

## Absolute triply differential cross sections for double photoionization of helium at 10, 20, and 52.9 eV above threshold

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We have calculated the coplanar triply differential cross section (TDCS) for double ionization of helium by one photon at total energies of 10, 20, and 52.9 eV above threshold. Our results agree very well with recently measured and calculated angular distributions [O. Schwarzkopf, B. Krässig, J. Elmiger, and V. Schmidt, *Phys. Rev. Lett.* **70**, 3008 (1993); O. Schwarzkopf, B. Krässig, V. Schmidt, F. Maulbetsch, and J. S. Briggs, *J. Phys. B* **27**, L347 (1994); F. Maulbetsch and J. S. Briggs, *ibid.* **26**, L647 (1993)] for the cases of equal energy sharing (photoelectron energies  $E_1$  and  $E_2$ , such that  $E_1=E_2=5$  eV and  $E_1=E_2=10$  eV) and unequal energy sharing ( $E_1=5$  eV,  $E_2=47.9$  eV). In contrast to previous calculations of the TDCS, our calculation is based on a basis-set method that is, in principle, exact, and we obtain *absolute* values of the TDCS that are fairly well converged with respect to the basis size.

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The triply differential cross section (TDCS) for double ionization of helium by one photon provides a sensitive tool for exploring electron-electron correlation in photoionization processes. However, this quantity is neither easy to calculate nor easy to measure. Nevertheless, Schwarzkopf *et al.* [1–3] recently measured, for the first time, relative values of the coplanar TDCS for double photoionization of helium at total energies from 10 to 53 eV above threshold. They investigated both equal energy sharing, where the photoelectron energies  $E_1$  and  $E_2$  were such that  $E_1=E_2=5$  eV or  $E_1=E_2=10$  eV, and unequal energy sharing, where  $E_1=5$  eV,  $E_2=47.9$  eV. Maulbetsch and Briggs [3,4], using an approximate final-state wave function, which is a product of three Coulomb wave functions [5,6], succeeded in calculating angular distributions whose shapes are in very good agreement with the measured data. However, while this three-Coulomb wave function is correct at large distances [i.e.,  $(k_1 r_1)(k_{12} r_{12}) \gg 1$  and  $(k_2 r_2)(k_{12} r_{12}) \gg 1$ , where these quantities are defined below], it is incorrect at small distances, and since the absolute probability for ionization is determined in the region near the nucleus one cannot expect to obtain accurate absolute probabilities; indeed, the three-Coulomb wave function yields absolute values of the TDCS that are believed to be substantially incorrect over a region of a hundred or so eV (perhaps more) above threshold [7,8]. In this paper we present results of basis-set calculations of the TDCS; the angular distributions are also in very good agreement with the measured data, and in addition the absolute values of the TDCS are fairly well converged with respect to the size of the basis.

The method used here was used earlier to calculate the asymmetry parameter and energy distribution for double photoionization of helium [9]. The calculation of the TDCS poses a far bigger challenge, since convergence with respect to basis size is more difficult to achieve. We sketch the method only briefly here, and with a slightly different emphasis from Ref. [9] (in particular, we pay attention to the possibility of spurious contributions from single ionization). We work in the velocity gauge, use atomic units, and ignore

spin (we factor out the spin). The Hamiltonian of the atom is  $H_a \equiv \mathbf{p}_1^2/2 + \mathbf{p}_2^2/2 - Z/r_1 - Z/r_2 + 1/r_{12}$ , where  $\mathbf{p}_1$  and  $\mathbf{p}_2$  are the canonical momentum operators for the electrons,  $Z$  is the atomic number of the atom, and  $\mathbf{r}_1$  and  $\mathbf{r}_2$  are the electron coordinates relative to the nucleus, with  $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2$ ,  $r_1 = |\mathbf{r}_1|$ ,  $r_2 = |\mathbf{r}_2|$ , and  $r_{12} = |\mathbf{r}_{12}|$ . Let  $\mathbf{k}_1$  and  $\mathbf{k}_2$  be the final momenta of the two electrons, with  $E_1 \equiv k_1^2/2$  and  $E_2 \equiv k_2^2/2$  their final energies. The triply differential cross section for the atom to absorb one photon, of frequency  $\omega$ , and for the two electrons to emerge into solid angles  $d\Omega_1$  and  $d\Omega_2$  is

$$\frac{d\sigma}{dE_1 d\Omega_1 d\Omega_2} = \frac{4\pi^2}{\omega c} k_1 k_2 |f(\mathbf{k}_1, \mathbf{k}_2)|^2. \quad (1)$$

