Ultrahigh resolution spectroscopy based on photon echo

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Photon echo was investigated in a gas containing atoms with free resonant energy levels, two of which result from hyperfine splitting or from some other cause and are very close. The frequencies of the resonant atomic transitions lie within the limits of the Doppler width of the spectral line. The role of degeneracy of the resonant level is determined. It is established that in the case of a narrow spectral line it is possible to construct an envelope passing through the maxima of the photon-echo amplitudes produced with different time intervals between the exciting pulses. This envelope oscillates with a frequency that is directly connected with the distance between the close levels. The planes of polarizations of the photon echo, which correspond to the maxima of the amplitudes, also oscillate under certain conditions, and at the same frequency. In the case of a broad spectral line it is possible to obtain, besides the phenomenon noted above, also oscillations of the average intensity of an individual photon echo, and sometimes oscillations of the plane of its polarization, with a frequency likewise connected with the indicated energy distance. The experimental observation of these observations makes it possible to determine the distance between close levels, as well as the dipole moments of the resonances of the atomic transitions.

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If two exciting ultrashort light pulses separated by a time interval τ_s pass through a resonant medium, then after an approximate time interval τ_s, following the second exciting pulse, spontaneous coherent emission is produced by the excited atoms of the medium, and is called photon echo[1] (see the review).[2]

The photon-echo phenomenon was first used for an experimental determination of the distance between two very close nodegenerate levels in[3], where the hyperfine splitting of energy levels of chromium impurity ions in a ruby crystal lattice was investigated. In the case of a solid, analogous problems were discussed in[4-10]. The hyperfine splitting of gas-molecule levels was investigated experimentally with the aid of photon echo in[11]. The possibility of a spectroscopic resolution of very close levels of gas atoms by the photon-echo method was considered also in[12-15] without allowance for the degeneracy of the quantum states. However, allowance for the degeneracy of the energy levels is essential for a correct formulation of the problem and for its subsequent solution, particularly in those cases when the polarization properties of the photon echo are investigated besides the intensity.

We consider below photon echo in a degenerate three-level system with energies $E_0<E_1<E_2$, in which two levels are very close, $E_2-E_1\ll E_1-E_0$. The degeneracy is due to the different orientation of the total angular momentum. The intensity of the photon echo is determined in general form for any multiplicity of the level degeneracy, and its polarization properties are established for atomic transitions with small values of the total angular momentum. The results show that the use of photon echo in spectroscopy within the Doppler width can offer some advantages over traditional methods, since the intensity of the echo is proportional to the square of the density of the excited atoms. This increases noticeably the sensitivity of the measurement and makes it possible to investigate excited levels whose populations are greatly weakened by the Boltzmann factor.

1. FUNDAMENTAL EQUATIONS

We choose as the basis the d'Alambert equation for the vector potential **A**

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{A}=-\frac{4\pi}{c}\int d\mathbf{v}\,\mathrm{Sp}\,\rho\dot{\mathbf{d}} \tag{1}$$

and the quantum-mechanical equation for the density matrix ρ of an atom moving with velocity $\mathbf{v}$:

$$i\hbar\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho=\left(H-\frac{1}{c}\mathbf{A}\dot{\mathbf{d}}\right)\rho-\rho\left(H-\frac{1}{c}\mathbf{A}\dot{\mathbf{d}}\right), \tag{2}$$

where H is the Hamiltonian of the system in the c.m.s., and the superior dot over the electric dipole moment operator **d** denotes the operator of the time derivative of this quantity.

We introduce the convenient notation

$$(I_1{}^\alpha)_{\nu\nu'}=\rho_{\nu m}\dot{d}_{m\nu'}{}^\alpha,\quad (I_2{}^\alpha)_{\nu\nu'}=\rho_{\nu\mu}\dot{d}_{\mu\nu'}{}^\alpha,$$
$$(N_1{}^{\alpha\beta})_{\nu\nu'}=\rho_{\nu\nu''}\dot{d}_{\nu''m}{}^\alpha\dot{d}_{m\nu'}{}^\beta-\dot{d}_{\nu m'}{}^\alpha\rho_{m'm}\dot{d}_{m\nu'}{}^\beta,$$
$$(P_1{}^{\alpha\beta})_{\nu\nu'}=\dot{d}_{\nu\mu}{}^\alpha\rho_{\mu m}\dot{d}_{m\nu'}{}^\beta,\quad (\Pi_1{}^{\alpha\beta})_{\nu\nu'}=\dot{d}_{\nu m}{}^\alpha\dot{d}_{m\nu'}{}^\beta,$$

where ν, m, and μ are the projections of the total angular momentum **J** in the three considered states, which are characterized by the quantum numbers J_0, J_1, and J_2, and also by the respective energies E_0, E_1, and E_2. The summation convention is used for all the matrix and tensor dummy indices. The expressions for $(N_2^{\alpha\beta})_{\nu\nu'}$, $(P_2^{\alpha\beta})_{\nu\nu'}$ and $(\Pi_2^{\alpha\beta})_{\nu\nu'}$ are obtained from $(N_1^{\alpha\beta})_{\nu\nu'}$, $(P_1^{\alpha\beta})_{\nu\nu'}$ and $(\Pi_1^{\alpha\beta})_{\nu\nu'}$ by simultaneously replacing two indices $m\to\mu$, $\mu\to m$, and $1\to 2$. As a result the matrix indices of all the matrices become the projections of the angular momentum of the lower level, which will henceforth be omitted for the sake of brevity.

