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Multiphoton Ionization of One-Electron Atoms with Circularly Polarized Light[*]

P. Lambropoulos[†]
Argonne National Laboratory, Argonne, Illinois 60439

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We present simple expressions for the differential as well as the total cross sections for multiphoton ionization (of any order) of a one-electron atom with circularly polarized light. We discuss their usefulness in the interpretation of multiphoton ionization experiments.

In a previous paper[1] we discussed the effect of light polarization on the cross sections for two- and three-photon ionization of a one-electron model of an atom. It is the purpose of this paper to present an expression for the cross section for N-photon ionization with circularly polarized light of an s state of a one-electron atomic model. It turns out that this cross section can be expressed in a simple form in terms of only one dynamical coefficient. This result will be very useful in comparing experimental data with calculations, especially for the alkali atoms. Calculations[2-5] have usually been based on one-electron models of the atom and have invariably been performed for linearly polarized light. As will be shown subsequently, however, circularly polarized light provides a far more direct comparison between theory and experiment. This will be particularly useful in N-photon ionization for $N>3$, as is the case in some recent experiments.[6-8] All of these experiments can be performed equally easily with circularly polarized light.

In view of the recent surge of multiphoton ionization experiments,[6-11] the present results are immediately applicable to measurements of total cross sections. Although there do not seem to exist any measurements of photoelectron angular distributions, such studies are just beginning,[12] and are expected to receive increasing attention in the immediate future. It is through such experiments that the atomic parameters which determine the rate of the process can be delineated and their relation to atomic structure be determined.

Let $|nlm\rangle \equiv R_{nl}(r)Y_{lm}(\vartheta,\varphi)$ be the orthonormal set of the atomic wave functions, and

$$f_{\vec{K}}(\vec{r}) \equiv 4\pi \sum_{L=0}^{\infty} i^L \exp(-i\delta_L) F_{KL}(r) \times \sum_{M=-L}^{+L} Y_{LM}^*(\theta,\phi) Y_{LM}(\vartheta,\varphi) \qquad (1)$$

the final state of the emitted photoelectron.[13] The spherical coordinates of its wave vector $\vec{K}$, in the laboratory system, are K, θ, and ϕ, where θ is the angle between the z axis and $\vec{K}$, and ϕ the angle between the x axis and the projection of $\vec{K}$ on the xy plane of an (xyz) right-handed system of Cartesian coordinates. Similarly, the spherical coordinates of $\vec{r}$ are r, ϑ, φ. The function $F_{KL}(r)$ is the radial part of the Lth partial wave, and δ_L the corresponding phase shift. Both F_{KL} and R_{nl} can be assumed to be real; the Y_{lm} are spherical harmonics. Spin-orbit coupling is ignored, and the energy of $|nlm\rangle$ is denoted by $\hbar\omega_{nl}$.

The light is assumed traveling along the positive z axis. If it is circularly polarized, its polarization vector is $(\vec{\epsilon}_x \pm i\vec{\epsilon}_y)/\sqrt{2}$, the upper sign corresponding to left and the lower to right circular polarization, and where $\epsilon_x = \epsilon_y = 1$. Thus, in the dipole approximation, circularly polarized light interacts with the atom through the operators $x \pm iy$. On the basis of parity conservation, it is *a priori* expected that the total cross sections, as well as the angular distributions, will be the same for left and right circular polarization.

