Convergent calculations of double ionization of helium: from (γ,2e) to (e,3e) processes

A. S. Kheifets
Research School of Physical Sciences, The Australian National University, Canberra ACT 0200, Australia

Igor Bray
Centre for Atomic, Molecular, and Surface Physics,
School of Mathematical and Physical Sciences, Murdoch University, Perth, 6150 Australia
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The first absolute (e,3e) measurements, by Lahmam-Bennani *et al.* [Phys. Rev. A **59**, 3548 (1999)], have been recently approximately reproduced by Berakdar [Phys. Rev. Lett. **85**, 4036 (2000)] and supported by Jones and Madison [Phys. Rev. Lett. **91**, 07321 (2003)], but with widely differing conclusions. The former indirectly implied that the Born-CCC-based calculations of Kheifets *et al.* [J. Phys. B **32**, 5047 (1999) were invalid due to the reliance on the 1st Born approximation. The latter argued that the 1st Born approximation was valid, but the wrong initial state was used. We investigate these claims and find that the original calculations of Kheifets *et al.* are reproduced whether the 2nd Born approximation is incorporated or if we use a ground state similar to that of Jones and Madison, but appropriately corrected as done by Le Sech and co-workers [J. Phys. B **23**, L739 (1990)].

When attempting calculations of complex collision processes we have found it important to utilise a formalism where convergence of the results can be tested by substantial variation of as many of the input parameters as possible. The goal is to develop a predictive theory whose outcomes can be relied upon on scientific merit irrespective of agreement with experiment or other theory. The convergent close-coupling (CCC) method [1] was developed with this idea in mind. The total wavefunction is expanded utilising a complete Laguerre basis and so greater accuracy is ensured with increasing basis size. In the Coulomb three-body problems, such as e-H scattering, once convergence is reached there is no freedom left to alter the results. Hence, when there is disagreement with experiment as discussed in Ref. [1], that may be even explained by others [2] or improved upon [3], we find that new experiments [4, 5] are supportive of the original CCC results. The predictive power of the CCC approach to double photoionization (DPI or γ,2e) was demonstrated more recently when initial disagreement with experiment [6] was resolved in favor of the CCC calculations [7].

Whereas in e-H calculations convergence considerations relate primarily to the usage of the Laguerre basis, in (γ,2e) calculations there is additional consideration of convergence with respect to the description of the initial state. In the more complicated Coulomb four-body problem that is electron-impact double ionization of helium (e,3e) there are further considerations of convergence with respect to the Born approximation order used to treat the case of a fast projectile. It is these issues that are of interest to us in the present Letter.

The physics of the He (e,3e) reaction in the very fast projectile mode where the incident electron has an order of magnitude or more energy than the two "slow" ejected electrons is quite similar to the case of (γ,2e). In both cases the initial and final atomic states are three-body problems of the He^{2+} nucleus interacting with the two slow electrons. The application of the CCC method to (e,3e) process under these kinematical conditions is straightforward and expected to produce results as accurate as those for (γ,2e). However, the first Born CCC calculation of (e,3e) on He at 5.6 keV incident energy [8] was found significantly lower in magnitude (by factors of ~ 3 and ~ 12 for 10 and 4 eV ejected electrons, respectively) as compared with absolute measurements of Lahmam-Bennani et al. [9]. Such a strong disagreement could only be attributed to deviation from the first Born regime since the treatment of the initial and final states was the same in the CCC (γ,2e) and (e,3e) calculations. Some indications of that followed from the work of Berakdar [10] who reported a good agreement with absolute measurements [9] on the basis of a lowest-order implementation of a Faddeev-type approach. These calculations differed significantly for electron or positron impact (by about a factor of 2) thus invalidating the first Born approximation.

This conclusion, however, was challenged by the recent work of Jones and Madison [11] who managed to get good agreement, both in shape and magnitude, with the experiment of Lahmam-Bennani et al. [9] and the calculations of Berakdar [10] while staying entirely within the first Born formalism. As in the first Born calculations presented in Lahmam-Bennani et al. [9], Jones and Madison [11] used the asymptotically exact 3C final state, but with a different helium ground state. They argued that Hylleraas-based ground states, as employed earlier [8, 9], fail to satisfy the Kato cusp conditions [12] and are inaccurate when the two electrons are close together. In support of their argument, Jones and Madison [11] employed the Pluvinage ground state [13] which treated the inter-electron interaction to all orders of perturbation theory and satisfied Kato's cusp conditions exactly. This combination of the 3C final state and the Pluvinage ground state restored agreement with experiment of Lahmam-

Bennani et al. [9] within the first Born model.

