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Circular versus Linear Polarization in Multiphoton Ionization

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We evaluate the maximum allowed value of the ratio between the total N-photon ionization rates for circularly and linearly polarized incident light. It appears that this maximum increases very rapidly with N.

Recent experimental observations[1,2] indicating that the total two- and three-photon ionization rates of unpolarized cesium atoms depend on the state of polarization of the incident radiation have stimulated the theoretical interest in this new effect peculiar to multiphoton processes.[3-5] A similar effect was discussed a few years ago[6] in the case of molecular and crystalline anthracene irradiated with elliptically polarized light.

In this Letter we present a simple method for evaluating the maximum allowed value of the ratio $(\sigma'/\sigma)_N$ between the total N-photon ionization cross sections for circularly versus linearly polarized radiation. As prerequisites of our calculation we assume the validity of the nonrelativistic dipole approximation and of conventional perturbation theory. This is certainly justified in the optical range and for light intensities up to about 10^{11} W/cm^2. In addition, we shall use a simple one-electron central-field model, which provides a fairly good approximation for a large number of atomic systems, and restrict ourselves to photoelectrons ejected from a bound s state. It is very likely[7] that the complex internal structure of a many-electron atom will not appreciably affect the main result derived below.

Before considering the general case, it will be instructive to reexamine the situation for $N=2$ and $N=3$. The total two-photon ionization cross section for linearly polarized light is[4,8]

$$\sigma = 2\pi^2\alpha a_0^2(I/I_0)\omega(\tfrac{4}{45}|T_{1;2}|^2 + \tfrac{1}{9}|T_{1;0}|^2), \quad (1)$$

where α is the fine-structure constant, a_0 is the Bohr radius, I is the light intensity in W/cm^2, $I_0 = 7.019\times10^{16}$ W/cm^2, ω is the photon energy in atomic units, and $T_{1;2}$ and $T_{1;0}$ are reduced amplitudes for transitions from the initial s state to final d and s states, respectively, via intermediate p states, as allowed by the selection rule $\Delta l = \pm 1$. For circularly polarized radiation the total cross section is given by[4]

$$\sigma' = 2\pi^2\alpha a_0^2(I/I_0)\omega\tfrac{2}{15}|T_{1;2}|^2. \quad (2)$$

Comparison with Eq. (1) immediately yields $\max(\sigma'/\sigma)_2 = \frac{3}{2}$.

Although the total transition rates are, of course, independent of the coordinate system, the difference between Eqs. (1) and (2) will be better understood if the latter is conveniently chosen in each case. Thus, for linearly polarized incident radiation we shall use spherical coordinates with the polar axis along the unit

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polarization vector $\vec{\epsilon}$, whereas for circularly polarized light it is more suitable to take the polar axis along the wave vector of the incoming photons. The dipole interaction operator then assumes the forms $\vec{\epsilon}\cdot\vec{r} = (4\pi/3)^{1/2} r Y_{1,0}(\hat{r})$ in the first case, and $\vec{\epsilon}\cdot\vec{r} = \mp (4\pi/3)^{1/2} r Y_{1,\pm 1}(\hat{r})$ in the second. Correspondingly, the selection rule for the magnetic quantum number will be simply $\Delta m = 0$ for linear polarization, and $\Delta m = +1$ ($\Delta m = -1$) for left (right) circular polarization. It is obvious that from the two possible channels, $l = 0 \to 1 \to 2$ and $l = 0 \to 1 \to 0$, the second one is not compatible with the last selection rule.

Similarly, in the case of three-photon ionization one finds[5]

$$\sigma = \pi^2 \alpha a_0^2 (I/I_0)^2 \omega \left(\tfrac{4}{175}|T_{12;3}|^2 + \tfrac{1}{27}|T_{10;1} + \tfrac{4}{5}T_{12;1}|^2\right) \qquad (3)$$

for linearly polarized light, and

$$\sigma' = \pi^2 \alpha a_0^2 (I/I_0)^2 \omega \tfrac{2}{35}|T_{12;3}|^2 \qquad (4)$$

for circularly polarized light. Again, from the three channels $l = 0 \to 1 \to 2 \to 3$, $l = 0 \to 1 \to 0 \to 1$, and $l = 0 \to 1 \to 2 \to 1$ allowed by the selection rule $\Delta l = \pm 1$, those leading to a final p state are forbidden by the selection rule $\Delta m = +1$ (or $\Delta m = -1$) when the incident radiation is circularly polarized. From Eqs. (3) and (4), it readily follows that $\max(\sigma'/\sigma)_3 = \frac{5}{2}$.

It is an easy matter to generalize the above formulas for arbitrary N. For the present purposes it will be sufficient to notice that the total N-photon ionization cross section for linearly polarized light has the form

$$\sigma = 4\pi^2 \alpha a_0^2 (I/2I_0)^{N-1} \omega (C_N |T_{12\cdots;N}|^2 + \cdots), \qquad (5)$$

where the omitted terms are all positive and represent contributions of the channels leading to final states with angular momenta $L = N-2, N-4, \cdots$. On the other hand, for circularly polarized light the total N-photon ionization cross section reduces to

$$\sigma' = 4\pi^2 \alpha a_0^2 (I/2I_0)^{N-1} \omega C_N' |T_{12\cdots;N}|^2, \qquad (6)$$

