Coherence properties of radiation and multiphoton resonances

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(Submitted June 6, 1974)
Zh. Eksp. Teor. Fiz. **68**, 465–468 (February 1975)

It is shown that from the shape of the absorption line of the tuning-adjustment light in the presence of intense, low-frequency radiation, "absolute" information about the statistical properties of the latter can be recovered. The theory includes the case when the transition probability does not exhibit a power-law dependence on the intensity of the radiation, which is especially important for absorption in highly nonlinear media. The feasibility of the extraction of analogous information in resonant multiphoton ionization is also investigated.

Recently there has been a sharp increase of interest in resonance effects associated with multiphoton excitation and ionization of atoms, related first of all to the possibility of an investigation of the energy spectrum of an atom in an intense electromagnetic field. As is well known, resonant multiphoton effects allow an adequate theoretical description of the experimental situation. On the other hand, one of the urgent problems in statistical optics is utilization of the methods of multiphoton spectroscopy, enabling us to determine the higher-order correlation functions of the electromagnetic field variables. The utilization of direct (nonresonant) multiphoton ionization for these purposes was discussed in the review[1]. For a power-law dependence of the ionization probability on the intensity of the electromagnetic field, the probabilities of direct n-photon ionization for coherent (w_c) and incoherent (w_{inc}) radiation are related by the following equation:

$$w_{\text{нк}}=\chi_n w_{\text{к}}. \tag{1}$$

The statistical factor χ_n is the ratio of the corresponding correlation functions. The relative nature of the obtained information, mentioned in[1], follows immediately from here (we need a standard source with statistical properties known beforehand). The results of the well-known articles[2,3], which investigate the manifestation of the statistical properties of the electromagnetic fields in multiphoton bound-bound transitions, essentially correspond to this same "relative" case.

The goal of the present work is the development of a method for the extraction of "absolute" information about the statistical properties of an intense electromagnetic field, utilizing multiphoton spectroscopy. Two cases will be investigated.

A) The characteristic features of resonant multiphoton transitions in systems with nonvanishing average values of their dipole moments, permitting exact solutions in a strong electromagnetic field (the hydrogen atom, hydrogenlike local centers and excitons in crystals, noncentrosymmetric molecules, etc.).

B) The case of intermediate resonances associated with multiphoton ionization. (In this connection, for simplicity the transition through a resonance level, which does not possess a dipole moment, will be treated.)

A) Let us show that, from the shape of the absorption line of the "tuning" light of frequency Ω in the presence of an intense, low-frequency, nonresonant electromagnetic field of frequency ω, absolute information about the statistical properties of the latter can be reestablished. With regard to the electromagnetic field of frequency ω, for simplicity we assume that its statistical properties are determined by the intensity distribution $P(L)$. In the case when the level i with nonvanishing average dipole moment d_i is separated by a wide energy gap from the remaining part of the spectrum, the possibility arises of the derivation of the exact solution, describing the behavior of this dipole moment in an external electromagnetic field.[4,5] Using this fact and omitting the details of the calculation, we present the corresponding expression for the function $F(\Omega)$ characterizing the shape of the absorption line for the light of frequency Ω:

$$F(\Omega)=\sum_{n=n_0} F_n(\Omega), \tag{2}$$

$$F_n(\Omega)=A\int d\Omega\int dL\frac{\pi^{-1}\gamma J_n^2(\sqrt{L/L_0})\rho(\Omega)P(L)}{(\Delta_{12}+n\omega-\Omega+\varepsilon(L))^2+\gamma^2},$$
$$L_0=\frac{\hbar^2\omega^2 c}{4\pi(d_1-d_2)^2},\quad n_0=-\left[\frac{\Delta_{12}}{\omega}\right], \tag{3}$$

$\hbar\Delta_{12}$ is the energy gap separating the ground state (1) and the excited state (2) of the absorbing system, and $\hbar\gamma$ and $\hbar\epsilon(L)\equiv\hbar\alpha L$ denote the width and the Stark shift of the excited state. For simplicity we neglect the width and the shift of the ground state; $J_n(x)$ denotes the Bessel function of real argument, $\rho(\Omega)$ is the spectral intensity of the light of frequency Ω, c is the speed of light, and [. . .] denotes the integer part of a number.

a) If the light of frequency Ω possesses a broad spectral region $\delta\Omega\gg\gamma$ in which $\rho(\Omega)$ is a smooth function of Ω, then integration over Ω gives

$$F_n(\Omega)=A\bar\rho\int_0^\infty P(L)J_n^2\left(\sqrt{\frac{L}{L_0}}\right)dL. \tag{4}$$

($\rho(\Omega)$ is taken outside the integral sign at the average value $\bar\rho$.) Let us consider two limiting cases for the function $P(L)$: a "δ-shaped" distribution $P^{(\delta)}(L) = \delta(L-\bar L)$ and a Gaussian (with respect to the field strength E of the EMF, $E\sim\sqrt{L}$) distribution

$$P^{(G)}(L)=(\bar L)^{-1}\exp\{-L/\bar L\},\quad \bar L=\int_0^\infty LP(L)dL.$$

It is not difficult to derive the following expression for the statistical factor $\chi_n = F_n^{(G)}/F_n^{(\delta)}$:

$$\chi_n=\exp\{-\bar L/2L_0\}I_n(\bar L/2L_0)/J_n^2(\sqrt{\bar L/L_0}). \tag{5}$$