In this Letter, we review our earlier (e,3e) calculations [8] in the light of these new theoretical findings [10, 11]. Following the implementation of the 2nd Born term [14] we are able to test the contribution of this term for the kinematics considered here. We have also implemented the Pluvinage ground state and its improved version as prescribed by Le Sech and co-workers [15, 16], who argued that the former lacked the screening of two-particle interactions by the third particle. Finally, we used a much bigger CCC expansion, than that used previously [8], in describing the final state. The new initial and final states are first applied to helium DPI to check the gauge-dependence as a function of energy. Subsequently, we consider the corresponding (γ,2e) and (e,3e) cases with two 10 eV outgoing electrons. Given the relatively low momentum transfer in the (e,3e) experiment we expect it to be close to the optical limit.

The probability of the (e,3e) reaction is given by the fully-differential cross-section (FDCS):

$$\frac{d\sigma}{d\Omega' d\Omega_1 d\Omega_2\, dE_2} = (2\pi)^4 \frac{k' k_1 k_2}{k_0} \left|T_{fi}\right|^2, \qquad (1)$$

where indices 0,1 and 2 are asigned to the incident and two ejected electrons, respectively. We treat the projectile as a plane wave and write the double ionization amplitude

$$T_{fi} = \frac{4\pi}{q^2} \frac{1}{(2\pi)^3} \left\langle \Psi_{\boldsymbol{k}_1 \boldsymbol{k}_2} \middle| e^{i\boldsymbol{q}\cdot\boldsymbol{r}_1} + e^{i\boldsymbol{q}\cdot\boldsymbol{r}_2} - Z \middle| \Psi_0 \right\rangle \qquad (2)$$

as the Fourier transform of the Coulomb interaction $V = |\boldsymbol{r}_0 - \boldsymbol{r}_1|^{-1} + |\boldsymbol{r}_0 - \boldsymbol{r}_2|^{-1} - Z/r_0$ between the projectile and the target. Here $Z = 2$ is the nucleus charge and $\boldsymbol{q} = \boldsymbol{k}_0 - \boldsymbol{k}'$ is the momentum transfer. In writing Eqs. (1-2) we use a continuum wave normalization $\langle \boldsymbol{k} | \boldsymbol{k}' \rangle = \delta(\boldsymbol{k} - \boldsymbol{k}')$ which gives the incident flux $j = k_0/(2\pi)^3$. By setting $\boldsymbol{q} \to 0$ we reach the optical limit. In this limit angular correlations of the two ejected electrons are the same in (e,3e) and (γ,2e) reactions if we align the vector $\boldsymbol{q}$ with the polarization axis of light. The magnitudes of the corresponding cross-sections differ by a universal scaling factor.

In contrast to the projectile-target interaction, which is treated here in the first and second orders of the perturbation theory, the interaction of the two ejected electrons is taken into account fully. To obtain the two-electron wave function $\Psi_{\boldsymbol{k}_1 \boldsymbol{k}_2}(\boldsymbol{r}_1, \boldsymbol{r}_2)$ we employ the convergent close-coupling (CCC) formalism [8]. In brief, the final state wave function is expanded over the channel functions each of which is a product of a true continuum state and a positive energy pseudostate. The latter is obtained by diagonalizing the target Hamiltonian in the Laguerre square integrable basis. The multiple interaction between the two ejected electrons is included by solving the integral Lippmann-Schwinger equation. Following the successful application of the CCC formalism to (e,2e) equal energy-sharing cross sections [18] we take Laguerre basis exponential fall-off parameters to be the same for all $l \leq 4$ and basis sizes $N_l = 60 - l$. All open plus lowest three closed states were included in the calculations. Previously [8], with smaller computational resources, we were restricted to $N_l = 17 - l$ and had to vary the exponential fall-off parameters for each $l \leq 4$ to avoid inaccuracy associated with interpolation of the complex amplitudes. Some minor variation of the present results from those published previously is due to the very different choices of the Laguerre bases used here.

In our earlier paper [8] we calculated only the first Born amplitude (2) using two accurate ground state wave functions: a 15-term multi-configuration Hartree-Fock and a 20-term Hylleraas. These two wave functions produced nearly identical results in the two gauges of the Born operator - length and velocity. In the present investigation we begin with the Pluvinage ground state:

$$\Psi_0(\boldsymbol{r}_1, \boldsymbol{r}_2) = 8\pi^{-1} e^{-Z(r_1+r_2)}\, N(k)\, \Phi_0(\eta, kr_{12}), \qquad (3)$$

where

$$\Phi_0(\eta, kr_{12}) = e^{-ikr_{12}}\, {}_1F_1(1 - i\eta,\ 2,\ 2ikr_{12}).$$

The non-correlated part of the ground state (3) is the product of the two hydrogenic orbitals in the field $Z = 2$. The correlation factor $\Phi_0(\eta, kr_{12})$ with $\eta = (2k)^{-1}$ describes the center-of-mass motion of the electron pair unimpeded by the nucleus.