Here the amplitude  $f(\mathbf{k}_1, \mathbf{k}_2)$  is

$$f(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)} | \boldsymbol{\epsilon} \cdot (\mathbf{p}_1 + \mathbf{p}_2) | \Psi_i \rangle, \quad (2)$$

where  $\boldsymbol{\epsilon}$  is the unit polarization vector for the light, and where  $|\Psi_i\rangle$  (normalized to unity) and  $|\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}\rangle$  (symmetrized and normalized on the momentum scale) represent the initial and final states of the atom and are eigenvectors of  $H_a$  with eigenvalues  $E_i$  and  $E_f \equiv E_i + \omega = E_1 + E_2$ , respectively. The total cross section  $\sigma$  for double ionization is obtained by integrating over the full solid angle of each electron, and over  $E_1$  from 0 to  $E_f/2$ ; since we symmetrized  $|\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}\rangle$ , this vector describes the same state as  $|\Psi_{\mathbf{k}_2, \mathbf{k}_1}^{(-)}\rangle$ , and we must restrict  $E_1$  to be smaller than  $E_2$  to avoid double counting. For computational purposes it is convenient to first reexpress the amplitude as the flux formula

$$f(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)} | (H_a^\dagger - H_a) | \mathcal{F}^+ \rangle, \quad (3)$$

where  $|\mathcal{F}^+\rangle$  satisfies the inhomogeneous equation

$$(E_f - H_a) | \mathcal{F}^+ \rangle = \boldsymbol{\epsilon} \cdot (\mathbf{p}_1 + \mathbf{p}_2) | \Psi_i \rangle. \quad (4)$$

The wave function  $\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  describes two electrons that emerge with momenta  $\mathbf{k}_1$  and  $\mathbf{k}_2$  at asymptotically large

distances after scattering from each other and from the nucleus, whereas the wave function  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$  describes the initially bound atom after it has absorbed one photon. While  $\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  has both incoming- and outgoing-wave components,  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$  has only outgoing-wave components, and when  $r_1$  and  $r_2$  are both large  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$  has the asymptotic form

$$\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2) \rightarrow \sqrt{\pi K^3} f(\mathbf{k}_1, \mathbf{k}_2) \frac{e^{iKR - i\gamma \ln(2KR)}}{R^{5/2}}, \quad (5)$$

where  $R \equiv \sqrt{r_1^2 + r_2^2}$  is the hyperradius,  $K = \sqrt{k_1^2 + k_2^2}$ , and  $\gamma = (R/K)(-Z/r_1 - Z/r_2 + 1/r_{12})$ . Using Green's theorem the volume integral on the right-hand side of Eq. (3) can be expressed as a surface integral over a hypersphere of very large hyperradius  $R$ . The surface integral can be evaluated upon inserting the asymptotic forms of  $\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  and  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$ . Although  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$  and (the incoming wave part of)  $\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  both contain components that describe one free electron and one bound electron, it can be shown that there is no contribution to the surface integral from the region where  $\alpha \equiv \tan^{-1}(r_1/r_2)$  is near 0 or  $\pi/2$ , i.e., where the ratio  $r_1/r_2$  or  $r_2/r_1$  is very small. Hence there is no contribution from single ionization. This remains true if the asymptotic form of Eq. (5) is used over the entire hypersphere, even though it is invalid in the region  $\alpha \approx 0$  or  $\pi/2$ ; the "spurious" contribution from this region oscillates with  $R$  and averages to zero. The only contribution to the surface integral comes from the point of stationary phase of the integrand, which corresponds to the classical asymptotic motion  $\mathbf{r}_1 \approx \mathbf{k}_1 t$  and  $\mathbf{r}_2 \approx \mathbf{k}_2 t$ , where  $t \equiv R/K$  (the "time") is a large parameter [10,11]. Therefore we can tentatively replace  $\Psi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  by a trial wave function  $\chi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  that satisfies the correct boundary conditions in the directions of classical asymptotic motion of two free electrons, provided that the surface contribution from other directions oscillates with  $R$  and averages to zero. Following Rudge and Seaton [10] we choose  $\chi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2)$  to be a product of two one-particle wave functions that describes the electrons moving independently in the Coulomb potential  $-(Z_1/r_1) - (Z_2/r_2)$ , where the velocity-dependent effective charges