Let exciting pulses with linear or circular polarization propagate along the Z axis with a carrier frequency

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$\omega = kc$ and let them be described by the vector potentials

$$\mathbf{A}_1 = \mathbf{l}_1 a_1 \exp[i(kz - \omega t + \Phi_1)] + \text{c.c.}$$

at $0 \le t - z/c \le T_1$, and

$$\mathbf{A}_2 = \mathbf{l}_2 a_2 \exp[i(kz - \omega t + \Phi_2)] + \text{c.c.}$$

at $\tau_s + T_1 \le t - z/c \le \tau_s + T_1 + T_2$, where $\mathbf{l}_1$ and $\mathbf{l}_2$ are unit polarization vectors, a_1 and a_2 are constant real amplitudes, Φ_1 and Φ_2 are constant phase shifts, T_1 and T_2 are the pulse duration and τ_s is the time interval between them. The quantities T_1, T_2, and τ_s are so small that the irreversible relaxation in Eq. (2) during the time of the echo formation can be neglected. For the circular polarization we have $\mathbf{l}_n \cdot \mathbf{l}_n^* = 1$ at $n = 1$ and 2.

We separate in the sought quantities are rapidly oscillating phase factors

$$\mathbf{A} = \mathbf{a} \exp[i(kz - \omega t + \Phi)] + \text{c.c.},$$
$$I_1^\alpha = j_1^\alpha \exp[-i(kz - \omega t + \Phi)],\ I_2^\alpha = j_2^\alpha \exp[-i(kz - \omega t + \Phi)],$$

where the amplitudes $\mathbf{a}$, $\mathbf{j}_1$ and $\mathbf{j}_2$ are slow functions of the time, and Φ is a possible constant phase shift. The equations for the slow functions then take, according to (1) and (2), the form

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right) a_\alpha^* = -i\frac{2\pi c}{\omega}\int dv\, \mathrm{Sp}(j_1^\alpha + j_2^\alpha), \tag{3}$$

$$i\hbar\frac{\partial}{\partial t} j_1^\alpha = \hbar\Delta\omega_1 j_1^\alpha + \frac{1}{c} a_\gamma^* (N_1^{\gamma\alpha} - P_1^{\gamma\alpha}), \tag{4}$$

$$i\hbar\frac{\partial}{\partial t} N_1^{\alpha\beta} = \frac{1}{c} a_\gamma [(j_1^\gamma + j_2^\gamma)\Pi_1^{\alpha\beta} + \Pi_1^{\alpha\gamma} j_1^\beta]$$
$$- \frac{1}{c} a_\gamma^* [(j_1^{\gamma+} + j_2^{\gamma+})\Pi_1^{\alpha\beta} + j_1^{\alpha+}\Pi_1^{\gamma\beta}], \tag{5}$$

$$i\hbar\frac{\partial}{\partial t} P_1^{\alpha\beta} = \hbar\Delta\omega P_1^{\alpha\beta} - \frac{1}{c} a_\gamma \Pi_2^{\alpha\gamma} j_1^\beta + \frac{1}{c} a_\gamma^* j_2^{\alpha+}\Pi_1^{\gamma\beta}, \tag{6}$$

$$i\hbar\frac{\partial}{\partial t} j_2^\alpha = \hbar\Delta\omega_2 j_2^\alpha + \frac{1}{c} a_\gamma^* (N_2^{\gamma\alpha} - P_2^{\gamma\alpha}), \tag{7}$$

$$i\hbar\frac{\partial}{\partial t} N_2^{\alpha\beta} = \frac{1}{c} a_\gamma [(j_1^\gamma + j_2^\gamma)\Pi_2^{\alpha\beta} + \Pi_2^{\alpha\gamma} j_2^\beta]$$
$$- \frac{1}{c} a_\gamma^* [(j_1^{\gamma+} + j_2^{\gamma+})\Pi_2^{\alpha\beta} + j_2^{\alpha+}\Pi_2^{\gamma\beta}], \tag{8}$$

$$i\hbar\frac{\partial}{\partial t} P_2^{\alpha\beta} = -\hbar\Delta\omega P_2^{\alpha\beta} - \frac{1}{c} a_\gamma \Pi_1^{\alpha\gamma} j_2^\beta + \frac{1}{c} a_\gamma^* j_1^{\alpha+}\Pi_2^{\gamma\beta} \tag{9}$$

Here v is the projection of the thermal velocity on the direction of the propagation of the exciting pulse, and the other symbols are defined as

$$\Delta\omega_1 = \omega - \omega_1 - kv,\ \Delta\omega_2 = \omega - \omega_2 - kv,\ \Delta\omega = \omega_2 - \omega_1,$$
$$\hbar\omega_1 = E_1 - E_0,\ \hbar\omega_2 = E_2 - E_0.$$

