Polarization properties of photon echo at small areas of the exciting pulses

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The polarization characteristics of photon echo in a gas medium are calculated for transitions with arbitrary angular momenta of the levels. These results point to the possibility of performing new experiments for the purpose of identifying molecular (atomic) transitions.

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Experiments on photon echo in molecular gases (see, e.g., Refs. 1–3) are being performed on transitions with large angular momenta of the levels. Usually it is even impossible to identify the branch of the molecular transition (P, R, or Q) on which the photon echo is produced. The reason is that the theoretical investigations of the polarization properties of photon echo[4–7] are limited only to cases of small angular momenta of the transitions. We note that an attempt was made in Ref. 5 to consider the case of arbitrary angular momenta of the levels, but the calculation method used there did not yield results for either $j \to j$ or $j \rightleftarrows j+1$ transitions.

The purpose of the present paper is to calculate the polarization characteristics of photon echo in a gas medium with arbitrary angular momenta of the transition levels. We hope that this will serve as a stimulus for organizing new experiments capable of identifying the corresponding transitions.

Consider the formation of photon echo gas by two exciting light pulses of durations T_1 and T_2; the pulses are linearly polarized at an angle ψ to each other and are resonant to the molecular transition with frequency ω_0 and with level angular momenta j_b and j_a. We determine the intensity of the electric field of the photon echo by using the d'Alambert equation

$$\Box \mathbf{E} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\int \mathbf{P}\, d\mathbf{v}.$$

The material polarization vector $\mathbf{P}$ of a group of molecules moving with velocity $\mathbf{v}$ can be obtained by solving the equations for the corresponding components $\rho_{\mu m}$, $\rho_{\mu\mu'}$ and $\rho_{mm'}$ of the density matrix, pertaining to the considered molecular transition. Here μ and μ' denote the projections of the angular momentum j_a of the lower level, while m and m' denote the projections of the angular momentum j_b of the upper level. These equations are of the usual form and we solve them by expanding $\rho_{\mu m}$, $\rho_{\mu\mu'}$, and $\rho_{mm'}$ in irreducible tensor operators.[8] For example

$$\rho_{\mu m} = (-1)^{j_a-\mu}(2j_a+1)^{-1/2}\sum_{\kappa,q}(2\kappa+1)\begin{pmatrix} j_a & j_b & \kappa \\ \mu & -m & q \end{pmatrix}\psi_q^{(\kappa)}$$

The relations between $\rho_{mm'}$ and $f_q^{(\kappa)}$, as well as between $\rho_{\mu\mu'}$ and $\phi_q^{(\kappa)}$ are similar. We note that the circular components of the polarization vector $\mathbf{P}^q$ of the medium are expressed in terms of the component $\psi_q^{(1)}$. Equations for $\psi_q^{(\kappa)}, f_q^{(\kappa)}$ and $\phi_q^{(\kappa)}$ can be found, for example, in Ref. 9.

The solution of the system of equations for $\psi_q^{(\kappa)}, f_q^{(\kappa)}$, and $\phi_q^{(\kappa)}$ was obtained in an approximation in which the exciting pulses are assumed to have small areas. This implies satisfaction of the inequality

$$|d|\, e^{(i)} T_i / (2j_b + 1)^{1/2} \hbar << 1, \qquad i = 1, 2, \tag{1}$$

where d is the reduced matrix element of the dipole-moment operator for the considered transition $j_b \to j_a$, while $e^{(1)}$ and $e^{(2)}$ are the amplitudes of the first and second excited pulses. In addition, it was assumed that the durations T_1 and T_2 of the exciting pulses are small compared with the time interval τ_s between them and times of the irreversible relaxations. The procedure for the calculation of the intensity of the electric field of the photon echo is similar to that used in Ref. 4. For the case of a narrow spectral line ($1/T_0 \ll 1/T_i$, $i = 1,2$) and for exciting pulses propagating along the Y axis, we have

$$\mathbf{E}^e = -\frac{\pi}{\hbar^3}\, \omega_0 \frac{L}{c} |d|^4 e^{(1)} T_1 e^{(2)2} T_2^2 N_0 \exp\left[-\frac{(t' - \tau_s)^2}{4T_0^2}\right]$$