Le Sech and co-workers [15, 16] suggested that the original Pluvinage ground state [13] could be improved by introducing the screening of the inter-electron interaction by the nucleus. This can be implemented by changing the effective strength of the Coulomb interaction to $\eta = q\ (2k)^{-1}$ where $q < 1$. In addition, the screening of the electron-nucleus interaction by another electron can be accommodated by introducing the shielding factor $\cosh(\lambda r_1) + \cosh(\lambda r_2)$ in the non-correlated part of the ground state with $\lambda < 1$. This shielding effect is well known and can be incorporated into the simplest non-correlated ground state by introducing an effective charge $Z - 5/16$ where Z is the charge of the nucleus [17].

In Fig. 1 we show the double-to-single photoionization cross-section ratio for He from threshold to intermediate energies where it is highly sensitive to electron correlation in the ground state. We performed calculations with three different ground state wave functions: Pluvinage, Le Sech and Hylleraas. Should the ground and final states be exact, the calculations in the three gauges of the electromagnetic operator, the length, velocity and acceleration, would be identical. Numerical difference between the three gauges is an indicator of the lack of accuracy in the wave functions. As the final state is identical in all three calculations, gauge-dependence indicates the inaccuracy of the ground state. The gauge convergence is worst for the Pluvinage ground state. Only the acceleration gauge, which takes most of its strength at small distances near the nucleus, gives good results. The two other gauges, the length and velocity, which are saturated at large and intermediate distances respectively,

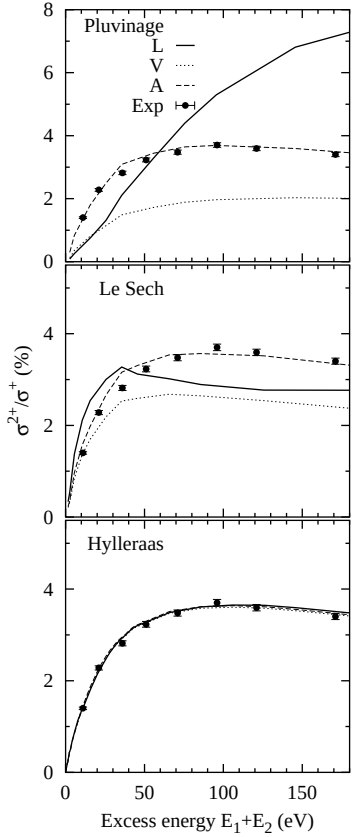

FIG. 1: Double-to-single photoionization cross-section ratio σ^{2+}/σ^{+} as a function of the electron pair energy $E_1 + E_2$. The CCC calculations with different ground states (top - Pluvinage, middle - Le Sech, bottom - Hylleraas) are shown in the length, velocity and acceleration (L - solid line, V - dotted line, and A - dashed line) gauges. The experimental data are from Dörner et al. [19].

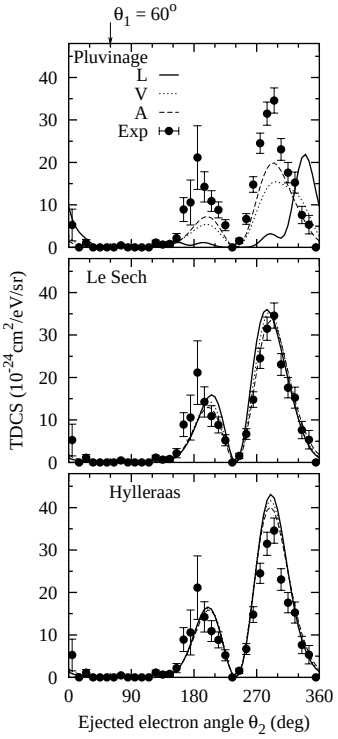

FIG. 2: Triply differential cross section (TDCS) in the polarization plane of light for (γ,2e) on He at $E_1 = E_2 = 10$ eV and the Stokes parameter $S_1 = 0.98$ As in Fig. 1, the CCC calculations with different ground states (top - Pluvinage, middle - Le Sech, bottom - Hylleraas) are shown in the length, velocity and acceleration (L, V, and A) gauges. The line styles are as in Fig. 1. The experimental data are from Bräuning et al. [20].

are in strong disagreement with each other and the experiment [19]. The Le Sech ground state brings significant reduction in the gauge variation, particularly at the energy of interest to us here of 20 eV above threshold. The Hylleraas ground state yields excellent agreement between the three gauges and the experiment.