For optically allowed atomic transitions, we obtain:
a) at $J_1 = J \to J_0 = J$

$$\frac{\Pi_1^{\alpha\beta}}{\omega_1^2 |d_J^J|^2} = \frac{\hat{J}_\alpha \hat{J}_\beta}{J(J+1)(2J+1)\hbar^2},$$

b) at $J_1 = J + 1 \to J_0 = J$

$$\frac{\Pi_1^{\alpha\beta}}{\omega_1^2 |d_J^{J+1}|^2} = \frac{-i(2J+3)\hbar\varepsilon_{\alpha\beta\gamma}\hat{J}_\gamma + 2(J+1)^2\hbar^2\delta_{\alpha\beta} - (\hat{J}_\alpha\hat{J}_\beta + \hat{J}_\beta\hat{J}_\alpha)}{2(J+1)(2J+1)(2J+3)\hbar^2}$$

c) at $J_1 = J \to J_0 = J + 1$

$$\frac{\Pi_1^{\alpha\beta}}{\omega_1^2 |d^J_{J+1}|^2} = \frac{i(2J+1)\hbar\varepsilon_{\alpha\beta\gamma}\hat{J}_\gamma + 2(J+1)^2\hbar^2\delta_{\alpha\beta} - (\hat{J}_\alpha\hat{J}_\beta + \hat{J}_\beta\hat{J}_\alpha)}{2(J+1)(2J+1)(2J+3)\hbar^2}.$$

In these formulas, $\hat{J}_\alpha$ is the operator of the total angular momentum of the lower level, $d_{J_0}^{J_1}$ is the reduced dipole moment of the $J_1 \to J_0$ transition, and $\epsilon_{\alpha\beta\gamma}$ is a fully antisymmetrical tensor. The quantity $\Pi_2^{\alpha\beta}$ is analogously defined, with the substitution $J_1 \to J_2$.

Prior to the passage of the exciting pulses we have

$$j_1^\alpha = j_2^\alpha = 0,\quad P_1^{\alpha\beta} = P_2^{\alpha\beta} = 0,\quad N_1^{\alpha\beta} = N_0 f(v)\Pi_1^{\alpha\beta},$$
$$N_2^{\alpha\beta} = N_0 f(v)\Pi_2^{\alpha\beta},$$

$$N_0 = \frac{n_0}{2J_0+1} - \frac{n_1}{2J_1+1} \approx \frac{n_0}{2J_0+1} - \frac{n_2}{2J_2+1},$$
$$f(v) = \frac{1}{\pi^{1/2}u}\exp\left(-\frac{v^2}{u^2}\right),$$

where n_0, n_1, and n_2 are the densities of the atoms on the lower and two upper levels at the initial instant of time, $f(v)$ is the Maxwellian distribution function, and u is the average thermal velocity of the atoms.

When solving Eqs. (3)–(9), we shall neglect the reaction of the emitted atoms on the passing light pulses, a procedure permissible if the following inequalities are satisfied[16,17]:

$$\omega |d_{J_0}^{J_1}|^2 L N_0 T_n / \hbar c \ll 1 \quad \text{if} \quad 1/ku \gg T_n,$$
$$|d_{J_0}^{J_1}|^2 L N_0 / \hbar u \ll 1 \quad \text{if} \quad 1/ku \ll T_n,$$

where $n = 1$ and 2, while L is length of the gas medium. Then the complex conjugate amplitude $\mathbf{a}_e^*$ of the photon echo is expressed in terms of the polarization current in simple fashion:

$$\mathbf{a}_e^* = -i2\pi\omega^{-1} L \int dv\, \mathrm{Sp}(\mathbf{j}_1 + \mathbf{j}_2),$$

the quantities $\mathbf{j}_1$ and $\mathbf{j}_2$ are determined from Eqs. (4)–(9), in which the amplitude a is constant and equal to $\mathbf{l}_1 a_1$ within the time of passage of the first exciting pulse, and to $\mathbf{a} = \mathbf{l}_2 a_2$ during the passage of the second pulse. In the time intervals between the exciting pulses and after these pulses we put $\mathbf{a} = 0$ in (4)–(9).

Eqs. (4)–(9) are necessary for the investigation of the polarization effects. In the simplest case, when the polarization vectors of the exciting pulses coincide, $\mathbf{l}_1 = \mathbf{l}_2 = \mathbf{l}$, the photon echo is produced with the same polarization, and greatest interest attaches to its intensity. At $\mathbf{l}_1 = \mathbf{l}_2 = \mathbf{l}$, the initial equations (4)–(9) become greatly simplified if the quantization axis is chosen along the vector $\mathbf{l}$ in the case of linear polarization and along the vector $\mathbf{k}$ for circular polarization. Then the basic matrix becomes diagonal

$$(l_\alpha^* \Pi_1^{\alpha\beta} l_\beta)_{\nu\nu'} = \omega_1^2 \Pi_{1\nu}\delta_{\nu\nu'}$$

with the following diagonal elements:
for linear polarizations and $J_1 = J \to J_0 = J$

$$\Pi_{1\nu} = \frac{\nu^2}{J(J+1)(2J+1)} |d_J^J|^2,$$

for linear polarizations and $J_1 = J + 1 \to J_0 = J$ or $J_1 = J \to J_0 = J + 1$

$$\Pi_{1\nu} = \frac{(J+1)^2 - \nu^2}{(J+1)(2J+1)(2J+3)} |d_J^{J+1}|^2,$$

for circular polarization and $J_1 = J \to J_0 = J$

$$\Pi_{1\nu}=\frac{(J+\nu)(J-\nu+1)}{2J(J+1)(2J+1)}|d_J{}^J|^2,$$

for circular polarization and $J_1 = J \to J_0 = J+1$

$$\Pi_{1\nu}=\frac{(J+\nu)(J+\nu+1)}{2(J+1)(2J+1)(2J+3)}|d_J^{J+1}|^2,$$

for circular polarization and $J_1 = J+1 \to J_0 = J$

$$\Pi_{1\nu}=\frac{(J-\nu+1)(J-\nu+2)}{2(J+1)(2J+1)(2J+3)}|d_J^{J+1}|^2.$$

Contributions to the photon echo are made only by diagonal elements of all the matrices of the equations, which we write in the form

