

Quantum beats of the coherent radiation emitted by atoms in a magnetic field

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It is shown that at a definite choice of the polarizations of exciting ultrashort light pulses and of the sequence of excitation of two adjacent atomic transitions the intensity of the coherent emission in a constant magnetic field \mathbf{H} depends on the precession of the magnetic moments of the excited atoms and undergoes harmonic quantum beats having a frequency that contains g factors of the resonant levels. A physical interpretation is offered for this phenomenon and it is established that, out of all modifications of coherent echo signals, only the three-level photon echo makes it possible to determine experimentally from the quantum beats the g factors of the levels of the one- and two-quantum atomic transitions. In a sufficiently strong field \mathbf{H} , Zeeman splitting leads to the appearance of a unique echo-signal structure that makes it possible to calculate the angular momenta and the difference of the g factors of the resonant levels.

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Theoretical^{1,2} and experimental³ investigations of coherent emission of two-level systems in a magnetic field \mathbf{H} have shown that following a two-pulse excitation of the gas the polarization of the produced photon echo is rotated through a definite angle that depends on \mathbf{H} , on the g factors of the levels, on the delay time between the two exciting pulses, and on the type of the resonant atomic transition. This rotation of the polarization differs considerably from the Faraday rotation. The specific rotation of the photon-echo polarization is attributed in Refs. 1–3 to precession of the induced atomic polarization in the field \mathbf{H} , and does not depend on the precession of the magnetic moments of the excited atoms.

We analyze in this paper the conditions under which coherent radiation in a field \mathbf{H} depends substantially also on the precession of the magnetic moments of the excited atoms. To this end we investigate the emission from the atoms in the form of a three-level photon echo (TPE) produced after a gas is irradiated by two ultrashort light pulses that are resonant to two adjacent optically allowed transitions. Under the influence of a constant longitudinal magnetic field \mathbf{H} , the polarization of the TPE is rotated and the intensity of the echo signal is changed. If one considers the intensity of the coherent emission of the atoms at the instant when the TPE is a maximum, this intensity undergoes oscillations (quantum beats) as a function of \mathbf{H} and of the delay time τ_{12} and τ_{13} between the exciting pulses; these beats are due to transitions of the atom between Zeeman sublevels. By virtue of the selection rules for photon emission and absorption, the quantum beats depend on the polarization of the exciting light pulses and on the sequence of the excitation of the adjacent optically allowed transitions. This makes possible the choice of conditions under which the quantum beats take the simplest form. If the first exciting pulse is linearly polarized in the direction of the vector \mathbf{l} while the second and third are right- or left-circularly polarized, the radiation intensity, time-averaged over the period of three light oscillations and corresponding to the projection of the electric field \mathbf{E} of the echo signal on the direction of \mathbf{l} , undergoes quantum beats when τ_{12} or τ_{23} or \mathbf{H} is changed. These beats are de-

scribed by a harmonic function. Depending on the form of the TPE, the frequency of the harmonic quantum beats is proportional to one g factor when the phenomenon depends on the precession of the magnetic moment of the excited atoms, or to the sum of two g factors if a contribution is made by the precession of the induced atomic polarization.

The obtained regularities of the quantum beats serve as a basis for the determination of the g factors of all three resonant levels, one of which belongs to an optically forbidden atomic transition and is not populated under ordinary conditions. The latter is of fundamental significance, since it makes it possible to study in equally simple fashion the g factors of individual levels belonging both to optically allowed and to forbidden (two-photon) atomic transitions. We note in this connection that in the known methods of studying the Zeeman effect, which are based on the use of double radio-optical resonance,⁴ of level crossing,⁵ of the Hanle effect,⁶ and of narrow resonances,⁷ one determines the g factors of only those excited levels that are connected with the fundamental optically allowed transition. We emphasize that photon echos are produced in two-level systems^{1–3} also in optically allowed transitions and do not make it possible to determine separately the g factors of each resonant level, except for the simplest transitions with total-angular-momentum change $1 \leftrightarrow 0$. Allowance for the influence of the precession of the magnetic moments of the excited atoms is thus not only of general physical but also of practical interest, since it leads to essentially new regularities that uncover additional capabilities of nonlinear spectroscopy. One of them is the determination of the level g factors independent of their degeneracy multiplicity.

