Angular distributions for double photoionization of helium: Discrepancies between theory and experiment

Marcel Pont and Robin Shakeshaft
Physics Department, University of Southern California, Los Angeles, California 90089-0484

Fritz Maulbetsch and John S. Briggs
Fakultät für Physik, University of Freiburg, Hermann-Herder-Strasse 3, 79104 Freiburg, Germany
(Received 18 September 1995; revised manuscript received 18 January 1996)

We apply two different methods to the calculation of the triply differential cross section (TDCS) for double ionization of helium. In one method, the 3C method, the final state is described by a product of 3 Coulomb continuum wave functions, while in the other method, the 2SC method, the final state is described by a product of 2 *screened* Coulomb wave functions employing effective charges. We present results at the energies and geometries covered in the recent experiment by Lablanquie *et al.* [Phys. Rev. Lett. **74**, 2192 (1995)]. There are substantial discrepancies. We also comment on a recent measurement of the *absolute* TDCS by Schwarzkopf and Schmidt [J. Phys. B **28**, 2847 (1995)]. The measured result, as originally reported, is a factor of two smaller than the value for the absolute TDCS calculated using the 2SC method. We discuss this discrepancy. [S1050-2947(96)08305-9]

PACS number(s): 32.80.Fb

Lablanquie *et al.* [1] have reported on measurements of the coplanar triply differential cross section (TDCS) for double photoionization of helium. Here we compare results of two different calculations of the TDCS with their data. The overall agreement between theory and experiment is fairly poor, particularly when contrasted with the good agreement found in a separate comparison [2] with the TDCS data from two recent experiments by Schwarzkopf and co-workers [3,4] and Dawber *et al.* [5]. On the other hand, there are also significant discrepancies between the results of the calculations — see especially Fig. 2(b) below — suggesting that the energy range studied by Lablanquie *et al.* remains a challenging one to theorists.

We address also in this paper a recent *absolute* measurement of the TDCS for double photoionization of helium by Schwarzkopf and Schmidt [4]. We comment on a factor of two discrepancy that was originally reported between the measured result and the result calculated by Pont and Shakeshaft [6].

The methods underlying the calculations have been described in detail elsewhere [2,6–8], and so we give only a brief sketch here. The calculations differ in the evaluation of the dipole matrix element, $f(\mathbf{k}_1,\mathbf{k}_2)$, for two electrons to emerge with momenta $\mathbf{k}_1$ and $\mathbf{k}_2$ and energies $E_1 \equiv k_1^2/2$ and $E_2 \equiv k_2^2/2$. In one approach [7] — the 3C method — the final-state wave function $\Psi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2}$ is approximated by a product of 3 Coulomb continuum wave functions [9], which take final-state correlation directly into account, and the dipole matrix element is evaluated directly as $f(\mathbf{k}_1,\mathbf{k}_2) = \langle \Psi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2} | V | \Psi_i \rangle$, where Ψ_i is the initial-state wave function and V is the atom-photon interaction. In the other approach [8] — the 2SC method — $\Psi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2}$ is replaced by a product of 2 *screened* Coulomb wave functions [10] employing effective charges [11], and is substituted into the flux formula $f(\mathbf{k}_1,\mathbf{k}_2) = \langle \Psi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2} | (H_a^\dagger - H_a) | \mathscr{F}^+ \rangle$, where H_a is the Hamiltonian of the bare atom and where $\mathscr{F}^+$ represents the *perturbed* initial state, satisfying the inhomogeneous equation

$$(E - H_a) | \mathscr{F}^+ \rangle = V | \Psi_i \rangle, \tag{1}$$

with $E \equiv E_1 + E_2$ the excess energy above threshold. Equation (1) is solved on a two-electron basis composed of products of one-electron complex radial Sturmian functions and spherical harmonics. The flux formula can be transformed to a surface integral whose value is determined in the region of classical asymptotic motion of both electrons, a region where the 2SC wave function is exact. Hence, in principle, though not in practice, the 2SC method can yield exact results.