To investigate the ground state effects further, we calculate the angular distribution of the two equal energy photoelectrons $E_1 = E_2 = 10$ eV in the form of the triply differential cross-section (TDCS) which is obtained from FDCS (1) in the optical limit $q \to 0$. This TDCS is the counterpart of the FDCS reported in the experiments of Lahmam-Bennani et al. [9]. In Fig. 2 we show the TDCS for a fixed angle of one of the photoelectrons whereas the second electron is detected on the full angular range. We choose $\theta_1 = 60^\circ$ where the TDCS is largest in magnitude. As in the case of the total DPI cross-section, the Pluvinage ground state gives the worst results, especially in the length form. Gauge invariance and agreement with experiment is much better with the Le Sech and Hylleraas ground states.

Complete failure of the length gauge with the Pluvinage ground state in DPI has severe implications for (e,3e) calculations which are also performed in the length gauge of the Born operator (2). Results of the CCC calculations of the (e,3e) FDCS is shown on the top row of panels in Fig. 3. As in Figs. 1 and 2, we show calculations with three ground state wave functions: Pluvinage, Le Sech and Hylleraas. In addition, the calculations were performed utilising the first and second Born approximations [14] and found to be indistinguishable (similar conclusion was reached in Ref. [21]). We choose the fixed ejected electron angles at the values where the four-body Green's function calculations of Berakdar [10] are available. Results of this calculation along with the 3C-Pluvinage calculation of Jones and Madison [11] are shown on the bottom panels.

In the top panel, scaling factors of 2.2 and 1.8 have to be applied to the CCC calculations with the Hylleraas and Le Sech ground states, respectively, to match the experiment. The shape of the FDCS calculated with the Pluvinage ground state is very different from the experiment and the other two calculations. A scaling factor of six was applied to this calculation to put it approximately on the same scale as the experiment. On the other

hand, in the bottom panel, the two 3C-based calculations of Berakdar [10] and Jones and Madison [11] agree between themselves and with the absolute measurements of Lahmam-Bennani et al. [9] without any additional scaling. However, we must recall that simply changing the charge of the projectile will destroy the good agreement between the two 3C-based calculations.

Thus, we encounter the extraordinary situation where we have confidence in our results even though they disagree with experiment and two other theories, all of which agree with each other. We do not agree with the implicit conclusion of Berakdar [10] that the first Born approximation is insufficient for the problem at hand. While Jones and Madison [11] also argue this, they use the Pluvinage ground state which we have shown here not to work for DPI or (e,3e) when combined with the CCC final state. However, when this ground state was improved to yield the Le Sech state, which still satisfies the Kato's cusp conditions, the results yielded good agreement with the originally used Hylleraas ground state. We do not accept that the 3C final state can work better than the CCC final state for the kinematics considered irrespective of the initial state. Finally, we have checked the absolute relations between the (γ,2e) and (e,3e) calculations and find them consistent. Consequently, we stand by the original approach to the problem [8]. Fundamentally, we argue that the physics of the (e,3e) process of the type considered here is closely related to the corresponding (γ,2e) process. We are hopeful that this work will stimulate further experimental and theoretical study of the subject.

