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Two-photon ionization of atomic hydrogen with elliptically polarized light

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The theory of two-photon ionization of a hydrogenic state in the nonrelativistic dipole approximation is generalized for elliptically polarized light. An application to the metastable 2S state of atomic hydrogen is made. Significant differences in the angular distribution of the outgoing electrons are found depending upon the polarization of the photons. It is claimed that two-photon ionization employing elliptically polarized photons from lasers may provide an additional test for the theories of multiphoton ionization.

It is well known that the most common multiphoton phenomenon that occurs when a strong enough radiation field, virtually of any frequency, is made to interact with gas atoms is ionization. The degree of ionization depends on the strength, polarization, and frequency of the field.

Though the theory of a two-photon process had been worked out a long time ago by Goeppert-Mayer,[1] its practical application had not been made until the advent of lasers. Since the publication of that paper, a number of papers on multiphoton ionization have appeared. A detailed description of the theory of multiphoton processes is given in Faisal's book.[2]

The theory of two-photon ionization of a hydrogenic state had been worked out by Zernik[3] in the nonrelativistic dipole approximation using linearly polarized photons. The theory had been generalized to circularly polarized photons amongst others by Grontier and Trahin.[4] To the best of our knowledge, no calculations on two-photon or multiphoton ionization using elliptical polarization have been reported.

In the present report, we generalize the work of Zernik on angular distribution and total cross section for elliptically polarized light. The same general notation, procedure, and coordinate system as followed by Zernik, and earlier by Rustgi, Zernik, Breit, and Andrews[5] for a calculation on the photodisintegration of the deuteron, is used.

Following Zernik's Fig. 1, two coordinate systems are used. The plane polarized light is taken as incident along the positive z axis of the unprimed coordinate system. The photoelectron travels along the negative z' axis of the primed coordinate system, with β and α being colatitude and azimuthal angles of z' with α referred to x. An elliptically polarized photon may be regarded as the superposition of two plane polarized photons, the planes of polarization being at right angles and the phases in general being different. In Zernik's paper[3] the amplitudes had been worked out for the state of polarization

$$E_x = a e^{i\kappa z}\ , \quad H_y = a e^{i\kappa z}\ . \tag{1}$$

One has to now add to the amplitudes the contribution for the perpendicular state of polarization

$$E_y = b e^{i\kappa z}\ , \quad H_x = -b e^{i\kappa z}\ . \tag{2}$$

Following Zernik, and using the second-order perturbation theory, the differential cross section per unit intensity may be written as

$$\frac{1}{I}\frac{d\sigma}{d\Omega} = \frac{\alpha}{4\pi I_0}\left|\sum_i \frac{(\hat{\epsilon}\cdot\mathbf{r})_{fi}(\hat{\epsilon}\cdot\mathbf{r})_{io}}{E_0 - E_i + E_p}\right|^2 E_p k_e a^2\ , \tag{3}$$

where α is the fine-structure constant, a is the Bohr radius, E_p is the energy of the photon, E_0 and E_i are the energies of the initial and intermediate atomic states, in units of $(me^4/\hbar^2)$, I_0 is 7.019×10^{16} W/cm^2, and k_e is the wave number of the emitted electron in units of $(me^2/\hbar^2)$. The rest of the symbols have their usual meaning.

Writing the initial, intermediate, and final states as in the paper by Zernik, one can write for elliptically polarized photons

$$\frac{1}{I}\frac{d\sigma}{d\Omega} = \frac{\pi^2\alpha}{8I_0}\,|\,M_{l+2}^{\chi} + M_l^{\chi} + M_{l-2}^{\chi}\,|^2 E_p a^2\ , \tag{4}$$

where

$$\begin{aligned} M_{l+2}^{\chi} = {} & P_{l+1,l+2}\, e^{i\eta_{l+2}}(i)^{l+2}[(2l+3)(2l+5)^{1/2}(2l+1)^{1/2}]^{-1} \\ & \times\{\cos^2\chi[C_{l+2,m-2}^{l+1}Y_{l+2,m-2}(\beta,\alpha) + C_{l+2,m+2}^{l+1}Y_{l+2,m+2}(\beta,\alpha) - 2C_{l+1,m}^{l+1}Y_{l+2,m}(\beta,\alpha)] \\ & + \sin^2\chi[C_{l+2,m-2}^{l+1}Y_{l+2,m-2}(\beta,\alpha) + C_{l+2,m+2}^{l+1}Y_{l+2,m+2}(\beta,\alpha) + 2C_{l+1,m}^{l+1}Y_{l+2,m}(\beta,\alpha)] \\ & - \sin(2\chi)[C_{l+2,m-2}^{l+1}Y_{l+2,m-2}(\beta,\alpha) - C_{l+2,m+2}^{l+1}Y_{l+2,m+2}(\beta,\alpha)]\}\ , \end{aligned} \tag{5}$$

