RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 48, NUMBER 2 AUGUST 1993

Double ionization of helium by a single photon with energy 89–140 eV

Daniel Proulx and Robin Shakeshaft
Physics Department, University of Southern California, Los Angeles, California 90089-0484
(Received 22 April 1993)

We have calculated the energy and angular distributions for double ionization of He by one photon, over the range of photon energies 89–140 eV. Our results compare favorably with experimental data.

PACS number(s): 32.80.Fb

The theoretical treatment of double ionization of an atom at photon energies of a few eV or more above threshold remains a difficult challenge. The problem is all the more interesting in view of recent measurements of the energy distribution and the angular asymmetry parameter for double ionization of He by one photon. We report below the results of calculations of the energy distribution, the asymmetry parameter, the cross section for double ionization, and its ratio to the single-ionization cross section, for one-photon double ionization of He over the range of photon energies 89–140 eV. We compare our results with experimental data and other theoretical data.

Taking the light to be linearly polarized, along the z axis, and the atomic states to be spin-singlet (we factor out the spin), we work in the velocity gauge, and unless specified otherwise we use atomic units. 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. Let θ_1 and θ_2 be the angles which $\mathbf{k}_1$ and $\mathbf{k}_2$ make with the z axis, and let θ_{12} be the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$. The differential cross section for the atom to absorb one photon, of frequency ω, and for the two electrons to emerge into solid angles $d\Omega_1$ and $d\Omega_2$ is

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

where, since the atom absorbs only one unit of angular momentum and is initially in a spherically symmetric spin-singlet state, the amplitude $f(\mathbf{k}_1, \mathbf{k}_2)$ has the form

$$f(\mathbf{k}_1, \mathbf{k}_2) = g(k_1, k_2, \cos\theta_{12}) \cos\theta_1 + g(k_2, k_1, \cos\theta_{12}) \cos\theta_2, \tag{2}$$

where $k_1 = |\mathbf{k}_1|$ and $k_2 = |\mathbf{k}_2|$, and where the function $g(k_1, k_2, \cos\theta_{12})$ is to be determined. The energy distribution $d\sigma/dE_1$ and the angular asymmetry parameter $\beta(E_1)$ can be expressed in terms of the auxiliary functions [1]

$$u(k_1, k_2, l) = \left(\frac{2l+1}{2}\right) \int_{-1}^{1} d\mu \, |g(k_1, k_2, \mu)|^2 P_l(\mu), \tag{3}$$

$$v(k_1, k_2, l) = 2\left(\frac{2l+1}{2}\right) \int_{-1}^{1} d\mu \, \mathrm{Re}[g(k_1, k_2, \mu) g^*(k_2, k_1, \mu)] \times P_l(\mu). \tag{4}$$

Note that $v(k_1, k_2, l)$ is symmetric in k_1 and k_2 but that $u(k_1, k_2, l)$ is not. Integrating the right-hand side of Eq. (1) over all directions of $\mathbf{k}_2$, using Eq. (2), gives, after some algebra, the result [2]

$$\frac{d\sigma}{dE_1 d\Omega_1} = \frac{1}{4\pi} \frac{d\sigma}{dE_1} [1 + \beta(E_1) P_2(\cos\theta_1)], \tag{5}$$

where [1]

$$\frac{d\sigma}{dE_1} = \frac{64\pi^4 k_1 k_2}{3\omega c} \left[u(k_1, k_2, 0) + u(k_2, k_1, 0) + \frac{1}{3} v(k_1, k_2, 1) \right], \tag{6}$$

$$\beta(E_1) = \frac{2[15u(k_1, k_2, 0) + 3u(k_2, k_1, 2) + 5v(k_1, k_2, 1)]}{15[u(k_1, k_2, 0) + u(k_2, k_1, 0) + \frac{1}{3} v(k_1, k_2, 1)]}. \tag{7}$$

We see that $d\sigma/dE_1$ is symmetric about the midpoint $E_f/2$, where $E_f \equiv E_1 + E_2$ is the total final energy, but $\beta(E_1)$ is asymmetric. The function $g(k_1, k_2, \cos\theta_{12})$ may be calculated by evaluating $f(\mathbf{k}_1, \mathbf{k}_2)$ at selected values of $\mathbf{k}_1$ and $\mathbf{k}_2$.