$Z_1$  and  $Z_2$  are constrained by the stationary phase condition and may be chosen as [11–13]  $Z_i = Z - \Delta_i$ ,  $i=1,2$  with  $Z=2$  and

$$\Delta_i = \frac{(\mathbf{k}_i \cdot \mathbf{k}_{ij}) k_i}{k_{ij}^3}, \quad (6)$$

where  $\mathbf{k}_{ij} = \mathbf{k}_i - \mathbf{k}_j$ ,  $j \neq i$ , and  $k_{ij} = |\mathbf{k}_{ij}|$ ; we have

$$\begin{aligned} \chi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2) &= \sqrt{2} (2\pi)^{-3} e^{i\mathbf{k}_1 \cdot \mathbf{r}_1 + i\mathbf{k}_2 \cdot \mathbf{r}_2} \\ &\times \prod_{j=1,2} e^{-\pi\gamma_j/2} \Gamma(1 - i\gamma_j) \\ &\times {}_1F_1(i\gamma_j, 1, -ik_j r_j - i\mathbf{k}_j \cdot \mathbf{r}_j), \quad (7) \end{aligned}$$

where  $\gamma_j = -Z_j/k_j$  and where the prefactor  $\sqrt{2}$  compensates for not symmetrizing the trial wave function. Hence Eq. (3) simplifies to

$$f(\mathbf{k}_1, \mathbf{k}_2) = \langle \chi_{\mathbf{k}_1, \mathbf{k}_2}^{(-)} | (E_f + W - H_a) | \mathcal{F}^+ \rangle, \quad (8)$$

where  $W$  is the potential

$$W = \frac{1}{r_{12}} - \frac{\Delta_1}{r_1} - \frac{\Delta_2}{r_2}. \quad (9)$$

However, the right-hand side of Eq. (8) involves a volume integral that is formally divergent, since  $W$  falls off asymptotically as a Coulomb potential except along the lines  $r_1 = k_1 t$ ,  $r_2 = k_2 t$ , and  $r_{12} = k_{12} t$ , where  $W$  vanishes. Nevertheless, since the integrand oscillates the integral can be given meaning in the Lebesgue sense. Furthermore, it can be shown that the surface contribution from single ionization, from the region  $\alpha \approx 0$  or  $\pi/2$ , oscillates with  $R$  and averages to zero. To evaluate the integral on the right-hand side of Eq. (8) we represent the amplitude by a (divergent) series, and analytically continue this representation by Padé summation [14,15]. We obtain a series representation of  $f(\mathbf{k}_1, \mathbf{k}_2)$  by expanding  $\mathcal{F}^+(\mathbf{r}_1, \mathbf{r}_2)$  on a two-electron basis consisting of terms  $S_{n_l}^K(r_1) S_{n'_l}^K(r_2) Y_{ll'}^{LM}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)$ , where  $Y_{ll'}^{LM}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)$  couples spherical harmonics and where  $S_{n_l}^K(r)$  is a radial Sturmian function that is a polynomial of degree  $n_r \equiv n - l - 1$  multiplied by  $r^l e^{iKr}$