In Figs. 1 and 2, we show our results along with the data of Lablanquie *et al.*, for both (a) equal and (b) and (c) unequal energy sharing. The different plots have been rescaled so that the TDCS has the same value at its maximum for all sets of data in a given case. In Table I we give the absolute value, as predicted by the 2SC calculation, of the TDCS at its maximum, for each case shown in Figs. 1 and 2. The 2SC calculation was done in the velocity gauge.

TABLE I. 2SC estimate of absolute TDCS at its maximum for each case in Figs. 1 and 2.

Fig.	TDCS
1(a)	2.5(-5)
1(b)	2.3(-5)
1(c)	2.4(-5)
2(a)	1.1(-5)
2(b)	1.2(-5)
2(c)	1.2(-5)

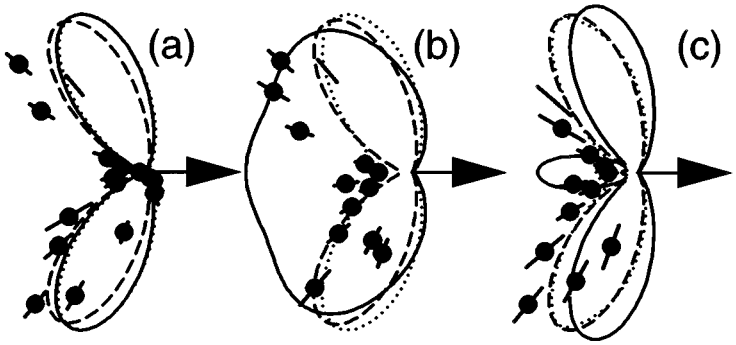

FIG. 1. Polar plots of coplanar TDCS. Electron 1 emerges along the polarization axis (see arrow), and $E=4.0$ eV, with (a) $E_1=E_2=2.0$ eV; (b) $E_1=3.3$ eV, $E_2=0.7$ eV; and (c) $E_1=0.7$ eV, $E_2=3.3$ eV. Experimental data are from Ref. [1]. Solid and dashed lines are from velocity- and length-gauge versions of 3C theory, respectively, and the dotted line is from 2SC theory. Plots have been rescaled so that TDCS has same value at its maximum.

The agreement between the (rescaled) results of the 2SC calculation and the length-gauge version of the 3C calculation is good in all cases apart from the two unequal-energy-sharing cases at higher excess energy, Figs. 2(b) and 2(c), but the substantial discrepancies between the results of the length- and velocity-gauge versions of the 3C calculations indicate that the 3C method is less reliable for the energies covered by the experiment of Lablanquie *et al.* [1] than for the energies covered by the experiments of Schwarzkopf and co-workers [3,4] and Dawber *et al.* [5]. The 2SC calculation was not performed in the length gauge since the 2SC method involves a surface integral of (asymptotically) large radius, and in the length gauge the atom-field interaction diverges on this surface; consequently, convergence would be very difficult, perhaps impossible, to achieve. Finally, we observe that the qualitative agreement between the results of the 2SC calculation and the experimental data is rather poor; the 2SC results lie, for the most part, well outside the error bars of the experiment — see Fig. 2(b) in particular.

In the absence of further theoretical and experimental work, it is not possible to decide where the error lies. We note, however, that the 2SC calculations yield values for the integrated cross section for double escape that agree almost perfectly [12] in the near-threshold region (at energies of 2 eV and not far above) with the data from the experiments of Kossmann, Schmidt, and Andersen [13] and Bizau and Wuilleumier [14]. (The 2SC calculations do not converge to satisfactory accuracy below 2 eV above threshold, and we chose to ignore them, but if a spline fit is made to the 2SC estimates of the cross section at 2 eV and above, and to the accepted result of zero at 0 eV, the threshold energy dependence predicted by Wannier [15] is confirmed to within 2%.) Furthermore, data from two recent measurements [16,17] of the ratio of the cross sections for double to single (or total) ionization agree to within a few percent with the 2SC results over the energy range 2–80 eV above threshold (the 2SC calculations do not converge to high accuracy at energies in excess of 80 eV above threshold).

