PHYSICAL REVIEW A VOLUME 20, NUMBER 5 NOVEMBER 1979

Interference effects in the two-photon ionization of metastable helium

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(Received 3 October 1978)

Multipole interference effects are predicted in the generalized cross sections, and in the ratio $\hat{\sigma}_C/\hat{\sigma}_L$ of the cross sections for circularly and linearly polarized laser light for the case of the two-photon ionization of the $n = 2$ singlet metastable helium atoms in the wavelength range 5036–5050 Å.

The interaction of high-intensity radiation with atomic and molecular systems allows the observation of multiphoton absorption processes, which have attracted considerable interest because of their importance in the understanding of various physical processes associated with the development and the application of laser technology. In a multiphoton process (ionization), the final state is in the continuum and can therefore be written as a superposition of partial waves; i.e., states of well-defined angular momentum. Then for a given continuum final-state energy (which is determined by the number of photons needed to ionize) all angular momenta are available. As a result, light of any arbitrary polarization will lead to ionization. The total rate, however, depends on the light polarization. The light polarization influences not only the angular distribution, but also the total photoionization rate, as noted by Fox *et al.*[1] and Kogan *et al.*[2] in the case of two- and three-photon ionization of caesium.

In an earlier paper[3] we studied the two-photon ionization of helium from the 2^1S state in the frame work of the perturbation theory and the electric dipole approximation.

Recently, Mathur[4] calculated the cross sections for the two-photon ionization from 2^1S state of the helium atom, for linearly polarized incident light, in the electric-dipole-plus-quadrupole approximation. Here we extend this calculation to obtain the cross sections for the circularly polarized light. We also report the ratio $\hat{\sigma}_C/\hat{\sigma}_L$ of the two cross sections obtained for circular and linear polarizations respectively. We show the effect of the interference of multipoles in the cross sections and in the ratio $\hat{\sigma}_C/\hat{\sigma}_L$.

The generalized cross section for the two-photon ionization from an initial state a_g to a final state a_f of an atom is given by[5] (the notation used here are the same as in Ref. 5)

$$\hat{\sigma} = (2\pi\alpha\omega)^2 \frac{m}{\hbar} \frac{k}{(2\pi)^2} \int |K^{(2)}_{a_f,a_g}|^2 d\Omega_k \ . \tag{1}$$

The transition matrix $K^{(2)}_{a_f,a_g}$ involves an infinite summation over intermediate states. If we carry out the summation by including explicitly the contribution of N intermediate states and accounting for the remaining states by closure and assuming an average intermediate state energy equal to $\hbar\omega_{N+1}$, we get

$$K^{(2)}_{a_f,a_g} = \sum_{a_j=1}^{N} \langle a_f|R|a_j\rangle\langle a_j|R|a_g\rangle \left[(\omega_{a_j,a_g} - \omega + \tfrac{1}{2}i\gamma)^{-1} - (\omega_{N+1,a_g} - \omega + \tfrac{1}{2}i\gamma)^{-1}\right] + \langle a_f|R^2|a_g\rangle(\omega_{N+1,a_g} - \omega + \tfrac{1}{2}i\gamma)^{-1} , \tag{2}$$

where $R = V(\vec{r}_1) + V(\vec{r}_2)$, and $\vec{r}_1$ and $\vec{r}_2$ are the position coordinates of the target electrons. The interaction operator $V(\vec{r})$, when both dipole and quadrupole terms are included in the interaction of the radiation field with the target atom, is already defined in the case of linear and circular polarization.[6-9]

In the present calculations we have included explicitly three dipole allowed states (2^1P, 3^1P, and 4^1P) and two quadrupole allowed states (3^1D and 4^1D) in the summation in Eq. (2). The wave functions for these bound states and the final continuum state are the same as used earlier.[4]