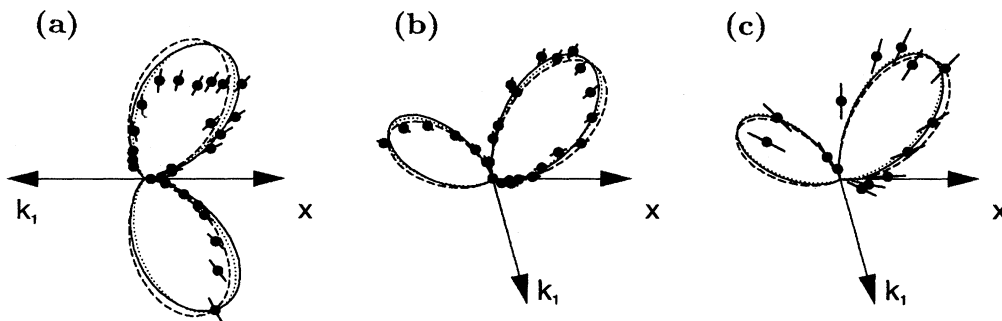


FIG. 1. The coplanar triply differential cross section (TDCS) for equal energy sharing when  $\theta_1$  is fixed: (a)  $\theta_1 = 180^\circ$ ,  $E_1 = 10$  eV; (b)  $\theta_1 = 284^\circ$ ,  $E_1 = 10$  eV; and (c)  $\theta_1 = 284^\circ$ ,  $E_1 = 5$  eV. The measured data are from Schwarzkopf *et al.* [1,2]; the dashed and dotted lines are from the velocity and length gauge versions, respectively, of the theory of Maulbetsch and Briggs [4]; and the solid line represents the present results.

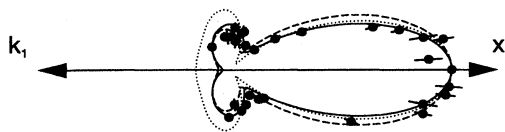


FIG. 2. The coplanar TDCS for unequal energy sharing when  $\theta_1$  is fixed:  $\theta_1 = 180^\circ$ ,  $E_1 = 5$  eV, and  $E_2 = 47.9$  eV. The measured data are from Schwarzkopf *et al.* [3]; the dashed and dotted lines are from the velocity and length gauge versions, respectively, of the theory of Maulbetsch and Briggs [3]; and the solid line represents the present results.

We now show results of calculations for the TDCS for one-photon ionization of ground-state helium. In our calculations we employed a very large basis, consisting of 34 radial functions  $S_{nl}^{\kappa}(r)$  per  $l$  (per electron) with  $0 \leq l \leq 5$ . The “wave number”  $\kappa$  was chosen to lie in the upper right quadrant of the complex  $\kappa$  plane ( $|\kappa| = 1.2$ ,  $\arg \kappa = 80^\circ$ ) so as to accommodate both outgoing-wave open channels and exponentially decaying closed channels. In order to reduce round-off error in matrix elements of the electron-electron repulsion, which can accumulate rapidly as the basis size increases, we devised a numerical method (to be described elsewhere) for accurately evaluating these matrix elements. This method is especially suited to parallel computation, and our calculation was carried out on the massively parallel Connection Machine, CM-5. In the experiments of Schwarzkopf *et al.* the synchrotron light that was used was not completely linearly polarized. The light is characterized by a Stokes parameter  $S_1$  such that the observed TDCS is a weighted average of cross sections corresponding to polarizations along the  $x$  and  $y$  axes, with weights  $(1 + S_1)/2$  and  $(1 - S_1)/2$ , respectively [1]. The major polarization component (at least 77–80%) is along the  $x$  axis. In Figs. 1 and 2 we show polar plots of the TDCS in the  $xy$  plane; the direction of  $\mathbf{k}_1$  is fixed while the direction of  $\mathbf{k}_2$  is allowed to vary in the  $xy$  plane. We compare our results with the data from the experiments and also with the results calculated, in the length and velocity gauges, by Maulbetsch and Briggs [3,4]. We have rescaled all of the results so that they agree at the maximum value of the TDCS. Let  $\theta_1$  and  $\theta_2$  be the angles that  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , respectively, make with the  $x$  axis. Figure 1 is for the case of equal energy sharing, i.e.,  $E_1 = E_2$ , with (a)  $\theta_1 = 180^\circ$ ,  $E_1 = 10$  eV,  $S_1 = 0.554$ ; (b)  $\theta_1 = 284^\circ$ ,  $E_1 = 10$  eV,  $S_1 = 0.564$ ; and (c)  $\theta_1 = 284^\circ$ ,  $E_1 = 5$  eV,  $S_1 = 0.595$ . Figure 2 is for the case of unequal energy sharing, i.e.,  $E_1 = 5$  eV and  $E_2 = 47.9$  eV, with  $\theta_1 = 180^\circ$  and  $S_1 = 0.99$ . The agreement between all of the results is rather good; the only significant discrepancy, where the theoretical results agree with each other but lie well outside the error bars of the experiment, is in the top lobe of Fig. 1(a). In Fig. 3 we show absolute values of the TDCS for the same cases considered in Figs. 1 and 2. The convergence of the absolute values with respect to basis size is best when the two electrons emerge in opposite, or nearly opposite, directions, and is worst when the two electrons emerge in similar directions. On average, the relative convergence error is very roughly 5%.

