Photon echo on adjacent optically allowed transitions

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We examine the formation of a photon echo in a three-level system in two adjacent optically allowed transitions. In the limit of small areas of the exciting pulses, we obtain the intensity and polarization of the echo as a function of the polarization of the exciting pulses. The calculations are valid for arbitrary angular momenta of the levels and for arbitrary ratio between the durations of the exciting light pulses and the time of the reversible Doppler relaxation. It is shown that the considered three-level systems can be divided, in accord with their polarization properties, into four groups, each of which can be identified by the photon-echo method. The indicated groups differ from one another by the values of the total angular momenta of the resonance levels and constitute transitions of the form $j \to j \to j$, $j \to j+1 \to j$, $j \to j \to j+1$, and $j \to j+1 \to j+2$.

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The photon echo phenomenon is being intensively investigated of late both experimentally and theoretically, since it permits the study of rapid relaxation processes in media, as well as the identification of the corresponding resonant transitions.

To describe the photon echo in a gas medium it is necessary in principle to take into account the degeneracy of the levels of the considered transitions, a degeneracy due to the different orientation of the total angular momentum j of the atom (molecule). A two-level model that takes this degeneracy into account was used in the theoretical papers[1-5] to describe the polarization properties of photon echo in gases. It is shown in these papers that the dependence of the polarization properties of the photon echo on the angular momenta of the resonant-transition levels makes it possible to identify these levels by the photon-echo method. At small j it is possible to establish the angular momenta of the transition levels exactly, and at $j \gg 1$ it is possible to determine the type of transition ($j \to j$ or $j \rightleftarrows j+1$).

Experimental investigations of the polarization properties of photon echo in molecular gases[6-11] were made on transitions with large level angular momenta. A comparison of the experimental results with the theory makes it possible in a number of cases to identify unambiguously the type of the transition. In other cases, however, the polarization properties of the photon echo differ from the results that follow from the two-level model. This disagreement is due apparently to the fact that the echo is formed in these cases either on an aggregate of independent transitions, one of which is $j_1 \to j_1$ and the other $j_2 \to j_2+1$, or on adjacent transitions with one common level.

Formation of photon echo in the three-level system with one common level b and two levels a and c that are close to each other was discussed in the literature many times (see, e.g., Refs. 12–17). We note that in all the papers devoted to such systems the degeneracy of the levels was neglected. This made it impossible to consider the polarization properties of the photon echo. An exception was the work of Alekssev and Basharov,[17] who took the level degeneracy into account. However, the analysis of the polarization properties of the photon echo in Ref. 17 was limited to the case of a narrow spectral line, $1/T_0 \ll 1/T_i$ ($i=1,2$), and small angular momenta of the levels ($j_a=1 \to j_b=0 \to j_c=1$, $1 \to 1 \to 0$, $\frac{1}{2} \to \frac{1}{2} \to \frac{3}{2}$ and $\frac{1}{2} \to \frac{3}{2} \to \frac{1}{2}$). Here T_1 and T_2 are the durations of the first and second exciting light pulses, and T_0 is the time of the reversible Doppler relaxation.

The present paper is devoted to an investigation of the formation of a photon echo on two adjacent optically allowed transitions $j_a \to j_b \to j_c$ at arbitrary values of the angular momenta of the levels and at an arbitrary ratio of T_0 and T_i. Obviously, without simplifying assumptions it is apparently impossible to obtain an analytic solution for such a problem. We have therefore used an approximation in which the areas of the exciting pulses are assumed small.[5] This has made it possible to investigate the polarization properties of the photon echo at arbitrary angular momenta of the levels for the case of both narrow and broad ($1/T_0 \gg 1/T_i$, $i=1,2$) spectral lines.