$$(j_1 l)_{\nu\nu}=j_{1\nu},\quad (l_\alpha{}^* N_1{}^{\alpha\beta} l_\beta)_{\nu\nu}=\omega_1{}^2 N_{1\nu},$$
$$(l_\alpha{}^* P_1{}^{\alpha\beta} l_\beta)_{\nu\nu}=\omega_1\omega_2 P_\nu.$$

This reduces the problem of the photon echo in a degenerate three-level system to a simultaneous solution of the equations

$$i\hbar\frac{\partial}{\partial t}j_{1\nu}=\hbar\Delta\omega_1 j_{1\nu}+\frac{a\omega_1{}^2}{c}\left(N_{1\nu}-\frac{\omega_2}{\omega_1}P_\nu\right), \tag{10}$$

$$i\hbar\frac{\partial}{\partial t}j_{2\nu}=\hbar\Delta\omega_2 j_{2\nu}+\frac{a\omega_2{}^2}{c}\left(N_{2\nu}-\frac{\omega_1}{\omega_2}P_\nu{}^*\right), \tag{11}$$

$$i\hbar\frac{\partial}{\partial t}P_\nu=\hbar\Delta\omega P_\nu+\frac{a}{c}\left(\frac{\omega_2}{\omega_1}j_{1\nu}{}^*\Pi_{2\nu}-\frac{\omega_1}{\omega_2}j_{2\nu}\Pi_{1\nu}\right), \tag{12}$$

$$i\hbar\frac{\partial}{\partial t}N_{1\nu}=\frac{a}{c}\Pi_{1\nu}[2(j_{1\nu}-j_{1\nu}{}^*)+j_{2\nu}-j_{2\nu}{}^*], \tag{13}$$

$$i\hbar\frac{\partial}{\partial t}N_{2\nu}=\frac{a}{c}\Pi_{2\nu}[2(j_{2\nu}-j_{2\nu}{}^*)+j_{1\nu}-j_{1\nu}{}^*], \tag{14}$$

where $a = \mathbf{a}\cdot\mathbf{l}^* = \mathbf{a}^*\cdot\mathbf{l}$. In (12)–(14) there is no summation over the repeated symbol, since here and hereafter it is neither a vector nor a matrix index.

2. PHOTON ECHO ON A NARROW SPECTRAL LINE

We say that a spectral line of free-atom emission is narrow relative to the energy spectrum of the n-th exciting pulse of duration T_n if it lies within the spectral energy distribution of this pulse:

$$1/T_0 \ll 1/T_n, \tag{15}$$

where $T_0 = 1/ku$ is the time of the reversible Doppler relaxation, and the index $n = 1, 2$ corresponds to two successively applied light pulses with carrier frequency ω between the limits $|\omega - \omega_1| \ll 1/T_n$ and $|\omega - \omega_2| \ll 1/T_n$. In this case the passing light pulses excite all the atoms within the Doppler width of the spectral line.

If the inequality (15) is satisfied, then the quantities $\Delta\omega_1$, $\Delta\omega_2$, and $\Delta\omega$ should be neglected in Eqs. (10)–(14) during the time of passage of the exciting pulses, and it is necessary to put $\omega_1 = = \omega_2 = \omega$ in all the coefficients. Taking the foregoing into account, we obtain from (10)–(14)

$$i\hbar\frac{\partial}{\partial t}j_\nu=\frac{a\omega^2}{c}N_\nu,\quad i\hbar\frac{\partial}{\partial t}N_\nu=\frac{2a}{c}(j_\nu-j_\nu{}^*)(\Pi_{1\nu}+\Pi_{2\nu}), \tag{16}$$

$$i\hbar\frac{\partial}{\partial t}q_\nu=\frac{a\omega^2}{c}p_\nu,\quad i\hbar\frac{\partial}{\partial t}p_\nu=\frac{a}{c}q_\nu(\Pi_{1\nu}+\Pi_{2\nu}), \tag{17}$$

where

$$j_\nu=j_{1\nu}+j_{2\nu},\quad N_\nu=N_{1\nu}+N_{2\nu}-P_\nu-P_\nu{}^*,$$
$$q_\nu=j_{1\nu}\Pi_{2\nu}-j_{2\nu}\Pi_{1\nu},\quad p_\nu=(N_{1\nu}-P_\nu)\Pi_{2\nu}-(N_{2\nu}-P_\nu{}^*)\Pi_{1\nu}.$$

As follows from (17), the quantity q_ν does not contribute to the photon echo. In the subsequent calculations we can therefore omit the terms proportional to q_ν in the expressions

$$j_{1\nu}=\frac{\Pi_{1\nu}j_\nu+q_\nu}{\Pi_{1\nu}+\Pi_{2\nu}},\quad j_{2\nu}=\frac{\Pi_{2\nu}j_\nu-q_\nu}{\Pi_{1\nu}+\Pi_{2\nu}},$$

where j_ν and q_ν are solutions of Eqs. (16) and (17). In the free-precession time intervals, the quantities $j_{1\nu}$ and $j_{2\nu}$ are determined from Eqs. (10) and (11) at $a = 0$, but with allowance for $\Delta\omega_1$ and $\Delta\omega_2$, inasmuch as $\tau_s > T_1$ and $\tau_s > T_2$.