Despite the evidence, quoted in the preceding paragraph, in support of the accuracy of the 2SC method, a troubling factor of 2 discrepancy with a measurement (the first and only such measurement, as far as we are aware) of the *absolute* coplanar TDCS was recently reported on by Schwarzkopf and Schmidt [4]. The shape of the measured TDCS is in very good agreement with the shape calculated [6] using the 2SC method, but Schwarzkopf and Schmidt [4] reported the absolute values of the TDCS to be about a factor of 2 smaller than those calculated using the 2SC theory. We now argue that one can estimate, to an accuracy of well within a factor of 2, the *integrated* cross section by using the analytic form taken by Schwarzkopf and Schmidt to fit their data for the TDCS. The result thereby obtained turns out to be smaller than the measured result [13,14,17] for the integrated cross section by about a factor of 2. We begin by reproducing Fig. 3 of the paper by Schwarzkopf and Schmidt [4]. This figure, which is also Fig. 3 of the present paper, shows the TDCS in the case of equal energy sharing when the excess energy E is 20 eV, when the Stokes parameter S_1 of the light is 0.59, and when electron 1 emerges along the major axis of polarization (see arrow). The scale is absolute, i.e., the TDCS has *not* been rescaled so that it has the same value at its maximum for all sets of data. The solid curve represents the 3C

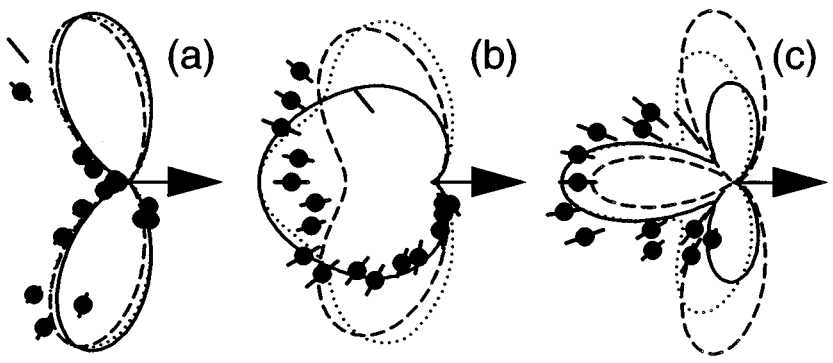

FIG. 2. Same as Fig. 1 but with $E=18.6$ eV and (a) $E_1=E_2=9.3$ eV; (b) $E_1=15.6$ eV, $E_2=3.0$ eV; and (c) $E_1=3.0$ eV, $E_2=15.6$ eV.

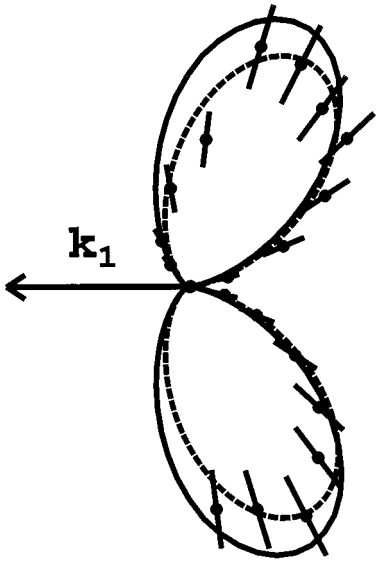

FIG. 3. Polar plots of coplanar TDCS on an absolute scale. The Stokes parameter S_1 is 0.59, electron 1 emerges along the major polarization axis (see arrow), and $E_1=E_2=10.0$ eV. Experimental data are from Ref. [4]. Solid and broken lines are from the velocity-gauge version of 3C theory and fit to experimental data (see text), respectively.