1. BASIC EQUATIONS AND RELATIONS

We use as our basis the d'Alambert equation

$$\Box \mathbf{E} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\int \mathbf{P}\, d\mathbf{v} \tag{1}$$

and the quantum mechanical equations for the components of the density matrix, in which account is taken of the interaction of the atoms (molecules) of the gas with the electromagnetic field and of the irreversible relaxation:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_b\right)\rho_{mm'}=\frac{i}{\hbar}[(\mathbf{Ed})_{m\mu}\rho_{\mu m'}-\rho_{m\mu}(\mathbf{Ed})_{\mu m'}+(\mathbf{Ed})_{m\nu}\rho_{\nu m'}-\rho_{m\nu}(\mathbf{Ed})_{\nu m'}], \tag{2}$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_c\right)\rho_{\nu\nu'}=\frac{i}{\hbar}[(\mathbf{Ed})_{\nu m}\rho_{m\nu'}-\rho_{\nu m}(\mathbf{Ed})_{m\nu'}], \tag{3}$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_{ab}-i\omega_0\right)\rho_{\mu m}=\frac{i}{\hbar}[(\mathbf{Ed})_{\mu m'}\rho_{m'm}-\rho_{\mu\mu'}(\mathbf{Ed})_{\mu' m}-\rho_{\mu\nu'}(\mathbf{Ed})_{\nu' m}], \tag{4}$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_{ac}-i\Delta\right)\rho_{\mu\nu}=\frac{i}{\hbar}[(\mathbf{Ed})_{\mu m}\rho_{m\nu}-\rho_{\mu m}(\mathbf{Ed})_{m\nu}]; \tag{5}$$

The subscripts μ, m, and ν label here the projections of the total angular momentum j_a, j_b, and j_c of the considered levels a, b, and c. The equation for the density-matrix components $\rho_{\mu\mu'}$ of the level a are next obtained from (3) by making the substitutions $\gamma_c \to \gamma_a$ and $\nu \to \mu$, and the equation for the components of the den-

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sity matrix $\rho_{\nu m}$ of the transition between the levels b and c is obtained from Eq. (4) by using the change of notation $\gamma_{ab} \to \gamma_{cb}$, $\omega_0 \to \bar{\omega}_0$ and $\mu \rightleftharpoons \nu$. Here ω_0 is the frequency of the optically allowed transition with change of total angular momentum $j_b \to j_a$, $\bar{\omega}_0$ is the analogous quantity for the optically allowed transition, $j_b \to j_c$, and $\Delta = \omega_0 - \bar{\omega}_0$ is the frequency of the optically forbidden transition $j_c \to j_a$. The vectors $\mathbf{d}_{m\mu}$ and $\mathbf{d}_{m\nu}$ are the matrix elements of the operator of the dipole moment of the atom for the transitions between the level b and the levels a and c, respectively. Finally, $1/\gamma_a$, $1/\gamma_b$ and $1/\gamma_c$ are the times of the irreversible relaxation of the populations of the levels a, b, and c, while $1/\gamma_{ab}$, $1/\gamma_{cb}$, and $1/\gamma_{ac}$ characterizes the relaxation times of the corresponding components of the density matrix. Summation over repeated indices is implied in Eqs. (2)–(5).

The medium-polarization vector $\mathbf{P}$ in (1) pertains to a group of atoms moving with velocity $\mathbf{v}$ and is connected with the components $\rho_{\mu m}$ and $\rho_{\nu m}$ of the density matrix by the relation

$$\mathbf{P}=\sum_{\mu,m}\rho_{\mu m}\,\mathbf{d}_{m\mu}+\sum_{\nu,m}\rho_{\nu m}\,\mathbf{d}_{m\nu}+\text{c.c.},\tag{6}$$

Let the exciting light pulses, which are linearly polarized at an angle ψ to each other, propagate along the Y axis with a carrier frequency ω and with electric field intensities

$$\mathbf{E}_1=(\mathbf{l}_x\sin\psi+\mathbf{l}_z\cos\psi)\,e^{(1)}\exp[i(\omega t-ky+\Phi_1)]+\text{c.c.},\tag{7}$$

$$\mathbf{E}_2=\mathbf{l}_z e^{(2)}\exp[i(\omega t-ky+\Phi_2)]+\text{c.c.},\tag{8}$$

where $e^{(1)}$ and $e^{(2)}$ are the constant real amplitudes, Φ_1 and Φ_2 are constant phase shifts, and $\mathbf{l}_x$ and $\mathbf{l}_z$ are the unit vectors of the corresponding Cartesian axes.