Eqs. (16) are the same as for the case of two-level atoms. To solve them we shall use the method described above.[16] Omitting the intermediate manipulations, we present a final expression for the amplitude $\mathbf{a}_e$ of the light echo

$$\mathbf{a}_e=\mathbf{l}\frac{\pi}{2}LN_0\sum_{\nu=-J_0}^{J_0}\sin\chi_{1\nu}T_1(1-\cos\chi_{2\nu}T_2)\,b_\nu(t')$$
$$\times\exp\left[-\frac{(t'-\tau_s)^2}{4T_0{}^2}+i\omega(t'-\tau_s)\right], \tag{18}$$

$$b_\nu(t')=[\Pi_{1\nu}\exp(i\omega_1\tau_s)+\Pi_{2\nu}\exp(i\omega_2\tau_s)]$$
$$\times[\Pi_{1\nu}\exp(-i\omega_1 t')+\Pi_{2\nu}\exp(-i\omega_2 t')](\Pi_{1\nu}+\Pi_{2\nu})^{-3/2},$$
$$t'=t-\tau_s-T_1-T_2-z/c,\quad \chi_{n\nu}=2\omega a_n(\Pi_{1\nu}+\Pi_{2\nu})^{1/2}/\hbar c,\quad n=1,2. \tag{19}$$

The inessential phase factor $\exp[i(\Phi_1 - 2\Phi_2)]$ is omitted throughout from the photon-echo amplitude.

When the levels E_1 and E_2 are significantly separated, $2\pi/T_0 < \Delta\omega \ll 1/T_n$, the echo intensity (8) undergoes modulation oscillations with frequency $\Delta\omega$, which were predicted in [12] from qualitative considerations. For very close levels $\Delta\omega T_0 < 2\pi$ the modulation oscillations of the individual echoes drop out and interest attaches to the value of the amplitude (18) at the instant $t' = \tau_s$ when the maximum is reached, since it contains the characteristic factor

$$b_\nu(\tau_s)=(\Pi_{1\nu}{}^2+\Pi_{2\nu}{}^2+2\Pi_{1\nu}\Pi_{2\nu}\cos\Delta\omega\tau_s)/(\Pi_{1\nu}+\Pi_{2\nu})^{3/2}. \tag{20}$$

According to (20) the envelope passing through the echo-amplitude maxima produced at different values of τ_s oscillates at a frequency $\Delta\omega = (E_2 - E_1)/\hbar$, proportional to the energy difference between the close levels. In practice this makes it possible to identify levels that are masked by the Doppler contour.

We note that the formation of photon echo in a three-level system differs substantially from the analogous phenomenon in a two-component gas mixture of two-level atoms, in which the resonant atomic frequencies ω_1 and ω_2 are shifted by a small amount $|\omega_2 - \omega_1| \ll 1/T_n$. This takes place, for example, in the presence of isotopes. If the inequality (15) is satisfied, then the light echo is produced independently on each component of the gas mixture. As a result, there appear immediately two echo signals with frequencies ω_1 and ω_2, and the intensity of the summary echo undergoes modulation oscillations with frequency $|\omega_2 - \omega_1|$ if $2\pi/T_0 < |\omega_2 - \omega_1| \ll 1/T_n$ (see Refs. 4 and 10). However, the envelope passing through the maxima of the intensities of the summary echo, as a function of τ_s, does not oscillate under any condition. Yet in a three-level system one of the working levels is common to two resonant atomic transitions, and it is this which leads to the above-mentioned envelope oscillations (quantum beats).

3. CASE OF BROAD SPECTRAL LINE

We regard a spectral line of free-atom radiation as broad if the frequency distribution of the energy of the n-th exciting pulse of duration T_n lies within the Doppler width

$$1/T_0 > 1/T_n, \tag{21}$$

where the subscript $n = 1, 2$ numbers two successive light pulses whose carrier frequency ω lies between ω_1 and ω_2, or is close to these frequencies ($1/T_0 \gtrsim \Delta\omega$). If the condition (21) is satisfied, only part of the atoms inside the Doppler width of the spectral line is excited. Since usually $\chi_{n\nu}T_n = m\pi$, where m is a number of the order of unity, the inequality (21) means that the amplitude of the exciting pulses should be low enough. This corresponds to a low-intensity regime of laser radiation. To the contrary, the inequality (15) corresponds to a high-intensity regime.

