^-\) cross sections are predicted to be much larger than for helium, while for helium-like positive ions the magnitudes decrease fast with increasing nuclear charges. These results were to be expected, and have been observed in previous publications with, however, only poor models of \(\Psi_i\) and/or \(\Psi_f^\perp\) (see references in [4]). Moreover, thanks to the analytical character of the 3C final state and by adequately scaling the incident and the ejected energies, an approximate scaling law for \((e, 3e)\) FDCS has been mathematically identified along the helium isoelectronic sequence [4]. The scaling law is rather well verified, and can be used to easily predict cross sections which are extremely difficult to measure.

3. Out of plane geometry

Let us now turn to the new out of plane arrangement, recently proposed by Choubisa and Sud [26], which supposedly would allow one to observe second Born effects. In this quasi-binary geometry the two electrons are ejected with the same energy out of the scattering plane, and are such that the momentum of the centre of mass is constant and lies in the scattering plane.

In [26] the authors obtained, within a "3C approach", a series of cross sections for helium which are peculiar. Indeed, in coplanar geometry their results, in first and second Born approximations, are at odds with all other 3C previously published calculations obtained with independent numerical codes [11, 12, 15, 21, 14]: the FDCS differ substantially in both shapes and magnitudes, and moreover show a much larger sensitivity to the choice of the initial state wavefunction. As demonstrated in [27], their FBA calculations do not correspond to the 3C model but an approximate version of it, as later acknowledged by the authors [28]. As it is now known, the model they used (named 2CG in [27]) in which the electron-electron interaction is described only by a Gamow factor, is a poor description of the double continuum. It yields large differences when compared to the 3C model, and cannot reproduce the proper shapes and order of magnitude (it largely underestimates the cross sections) of experimental data. We should also point out that there is in Ref. [26] a magnitude problem related to the initial state normalization which brings in further confusion. As a consequence, Choubisa and Sud [26] observed also important second Born effects which have not been found before in the same kinematics with the 3C model [21] (or with the CCC approach [19]). In summary, in [27] it was made clear that the first (and second) Born calculations presented in [26] (i) do not correspond to a 3C but a 2CG calculation; (ii) show a systematic magnitude problem.

The investigations in the new out of plane geometry [26], whether in first or second Born approximations, are equally flawed. Again, their results are 2CG calculations, and the geometry effects observed were not confirmed within the 3C model. Hence, the new arrangement does not seem, unfortunately, to provide a useful tool to investigate geometrical or second order effects.

4. Concluding remarks

None of the theoretical models discussed here are satisfactory for both \((e, 3e)\) and \((\gamma, 2e)\) processes, so the study of differential double ionization processes cannot be considered as resolved from a theoretical point of view. Further studies are clearly needed to understand the interplay of initial and final states. Numerical approaches, like the CCC or the wave-packet
evolution approach, while working well for \((\gamma,2e)\) processes do not seem to work as well for the \((\epsilon,3e)\) processes, in particular as the absolute scale is concerned. If agreement between these numerically exact approaches were found, and still in disagreement with the experimental data, then one of the two (or the two) statements would emerge: either higher Born orders are necessary or the experimental data are not on the right absolute scale. However, agreement between theories is not observed, bringing more confusion to the subject. The description of the final state by the 3C model manages to reproduce rather well the shapes; however severe magnitude problems appear for low energy ejected electrons. The agreement for two electrons ejected at 10 eV each, in combination with simple initial state wavefunctions, is fortuitous.

In view of the present analysis of \((\epsilon,3e)\) processes, and in order to elucidate the absolute scale divergencies, it would be very useful to have new and independent absolute experimental data (i) in the same kinematical conditions discussed here in order to confirm the measurements of [1, 2]; (ii) for asymmetric ejected energies \(E_1 \neq E_2\) since this would decrease electron-electron correlation effects in the final channel; and (iii) at higher ejected energies \(E_1 = E_2\) as proposed in [3] (this would allow us to (a) confirm the fortuitous character of the 10 eV agreement observed with the 3C model combined with simple functions \(\Psi_i\); (b) to see from which value \(E_1 = E_2\) the 3C model is adequate to describe the double electron continuum).

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References

[1] Kheifets A, Bray I, Lahmam-Bennani, Duguet A and Taouil I 1999 J. Phys. B 32 5047
[2] Lahmam-Bennani A et al. 1999 Phys. Rev. A 59 3548
[3] Ancarani L U, Gasaneo G, Colavecchia F D and Dal Cappello C 2008 Phys. Rev. A. 77 062712
[4] Ancarani L U, Dal Cappello C, Charpentier I, Rodriguez K V and Gasaneo G 2008 Phys. Rev. A 78 062709
[5] Kheifets A S and Bray I, 2004 Phys. Rev. A 69 050701(R)
[6] Knyr V A, Nasirov V V and Popov Yu V 2003 Correlation and Polarization in Photonic, Electronic, and Atomic Collisions (AIP Conf. Proc. Vol. 697) ed G F Hanne et al. (New York: AIP, Melville) p 76
[7] Zaytsev S A, Knyr V A and Popov Yu V 2007 Phys. At. Nucl. 70 676
[8] Serov V V, Derbov V L, Joulakian B B and Vinitsky S I 2007 Phys. Rev. A. 75 012715
[9] Serov V V, private communication
[10] Berakdar J 2000 Phys. Rev. Lett. 85 4036
[11] Jones S and Madison D H 2003 Phys. Rev. Lett. 91, 73201
[12] Ancarani L U, Montagnese T and Dal Cappello C 2004 Phys. Rev. A 70 12711
[13] Jones S, Macek J H and Madison D H 2004 Phys. Rev. A 70 12712
[14] Chulumbaatar O et al. 2006 Phys. Rev. A 74 14703
[15] Colavecchia F D, Gasaneo G and Rodriguez K V 2007 J. Electron Spectrosc. Relat. Phenom. 161 73
[16] Ancarani L U and Dal Cappello C 2007 J. Electron Spectrosc. Relat. Phenom. 161 22
[17] Gasaneo G, Otranto S and Rodriguez K V 2006 Proc. XXIV Int. Conf. on Photonic, Electronic and Atomic Collisions (Singapore: World Scientific) p 360
[18] Ancarani L U, Dal Cappello C and Montagnese T 2006 Ionization, Correlation, and Polarization in Atomic Collisions (AIP Conf. Proc. Vol. 811) ed A Lahmam-Bennani et al. (New York: AIP, Melville) p 1
[19] Kheifets A S 2004 Phys. Rev. A 69 032712
[20] Grim M, Dal Cappello C, El Mkhant R and Rasch J 2000 J. Phys. B 33 131
[21] Ancarani L U, Montagnese T and Dal Cappello C 2005 Electron and Photon Impact Ionization and Related Topics (IOP Conf. Proc. Vol 183) ed B Piraux (London: IOP) p 21
[22] Popov Yu V, private communication
[23] Joulakian B, Dal Cappello C and Brauner M 1992 J. Phys. B 25 2863
[24] Ancarani L U and Gasaneo G 2007 Phys. Rev. A 75 032706
[25] Kato T 1957 Commun. Pure Appl. Math. 10 151
[26] Choubisa R and Sud K K 2008 J. Phys. B 41 035202.
[27] Dal Cappello C and Ancarani L U 2008 J. Phys. B 41 208001
[28] Choubisa R and Sud K K 2008 J. Phys. B 41 208002