Prior to the incidence of the first exciting pulse on the medium we have

$$\rho_{\mu m}=\rho_{\nu m}=\rho_{\mu\nu}=0,\quad \rho_{\mu\mu'}=n_a f(v)\,\delta_{\mu\mu'},$$

where $(2j_a+1)n_a$ is the density of the atoms on the level a at $t-y/c\leqslant 0$, and $f(v)$ is the Maxwellian distribution function. The initial levels for $\rho_{mm'}$ and $\rho_{\nu\nu'}$ are obtained from the initial condition for $\rho_{\mu\mu'}$ by making respectively the substitutions $n_a \to n_b$, $\mu \to m$ and $n_a \to n_c$, $\mu \to \nu$.

The solution of the system of equations (2)–(5) for the components of the density matrix is best carried out by expanding them in irreducible tensor operators.[18] For example, for $\rho_{\mu m}$ we have

$$\rho_{\mu m}=(-1)^{j_a-\mu}(2j_a+1)^{-1/2}\sum_{\varkappa,q}(2\varkappa+1)\begin{pmatrix} j_a & j_b & \varkappa\\ \mu & -m & q\end{pmatrix}\psi_q^{(\varkappa)},\tag{9}$$

and the connection of $\rho_{\nu m}$ with $\chi_q^{(\varkappa)}$ is obtained from (9) by making the changes

$$j_a\to j_c,\quad \mu\to\nu.$$

Similar relations hold also for the changes of $\rho_{mm'}$ with $f_q^{(\varkappa)}$, $\rho_{\mu\mu'}$ with $\varphi_q^{(\varkappa)}$, $\rho_{\nu\nu'}$ with $\xi_q^{(\varkappa)}$ and $\rho_{\mu\nu}$ with $\beta_q^{(\varkappa)}$. We note that the circular components of the polarization vector of the medium, P, defined by formula (6) are connected with the quantities $\psi_q^{(1)}$ with $\chi_q^{(1)}$.

The procedure of calculating the electric field intensity of a photon echo produced on adjacent optically allowed transitions is similar to that used in Ref. 5 for the case of a two-level system. The system of equations for the slow functions is solved in the given-field approximation (7) and (8). As a result we have for $\psi_q^{(\varkappa)}(t-y/c)$ and $\chi_q^{(\varkappa)}(t-y/c)$ at the instant of time $t=T_1+y/c$ when the first exciting pulse leaves the point y of the gas medium

$$\psi_q^{(\varkappa)}(T_1)=(-1)^{1+j_a-j_b}(2j_a+1)^{1/2}\frac{p_1}{d_1}C_q^{(\varkappa)}(\psi)\exp[i(\omega T_1+\Phi_1)],\tag{10}$$

where

$$p_1=-i|d_1|^2e^{(1)}N_0f(v)\,[\sin\delta_1T_1+i(1-\cos\delta_1T_1)]/3\hbar\delta_1,$$
$$N_0=n_b-n_a,\quad \delta_1=kv-\Delta\omega,\quad \Delta\omega=\omega-\omega_0,\quad \Delta\bar{\omega}=\omega-\bar{\omega}_0,\tag{11}$$

and the quantity $C_q^{(\varkappa)}(\psi)$ is given in Ref. 5. The expression for $\chi_q^{(\varkappa)}(T_1)$ is obtained from (10) by making the substitutions $j_a\to j_c$, $d_1\to d_2$ and $p_1\to p_2$, where p_2 is obtained from (11) by the transformations

$$d_1\to d_2,\quad \delta_1\to\delta_2=kv-\Delta\bar{\omega},\quad N_0\to\bar{N}_0=n_b-n_c.$$

Here d_1 and d_2 are the reduced matrix elements of the dipole moment of the atom for the transitions $j_b\to j_a$ and $j_b\to j_c$, respectively.

Expression (10) and the corresponding expression for $\chi_q^{(\varkappa)}(T_1)$ serve as the initial conditions in the solution of the equations for the slow functions in the time domain $T_1\leqslant t-y/c\leqslant\tau_s+T_1$, where τ_s is the time interval between the exciting light pulses. In this region, dephasing of the individual radiators takes place on account of the thermal motion of the atoms, but the medium retains the phase memory of the first exciting pulse. In the subsequent calculations we shall assume that τ_s is so small that the irreversible relaxation during the time of formation of the echo can be neglected.