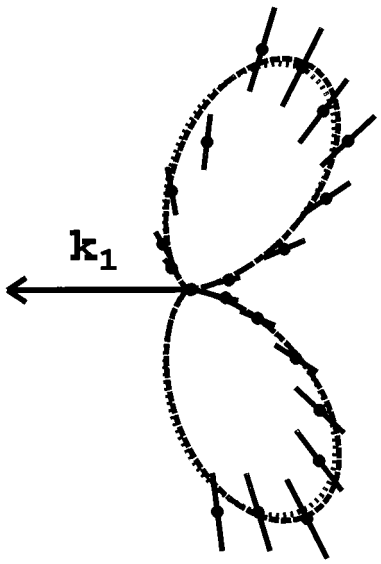

FIG. 4. Same as Fig. 3 but with experimental data, and its fit, multiplied by 2.2, and with 3C data omitted. The dotted line is the 2SC result from Ref. [6] but with $S_1=0.59$.

velocity-gauge results of Maulbetsch and Briggs, and the broken line is a fit to the experimental data (see below). In the case of equal energy sharing, the TDCS for double photoionization can be expressed as [4]

$$\frac{d^3\sigma^{2+}(E_1=E/2)}{dE_1 d\Omega_1 d\Omega_2}=\left[\left(\frac{1+S_1}{2}\right)(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{e}}_x+\hat{\mathbf{k}}_2\cdot\hat{\mathbf{e}}_x)^2+\left(\frac{1-S_1}{2}\right)(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{e}}_y+\hat{\mathbf{k}}_2\cdot\hat{\mathbf{e}}_y)^2\right]|\tilde{f}(\mu)|^2, \quad (2)$$

where $\hat{\mathbf{e}}_y$ and $\hat{\mathbf{e}}_y$ are orthogonal unit vectors in the plane of polarization of the light, $\hat{\mathbf{k}}_1$ and $\hat{\mathbf{k}}_2$ are unit vectors along the directions of emission of the electrons, $\mu=\cos\theta_{12}$, with θ_{12} the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$, and $|\tilde{f}(\mu)|^2$ is a quantity which is independent of S_1 and, following Huetz *et al.* [18], can be approximated by the analytic form [4]

$$|\tilde{f}(\mu)|^2=a\exp[-4\ln 2(180°-\theta_{12})^2/(\theta_0 E^{1/4})^2], \quad (3)$$

where a and θ_0 are parameters which were chosen to optimize the fit to the experimental data, i.e., $a=57(9)$ b cm^2 sr /eV and $\theta_0=43(1)°$ (eV)$^{-1/4}$ [4]. The fit to the experimental data, in Fig. 3, is seen to be very good. Now, integrating the right-hand side of Eq. (2) over solid angles of both electrons, the singly differential cross section can, in the cases of equal energy sharing and $S_1=1$, be expressed as

$$\frac{d\sigma^{2+}(E_1=E/2)}{dE_1}=\frac{(4\pi)^2}{3}\int_{-1}^{1}d\mu(1+\mu)|\tilde{f}(\mu)|^2, \quad (4)$$