$$M_{l-2}^{\chi}=P_{l-1,l-2}e^{i\eta_{l-2}}(i)^{l-2}[(2l-1)(2l+1)^{1/2}(2l-3)^{1/2}]^{-1}$$
$$\times\{\cos^2\chi[C_{l-2,m-2}^{l-1}Y_{l-2,m-2}(\beta,\alpha)+C_{l-2,m+2}^{l-1}Y_{l-2,m+2}(\beta,\alpha)-2C_{l-2,m}^{l-1}Y_{l-2,m}(\beta,\alpha)]$$
$$+\sin^2\chi[C_{l-2,m-2}^{l-1}Y_{l-2,m-2}(\beta,\alpha)+C_{l-2,m+2}^{l-1}Y_{l-2,m+2}(\beta,\alpha)+2C_{l-2,m}^{l-1}Y_{l-2,m}(\beta,\alpha)]$$
$$-\sin(2\chi)[C_{l-2,m-2}^{l-1}Y_{l-2,m-2}(\beta,\alpha)-C_{l-2,m+2}^{l-1}Y_{l-2,m+2}(\beta,\alpha)]\}\ , \tag{6}$$

$$M_{l}^{\chi}=-P_{l+1,l}e^{i\eta_{l}}(i)^{l}[(2l+1)(2l+3)]^{-1}$$
$$\times\{\cos^2\chi[C_{l,m-2}^{l+1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l+1}Y_{l,m+2}(\beta,\alpha)-C_{l,m}^{l+1}Y_{l,m}(\beta,\alpha)]$$
$$+\sin^2\chi[C_{l,m-2}^{l+1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l+1}Y_{l,m+2}(\beta,\alpha)+C_{l,m}^{l+1}Y_{l,m}(\beta,\alpha)]$$
$$+\sin(2\chi)[-C_{l,m-2}^{l+1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l+1}Y_{l,m+2}(\beta,\alpha)]\}-P_{l-1,l}e^{i\eta_{l}}(i)^{l}[(2l-1)(2l+1)]^{-1}$$
$$\times\{\cos^2\chi[C_{l,m-2}^{l-1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l-1}Y_{l,m+2}(\beta,\alpha)-C_{l,m}^{l-1}Y_{l,m}(\beta,\alpha)]$$
$$+\sin^2\chi[C_{l,m-2}^{l-1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l-1}Y_{l,m+2}(\beta,\alpha)+C_{l,m}^{l-1}Y_{l,m}(\beta,\alpha)]$$
$$+\sin(2\chi)[-C_{l,m-2}^{l-1}Y_{l,m-2}(\beta,\alpha)+C_{l,m+2}^{l-1}Y_{l,m+2}(\beta,\alpha]\}\ , \tag{7}$$

where χ is a parameter which gives a photon polarized along the x or along the y axis when it is set equal to zero or $\pi/2$, respectively. When $\chi=\pi/4$, it gives circularly polarized photons. For other values, it describes elliptically polarized photons. The quantities $P_{\lambda L}(E_P)$ and η_L are defined by Eqs. (14) and (7) in Ref. 3. The coefficients $C_{L,M}^{\lambda}$ can be obtained from Eqs. (11), (12), and (13) of Ref. 3 by putting $\chi=0$.

By substituting $l=0$, $m=0$ in Eqs. (7) and (5) and dropping the subscript λ on $P_{\lambda L}$, one obtains

$$M_0^{\chi}=\tfrac{2}{3}\pi^{-1/2}P_0(E_p)e^{i\eta_0}\cos(2\chi)\ , \tag{8}$$

TABLE I. The total cross section per unit intensity for linearly ($\chi=0$), circularly ($\chi=45°$), and elliptically polarized light ($\chi=60°$). The numbers in square brackets indicate powers of ten.