In the time interval $\tau_s+T_1\leqslant t-y/c\leqslant\tau_s+T_1+T_2$, a second exciting pulse (8) passes through the medium. At the instant of time $t=\tau_s+T_1+T_2+y/c$, when it leaves the point y of the gas medium, the part of $\psi_q^{(\varkappa)}(T_2)$ that contributes to the echo is of the form

$$\psi_q^{(1)}(T_2)=(-1)^{j_a-j_b}(2j_a+1)^{1/2}\frac{3e^{(2)2}}{\hbar^2d_1}F_{1q}\cdot$$
$$\cdot\exp\{i[\omega(\tau_s+T_1+T_2)-kv\tau_s+2\Phi_2-\Phi_1]\},\tag{12}$$

where

$$F_{1q}=\frac{|d_1|^2}{\delta_1}\Big\{\alpha_{1q}\frac{1}{\delta_1}(1-\cos\delta_1T_2)+\gamma_{1q}\Big[\frac{2}{\delta_1+\delta_2}(e^{-i\delta_2T_2}-e^{i\delta_1T_2})$$
$$-\frac{2}{\delta_2}(e^{-i\delta_2T_2}-1)+\frac{1}{\delta_1}(e^{i\Delta T_2}-1)+\frac{\Delta}{\delta_1\delta_2}(e^{-i\delta_2T_2}-1+e^{i\Delta T_2}-e^{i\delta_1T_2})$$
$$-\frac{\Delta}{\delta_1(\delta_1+\delta_2)}(e^{-i\delta_2T_2}-e^{i\delta_1T_2})\Big]\Big\},\tag{13}$$

$$\alpha_{1q}=-p_1^*\exp(i\Delta\omega\tau_s)\Big[\frac{1}{3}\delta_{q,0}A(j_b,j_a)\cos\psi+\frac{1}{2\sqrt{2}}(\delta_{q,1}-\delta_{q,-1})B(j_b,j_a)\sin\psi\Big].\tag{14}$$

The expression for γ_{1q} is obtained from α_{1q} by the substitutions

$$p_1\to p_2,\ \Delta\omega\to\Delta\bar{\omega},\ A(j_b,j_a)\to F(j_b,j_a,j_c),$$
$$B(j_b,j_a)\to G(j_b,j_a,j_c).$$

Next, the expression for $\chi_q^{(\varkappa)}(T_2)$ is obtained from (12) by the substitutions

$$j_a\to j_c,\quad d_1\to d_2,\quad F_{1q}\to F_{2q},$$

and the quantities F_{2q}, α_{2q} and γ_{2q} are obtained from F_{1q}, α_{1q} and γ_{1q} by the change

$$j_a \rightleftarrows j_c, \quad \delta_1 \rightleftarrows \delta_2, \quad d_1 \to d_2, \quad p_1 \rightleftarrows p_2,$$

of notation

$$\Delta\omega \rightleftarrows \Delta\tilde{\omega}, \ \Delta \to -\Delta.$$

Finally, the quantities $A(j_b, j_a)$ and $B(j_b, j_a)$ are given in Ref. 5, and $F(j_b, j_a, j_c) = F(j_b, j_c, j_a)$ and $G(j_b, j_a, j_c) = G(j_b, j_c, j_a)$ for different groups of transitions take the form

$$F(j,j,j) = \frac{3j^2+3j-1}{5j(j+1)(2j+1)}, \quad G(j,j,j) = \frac{2(j-1)(j+2)}{15j(j+1)(2j+1)}, \tag{15}$$

$$F(j+1,j,j) = \frac{4j^2+8j+5}{5(j+1)(2j+1)(2j+3)},$$
$$G(j+1,j,j) = -\frac{4j(j+2)}{15(j+1)(2j+1)(2j+3)}, \tag{16}$$

$$F(j,j,j+1) = \frac{j+2}{5(j+1)(2j+1)}, \quad G(j,j,j+1) = -\frac{j-3}{15(j+1)(2j+1)}, \tag{17}$$

$$F(j+1,j,j+2) = \frac{2}{5(2j+3)}, \quad G(j+1,j,j+2) = \frac{1}{5(2j+3)}. \tag{18}$$