We consider first the case

$$1/T_1 \gg \Delta\omega, \quad 1/T_2 \gg \Delta\omega, \tag{22}$$

when the energy differency $\hbar\Delta\omega$ can be neglected during the time of passage of the exciting pulses. Then Eqs. (10)–(14) can be solved in analogy with (16) and (17). The amplitude $\mathbf{a}_e$ of the photon echo takes the form

$$\mathbf{a}_e = \mathbf{l}\frac{\pi^{1/2}}{2}LN_0T_0\int_{-\infty}^{\infty} d\eta \exp[-(\eta T_0)^2]\sum_{\nu=-J_0}^{J_0}\frac{\chi_{1\nu}\chi_{2\nu}^2}{\Omega_{1\nu}\Omega_{2\nu}^2}(1-\cos\Omega_{2\nu}T_2)$$
$$\times\left[\sin\Omega_{1\nu}T_1 + i\frac{\eta}{\Omega_{1\nu}}(1-\cos\Omega_{1\nu}T_1)\right] b_\nu(t')\exp[i(\omega-\eta)(t'-\tau_s)], \tag{23}$$
$$\Omega_{n\nu} = (\eta^2+\chi_{n\nu}^2)^{1/2}, \quad n = 1.\ 2,$$

where $\eta = kv$ and the function $b_\nu\ (t')$ is given by (19).

The echo-signal duration is in this case of the same order as the durations of the exciting pulses. The maximum of the amplitude (23) is shifted slightly relative to the instant $t' = \tau_s$, and, depending on the values of $x_{1\nu}T_1$ and $x_{2\nu}T_2$, the amplitude can have several maxima. If the amplitude (23) is measured at the instant $t' = \tau_s$, then it experiences, as a function of τ_s, oscillations of the same form as the preceding ones in (20).

A recent paper[15] was devoted to an investigation of a nondegenerate three-level system for the special case when the photon echo is produced as a result of successive applications of two ultrashort Stark pulses, which shift the levels in such a way that the two considered atomic transitions are at resonance for a short time with the monochromatic light wave of frequency ω. The reaction of the atoms is in this case the same as if they were irradiated with two ultrashort light pulses of frequency ω. It was suggested in[15] that the close working levels, owing to the Stark effect, combine into one doubly degenerate level, and therefore the quantum system behaves like a three-level system only in the absence of Stark pulses. Expression (23) is a generalization of[15] to the case of a degenerate three-level system. If the close levels E_1 and E_2 not only move apart during the action of the Stark pulses, but also come closer together, so that the following inequalities are satisfied for all the split Stark components E_1' and E_2'

$$|E_2'-E_1'|T_1/\hbar \ll 1, \quad |E_2'-E_1'|T_2/\hbar \ll 1, \tag{24}$$

then these relations replace the condition (22). In this case, in the absence of Stark pulses, the inverse inequalities $\Delta\omega T_1 > 2\pi$ and $\Delta\omega T_2 > 2\pi$ may be satisfied. The intensity of the echo (23) then undergoes modulation oscillations with frequency $\Delta\omega$.

If the inequalities

$$\Delta\omega \gtrsim 1/T_1, \quad \Delta\omega \gtrsim 1/T_2 \tag{25}$$

are satisfied during the course of the entire experiment, then the quantity $\Delta\omega$ cannot be neglected in (10)–(14), and the intensity of the photon echo in the region (21) can be obtained only by numerical methods. It undergoes modulation oscillations with frequency $\Delta\omega$.[12]

By way of example, we consider a three-level system with $J_0 = J_1 = \frac{1}{2}$ and $J_2 = \frac{2}{3}$, produced as a result of hyperfine interaction of the nuclear spin $S = \frac{1}{2}$ with electrons from two atomic shells, each characterized by a total angular momentum 1 and 0. The reduced dipole moments of the considered atomic transitions are connected by the relations

$$d_{1/2}^{1/2} = -\left(\frac{2}{3}\right)^{1/2} d_0^1, \quad d_{1/2}^{3/2} = \frac{2}{3^{1/2}} d_0^1, \tag{26}$$

where d_0^1 is the reduced dipole moment of the atomic transition following a change $1 \to 0$ in the total angular momentum of the electron system, without allowance for the nuclear spin. We denote the frequency of this atomic transition by ω_0. The results of the calculation at $\Delta\omega T_1 > 2\pi$ and $\Delta\omega T_2 > 2\pi$ are shown in Figs. 1 and 2. The units on the abscissa and ordinate axes corresponds to

$$\tau_0 = 3\hbar c/a_1\omega_0|d_0^1|, \quad I_0 = 8\pi(\omega_0 LN_0|d_0^1|Q)^2/9c,$$

and the zero of the abscissa axis corresponds to the instant of time $t_0' = \tau_s$. In addition, we have introduced the dimensionless parameters

$$\delta = \tau_0\Delta\omega, \quad \beta = \tau_0(\omega-\omega_2), \quad Q = T_0/\tau_0.$$

As seen from Figs. 1 and 2, the intensity of the photon echo averaged over the period $2\pi/\omega$, oscillates slowly with time. The time interval between neighboring minima (maxima) on the averaged-intensity curve (i.e., the period of these oscillations) is $2\pi/\Delta\omega$. The experimental observation of the oscillations of the average intensity of an individual photon echo makes it possible to determine the hyperfine splitting of the atomic levels.