λ(A°)	$\chi=0°$	$\chi=45°$	$\chi=60°$
7290.1	0.2427[−28]	0.3465[−28]	0.3206[−28]
7119.2	0.3278[−28]	0.4621[−28]	0.4285[−28]
7000.0	0.4437[−28]	0.6193[−28]	0.5755[−28]
6943.5	0.5346[−28]	0.7423[−28]	0.6904[−28]
6800.4	0.1073[−27]	0.1464[−27]	0.1366[−27]
6661.3	0.2984[−26]	0.6317[−27]	0.1220[−26]
6472.0	0.3987[−27]	0.5129[−27]	0.4843[−27]
6300.0	0.2991[−28]	0.3638[−28]	0.3476[−28]
6200.0	0.1160[−28]	0.1341[−28]	0.1296[−28]
6075.1	0.4140[−29]	0.4341[−29]	0.4291[−29]
6000.0	0.2288[−29]	0.2180[−29]	0.2208[−29]
5900.0	0.1026[−29]	0.7796[−30]	0.8425[−30]
5800.0	0.4422[−30]	0.1980[−30]	0.2591[−30]
5695.4	0.1882[−30]	0.8419[−32]	0.5336[−31]
5600.0	0.1266[−30]	0.2897[−31]	0.5335[−31]
5500.0	0.1650[−30]	0.1651[−30]	0.1651[−30]
5360.4	0.3460[−30]	0.5003[−30]	0.4618[−30]
5300.0	0.4734[−30]	0.7057[−30]	0.6477[−30]
5200.0	0.7948[−30]	0.1190[−29]	0.1091[−29]
5100.0	0.1432[−29]	0.2096[−29]	0.1930[−29]
5000.0	0.3364[−29]	0.4737[−29]	0.4394[−29]
4915.1	0.1613[−28]	0.2153[−28]	0.2019[−28]
4873.0	0.2380[−27]	0.3053[−27]	0.2885[−27]
4852.3	0.5970[−27]	0.7464[−27]	0.7090[−27]
4811.3	0.1171[−28]	0.1374[−28]	0.1323[−28]
4700.0	0.3774[−30]	0.2590[−30]	0.2886[−30]
4649.3	0.1084[−30]	0.1722[−31]	0.4001[−31]
4600.0	0.5590[−31]	0.1565[−31]	0.2571[−31]
4556.3	0.8519[−31]	0.9860[−31]	0.9526[−31]

$$M_2^{\chi}=\frac{2}{3}\pi^{-1/2}P_2(E_P)e^{i\eta_2}[\cos^2\chi(1-3\sin^2\alpha\cos^2\beta)-\sin^2\chi(1-3\sin^2\alpha\sin^2\beta)-\tfrac{3}{2}i\sin^2\beta\sin(2\alpha)\sin(2\chi)]\ . \tag{9}$$

On substituting Eqs. (8) and (9) into Eq. (4), the differential cross section per unit intensity may be written as

$$\frac{1}{I}\frac{d\sigma}{d\Omega}=\frac{\alpha}{18I_0}\pi a^2\,|M^{\chi}|^2E_p\ , \tag{10}$$

where

$$|M^{\chi}|^2=a[1-\sin^2 2\chi]+b\sin^2\beta\cos(2\chi)[\cos^2\alpha\cos^2\chi-\sin^2\alpha\sin^2\chi]$$
$$+c\sin^4\beta[\cos^4\alpha\cos^4\chi+\sin^4\alpha\sin^4\chi+\tfrac{1}{8}\sin^2\alpha\sin^2(2\chi)]+d\cos(2\chi)\sin(2\chi)\sin^2\beta\sin(2\alpha)\ , \tag{11}$$

and

$$a=P_0^2+P_2^2+2P_0P_2\cos(\eta_2-\eta_0)\ , \tag{12}$$

$$b=-6P_0P_2\cos(\eta_2-\eta_0)-6P_2^2\ , \tag{13}$$

$$c=9P_2^2\ , \tag{14}$$

$$d=3P_0P_2\sin(\eta_2-\eta_0)\ . \tag{15}$$

It is seen that the differential cross section is symmetrical about $\beta=90°$. The total cross section per unit intensity is found to be

$$\frac{\sigma}{I}=\left[\frac{\sigma}{I}\right]_{\rm Lin}\cos^2(2\chi)+\frac{8\pi}{15}C\sin^2(2\chi)\ , \tag{16}$$

where

$$C=\frac{\alpha}{18I_0}\pi a^2 c\ , \tag{17}$$

and $(\alpha/I)_{\rm Lin}$ denotes the total cross section per unit intensity for the linearly polarized ($\chi=0$) photons. For the case of linearly polarized photons ($\chi=0$), Eq. (10) reduces to Eq. (21) in Ref. 3. For circular polarization, the angular distribution is azimuthally isotropic, as expected.