Expression (12) and the corresponding expression for $\chi_q^{(1)}(T_2)$ serve as the initial conditions for the solution of the equations for the slow functions in the region $\tau_s + T_1 + T_2 \leq t - y/c$ after the passage of the second exciting pulse. Omitting the intermediate calculations, we write down the final equation for the intensity of the electric field of the photon echo:

$$\mathbf{E}^e = -6i\pi\omega \frac{Le^{(2)2}}{c\hbar^2} \exp\left\{ i\left[\omega_0\left(t - \frac{y}{c}\right) + \Delta\omega(\tau_s + T_1 + T_2) + 2\Phi_2 - \Phi_1 \right]\right\} \int dv \exp\{ikv(t' - \tau_s)\}[\mathbf{F}_1 + \mathbf{F}_2 \exp\{-i\Delta t'\}] + \text{c.c.}, \tag{19}$$

where

$$t' = t - \tau_s - T_1 - T_2 - y/c.$$

The components of the vetors F_1 and F_2 are connected with F_{1q} and F_{2q} by the relations

$$F_{ix} = \sqrt{2}(F_{i1} - F_{i,-1}), \quad F_{iy} = 0, \quad F_{iz} = F_{i0}; \quad i = 1, 2.$$

The photon echo (19) is in the general case elliptically polarized. At $\Delta\tau_s > 1$ the envelope passing through the maxima of the echo intensity oscillates as a function τ_s (quantum beats), with a frequency Δ. These beats were predicted from qualitative considerations in Ref. 13.

2. DISCUSSION OF RESULTS

To simplify the discussion, we consider the case of symmetrical tuning of the frequency of the exciting pulses relative to the central frequencies of the considered transitions $\Delta\omega = -\Delta\bar{\omega} = -\Delta\sqrt{2}$. In addition, we put $T_1 = T_2 = T$, as is usually the case in experiment.[6-11]

An important role in the theoretical investigation of the photon echo is played by the relation between the duration T of the exciting light pulses and the time of reversible Doppler relaxation $T_0 = 1/ku$, where u is the average thermal velocity of the atoms (molecules) of the gas. We consider two limiting cases of a narrow $(1/T_0 \ll 1/T)$ and a broad $(1/T_0 \gg 1/T)$ spectral line.

When the echo is formed on a narrow spectral line, the integral in (19) can be calculated analytically for various combinations of the parameters Δ, T_0, and T in all cases with the exception of $\Delta \sim 1/T_0$. As a result we have the following expression for the electric field intensity of the photon echo formed on a narrow spectral line:

$$\mathbf{E}^e = AI \exp\{i[\omega_0(t - y/c) - {}^1/_2\Delta(\tau_s + 2T) + 2\Phi_2 - \Phi_1]\} \cdot [\mathbf{G}_1 + \mathbf{G}_2 \exp(-i\Delta t')] + \text{c.c.}, \tag{20}$$

where

$$A = -\pi\omega Le^{(1)}e^{(2)2}T^3/c\hbar^3, \quad I = \exp[-(t' - \tau_s)^2/4T_0^2],$$

$$\mathbf{G}_1 = \frac{16}{(\Delta T)^3}|d_1|^2 \left\{ \left[\frac{1}{3}A(j_b, j_a)\cos\psi \mathbf{l}_z + \frac{1}{2}B(j_b, j_a)\sin\psi \mathbf{l}_x \right] \cdot \left(1 - \cos\frac{\Delta}{2}T\right)|d_1|^2 N_0 \sigma \exp\left(-i\frac{\Delta}{2}\tau_s\right) - \left[\frac{1}{3}F(j_b, j_a, j_c)\cos\psi \mathbf{l}_z + \frac{1}{2}G(j_b, j_a, j_c)\sin\psi \mathbf{l}_x \right] |d_2|^2 \bar{N}_0 \sigma^* \exp\left(i\frac{\Delta}{2}\tau_s\right)\left(1 - 2\cos\frac{\Delta}{2}T + \cos\Delta T + i\sin\Delta T - 2i\sin\frac{\Delta}{2}T\right)\right\}, \tag{21}$$

$$\sigma = \sin\frac{\Delta}{2}T - i\left(1 - \cos\frac{\Delta}{2}T\right),$$

and the vector G_2 is obtained from (21) by the substitutions

$$d_1 \rightleftarrows d_2, \quad N_0 \rightleftarrows \bar{N}_0, \quad j_a \rightleftarrows j_c, \quad \Delta \to -\Delta.$$

We note that the case $\Delta \sim 1/T_0$ can be investigated by numerically integrating (19).