For computational purposes it is convenient to start from the following expression [3] for the amplitude:

$$f(\mathbf{k}_1, \mathbf{k}_2) = \langle \chi^{(-)}_{\mathbf{k}_1, \mathbf{k}_2} | (E_f - H_0) | \mathcal{F}^+ \rangle, \tag{8}$$

where $\chi^{(-)}_{\mathbf{k}_1, \mathbf{k}_2}(\mathbf{r}_1, \mathbf{r}_2)$ is *any* trial wave function that correctly describes two outgoing electrons at asymptotically large distances [4] and that is an eigenvector, with eigenvalue E_f, of some (possibly nonlocal) trial operator H_0 (whose form may depend on $\mathbf{k}_1$ and $\mathbf{k}_2$) and where $|\mathcal{F}^+\rangle$ satisfies the inhomogeneous equation

$$(E_f - H_a) |\mathcal{F}^+\rangle = \left(\frac{d}{dz_1} + \frac{d}{dz_2}\right) |\Psi_i\rangle, \tag{9}$$

with H_a the full Hamiltonian of the atom and $|\Psi_i\rangle$ the (fully correlated) ground-state eigenvector. The operator H_0 is the asymptotic Hamiltonian of the doubly ionized atom in the sense that $(H_a - H_0)|\chi^{(-)}_{\mathbf{k}_1, \mathbf{k}_2}\rangle$ falls off faster than a Coulomb potential in position space; in this sense, the potential $W \equiv H_a - H_0$ is "short" range [5]. Unfortunately, a trial wave function having the correct asymptotic form is not easy to handle [4], and therefore

we invoke the following simplification: We first observe that when the right-hand side of Eq. (8) is written as an integral over position space, and the volume integral is converted to a surface integral, the main contribution comes from the point of stationary phase of the integrand, which corresponds to the classical asymptotic motion of the electrons. Thus, following Rudge and Seaton [6] and Jetzke and Faisal [7], we introduce a quasiclassical wave function that describes the electrons moving asymptotically in the Coulomb potential $-(Z_1/r_1) - (Z_2/r_2)$ where Z_1 and Z_2 are velocity-dependent effective charges given by $Z_i = Z - \Delta_i$, $i = 1, 2$ with $Z = 2$ and

$$\Delta_i = \frac{(\mathbf{k}_i \cdot \mathbf{k}_{ij})k_i}{k_{ij}^3}, \tag{10}$$

where $\mathbf{k}_{ij} = \mathbf{k}_i - \mathbf{k}_j$, $j \neq i$, and $k_{ij} = |\mathbf{k}_{ij}|$. We therefore choose the trial wave function [6, 7]

$$\chi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2}(\mathbf{r}_1, \mathbf{r}_2) \approx (2\pi)^{-3} e^{i\mathbf{k}_1\cdot\mathbf{r}_1 + i\mathbf{k}_2\cdot\mathbf{r}_2} \times \prod_{j=1,2} e^{-\pi\gamma_j/2}\Gamma(1 - i\gamma_j) \times {}_1F_1(i\gamma_j, 1, -ik_jr_j - i\mathbf{k}_j \cdot \mathbf{r}_j), \tag{11}$$

where $\gamma_j = -Z_j/k_j$. Thus H_0 simplifies to a local (but velocity-dependent) operator $H_a - W$ with

$$W = \frac{1}{r_{12}} - \frac{\Delta_1}{r_1} - \frac{\Delta_2}{r_2}, \tag{12}$$

where $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$. We note that Pan and Starace [8] recently made use of the same velocity-dependent effective charges in their study of $(e, 2e)$ collisions. Putting $r_1 = k_1 t$, $r_2 = k_2 t$, and $r_{12} = k_{12} t$—we call this the "stationary phase limit"—gives $W = 0$. However, substituting the form (11) for $|\chi^{(-)}_{\mathbf{k}_1,\mathbf{k}_2}\rangle$ into Eq. (8) gives a divergent integral over $\mathbf{r}_1$ and $\mathbf{r}_2$, since the form (12) for W falls off asymptotically as a Coulomb potential; the trial wave function of Eq. (11) has the correct asymptotic form *only* in the stationary phase limit. To obtain a convergent integral we represent the (divergent) amplitude by a divergent series, and then we attempt to continue analytically this representation, that is, we attempt to sum the series by using one of Levin's algorithms [9]. Since the essential physics has been included in Eqs. (8), (9), and (11), we conjecture that if the series can be summed to a converged finite number, that number is the correct value of the amplitude. To obtain a series representation of $f(\mathbf{k}_1, \mathbf{k}_2)$ we expand $|\mathcal{F}^+\rangle$ on a finite basis. As in our previous work [3], we used a two-electron basis consisting of terms $S^\kappa_{nl}(r_1) S^\kappa_{n'l'}(r_2) Y^{LM}_{ll'}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)$, where $Y^{LM}_{ll'}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)$ couples spherical harmonics and where $S^\kappa_{nl}(r)$ is a radial Sturmian function that is a polynomial of degree $n_r \equiv n - l - 1$ multiplied by $r^{l+1}e^{i\kappa r}$. The "wave number" κ was chosen to lie in the upper right quadrant of the complex κ plane so as to simulate both outgoing-wave open channels and exponentially decaying closed channels [10]. We have applied this method to double ionization of He, and we now show results.