In the case of a narrow spectral line, the amplitude

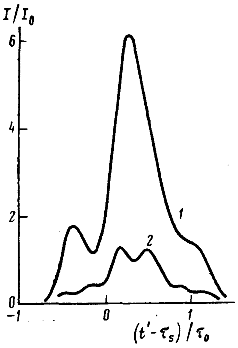

FIG. 1. Photon-echo intensity averaged over the period $2\pi/\omega$ of the fast oscillations. It is assumed that $\tau_s = 5\tau_0$, $Q = 0.1$, $\omega = (\omega_1 + \omega_2)/2$, $a_1 = a_2$, $2T_1 = T_2 = 1.2\tau_0$, and $2\chi_1 T_1 = \chi_2 T_2 = 4.1$. Curves 1 and 2 correspond to $\delta = 10$ and $\beta = -5$ and to $\delta = 20$ and $\beta = -10$, respectively. The second curve lies lower than the first because a smaller faction of the atoms are excited within the Doppler width in the second case than in the first.

of the photon echo (18) for the given system with $J_0 = J_1 = \frac{1}{2}$ and $J_2 = \frac{3}{2}$ at the instant of time $t' = \tau_s$, with allowance for (20) and (26), takes the form

$$\mathbf{a}_e = \mathbf{l}\frac{\pi}{3^{5/2}} LN_0 |d_0{}^1| \sin \chi_1 T_1 (1 - \cos \chi_2 T_2)(5 + 4\cos\Delta\omega\tau_s),$$
$$\chi_n = 2|d_0{}^1|\omega_0 a_n / 3^{1/2}\hbar c, \quad n = 1, 2. \tag{27}$$

The amplitude (27) is maximal when the first exciting pulse is a 90° pulse, $\chi_1 T_1 = \pi/2$, and the second is a 180° pulse, $\chi_2 T_2 = \pi$. If the spectral line is broad, then the parameters $\chi_1 T_1$ and $\chi_2 T_2$ influence the photon echo differently. At $2\chi_1 T_1 = \chi_2 T_2 = 4.1$ there is one broad maximum (Fig. 1), and at $\chi_1 T_1 = \chi_2 T_2 = 4{,}1$ the averaged echo intensity has two broad maxima, on which oscillations due to the proximity of the two upper levels of energies E_1 and E_2 are superimposed (Fig. 2). With increasing parameters $X_1 T_1$ and $X_2 T_2$, additional broad maxima appear and the picture becomes more complicated.

In the case (25), the results are another possibility of determining the hyperfine splitting from the oscillations of the average intensity I of the photon echo as a function of τ_s:

$$I = I_1 \cos(\delta\tau_s/\tau_0 + \varphi_1) + I_2 \cos(2\delta\tau_s/\tau_0 + \varphi_2) + I_3. \tag{28}$$

It is assumed here that the intensities of all the produced echo signals are measured after the lapse of one and the same time interval $\Delta t = t' - \tau_s$ following the characteristic instant of time $t'_0 = \tau_s$. The coefficients $X_1 T_1 = X_2 T_2 = 4{,}1$ and φ_2 in (28) do not depend on τ_s and are calculated with a computer for each particular case. For the parameter values $\delta = 10$, $\beta = -5$, $Q = 0{,}1$ and $T_1 = T_2 = 1.2\tau_0$ at $\Delta t = \tau^0$, these coefficients are $I_1 = 0.56$, $I_2 = 0.03$, $I_3 = 1.37$, $\varphi_1 = 0.76$ and $\varphi_2 = 1.57$.

If the condition $1/T_0 > 1/T_n > \Delta\omega > 2\pi/\tau_s$, where $n = 1, 2$, is satisfied, then it is necessary to observe the oscillation of the envelope passing through the photon-echo intensity maxima produced at different values of τ_s. This oscillation is also described by formula (28), in which a fixed time interval Δt corresponds to the maximum intensity of the photon echo.

We emphasize that in a two component gas mixture of two-level atoms with slightly shifted resonant atomic frequencies $\Delta\omega = |\omega_2 - \omega_1| \lesssim 1/T_0$, the intensity of photon echo on a broad spectral line, averaged over the period $2\pi/\omega$, is not subject to any oscillations, in contrast to the three-level systems considered above. There is likewise no oscillation of the envelope passing through the echo-intensity maxima produced at different values of τ_s.

4. ECHO POLARIZATION IN A THREE-LEVEL SYSTEM

Let the polarization vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ of linearly polarized exciting pulses make an angle ψ. In three-level systems with $J_0 = 1$ and $J_1 = J_2 + 1$, and also with $J_0 + J_1 = 1$ and $J_2 = 0$ or $J_0 = J_2 = 1$ and $J_1 = 0$ the intensity of the light echo is proportional to $\cos^2\psi$, and its polarization is the same as in the second exciting pulse, in full analogy with the two-level systems with atomic transitions $1 \rightleftharpoons 0$ and $1 \to 1$.

We have considered also a three-level system with $J_0 = j_1 = \frac{1}{2}$ and $J_2 = \frac{3}{2}$, in which the close levels are produced in arbitrary manner. In contrast to the preceding case, all the matrices of Eqs. (3)–(9) are expressed in terms of Pauli matrices, and this greatly simplifies the calculations. We write down the amplitude of the photon echo on a narrow spectral line

$$\mathbf{a}_e = \pi LN_0 \sin\chi_1 T_1 (1 - \cos\chi_2 T_2)\,\mathbf{e}(t') \times \exp[-(t'-\tau_s)^2/4T_0{}^2 + i\omega(t'-\tau_s)],$$
$$e_x(t') = \cos\psi[\Pi_1 \exp(i\omega_1\tau_s) + \Pi_2 \exp(i\omega_2\tau_s)] \times [\Pi_1 \exp(-i\omega_1 t') + \Pi_2 \exp(-i\omega_2 t')](\Pi_1 + \Pi_2)^{-3/2},$$
$$e_y(t') = \sin\psi[\Pi_1 \exp(i\omega_1\tau_s) - {}^1/{}_2\Pi_2 \exp(i\omega_2\tau_s)] \times [\Pi_1 \exp(-i\omega_1 t') - {}^1/{}_2\Pi_2 \exp(-i\omega_2 t')](\Pi_1 + \Pi_2)^{-3/2},$$
$$\chi_{1,2} = 2\omega a_{1,2}(\Pi_1 + \Pi_2)^{1/2}/\hbar c, \quad \Pi_1 = |d_{1/2}{}^{1/2}|^2/6, \quad \Pi_2 = |d_{1/2}{}^{3/2}|^2/6.$$