It follows from (20) and (21) that the maximum of the amplitude of an echo on a narrow spectral line is reached at the instant of time $t = 2\tau_s + 2T + y/c$, and the duration of the echo pulse is of the order of T_0. At $\Delta \gtrsim 1/T$, as follows from (20) and (21), modulation oscillations of the intensity with frequency Δ take place within the photon-echo pulse. In the case $1/T_0 \ll \Delta \ll 1/T$ there are likewise modulation oscillations, but (20) takes a much simpler form

$$\mathbf{E}^e = AIN_0 \exp\{i[\omega_0(t - y/c) - \Delta\tau_s + 2\Phi_2 - \Phi_1]\} \cdot \left\{ \frac{1}{3}\cos\psi[A(j_b, j_a)|d_1|^4 + A(j_b, j_c)|d_2|^4 \exp[-i\Delta(t' - \tau_s)] + 2F(j_b, j_a, j_c)|d_1|^2|d_2|^2(e^{i\Delta\tau_s} + e^{-i\Delta t'})]\mathbf{l}_z + \frac{1}{2}\sin\psi[B(j_b, j_a)|d_1|^4 + B(j_b, j_c)|d_2|^4 \exp[-i\Delta(t' - \tau_s)] + 2G(j_b, j_a, j_c)|d_1|^2|d_2|^2(e^{i\Delta\tau_s} + e^{-i\Delta t'})\mathbf{l}_x \right\} + \text{c.c.}, \tag{22}$$

where we have put for simplicity $N_0 = \bar{N}_0$. We note that Eq. (22) describes the intensity of the electric field of the photon echo also at $\Delta \ll 1/T_0 \ll 1/T$, when no modulation oscillations are observed inside the echo pulse.

In the general case the echo (22) is electrically polarized. At $1/T_0 \ll \Delta \ll 1/T$ the polarization-ellipse axes oscillate at a frequency $\Delta/2$ over the length of the echo pulse. For very close levels $\Delta \ll 1/T_0 \ll 1/T$, the polarization ellipse contracts to a line.

We emphasize that the maximum of the echo amplitude (21) is always linearly polarized. Denoting by θ_e the angle between the echo polarization vector at the maximum of the amplitude and the polarization vector of the second exciting pulse, we obtain

$$\text{tg}\,\theta_e = {}^3/_2\, \text{tg}\,\psi[B(j_b, j_a)|d_1|^4 + B(j_b, j_c)|d_2|^4 + 4G(j_b, j_a, j_c)|d_1|^2|d_2|^2\cos\Delta\tau_s][A(j_b, j_a)|d_1|^4 + A(j_b, j_c)|d_2|^4 + 4F(j_b, j_a, j_c)|d_1|^2|d_2|^2\cos\Delta\tau_s]^{-1}. \tag{23}$$

This equation is valid for arbitrary angular momenta of the levels. For small angular momenta $j_a = 1 \to j_b = 0 \to j_c = 1$, $1 \to 1 \to 0$ and $\frac{1}{2} \to \frac{3}{2} \to \frac{1}{2}$ the polarization properties of the photon echo at the maximum, which follow from (23), agree with the corresponding results of Ref.

17. For the transition $\frac{1}{2}\to\frac{1}{2}\to\frac{3}{2}$, Eq. (23) also goes over in the result of Ref. 17 if we use the small-area approximation in the latter.

As follows from (15)–(18) and (23), with respect to the polarization properties of the photon echo we can divide the systems under consideration into four groups. For the first group ($j\to j\to j$), using (15) and the expressions for $A(j,j)$ and $B(j,j)$ from Ref. 5, we get

$$\operatorname{tg}\theta_e=(j-1)(j+2)\operatorname{tg}\psi/(3j^2+3j-1). \tag{24}$$

For the second group ($j\to j+1\to j$) we get from (16) and from the expressions for $A(j+1,j)$ and $B(j+1,j)$ (Ref. 5)

$$\operatorname{tg}\theta_e=-2j(j+2)\operatorname{tg}\psi/(4j^2+8j+5). \tag{25}$$

We emphasize that for the first group of transitions the dependence of θ_e on ψ is the same as when the echo is formed on an assembly of two independent transitions $j\to j$ and $j\to j$, while for the second group of transitions this dependence is the same as when the echo is formed on two independent transitions $j+1\to j$ and $j+1\to j$.