When the incident photon frequency is approximately equal to the energy difference between the two states of the helium atom, connected via a dipole transition ($2^1S - 3^1P$) or via a quadrupole transition ($2^1S - 3^1D$), resonance peaks will be obtained in the generalized cross section, corresponding to such photon frequencies. At a particular wavelength in the vicinity of a resonance, the nonresonant states contribute very little towards the ionization cross section and the contribution of the closure term is found to be negligible. Therefore, at resonance Eq. (1) will yield a simple expression for the ratio $\hat{\sigma}_C/\hat{\sigma}_L$ of the two cross sections for circular and linear polarizations. For example, the expression for $\hat{\sigma}_C/\hat{\sigma}_L$ at resonance corresponding to a quadrupole transition (2^1S-3^1D), which occurs at an incident photon frequency of 5043.5 Å, is given as

$$\hat{\sigma}_C/\hat{\sigma}_L = 1.43\, S_3^2/(S_1^2 + \tfrac{8}{7} S_3^2)\,,$$

where

$$S_l = \int_0^\infty e^{-\beta r_2} G_l^*(k, r_1)\, g_{3d}(r_1 r_2) r_1^3 r_2^2\, dr_1\, dr_2$$

and $G_l(k,r)$ is the radial part of the continuum wave function corresponding to the partial wave of angular momentum l. $g_{3d}(r_1 r_2)$ is the radial part of the $3d$ bound-state wave function, and $\beta = 2$.

Figures 1 and 2 show the variation with wavelength of the generalized cross section obtained for the circularly polarized light in the wavelength ranges 5016.2–5018 and 5042–5045 Å, respectively. Similar to the case of linear polarization,[4] these figures (full-line curves) show resonance peaks in the cross section at 5017.08 and 5043.5 Å corresponding to the dipole and quadrupole allowed 3^1P and 3^1D intermediate states, respectively. Again we see that in the range 5042–5045 Å the quadrupole contribution dominates over the usual pure dipole contribution (Fig. 2, broken line) which is seen to remain nearly constant in this region. The interference between the dipole and quadrupole contributions leads to a significant enhancement of the generalized cross section in the wavelength range 5042–5045 Å.

In Fig. 3, we show the variation of the ratio of the generalized cross sections for circularly and linearly polarized light ($\hat{\sigma}_C/\hat{\sigma}_L$) in the wavelength range 5036–5050 Å. The upper broken-line curve shows the variation of $\hat{\sigma}_C/\hat{\sigma}_L$ corresponding to the contributions in Eq. (1) from the dipole terms only. We see that the ratio $\hat{\sigma}_C/\hat{\sigma}_L$ at first increases slowly and finally becomes constant giving a max-

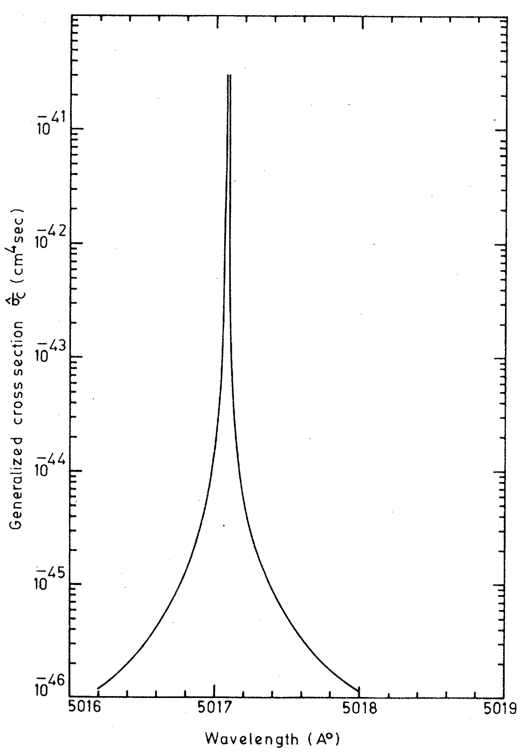

FIG. 1. Generalized cross section ($\hat{\sigma}_C$) for the two-photon ionization of He from the 2^1S state for circularly polarized incident light. Solid line, dipole-plus-quadrupole contribution.

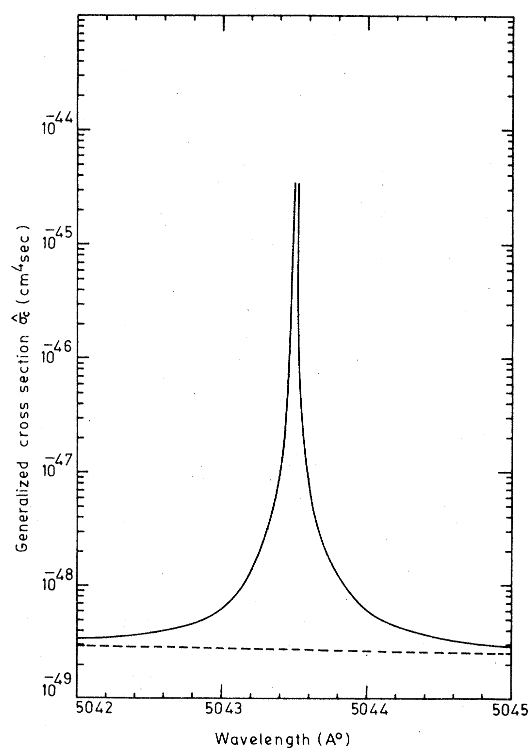

FIG. 2. Generalized cross section ($\hat{\sigma}_C$) for the two-photon ionization of He in the 2^1S state for circularly polarized incident light. Solid line, dipole-plus-quadrupole contribution; dashed line, pure dipole contribution.