The X axis is chosen here along the polarization vector $\mathbf{l}_2$ of the second exciting pulse, and the Z axis is directed towards the wave propagation. The photon echo (29) is elliptically polarized. At $2\pi/T_0 < \Delta\omega \ll 1/T_n$ the axis of the polarization of this oscillates with time at a frequency $\Delta\omega/2$. For very close levels $\Delta\omega T_0 \ll 1$, the polarization ellipse shrinks and lengthens into a line segment, and the photon echo becomes linearly polarized.

At the instant $t' = \tau_s$ of the maximum of the amplitude (29), the photon echo is linearly polarized, and its polarization vector lies outside the angle ψ between the polarization vectors of the exciting pulses. Designating by φ the angle of inclination of the echo-polarization

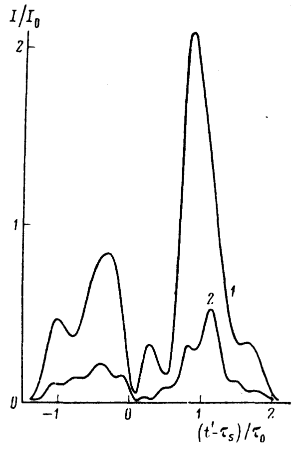

FIG. 2. The notation and the numerical values of the parameters are the same as in Fig. 1, with the exceptions $T_1 = T_2 = 1.2\tau_0$ and $\chi_1 T_1 = \chi_2 T_2 = 4.1$.

to the X axis, we have

$$\operatorname{tg}\varphi=\frac{\Pi_1^2+\Pi_2^2/4-\Pi_1\Pi_2\cos\Delta\omega\tau_s}{\Pi_1^2+\Pi_2^2+2\Pi_1\Pi_2\cos\Delta\omega\tau_s}\operatorname{tg}\psi. \quad (30)$$

As a function of τ_s, the plane of polarization of the echo executes quantum beats analagous to the quantum beats of the amplitude (20). By measuring the oscillations of the plane of polarization (30) and of the amplitude (20) we can determine not only the energy difference, $E_2 - E_1$, but also the dipole moments of the atomic transitions.

In the case of the broad spectral line in the regions (22) and (24) the polarization plane of the echo at $t' = \tau_s$ oscillates as a function of τ_s and the oscillations are likewise described by formula (30). In the region of (25), the obtained effect can be calculated by numerical methods.

The polarization properties of the light echo in a three-level system with $J_0=\frac{3}{2}$ and $J_1=J_2=\frac{1}{2}$ are the same as in a two-level system with atomic transition $\frac{1}{2}\to\frac{3}{2}$. Putting $\pi_1=0$ or $\pi_2=0$ in (29) we obtain the amplitude of the photon echo for two-level systems with atomic transitions $\frac{3}{2}\to\frac{1}{2}$ or $\frac{1}{2}\to\frac{1}{2}$, respectively.

If the close levels E_1 and E_2 are formed as a result of hyperfine interaction, then according to (26) the ratio (30) takes the simple form

$$\operatorname{tg}\varphi=2(1-\cos\Delta\omega\tau_s)\operatorname{tg}\psi/(5+4\cos\Delta\omega\tau_s),$$

from which it follows that the amplitude of the oscillations of the plane of polarization of the echo is large enough for experimental observation of the obtained effect.

Radiative decay and atomic collisions cause the amplitude of the photon echo to attenuate with time t, mainly in accord with the exponential law $\propto\exp(-t/2t_r)$, where t_r is the time of irreversible relaxation. If $\tau_s \sim t_r$, then the echo-amplitude ascillations determined above will be superimposed on this damped component. This amplitude damping, however, does not affect the oscillations (30) of the echo-polarization plane.

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Polarization phenomena in an oriented helium plasma

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The collisions of metastable oriented $He(2^3S)$ atoms, accompanied by elastic scattering and by Penning ionization, is analyzed. The amplitude collision matrices are obtained. The cross sections and polarizations of all the particles are calculated, and schemes are proposed for a complete experiment, i.e., for a set of experiments that determine the amplitudes that characterize the scattering process. The obtained formulas are used for numerical estimates of the effects of polarization in the Penning ionization process.

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Spin-polarization phenomena in electron and atom collisions have been experimentally investigated in recent years. The reasons for the noticeable interest in polarization phenomena are in all cases of interference origin, and therefore provide a subtle and most sensitive means of investigating the structure and properties of matter and of analyzing physico-chemical processes. Even the very first experiments with polarized electrons[1-3] have confirmed that investigations of polarization effects are promising. It has immediately become clear that polarization phenomena constitute a new means of collision spectroscopy of atoms and molecules,