$$\times\, \mathbf{e}^e \exp[-\gamma^{(1)}(t' + \tau_s)] \exp[i(\omega_0 t - k y + 2\Phi_2 - \Phi_1)] + \text{c.c.}, \tag{2}$$

$$e_z^e = \frac{1}{3} A(j_b, j_a) \cos\psi, \qquad e_x^e = \frac{1}{2} B(j_b, j_a) \sin\psi, \tag{3}$$

$$A(j, j) = \frac{2(3j^2 + 3j - 1)}{5j(j + 1)(2j + 1)}, \qquad B(j, j) = \frac{4(j - 1)(j + 2)}{15j(j + 1)(2j + 1)}, \tag{4}$$

$$\begin{aligned} A(j + 1, j) = A(j, j + 1) &= \frac{2(4j^2 + 8j + 5)}{5(j + 1)(2j + 1)(2j + 3)}, \\ B(j + 1, j) = B(j, j + 1) &= -\frac{8j(j + 2)}{15(j + 1)(2j + 1)(2j + 3)}, \end{aligned} \tag{5}$$

$$t' = t - \tau_s - T_1 - T_2 - y/c, \quad N_0 = n_b - n_a, \quad \gamma^{(1)} = (\gamma_a^{(0)} + \gamma_b^{(0)})/2 + \Gamma^{(1)}.$$

Here T_0 is the time of the reversible Doppler relaxation, which is expressed in terms of the average thermal velocity u of the gas molecules; $T_0 = 1/ku$; L is the dimension of the gas medium, Φ_1 and Φ_2 are the constant phase shifts of the first and second exciting pulses, and $(2j_b + 1)n_b$ and $(2j_a + 1)n_a$ are the densities of the molecules on the upper and lower levels prior to the incidence of the exciting pulses on the medium. Next, $1/\gamma_a^{(0)}$ and $1/\gamma_b^{(0)}$ are the relaxation times of levels a and b via gaskinetic inelastic collisions and radiative decay, and $\Gamma^{(1)}$ characterizes the broadening of the spectral line of the $j_b \to j_a$ transition under the influence of elastic depolarizing collisions. In (2) and (3) the Z axis coincides with the polarization of the second exciting pulse, while the polarization of the first pulse makes an angle $\pi/2 - \psi$ with the positive X axis. Formulas (2)–(5) describe the direction of polarization of the photon echo both for $j \to j$ $j \rightleftarrows j+1$ transitions at arbitrary values of j. In particular, for transitions with small angular momenta of the levels, $0 \rightleftarrows 1$, $1/2 \to 1/2$, $1 \to 1$, and $1/2 \rightleftarrows 3/2$, the photon-echo polarizaion properties that follow from (2)–(5) coincide with those obtained in Refs. 4–6. We note that the polarization properties of the echo on these transitions do not change even if the inequality (1) is violated.

For experiments performed with molecular gases, the case of large angular momenta ($j \gg 1$) is especially important. In this case we have from (3)–(5)

$$e_z^e = \frac{1}{5j}\cos\psi, \quad e_x^e = \frac{1}{15j}\sin\psi \quad \text{for transitions} \quad j \to j,$$

$$e_z^e = \frac{2}{15j}\cos\psi, \quad e_x^e = -\frac{1}{15j}\sin\psi \quad \text{for transitions} \quad j \rightleftarrows j+1.$$

This means that for the $j \to j$ transitions (Q branch) the echo polarization vector lies between the polarization vectors of the exciting pulses, and its angle θ_e with the Z axis is defined by the equation $\tan\theta_e = (\tan\psi)/3$. For $j \rightleftarrows j+1$ transitions (P and R branches) the echo polarization vector lies outside the angle between the polarization vectors of the exciting pulses, and the angle θ_e is determined for $\tan\theta_e = -(\tan\psi)/2$.

The simple relations obtained for the photon-echo polarization indicate that photon-echo experiments in gases should be performed with exciting pulses that have small areas. This makes it easy to identify the type of transition, $j \to j$ or $j \rightleftarrows j+1$, on which the photon echo is produced. We note that formulas (3)–(5) are valid also in the case of a broad spectral line ($1/T_0 \gg 1/T_i$).

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