For the two other groups of transitions, as follows from (23), the echo polarization plane, as a function of τ_s, executes quantum beats similar to the quantum beats of the echo intensity. We note that for the case $\frac{1}{2}\to\frac{1}{2}\to\frac{3}{2}$ this circumstance was pointed out in Ref. 17.

For experiments in molecular gases, a particular important role is played by the case of large angular momenta ($j\gg 1$). In this case we have from (24) and (25) for the first two groups of transitions

$$\operatorname{tg}\theta_e={}^1/_3\operatorname{tg}\psi,\quad j\to j\to j, \tag{26}$$

$$\operatorname{tg}\theta_e=-{}^1/_2\operatorname{tg}\psi,\quad j\to j+1\to j. \tag{27}$$

For the two other groups of the transitions we get from (17), (18), and (23) at $j\gg 1$

$$\operatorname{tg}\theta=\operatorname{tg}\psi(|d_1|^4-|d_2|^4-|d_1|^2|d_2|^2\cos\Delta\tau_s)(3|d_1|^4+2|d_2|^4+2|d_1|^2|d_2|^2\cos\Delta\tau_s)^{-1},\quad j\to j\to j+1, \tag{28}$$

$$\operatorname{tg}\theta_e=-\operatorname{tg}\psi(|d_1|^4+|d_2|^4-3|d_1|^2|d_2|^2\cos\Delta\tau_s)\cdot[2(|d_1|^4+|d_2|^4+2|d_1|^2|d_2|^2\cos\Delta\tau_s)]^{-1},\quad j\to j+1\to j+2. \tag{29}$$

In the case $|d_1|=|d_2|$ and $\Delta\tau_s\ll 1$ we have from (28) and (29)

$$\operatorname{tg}\theta_e=-{}^1/_7\operatorname{tg}\psi,\quad j\to j\to j+1, \tag{30}$$

$$\operatorname{tg}\theta_e={}^1/_8\operatorname{tg}\psi,\quad j\to j+1\to j+2. \tag{31}$$

We consider now the case of echo formation on a broad spectral line. We note that it is precisely this case which takes place in experiments on photon echo in molecular gases.[6-11] In the case of echo formation on a broad spectral line, the maximum of the echo amplitude is usually shifted relative to the instant of time $t=2\tau_s+2T+y/c$, and the duration of the echo pulse is of the order of the duration T of the exciting pulses. Therefore at $\Delta T>1$, as follows from (19), in the case of a broad spectral line modulation oscillations of the intensity take place within the echo pulse. At different relations between the parameters Δ, T_0, and T, Eq. (19) can be simplified. We shall dwell on the case $\Delta T\ll 1$, when the integral in (19) can be relatively easily calculated. The intensity of the electric field of the photon echo at $N_0=\overline{N}_0$ is given in the case by expression (22), in which I is of the form

$$I=\sqrt{\pi}\frac{T_0}{T}\{(a+1)^2[\Theta(a+1)-\Theta(a)]-(2a^2-2a-1)[\Theta(a)-\Theta(a-1)]+(a-2)^2[\Theta(a-1)-\Theta(a-2)]\}, \tag{32}$$

where

$$a=\frac{t'-\tau_s}{T},\quad \Theta(x)=\begin{cases}1, & x\geqslant 0\\ 0, & x<0\end{cases}.$$

As follows from (32), the maximum amplitude of an echo on a broad spectral line is shifted at $\Delta T\ll 1$ relative to the instant of time $t=2\tau_s+2T+y/c$ by $0.5T$, and the echo-pulse duration is of the order T. Thus, it follows from (22) and (32) that on a broad spectral line at $\Delta T\ll 1$ the echo is linearly polarized, and Eqs. (23)–(31) are valid not only at the maximum of the amplitude, just as for a narrow spectral line, but for the entire echo pulse.

We shall now dwell in greater detail on the case of echo formation in molecular gases on transitions with large angular momenta of the levels. If the echo is formed on two adjacent optically allowed transitions, then these systems can be divided, with respect to the polarization properties of the echo, into the following four groups. The first group includes systems in which each of the optically allowed transitions pertains to the Q branch, the second group includes systems in which each of the optically allowed transitions pertains to the $P(R)$ branch. The third group consists of the systems in which one of the optically allowed transitions pertains to the Q branch and the other to the P or R branch. Finally, the fourth group includes systems in which one of the optically allowed transitions pertains to the branch and the other to the R branch.