Here $I_n(x)$ is a modified Bessel function. As follows from the derived formula, the statistical factor χ_n tends to infinity at those values of the intensity $\overline{L}$ corresponding to zeros of the Bessel function $J_n(\sqrt{\overline{L}/L_0})$. (Of course, for small intensities ($\overline{L}/L_0 \ll 1$) $\chi_n = n!$). The divergence of the statistical factor associated with specific intensities is related to the different nature of the dependences of the shape functions on $\overline{L}$ (a smooth dependence for $F_n^{(G)}$ and a pulsing dependence for $F_n^{(\delta)}$). This circumstance may be utilized in order to obtain "absolute" information about the statistical properties of the EMF of frequency ω.

b) For a narrow spectral line ($\delta\Omega \ll \gamma$), assuming $\rho(\Omega) = \rho_0\delta(\Omega - \Omega_0)$ we find

$$F_n(\Omega) = A\rho_0 \int_0^\infty dL \frac{\pi^{-1}\gamma J_n^2(\sqrt{L/L_0})P(L)}{(\Delta_{12}+n\omega-\Omega+\varepsilon(L))^2+\gamma^2}. \tag{6}$$

In this connection, if $\epsilon(\overline{L}) \gg \gamma$ (which is usually satisfied under the conditions of a contemporary experiment), the Lorentzian in formula (6) may turn out to be a more abrupt function of L than P(L) (for symmetric distributions P(L); the latter implies that the halfwidth of such a distribution is greater than γ/α; the fulfilment of the condition $\epsilon(\overline{L}) \gg \gamma$ is sufficient for $P^{(G)}(L)$). We find

$$F_n(\Omega_0) = A\rho_0(\alpha)^{-1}P(x)J_n^2(\sqrt{x/L_0}), \qquad \alpha x = \Delta_{12}+n\omega-\Omega_0 > 0. \tag{7}$$

As follows from formula (7), a direct connection exists between the shape of the absorption line for the light of frequency Ω_0 and the form of the distribution function P(L). Experimentally plotting the dependence $F_n(\Omega_0)$, one can immediately reestablish P(L). It is especially important to emphasize that formula (7) may be used in order to reestablish the form of P(L) associated with a non-power-law dependence of the transition probability on the intensity of the electromagnetic field, that is, it applies in practice to the region of intense fields. In connection with the development of picosecond lasers, the investigation of nonlinear effects in solids associated with electromagnetic field strengths of 10^6 to 10^7 V/cm is possible. In this connection the properties of the medium may be modified and the function P(L), determined from such experiments, gives "absolute" information about the actual distribution of the electromagnetic-field intensity in the medium. (It is obvious that the chosen example of a non-power-law dependence of the transition probability on L due to a strong perturbation of the states with nonvanishing average dipole moment is not the only possibility (see, for example,[6]).) An interesting consequence of the derived formula is the appearance on one side of the line profile of zeros in the absorption, i.e., the shape of the absorption line corresponds to a one-sided pulse curve. The position of the zeros in Eq. (7) allows one, in particular, to determine the dynamic polarizability in the excited state if its dipole moment is known, or to determine the dipole moment if the polarizability is known. For certain solid state and molecular systems, the latter quantity is of considerable interest.

We note that the question of the manifestation of the coherence properties of radiation in a double optical resonance was treated in similar aspects by Zusman and Burshteĭn.[7]

B) In conclusion let us investigate the case of resonant multiphoton ionization. Let us show that the realization of resonances in multiphoton ionization may also serve as a source of "absolute" information about the statistical properties of the ionizing radiation. The formula for the probability of k_0-photon ionization in the presence of a single intermediate r-photon resonance of the p-th discrete level has the form

$$w_{k_0} = B\int P(L)L^r\mathscr{L}_r(L)\,dL, \tag{8}$$

$$\mathscr{L}_r(L) = \frac{\pi^{-1}\gamma_p(L)}{(\Delta_p - r\omega+\varepsilon_p)^2+\Gamma_p^2}, \quad \Gamma_p = \gamma_p^0+\gamma_p(L), \quad \gamma_p(L) = \beta_p L^{k_0-r}, \quad \varepsilon_p = \alpha_p L, \quad k_0 = [\mathscr{E}/\hbar\omega]+1, \tag{9}$$

$\mathscr{E}$ is the ionization potential of the atom, $\hbar\Delta_p$ and $\hbar\gamma_p^0$ denote the excitation energy and radiative width of the resonance level, and $\hbar\gamma_p(L)$ and $\hbar\epsilon_p(L)$ denote its ionization width and Stark shift. If $\epsilon_p(\overline{L}) \gg \gamma_p^0, \gamma_p(\overline{L})$, which is often realized experimentally, then for the average number N of photoelectrons we find (for $\gamma_p(\overline{L}) \ll \gamma_p^0$)

$$\overline{N} = N_0TB(\alpha_p\gamma_p^0)^{-1}\beta_p P(y)y^{k_0}, \qquad \alpha_p y = \Delta_p - r\omega > 0; \tag{10}$$

for $\gamma_p(\overline{L}) \gg \gamma_p^0$ we have

$$\overline{N} = N_0TB(\alpha_p)^{-1}P(y)y^r, \tag{11}$$

N_0 is the average number of neutral atoms per unit volume, and T is the duration of the laser pulse. As follows from formulas (10) and (11), an investigation of resonant multiphoton ionization enables one to reestablish the distribution function for the intensity of the laser radiation from the frequency dependence of the average number of photoelectrons.

The case $\epsilon(\overline{L}) \ll \gamma_p^0, \gamma_p(\overline{L})$ only gives relative information:

$$\frac{\overline{N}^{(G)}}{\overline{N}^{(\delta)}} = k_0! \quad \gamma_p(\overline{L}) \ll \gamma_p^0; \qquad \frac{\overline{N}^{(G)}}{\overline{N}^{(\delta)}} = r! \quad \gamma_p(\overline{L}) \gg \gamma_p^0. \tag{12}$$

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Translated by H. H. Nickle
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