In Fig. 1 we show estimates of $d\sigma/dE_1$ vs E_1, for various photon energies. Recall that $d\sigma/dE_1$ is symmetric about $(E_1 + E_2)/2$. The solid circles, in the upper box, are our calculated points. We were able to obtain results

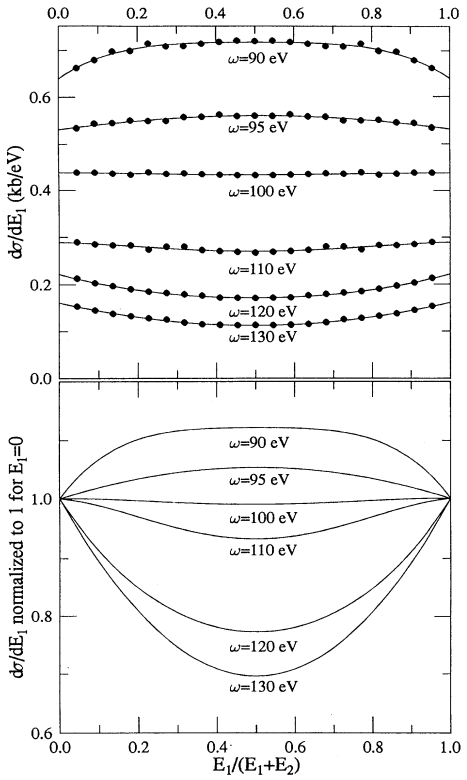

FIG. 1. Unnormalized (upper box) and normalized (lower box) energy distributions at various photon energies.

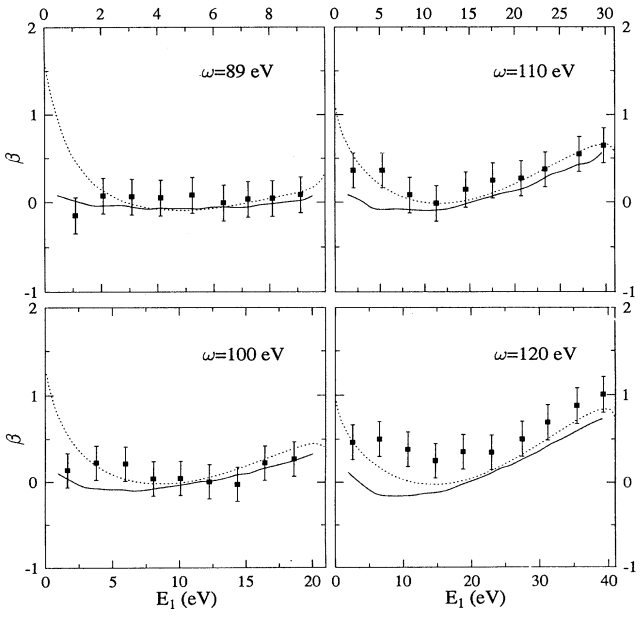

FIG. 2. Asymmetry parameter at various photon energies. Broken curve: theoretical results of Ref. [16]. Solid curve: present results. The experimental data are revised [15] from Ref. [11].

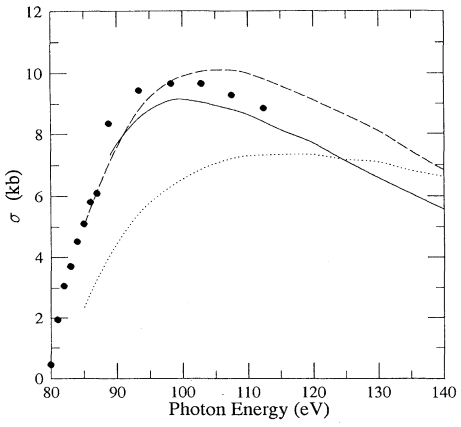

FIG. 3. Absolute integrated cross section. Solid circles: experimental data from Ref. [18], revised as noted in text. Short and long broken lines: Theoretical results of Tiwary [22] and Carter and Kelly [21], respectively. Solid line: present results (the slight jaggedness is probably due to numerical noise—see text).