The figure shows the dependences of the angle θ_e on ψ, plotted in accordance with formulas (27), (30), (31), and (26) (curves 1–4, respectively). We note that in the figure the angle θ_e is assumed positive if the echo polarization vector lies outside the angle ψ, and negative if it is inside the angle ψ. This choice of the sign agrees with the work of Alimpiev and Karlov.[7,8] As follows from the figure, there is a large region of values of ψ where the curves are significantly separated, so that it is possible to identify experimentally each of the group of the adjacent optically allowed transitions. It must be borne in mind, however, that, as follows from Ref. 5, the first group of transitions is indis-

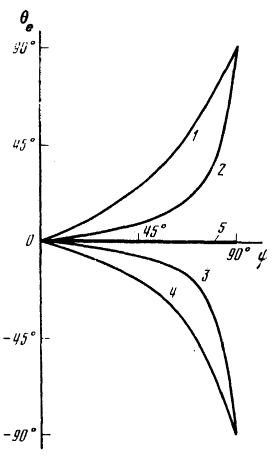

FIG. 1. Dependence of the angle θ_e on ψ. The angle θ_e is positive or negative, depending on whether the vector of the echo polarization lies outside or inside the angle ψ.

tinguishable with respect to the polarization properties from the case when the echo is formed in two independent transitions, each of which belongs to the Q branch. A similar remark holds also for the second group of transitions and for the case of formation of an echo on two independent transitions, each of which belongs to the $P(R)$ branch, or one of them belongs to the P branch and the other to the R branch.

The line 5 in the figure is a plot of θ_e against ψ when the echo is formed on two independent transitions, one of which belongs to the Q branch and the other to the P or R branch. This polarization dependence is obtained from Ref. 5 if the echo is formed on two independent transitions: $j_1 \to j_1 (j_1 \gg 1)$ and $j_2 \rightleftarrows j_2+1 (j_2 \gg 1)$ at $N_{01} = N_{02}$, $|d_1| = |d_2|$ and $(\omega_{01} - \omega_{02})\tau_s \ll 1$. Here N_0, d_1, and ω_{01} are the density of the excess population, the reduced dipole moment, and the frequency for the $j_1 \to j_1$ transition, while N_{02}, d_2, and ω_{02} are the analogous quantities for the transition $j_2 \rightleftarrows j_2+1$.

We note that the comparison of the experimental curves with those of the figure should be made at small areas of the exciting pulses, when the theory considered here is valid. This points for the need of organizing new experiments aimed at identifying molecular transitions by the photon-echo method.

We note in conclusion that if τ_s is comparable with the times of the irreversible relaxation, then the intensity of the echo will attenuate with time in accord with an exponential law that takes at $\gamma_{ab} = \gamma_{ab}$ the form $\exp(-2\gamma_{ab} t)$.

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Laser fluorescence detection of single atoms

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Results are presented of an investigation of the detection of single sodium atoms by means of a fluorescence method under conditions of cyclic interaction between the sodium atoms and a circularly polarized light wave of resonant frequency. To ensure multiple interaction between the atoms and laser radiation, the atoms are preliminarily oriented optically, as a result of which they occupy the $F=2$, $m_F=2$ sublevel from which only a transition to the $F'=3$, $m'_F=3$ sublevel is possible. The maximum mean signal from each sodium atom interacting with the radiation is $\bar{n}_{max}=2$ photoelectrons. The minimum recorded sodium atom flux in the experiment is ≈ 1 atom/sec for an atomic registration probability of 0.4. Strong suppression of the atomic absorption spectral line wings occurs on registration of single atoms when the fluorescence signal in the center of the absorption line is $\bar{n}_{ph} > 1$ photoelectron. It is shown that this method of suppression of spectral line wings can be used to detect low concentrations of the atoms of a certain element in the presence of a high concentration of another element when the distance between the spectral absorption lines of the elements is $\approx$0.1–100 cm^{-1}.

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INTRODUCTION

One of the pressing problems in the development of laser spectroscopy methods is the attainment of maximum sensitivity, up to a limit of spectroscopy of quantum states using single atoms and molecules.[1,2] In investigations of ultrasmall concentrations of atoms, the most effective are the fluorescence and photoionization methods, which have yielded the best results. Using cw dye lasers and a standard fluorescence pro-

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