Polarization Effects in High-Order Multiphoton Ionization

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Linear polarization is shown to dominate circular polarization cross sections strongly for high-order multiphoton ionization of atoms.

Experiments[1,2] on two- and three-photon ionization of atoms have shown that circularly polarized light gives larger cross sections than linearly polarized light. Theoretical treatments[3,4] of low-order multiphoton ionization are consistent with these results. This has stimulated interest in high-order effects with circular polarization. Klarsfeld and Maquet[5] have found an upper bound for the ratio of circular- to linear-polarization cross sections for multiphoton ionization of atoms. The bound they find shows a strong dominance of circular over linear polarization for large N, where N is the multiphoton order. Lambropoulos[6] has pointed out the theoretical simplicity of the circular polarization case.

It is shown here that a greatly improved upper bound can be found for the ratio of circular- to linear-polarization cross sections. This new bound is very much less than unity for large N, which thus reverses the earlier conclusions. The strong dominance of linear over circular polarization for large N discourages the use of circular polarization in experiments, and attenuates the advantages attendant upon a simple theoretical analysis of circular polarization.

The analysis performed here will employ the momentum-translation method,[7] which is suited to the treatment of high-order transitions, and which gives simple, analytical results.

Klarsfeld and Maquet found their upper bound on the ratio of circular- to linear-polarization cross sections by examining the ionization cross sections from an initial s state to a final state with angular momentum $l=N$. This is the only final state possible for circular polarization, but for linear polarization there are $\frac{1}{2}N+1$ final states allowed if N is even, and $\frac{1}{2}(N+1)$ if N is odd. The reason that Klarsfeld and Maquet's result is misleading is that $l=N$ is the least important of the many allowed states in the linear polarization case.

For intensity in the photon field not too high for perturbation theory to be valid, the momentum-translation method[7,8] gives a transition matrix element for a central binding potential which can be written as

$$(\phi_f, [\vec{\rho}\cdot\vec{\epsilon}]^N\phi_i) = \int_0^\infty d\rho\,\rho^2 R_f^*(\rho) R_i(\rho) \int d\Omega\, Y_{l_f}^{m_f\,*}(\theta,\varphi) Y_{l_i}^{m_i}(\theta,\varphi)(\vec{\rho}\cdot\vec{\epsilon})^N, \tag{1}$$

where $\vec{\rho}=\vec{x}/a_0$ is a dimensionless radius vector, $\rho=|\vec{\rho}|$, a_0 is the Bohr radius,[9] $\vec{\epsilon}$ is the polarization vector of the ionizing field, ϕ is a state vector, Y_l^m is a spherical harmonic, θ, φ are spherical polar coordinates, and the subscripts f and i refer to final and initial states. It is convenient to take the polar axis of spherical coordinates along the propagation direction of the photon field for circular polarization, while for linear polarization it is better to take the axis along the direction of polarization. For circular polarization,

$$(\vec{\rho}\cdot\vec{\epsilon})^N = \rho^N 2^{-N/2}\sin^N\theta\, e^{\pm iN\varphi}, \tag{2}$$

where the ambiguous sign refers to right-hand or left-hand cases. Linear polarization gives simply

$$(\vec{\rho}\cdot\vec{\epsilon})^N = \rho^N\cos^N\theta. \tag{3}$$

The case $l_i=0$ will be examined for the sake of simplicity. The subscripts f can be dropped from the l_f, m_f quantum numbers, since there is now no ambiguity. There are two angular integrals to be evaluated,

$$A' = \int d\Omega\, Y_l^{m*} Y_0^0 2^{-N/2}\sin^N\theta\, e^{\pm iN\varphi} \tag{4}$$

for circular polarization, and

$$A_l = \int d\Omega\, Y_l^{m*} Y_0^0 \cos^N\theta \tag{5}$$

for linear polarization, where the subscript l on A in Eq. (5) refers to the final angular momentum quantum number. The evaluation of the angular integrals is done more easily with the spherical harmonics $C_l^m(\theta,\varphi)$ than with the more conventional $Y_l^m(\theta,\varphi)$, where $C_l^m = (4\pi)^{1/2}(2l+1)^{-1/2}Y_l^m$. Then since

$$C_1^{\pm 1}(\theta,\varphi) = -2^{-1/2}\sin\theta\, e^{\pm i\varphi}, \tag{6}$$

evaluation of Eq. (4) requires knowledge of $(C_1^{\pm 1})^N$. This is given by the theorem

$$(C_1^{\pm 1})^N = (N!)^{1/2}[(2N-1)!!]^{-1/2}C_N^{\pm N} \tag{7}$$

which can be proven readily from the lemma

$$C_1^{\pm 1}C_{N-1}^{\pm(N-1)} = N^{1/2}(2N-1)^{-1/2}C_N^{\pm N}.$$

With Eqs. (6) and (7), Eq. (4) is

$$A' = (-1)^N(2l+1)^{1/2}(4\pi)^{-1}(N!)^{1/2}[(2N-1)!!]^{-1/2}\int d\Omega\, C_l^{m*}C_N^{\pm N} = (-1)^N(N!)^{1/2}[(2N+1)!!]^{-1/2}, \tag{8}$$

with the conditions $l=N$, $m=\pm N$. To evaluate Eq. (5), the expansion

$$\cos^N\theta = \sum_j b_j^{(N)} C_j^0(\theta,\varphi) \tag{9}$$

is useful, where

$$b_j = \begin{cases} 0, & N-j \text{ odd or negative} \\ \dfrac{(2j+1)2^j N!\,(\frac{1}{2}N+\frac{1}{2}j)!}{(\frac{1}{2}N-\frac{1}{2}j)!\,(N+j+1)!}, & N-j \text{ even and non-negative.} \end{cases} \tag{10}$$

Equation (5) is thus

$$A_l = \sum_j b_j^{(N)}(2l+1)^{1/2}(4\pi)^{-1}\int d\Omega\, C_l^{m*}C_j^0 = (2l+1)^{-1/2}b_l^{(N)}, \tag{11}$$

where $j=l$, $m=0$.