In general, the probability per unit time that a photoelectron due to N-photon ionization will be emitted within a solid angle $d\Omega \equiv \sin\theta \, d\theta \, d\phi$ can be written[3]

$$dW_N/d\Omega = I_1 I_2 \cdots I_N \, d\hat{\sigma}_N/d\Omega.$$

The quantity $d\hat{\sigma}_N/d\Omega$ has the dimensions cm^{2N} sec^{N-1} and shall be referred to as "generalized cross section" (gcs), while $I_1, I_2, \cdots, I_N$ are the intensities of N light beams with respective frequencies $\omega_1, \omega_2, \cdots, \omega_N$. Light intensities are here measured in photons/cm^2 sec. Strictly speaking, the above formula for $dW_N/d\Omega$ is valid only when the intensities are δ functions in frequency. For the more realistic case of beams with nonzero frequency width, integrations over $\omega_1, \omega_2, \cdots, \omega_N$ will be involved. This is particularly important if resonances with intermediate states exist. In most actual experiments, all N photons are absorbed from the same light beam. Subsequently, we shall indicate how the equations should be modified in that case. It should perhaps be emphasized that resonances alone do not alter the angular distributions. Only when the resonant intermediate state is influenced by other interactions which couple the m states, may the angular distribution be affected.[12]

Using perturbation theory of the appropriate order, we obtain

$$d\hat{\sigma}_N{}^c/d\Omega = (\pi\alpha)^N (m/\pi\hbar) K \omega_1 \omega_2 \cdots \omega_N A_{2N}{}^{(N)} \sin^{2N}\theta, \tag{2}$$

where α is the fine-structure constant, m the electron mass, and the superscript c indicates circularly polarized light. The wave vector K is given by

$$K = [2m(\omega_1 + \omega_2 + \cdots + \omega_N - \omega_I)/\hbar]^{1/2},$$

where $\hbar\omega_I$ is the ionization energy from the initial state $|i00\rangle$. The coefficient $A_{2N}{}^{(N)}$ is given by

$$A_{2N}{}^{(N)} = \Big| \sum_{n_{N-1} \cdots n_2 n_1} \; \sum_{P(\omega_1, \omega_2, \cdots, \omega_N)} F(KN; n_{N-1}(N-1)) R(n_{N-1}(N-1); n_{N-2}(N-2)) \cdots R(n_2 2; n_1 1) R(n_1 1; i0)$$
$$\times [\omega_{n_{N-1}(N-1)} - \omega_{i0} - \omega_1 - \omega_2 \cdots - \omega_{N-1}]^{-1} \cdots [\omega_{n_2 2} - \omega_{i0} - \omega_1 - \omega_2]^{-1} [\omega_{n_1 1} - \omega_{i0} - \omega_1]^{-1} \Big|^2. \tag{3}$$

The R's are radial matrix elements defined by

$$R(n'l'; nl) \equiv \int_0^\infty R_{n'l'}(r) r^3 R_{nl}(r) \, dr,$$
$$F(KN; n_{N-1}(N-1)) \equiv \int_0^\infty F_{KN}(r) r^3 R_{n_{N-1}(N-1)}(r) \, dr.$$

The first sum in Eq. (3) extends over all intermediate states. Each n_j $(j = 1, 2, \cdots, N-1)$ runs over all atomic states including the continuum. This summation is only over the quantum number n because the summations over l and m have been performed during the calculation. In each step, these summations reduce to only a few terms as a result of the selection rules. The second sum in Eq. (3) is over all permutations $P(\omega_1 \omega_2 \cdots \omega_N)$ of the frequencies $\omega_1, \omega_2, \cdots, \omega_N$ taking $N-1$ of them each time. This leads to $N!$ terms.

In the case of only one light beam of frequency ω_0 and of small, but not zero, frequency width, one can set $\omega_1 = \omega_2 = \cdots = \omega_N = \omega_0$ and $I_1 I_2 \cdots I_n = I^N$, where I is the intensity of the beam. The sum over $P(\omega_1 \omega_2 \cdots \omega_N)$ is then replaced by a factor $N!$. This factor has not been included in previous calculations,[2-5] which have assumed that only one mode of the radiation field is excited. Note that, in the present calculation, the field was expanded in terms of plane waves inside a box of volume V and the limit $V \to \infty$ was taken. The beam of a pulsed or Q-switched laser contains a large number of such plane-wave modes, in fact, a continuum with a frequency width determined by the particular laser. For example, for a Q-switched Nd$^+$-glass laser, the width may be as high as 100 Å. Under such multimode condition, a factor $(N!)^2$ would be involved in the expression for $A_{2N}{}^{(N)}$. The omission of this factor may well be one of the reasons for the large discrepancy between experiment and calculations found in Ref. 7. For the sake of simplicity, we have bypassed all questions of photon statistics[14,15] and spatiotemporal structure[16,17] of the light beam. These effects, which have been discussed elsewhere, would introduce additional factors in the expression for $dW_N/d\Omega$.