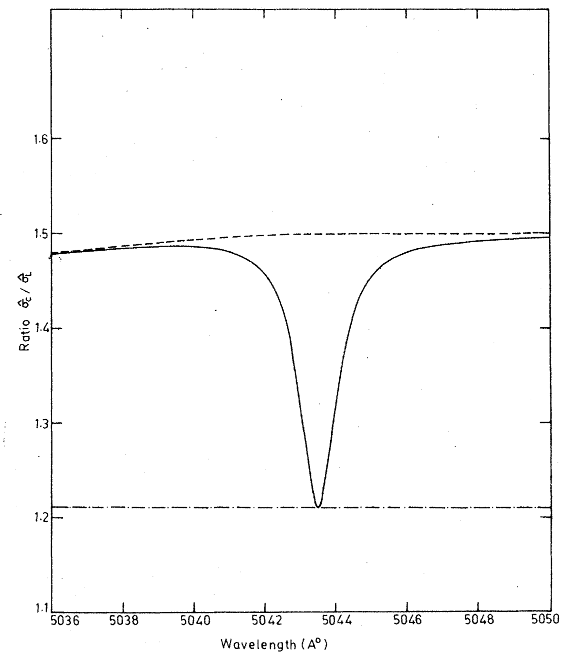

FIG. 3. Ratio $\hat{\sigma}_C/\hat{\sigma}_L$ for circularly and linearly polarized incident light for the two-photon ionization of He from 2^1S state. Dashed line, pure dipole contribution; dot-dashed line, pure quadrupole contribution; solid line, dipole-plus-quadrupole contribution.

imum value of 1.5. This is in agreement with the theory of Klarsfeld and Maquet,[10] in which they have shown that the maximum ratio between the total N-photon ionization rates for circularly and linearly polarized incident light, from a bound S-state has a value $(2N-1)!!/N!$.

The lower dot-dashed curve gives the variation of $\hat{\sigma}_C/\hat{\sigma}_L$ when the contribution from quadrupole term alone is considered. We observe that it remains constant over the whole wavelength range under consideration.

The full-line curve shows the variation of the ratio $\hat{\sigma}_C/\hat{\sigma}_L$ when both dipole and quadrupole contributions are taken into account in the calculation of the cross sections. From here we see that the ratio shows a sharp variation over the photon frequencies shown in Fig. 3. It is noted that the ratio decreases from its initial value of 1.478 at 5036 Å (which is nearly equal to the pure dipole value 1.479) and acquires a minimum value of 1.21 at 5043.5 Å (which is equal to the pure quadrupole value), and then rises and acquires a value of 1.493 at 5050 Å which again nearly coincides with the pure dipole value of the ratio at this wavelength. This variation in the ratio, in the wavelength region 5036–5050 Å is caused due to the multipole interference effects. In this wavelength region the interference is between dipole and quadrupole contributions. The higher multipoles (say octupole) do not effect, since their contribution is negligible. The matrix elements corresponding to the octupole allowed transitions are much smaller compared to the dipole and quadrupole transitions and also, the energy denominators for the octupole allowed states would be nonresonant in the wavelength range under consideration.

An experimental verification of the above result would be of great interest, as an accurate knowledge of the ratio would be of considerable importance in knowing the nature of interactions and in the understanding of the actual mechanism of the multiphoton processes. From the knowledge of the ratio, one can also get information about the matrix elements for the bound free transitions. Further in the wavelength range studied here the magnitude of the cross sections, with the inclusion of the quadrupole effects become substantial. The existence of tunable lasers would render such measurements feasible at the present time.

ACKNOWLEDGMENTS

We thank Dr. S. N. Gupta for some helpful discussion. We are also thankful to the University Grants Commission (UGC), India for providing financial assistance for this work. One of us (R.K.S.) is also thankful to the UGC for a fellowship.

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