Acknowledgments

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[1] I. Bray and A. T. Stelbovics, Phys. Rev. A **46**, 6995 (1992).
[2] L. B. Madsen and K. Taulbjerg, Phys. Rev. A **52**, 2429 (1995).
[3] D. P. Dewangan, J. Phys. B **30**, L467 (1997).
[4] H. Yalim, D. Cvejanovic, and A. Crowe, Phys. Rev. Lett. **79**, 2951 (1997).
[5] R. W. O'Neill, P. J. M. van der Burgt, D. Dziczek, P. Bowe, S. Chwirot, and J. A. Slevin, Phys. Rev. Lett. **80**, 1630 (1998).
[6] A. S. Kheifets and I. Bray, Phys. Rev. Lett. **81**, 4588 (1998).
[7] M. Achler, V. Mergel, L. Spielberger, R. Dörner, Y. Azuma, and H. Schmidt-Böcking, J. Phys. B **34**, 965 (2001).
[8] A. S. Kheifets, I. Bray, A. Duguet, A. Lahmam-Bennani, and I. Taouil, J. Phys. B **32**, 5047 (1999).
[9] A. Lahmam-Bennani, I. Taouil, A. Duguet, M. Lecas, L. Avaldi, and J. Berakdar, Phys. Rev. A **59**, 3548 (1999).
[10] J. Berakdar, Phys. Rev. Lett. **85**, 4036 (2000).
[11] S. Jones and D. H. Madison, Phys. Rev. Lett. **91** (2003).
[12] T. Kato, Commun. Pure Appl. Math. **10**, 151 (1957).
[13] P. Pluvinage, Ann. Phys. (N.Y.) **5**, 145 (1950).
[14] A. S. Kheifets, Phys. Rev. A p. submitted (2004).
[15] A. Moumeni, O. Dulieu, and C. L. Sech, J. Phys. B **23**, L739 (1990).
[16] L. D. A. Siebbeles, D. P. Marshall, and C. L. Sech, J. Phys. B **26**, L321 (1993).
[17] H. A. Bethe and E. E. Salpeter, *Quantum mechanics of one-and two-electron atoms* (Plenum, New York, 1977).
[18] I. Bray, Phys. Rev. Lett. **89**, 273201 (2002).
[19] R. Dörner, T. Vogt, V. Mergel, H. Khemliche, S. Kravis, and C. L. Cocke, Phys. Rev. Lett. **76**, 2654 (1996).
[20] H. Bräuning, R. Dörner, C. L. Cocke, M. H. Prior, B. Krässig, A. S. Kheifets, I. Bray, A. Bräuning-Demian, K. Carnes, S. Dreuil, et al., J. Phys. B **31**, 5149 (1998).
[21] J. Rasch, H. R. J. Walters, P. Marchalant, C. Whelan, and D. H. Madison, in *Photonic, Electronic, and Atomic Collisions (XXII ICPEAC)*, edited by J. Burgdorfer, J. Cohen, S. Datz, and C. Vane (Rinton Press, Princeton, USA, 2002), pp. 448–459.

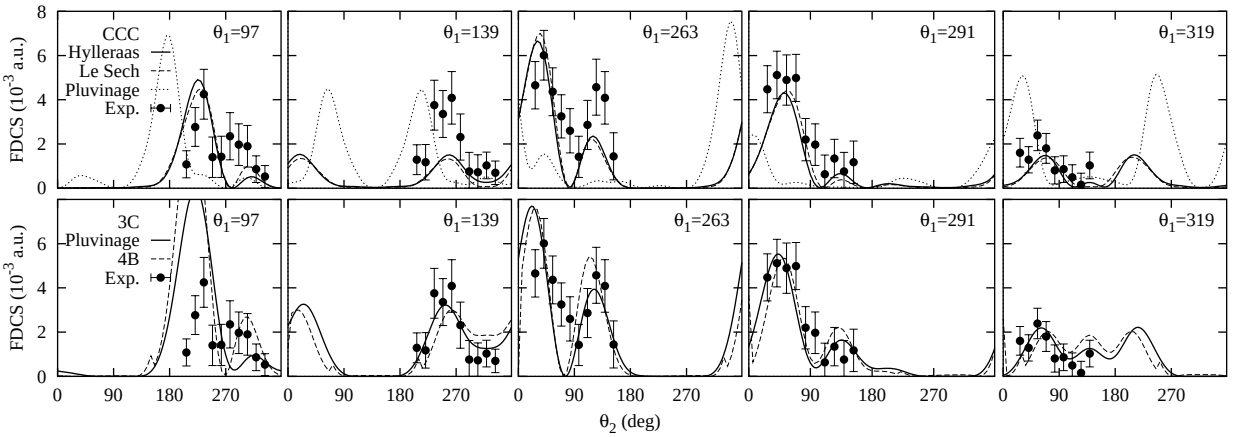

FIG. 3: Fully differential cross section (FDCS) in the scattering plane for (e,3e) on He at $E_0 = 5.6$ keV, $E_1 = E_2 = 10$ eV and $q = 0.24$ a.u. for selected fixed ejected electron angles θ_1. The CCC calculations with different ground state wave functions are shown on the top row of panels. The Pluvinage, Le Sech and Hylleraas ground state results are shown by the dotted, dashed and solid lines and multiplied by factors of 6, 1.8 and 2.2, respectively, to match the experiment of Lahmam-Bennani et al. [9]. On the bottom row of panels we show the Pluvinage-3C calculation of Jones and Madison [11] and the four-body (4B) Green's function calculation of Berakdar [10].