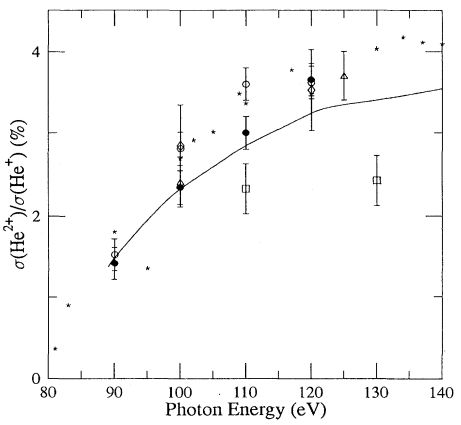

FIG. 4. Ratio of cross sections for double to single ionization vs photon energy ω, compared to the data of Refs. [11] (•), [24] (□), [25] (△), [26] (○), [27] (◇), and [28] (⋆).

that were usually converged, with respect to basis size, to within a few percent; the scatter of the solid circles is most probably due to numerical errors arising from imperfect convergence or numerical instability. The continuous lines are smooth interpolations. We see that the curvature of the energy distribution changes from being slightly negative to more strongly positive as the photon energy increases. Wehlitz *et al.* [11] have measured an energy distribution that is flat, but the relative uncertainty of the measurement is 20% so there is no conflict with our results. The negative curvature of the energy distribution, at lower photon energies, may be due in part to the phase-space factor $k_1 k_2$, which has a maximum at the midpoint [12]. At higher photon energies, the curvature changes sign owing to the difficulty of ejecting two *fast* electrons. As the photon energy increases the distribution becomes more deeply U shaped and at very high photon energies it may become W shaped [13]. Le Rouzo and Dal Cappello [14] have reported results for the curvature of the energy distribution at photon energies from 100 to 300 eV, but they find significantly greater curvature; e.g., at a photon energy of 100 eV they calculate about a 20% change in $d\sigma/dE_1$ over $0 \leq E_1 \leq E_f/2$, compared to our 1% change.

In Fig. 2 we show the asymmetry parameter $\beta(E_1)$ for four different photon energies. We compare with the measured results of Wehlitz *et al.* [11, 15], and with the recent theoretical results of Maulbetsch and Briggs [16] based on using a final-state wave function with the correct asymptotic form. At photon energies not far above threshold, when both E_1 and E_2 are small, $\beta(E_1)$ does not vary much, and remains small, over the allowed range of E_1; as the two slow electrons depart, after absorbing the photon, they continue to interact for a long time and evidently lose their memory of the electric field, so that there is no preferred direction into which one electron emerges when it is not detected with reference to the other. However, at larger photon energies, $\beta(E_1)$ increases with increasing E_1. Indeed, when E_1 is very large, $\beta(E_1) \approx 2$ [17, 13]; a fast electron has a $\cos^2(\theta_1)$ angular distribution (if it is not detected with reference to the slow electron) since it absorbs the photon primarily from a zero angular-momentum component of the ground state and therefore emerges with one unit of angular momentum.

In Fig. 3 we show the absolute integrated cross section. Lablanquie *et al.* [18] measured the ratio of the cross section for double-ionization to the total ionization cross-section, and they used the values of the total ionization cross section measured by Marr and West [20] to deduce the cross section for double ionization. We have revised the double-ionization cross-section results of Lablanquie *et al.* by using values of the total ionization cross section measured and compiled by Samson [19], which are believed to be more accurate. We show the revised results in Fig. 3, along with the theoretical results (calculated in the velocity gauge) of Tiwary [22] and Carter and Kelly [21] (similarly revised).

Finally, in Fig. 4, we show our estimate of the ratio of cross sections for double to single ionization [23], and we compare with various experimental data [11,24–28].

In conclusion, we have proposed a method, which is relatively easy to implement, for treating double ionization of two-electron systems. The soundness of the method is indicated, if not confirmed, by the reasonable agreement between the available experimental data and our calculated results in our application to He.