since the other channels are forbidden by the selection rule for m. Comparing Eqs. (5) and (6), one immediately sees that the maximum allowed value of the ratio σ'/σ is equal to the ratio of the coefficients C_N' and C_N. These are determined exclusively by the coupling of orbital momenta in the channel $l = 0 \to 1 \to 2 \to \cdots \to N$, according to the additional selection rules $\Delta m = +1$ (-1) or $\Delta m = 0$, respectively. More precisely, taking into account the expressions of the dipole interaction operator given above, one gets

$$\max\left(\frac{\sigma'}{\sigma}\right)_N = \frac{C_N'}{C_N} = \prod_{j=1}^{N} \left| \frac{\langle Y_{j,j} | Y_{1,1} | Y_{j-1,j-1}\rangle}{\langle Y_{j,0} | Y_{1,0} | Y_{j-1,0}\rangle} \right|, \qquad (7)$$

or, in terms of Wigner's 3-j symbols,[9]

$$\max\left(\frac{\sigma'}{\sigma}\right)_N = \prod_{j=1}^{N} \begin{pmatrix} j & 1 & j-1 \\ -j & 1 & j-1 \end{pmatrix}^2 \begin{pmatrix} j & 1 & j-1 \\ 0 & 0 & 0 \end{pmatrix}^{-2}, \qquad (8)$$

which explicitly means

$$\max(\sigma'/\sigma)_N = (2N-1)!!/N!. \qquad (9)$$

For $N=2$ and 2, this gives for the maximum ratio the values $\frac{3}{2}$ and $\frac{5}{2}$, respectively, obtained previously,[3-5] which happen to be very close to the experimental ratios of 1.28 ± 0.2 and 2.15 ± 0.4 reported in the case of cesium irradiated with ruby-laser light.[1,2] A very remarkable feature of Eq. (9) is that the right-hand side increases exponentially with the number of absorbed photons. For large N one can use the asymptotic formula $\max(\sigma'/\sigma)_N \sim 2^N (\pi N)^{-1/2}$, giving a maximum ratio of about 1.8×10^2 for $N=10$, and about 1.3×10^5 for $N=20$.

Of course, the result stated above represents only a theoretical maximum, and has been obtained by assuming an initial s state. However, the detailed analysis performed in the case of two-photon ionization[4] strongly suggests that it also holds for an arbitrary initial bound state, and that the actual ratio will stay in the zone of high values near its maximum for a very large range of incident photon energies. Thus, it may be expected that in practice circularly polarized radiation will often prove much more efficient in multiphoton ionization than linearly polarized radiation. This point certainly deserves further experimental investigation, especially in the case of rare gases, where absorption of a large number of photons occurs.[10-13] Accurate measurements of the ratio σ'/σ under various conditions should also contribute to the understanding of the actual mechanism of multiphoton transitions.

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10.6-μm Laser Scattering from Cyclotron-Harmonic Waves in a Plasma

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We report the scattering of CO_2-laser light from driven cyclotron-harmonic waves in a collisionless plasma of density 4×10^{10} cm^{-3}. Using light-mixing spectroscopy, we observe scattering from waves with frequencies of several hundred megahertz, wavelengths from 2.0 to 0.75 mm, and associated fractional electron-density modulations as small as 10^{-5}.

Microwave scattering and electrostatic probes have a limited range of utility as probes of short-wavelength fluctuations in plasmas. Electrostatic probes disturb the plasma for a distance of the order of the sheath size and are impractical in high-temperature plasmas. Microwave scattering is capable of resolving only wavelengths larger than half the microwave wavelength ($\gtrsim 1$ mm), which is a serious limitation in many controllable laboratory plasmas and in plasmas suitable for fusion where the Debye wavelength λ_D is considerably less than 1 mm (i.e., in the Tokamak $\lambda_D = 5\times10^{-2}$ mm). In principle, scattering with a laser of suitable wavelength λ_l is a technique which is capable of resolving short-wavelength plasma fluctuations without perturbing the plasma. As we discuss below, in order to maximize the scattered power (which is proportional to λ_l^2) and still resolve the wavelengths of all collective phenomena, λ_l should be of the order of λ_D. In view of these considerations, the high-power CO_2 laser with λ_l of 1.06×10^{-2} mm is an excellent scattering source not previously utilized as a probe of collective phenomena in plasmas. Previously, light scattering from collective phenomena has only been done with visible lasers from highly transient, dense plasmas[1] whose properties are not easily controllable. In this Letter, we report CO_2-laser scattering from externally driven cyclotron-harmonic waves (Bernstein waves)[2] in a steady-state plasma whose properties are variable in a controlled manner. Using light-mixing spectroscopy,[3] we observe waves with frequencies of several hundred megahertz, levels of modulation of the electron density as low as 6×10^5 cm^{-3}, and plasma wavelengths between 2.0 and 0.75 mm (which was the shortest wavelength we were able to generate with an rf voltage applied to a probe). In principle, this technique is capable of resolving wavelengths between 2 mm and 5×10^{-3} mm (one half the laser wavelength).

The experimental apparatus is shown in Fig. 1.[4] The plasma is produced by a dc discharge between a filament and grid in 7×10^{-4} Torr of He gas. Plasma leaks into a region of uniform magnetic field between two magnetic mirrors located 90 cm apart. The plasma density is uniform to within $\pm15\%$ and the magnetic field $\vec{B}$ is uniform to within $\pm0.1\%$ over a region 5 cm in diameter and 5 cm long located halfway between the mirrors. Depending on the magnetic field, the electron temperature varies between 4 and 6 eV, and