To obtain the total gcs, Eq. (2) should be integrated over the solid angle $d\Omega$. The result is

$$\hat{\sigma}_N{}^c = (4\pi\alpha)^N \frac{m}{\hbar} K \omega_1 \omega_2 \cdots \omega_N \frac{4(N!)^2}{(2N+1)!} A_{2N}{}^{(N)}. \tag{4}$$

Combining Eqs. (2) and (4) one can write

$$\frac{d\hat{\sigma}_N{}^c}{d\Omega} = \hat{\sigma}_N{}^c \frac{(2N+1)!}{\pi 4^{N+1}(N!)^2} \sin^{2N}\theta \tag{5}$$

from which one can determine $\hat{\sigma}_N{}^c$ by measuring $d\hat{\sigma}_N{}^c/d\Omega$. Observing, for example, that $(N!)^2/(2N+1)! < 1$, one can always find an angle θ_N such that $(2N+1)! \sin^{2N}(\theta_N)/(N!)^2 = 1$. By measuring the differential gcs at that angle, one also obtains the total gcs through the equation $\hat{\sigma}_N{}^c = 4^{N+1}\pi(d\hat{\sigma}_N{}^c/d\Omega)_{\theta=\theta_N}$. The most remarkable feature of these results is that for circularly polarized light, the gcs contains only one atomic parameter $A_{2N}{}^{(N)}$. This is because the operators $x \pm iy$ have nonvanishing matrix elements only between (l, m) and $(l+1, m\pm 1)$ states, respectively.[18]

As a result, the emitted electron has angular momentum $L=N$, and only one partial wave from the expansion of Eq. (1) contributes. For linearly polarized light, $N+1$ or N parameters (depending on whether N is even or odd) are involved in the angular distribution.[19] These parameters contain contributions from $(N+2)/2$ [or $(N+1)/2$ if N is odd] partial waves of the final state. This involves interference between the phase shifts of the various partial waves; and, to calculate the cross section, all such phase shifts are needed.

For the hydrogen atom, the phase shifts are known.[2] For other atoms, however, for which the one-electron model is used, the phase shifts are not easily obtained.[20] Usually they must be estimated using,[4] for example, quantum defect theory. For such cases, which are the most interesting experimentally, Eqs. (2) and (4) present a great advantage because the phase shift δ_N does not appear in the expression for the coefficient $A_{2N}{}^{(N)}$. As a result, its value involves only the radial part $F_{KN}(r)$ (i.e., the absolute value) of the Nth partial wave which can be related to, or be inferred from, single-photon ionization data. In addition, the single-parameter angular distribution renders the experimental determination of this parameter considerably easier.

Finally, the simplicity of these results also facilitates their computation. This is particularly important for $N>3$ because then the expressions for the coefficients occurring in the angular distribution for linearly polarized light become increasingly complicated with increasing N. Expressions for such angular distributions, as well as the details of the present calculation, will be presented in a future publication.

Note added.—After submission of this Letter, a Letter by Klarsfeld and Maquet[21] was published discussing related aspects of the same subject. The main difference between the two Letters is that here are presented explicit expressions for angular distributions, while Ref. 21 presents maximum values of the ratio of the total cross sections for circular and linear polarization.

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†Address after 15 September 1972: Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colo. 80302.

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