We thank Zhong-jian Teng for many fruitful discussions, and Marcel Pont for his assistance in improving the numerical accuracy of matrix elements. We are grateful to Professor U. Becker, Professor J. Briggs, and Professor J. Samson for allowing us to present their results prior to publication, and to Professor A.R.P. Rau for his comments on our manuscript. This work was supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, the Department of Energy under Contract No. DE-FG03-92ER14266.

[1] Z.-j. Teng, Ph.D. thesis, University of Southern California (1993) (unpublished); Z.-j. Teng and R. Shakeshaft (unpublished).
[2] C. N. Yang, Phys. Rev. **74**, 764 (1948).
[3] D. Proulx and R. Shakeshaft, Phys. Rev. A **46**, R2221 (1992); J. Phys. B **26**, L7 (1993).
[4] M. Brauner, J. S. Briggs, and H. Klar, J. Phys. B **22**, 2265 (1989).
[5] In position space, the component of $(E_f - H_0)|\mathcal{F}^+\rangle$, which corresponds to the electrons leaving with momenta $\mathbf{k}_1$ and $\mathbf{k}_2$, falls off faster than a Coulomb potential. Hence when the right-hand side of Eq. (8) is expressed as an integral over $\mathbf{r}_1$ and $\mathbf{r}_2$, the nonoscillatory part of the integrand falls off faster than a Coulomb potential and the integral (or, rather, its analytic continuation) is finite.
[6] M. R. H. Rudge and M. J. Seaton, Proc. R. Soc. London Ser. A **283**, 262 (1965).
[7] S. Jetzke and F. H. M. Faisal, J. Phys. B **25**, 1543 (1992).
[8] C. Pan and A. F. Starace, Phys. Rev. Lett. **67**, 185 (1991).
[9] See E. J. Weniger, J. Math. Phys. **26**, 276 (1985).
[10] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **39**, 1545 (1989); M. Dörr, R. M. Potvliege, and R. Shakeshaft, J. Opt. Soc. Am. B **7**, 433 (1990).
[11] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[12] Very close to threshold, where the Wannier-Rau-Peterkop theory applies, the energy distribution is predicted to be flat. See, e.g., A. R. P. Rau, Phys. Rep. **110**, 369 (1984). However, there is experimental evidence that the energy distribution for the $(e, 2e)$ process has negative curvature just above threshold. See A. R. P. Rau, Comments At. Mol. Phys. **14**, 285 (1984).
[13] Ref. [1] and Z.-j. Teng and R. Shakeshaft, Phys. Rev. A **47**, 3487 (1993).
[14] H. Le Rouzo and C. Dal Cappello, Phys. Rev. A **43**, 318 (1991).
[15] The values of $\beta(E_1)$ published in Ref. [11] were based on erroneously fitting to a symmetric polynomial. The correctly revised values are shown in Fig. 2, courtesy of U. Becker.
[16] F. Maulbetsch and J. S. Briggs, J. Phys. B (to be published).
[17] F. Maulbetsch and J. S. Briggs, Phys. Rev. Lett. **68**, 2004 (1992).
[18] P. Lablanquie, K. Ito, P. Morin, I. Nenner, and J. H. D. Eland, Z. Phys. D **16**, 77 (1990).
[19] J. A. R. Samson (private communication).
[20] G. V. Marr and J. B. West, At. Nucl. Data Tables **18**, 497 (1976).
[21] S. L. Carter and H. P. Kelly, Phys. Rev. A **24**, 170 (1981).
[22] S. N. Tiwary, J. Phys. B **15**, L323 (1982).
[23] We used the total-cross-section data of Samson, Ref. [19], and subtracted the double-ionization contribution.
[24] T. Carlson, Phys. Rev. **156**, 142 (1967).
[25] V. Schmidt, N. Sandner, H. Kuntzemüller, P. Dhez, F. Wuilleumier, and E. Källne, Phys. Rev. A **13**, 1748 (1976).
[26] G. R. Wight and M. J. Van der Wiel, J. Phys. B **9**, 1319 (1976).
[27] D. M. P. Holland, K. Codling, J. B. West, and G. V. Marr, J. Phys. B **12**, 2465 (1979).
[28] R. J. Bartlett, P. J. Walsh, Z. X. He, Y. Chung, E. M. Lee, and J. A. R. Samson, Phys. Rev. A **46**, 5574 (1992).