The shapes of the angular distributions in the case of equal energy sharing have been commented on in detail by Maulbetsch and Briggs [4], and so we discuss here only the

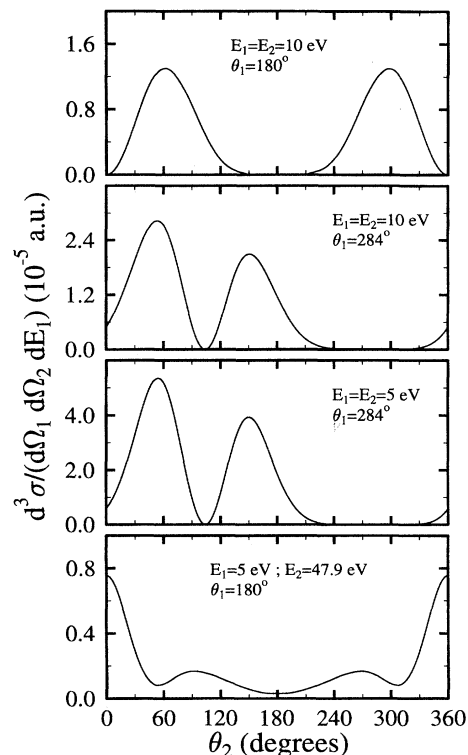


FIG. 3. The absolute values, in atomic units, of the coplanar TDCS for the four cases considered in Figs. 1 and 2.

case of unequal energy sharing. Focusing on the bottom figure of Fig. 3 we see that when one electron emerges rapidly and the other slowly there are two sets of peaks, one set at  $0^\circ$  and  $360^\circ$  and the other at  $90^\circ$  and  $270^\circ$ . The electron that emerges rapidly absorbs most of the energy; this is the electron that absorbs the photon. The slow electron is concomitantly either shaken off or knocked out (see, e.g., Ref. [16]). Prior to the absorption of the photon the two bound electrons undergo “soft” collisions with each other in such a manner as to maintain the equilibrium of the atom. Upon the swift removal of one of the electrons, the absence of further soft collisions may result in the shakeoff of the other (slow) electron. If this occurs, the slow electron is most likely to emerge in the direction opposite the fast electron, owing to the repulsion between electrons [17]. Thus the peaks at  $0^\circ$  and  $360^\circ$  may be interpreted as originating from shakeoff. On the other hand, after absorbing the photon the fast electron may undergo a “hard” binary collision with the other electron, and, on kinematical grounds, if the latter electron is initially at rest the two electrons will subsequently emerge with a relative angle of either  $90^\circ$  or  $270^\circ$ . Thus the peaks at  $90^\circ$  and  $270^\circ$  may be interpreted as originating from knockout.

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