which is in agreement with the expression used by Pont and Shakeshaft; see, e.g., [12]. If we assume that the energy distribution is flat — both experiment [19] and theory [12] indicate that, in the region of $E=20$ eV, this distribution is flat to within a few percent — we need only multiply the singly differential cross section by the factor $E/2$ to obtain the integrated cross section. It is important to note that we multiply by $E/2$ rather than E since the energy distribution is symmetric about $E/2$ and were we to multiply by E, we would be counting the same event twice. Inserting the analytic form of Eq. (3), with the experimentally deduced values of the parameters a and θ_0, into the right-hand side of Eq. (4) gives, for $E=20$ eV and now $S_1=1$, a singly differential cross section of 0.40 kb/eV; multiplying by $E/2$ gives, for the integrated cross section for double photoionization, the value 4.0 kb. On the other hand, the 2SC calculations yield a singly differential cross section of 0.89 kb/eV for $E=20$ eV and $S_1=1$; multiplying by $E/2$ gives, for the integrated cross section, the value 8.9 kb, compared to the value 9.1 kb which is obtained by integrating without further approximation over the 2SC energy distribution. The value 9.1 kb is close to the measured values [13,14,17] of the integrated cross section at $E=20$ eV. This indicates that the absolute measurement of the TDCS by Schwarzkopf and Schmidt [4] may be too small by a factor of two. Schwarzkopf and Schmidt have independently arrived at the same conclusion that their data should be revised upward by a factor of about 2 [20]. If we multiply the measured values of the TDCS, and also the fitted values, by 8.9/4.0 (= 2.2), and if we compare with the 2SC calculated values, again on an absolute scale, we obtain excellent agreement; see Fig. 4.

This work was supported by the DOE, Division of Chemical Sciences, by the NCSA at Urbana-Champaign, by the DFG in SFB 276, TP A2, and by a ''DAAD-Doktorandenstipendium.'' F.M. gratefully acknowledges the hospitality of the University of Southern California where he was a visitor during the time the work was completed.

[1] P. Lablanquie, J. Mazeau, L. Andric, P. Selles, and A. Huetz, Phys. Rev. Lett. **74**, 2192 (1995).
[2] F. Maulbetsch, J. S. Briggs, M. Pont, and R. Shakeshaft, J. Phys. B **28**, L341 (1995).
[3] O. Schwarzkopf, B. Krässig, J. Elmiger, and V. Schmidt, Phys. Rev. Lett. **70**, 3008 (1993); O. Schwarzkopf, Ph.D. thesis, University of Freiburg, 1995.
[4] O. Schwarzkopf and V. Schmidt, J. Phys. B **28**, 2847 (1995).
[5] G. Dawber, L. Avaldi, A. G. McConkey, H. Rojas, M. A. MacDonald, and G. C. King, J. Phys. B **28**, L271 (1995).
[6] M. Pont and R. Shakeshaft, Phys. Rev. A **51**, R2676 (1995).
[7] F. Maulbetsch and J. S. Briggs, J. Phys. B **26**, 1679 (1993); L647 (1993); **27**, 4095 (1994).
[8] D. Proulx and R. Shakeshaft, Phys. Rev. A **48**, R875 (1993).
[9] C. Garibotti C and J. E. Miraglia, Phys. Rev. A **21**, 572 (1980); M. Brauner, J. S. Briggs, and H. Klar, J. Phys. B **22**, 2265 (1989).
[10] M. R. H. Rudge and M. J. Seaton, Proc. R. Soc. London Ser. A **283**, 262 (1965).
[11] M. R. H. Rudge, Rev. Mod. Phys. **40**, 564 (1968); C. Pan and A. F. Starace, Phys. Rev. Lett. **67**, 185 (1991); S. Jetzke and F. H. M. Faisal, J. Phys. B **25**, 1543 (1992).
[12] M. Pont and R. Shakeshaft, J. Phys. B **28**, L571 (1995).
[13] H. Kossmann, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1266 (1988); H. Kossmann, Ph.D. thesis, Freiburg University, 1992.
[14] J. M. Bizau and F. J. Wuilleumier, J. Electron Spectrosc. Relat. Phenom. **71**, 205 (1995).
[15] G. H. Wannier, Phys. Rev. **90**, 817 (1953); R. Peterkop, J. Phys. B **4**, 513 (1971); A. R. P. Rau, Phys. Rev. A **4**, 207 (1971).
[16] R. Dörner (private communication).
[17] J. Samson (private communication).
[18] A. Huetz, P. Selles, D. Waymel, and J. Mazeau, J. Phys. B **24**, 1917 (1991).
[19] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[20] V. Schmidt (private communication).