If σ' is the cross section for ionization by circularly polarized light, and σ is the cross section for linear polarization, where all final angular momentum states are included, then a bound on the ratio is given by

$$\sigma'/\sigma < \sigma'/\sigma_l, \tag{12}$$

where σ_l is any of the partial cross sections for angular momentum l.

One possible bound suggested by Eq. (12) is realized if σ_N is employed on the right-hand side. This is the simplest case, since then all dependence on radial integrals cancels, and the ratio of σ' to σ_N is given simply by the angular integrals, i.e.,

$$\sigma'/\sigma_N = A'^2/A_N^2. \tag{13}$$

From Eqs. (8), (10), (11), and (13), the upper bound on the ratio of circular- to linear-polarization cross sections obtained in this way is

$$\frac{\sigma'}{\sigma} < \frac{\sigma'}{\sigma_N} = \frac{(2N-1)!!}{N!}, \tag{14}$$

which is the result of Klarsfeld and Maquet.[5] Since $(2N-1)!!/N!$ behaves as $2^N(\pi N)^{-1/2}$ for large N, Eq. (14) suggests (but does not demonstrate) a strong dominance of circular over linear polarization for high orders. However, calculations of high-order multiphoton ionization of hydrogen by linearly polarized light have shown[10] that, whereas many final angular momentum states can contribute significantly, the state with $l=N$ scarcely contributes at all.

A more reasonable upper bound to find from Eq. (12) should thus employ a relatively small value of l for σ_l. It is convenient to examine $l=0$. A comparison of the angular parts alone is striking. The ratio

$$\frac{A'^2}{A_0^2} = (N+1)(N+1)!/(2N+1)!! \tag{15}$$

behaves as $(\pi N^3)^{3/2}/2^{N+1}$ for large N, which is very small when N is large. What is really needed is the ratio

$$\frac{\sigma'}{\sigma_0} = \frac{|\mathfrak{R}'|^2}{|\mathfrak{R}_0|^2}\frac{A'^2}{A_0^2}, \tag{16}$$

where

$$\mathcal{R}_l = \int_0^\infty d\rho\, \rho^{N+2} R_f^*(\rho) R_i(\rho); \tag{17}$$

R_f and R_i are final- and initial-state radial wave functions, the dependence of $\mathcal{R}_l$ on l arises from $\mathcal{R}_f$, and $\mathcal{R}' = \mathcal{R}_N$. The implication of Eqs. (12), (16), and (15) is that linear polarization is much more important than circular polarization unless the radial integrals found from (17) can offset the very small result of Eq. (15). An examination of radial integrals involves explicit specification of potential and wave functions which has been avoided up to this point. All that is needed, however, is an indication for a simple (but realistic) special case that the radial integral comparison will not compensate the angular integral comparison of Eq. (15).

Consider the ionization of a hydrogen atom from the ground state to a Coulomb final state in the limit as the momentum k of the ionized electron goes to zero. This gives meaningful results for E_f (the energy of the ionized electron) such that $E_f \ll E_I$ (where E_I is the ionization energy). This inequality would generally be satisfied in high-order photoionization. With a normalization which gives a finite result as $k \to 0$, the initial-state radial wave function is

$$R_i(\rho) = 2e^{-\rho}$$

and the final-state wave function is

$$R_f(\rho) = (2/\rho)^{1/2} J_{2l+1}((8\rho)^{1/2}).$$

These wave functions lead to

$$\mathcal{R}_l = \frac{2(N+l+2)!}{e(2l+1)!} M_{N+2,l+1/2}(2), \tag{18}$$

where the function $M_{\mu,\nu}(z)$ is a Whittaker function. For $l=N$, Eq. (18) gives

$$\mathcal{R}_N = \mathcal{R}' = 2^{N+3} e^{-2} N, \tag{19}$$

while for $l=0$,

$$\mathcal{R}_0 = e^{-1} 2(N+2)!\, M_{N+2,1/2}(2) \sim \frac{2(N+1)!\,[2(N+2)]^{1/4}}{e\pi^{1/2}} \cos([8(N+2)]^{1/2} - \tfrac{3}{4}\pi), \tag{20}$$

where the asymptotic form for large N is given by the last expression in Eq. (20). The trigonometric function in (20) can never exactly vanish, and will be small only under very special circumstances. If a typical value of the square of the cosine function is taken to be $\frac{1}{2}$, then Eqs. (19) and (20) yield, in the large-N limit,

$$\frac{\mathcal{R}'^2}{\mathcal{R}_0^2} \sim \frac{16}{e^2(2N^3)^{1/2}} \left(\frac{2e}{N}\right)^{2N}. \tag{21}$$

This is a very small result, and emphasizes rather than compensates the small ratio given by (15). Hence, except for very special circumstances, the final conclusion is that

$$\frac{\sigma'}{\sigma} < \frac{\sigma'}{\sigma_0} \ll 1.$$

For the small-k limit cited here, one can estimate that the reversal from circular-polarization dominance to linear-polarization dominance would occur for only slightly larger N values than have already been investigated experimentally. That is, the reversal might occur at about $N=4$ or 5.

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[9] It is convenient to refer specifically to hydrogen atom parameters. However, the arguments given here are applicable to any single-electron atom with central potential.

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