The numerical results for the total cross section employing the radial integrals from Ref. 3, are presented in Table I. A graph of the differential and total cross section per unit intensity for the linearly polarized, circularly polarized, and elliptically polarized light are shown in Figs. 1 and 2, respectively. It is clear from Fig. 1 that the elliptically polarized light gives a very different angu-

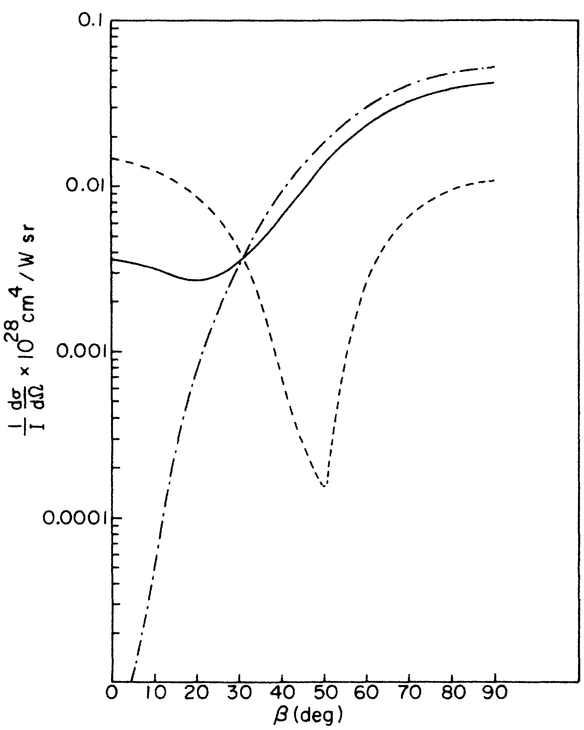

FIG. 1. The differential cross section per unit intensity as a function of β with $\alpha=45°$, for linearly polarized (dashed line), circularly polarized (dot-dashed line), and elliptically polarized (solid line, $\chi=60°$) light of $\lambda=7290.1$ Å.

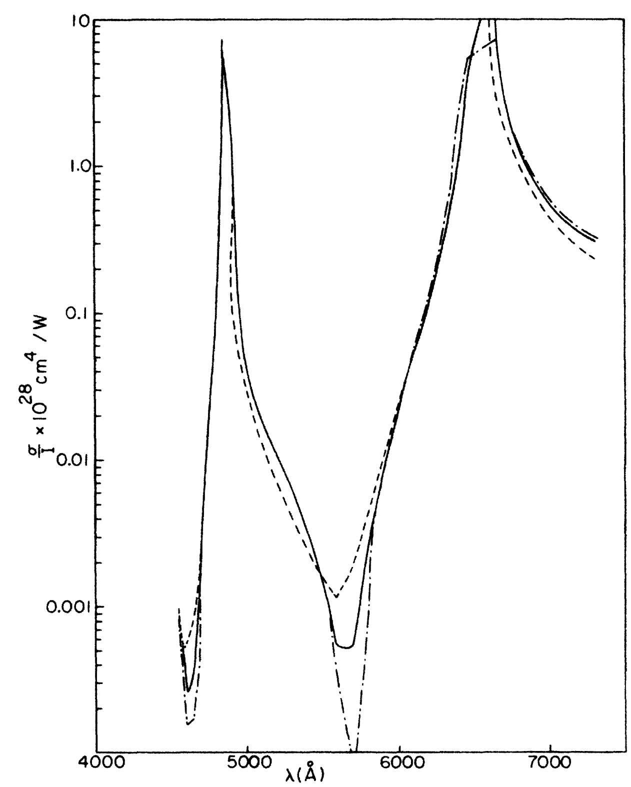

FIG. 2. Graph of σ/I, the total cross section per unit intensity vs the wavelength for the linearly polarized (dashed line), circularly polarized (dot-dashed line), and elliptically polarized (solid line, $\chi=60°$) light.

lar distribution in the forward direction, as compared to the linearly and circularly polarized light. Figure 2 shows that the total cross section per unit intensity have different values of the minimum and maximum cross sections as a function of the wavelength.

An experimental verification of these results would be desirable as experiments with elliptically polarized laser light have not been performed as yet. Since the completion of this work, experimental results on multiphoton ionization angular distributions for elliptically polarized laser radiation in xenon and krypton have been reported by Bashkansky, Bucksbaum, and Schumacher.[6]

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