1. CALCULATION METHOD

The interaction of gas atoms with a short light pulse

$$\mathbf{E} = \frac{1}{2} \mathbf{l}^{(s)} a \exp [i(kz - \omega t - \Phi)] + \text{c.c.} \quad (1)$$

of frequency ω and with linear ($s = 0$) or circular ($s = \pm 1$) polarization

$$\mathbf{l}^{(0)} = \mathbf{l}_x, \quad \mathbf{l}^{(\sigma)} = (i^{\sigma-1} \mathbf{l}_x + i \mathbf{l}_y) / \sqrt{2}, \quad \sigma = \pm 1,$$

is described by the d'Alambert equation for the electric field

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E} = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \int \sum_{\mu m} (\rho_{\mu m} \mathbf{d}_{\mu m} + \rho_{m\mu} \mathbf{d}_{\mu m}') d\mathbf{v} \quad (2)$$

and by the equations for the components of the density matrix ρ in the presence of a longitudinal magnetic field \mathbf{H}

$$\begin{aligned} & \left[\frac{\partial}{\partial t} + v_z \frac{\partial}{\partial z} + i(\omega_{ba} + \Omega_b \mu - \Omega_a m) + \gamma_{ba} \right] \rho_{\mu m} \\ &= \frac{i\mathbf{E}}{\hbar} \left(\sum_{m'} \mathbf{d}_{\mu m'} \rho_{m'm} - \sum_{\mu'} \rho_{\mu\mu'} \mathbf{d}_{\mu'm} \right), \end{aligned} \quad (3)$$

$$\begin{aligned} & \left[\frac{\partial}{\partial t} + v_z \frac{\partial}{\partial z} + i\Omega_a(m - m') + \gamma_a \right] \rho_{mm'} \\ &= \frac{i\mathbf{E}}{\hbar} \sum_{\mu} (\mathbf{d}_{m\mu} \rho_{\mu m'} - \rho_{m\mu} \mathbf{d}_{\mu m'}) + \frac{\gamma_a N_a f(v)}{2j_a + 1} \delta_{mm'}, \end{aligned}$$

$$\Omega_a = g_a \mu_0 H / \hbar, \quad \Omega_b = g_b \mu_0 H / \hbar, \quad f(v) = (\pi u^2)^{-1/2} \exp(-v^2/u^2), \quad (4)$$

while the equation for $\rho_{\mu\mu'}$ is obtained from (4) by interchanging the indices $m \rightarrow \mu$, $m' \rightarrow \mu'$, $\mu \rightarrow m$ and $a \rightarrow b$. Here $\mathbf{d}_{\mu m}$ is the dipole-moment operator matrix, v_z is the projection of the atom velocity \mathbf{v} on the Z axis, $f(v)$ is the Maxwell distribution, u is the most probable velocity, μ_0 is the Bohr magneton, γ_{ba} , γ_a , and γ_b are the irreversible relaxation constants, N_a and N_b are the stationary densities of the atoms on the lower and upper levels at $\mathbf{E} = \mathbf{H} = 0$, \mathbf{l}_x and \mathbf{l}_y are unit vectors along the Cartesian axes X and Y , and $\omega_{ba} = (E_b - E_a)/\hbar$ is the frequency of the resonant atomic transition between states that are characterized, besides by the energies E_a and E_b , also by total angular momenta j_a and j_b and their projections m and μ on the quantization axis. The gyromagnetic factors g_a and g_b of the resonant levels are defined in a region in which the Zeeman splitting ΔE_H is small compared with the hyperfine-structure intervals ΔE_{hf} , and the g factor of each level is connected with the electronic g_j factor by the relation

$$g = g_j [j(j+1) + J(J+1) - I(I+1)] / 2j(j+1),$$

where i , J , and I are the quantum numbers of the total angular momentum, of the electron angular momentum of the atom, and the spin of the nucleus. When the nuclear spin is zero and $\Delta E_{hf} = 0$, the interaction with the field in (3) and (4) remains valid provided only that ΔE_H is small compared with the intervals ΔE_{fs} of the fine structure of the level. At $\Delta E_H \ll \Delta E_{fs}$ Eqs. (3) and (4), as well as the conclusions that follow, are valid also for the vibrational-rotational levels of the molecule.

We solve Eqs. (3) and (4) for optically thin media, when the reaction of the medium on the exciting pulse is small and the field \mathbf{E} in (3) and (4) can be regarded as a given function (1)

with $\omega = kc$ and with constant values of a and Φ in the time interval $t_0 \leq t - z/c \leq t_0 + \tau_p$ and with $a = 0$ outside this interval; here t_0 is the initial instant of entry of the pulse (1) into the gas medium at the point $z = 0$, and the duration τ_p of this pulse is short compared with the irreversible relaxation times, $\gamma_{ba} \tau_p \ll 1$, $\gamma_a \tau_p \ll 1$ and $\gamma_b \tau_p \ll 1$. We impose in addition on the Zeeman splitting the restriction

$$j_a \Omega_a \tau_p \ll 1, \quad j_b \Omega_b \tau_p \ll 1, \quad (5)$$

so as to neglect the field \mathbf{H} when the pulse (1) passes through an arbitrary point z of the gas in question. The solution of Eqs. (3) and (4) in the region $t_0 \leq t - z/c \leq t_0 + \tau_p$ takes then the form

$$\begin{aligned} \rho_{\mu m} &= r_{\mu m}(t - z/c) \exp[i(kz - \omega t - \Phi)], \\ \rho_{mm'} &= r_{mm'}(t - z/c), \quad \rho_{\mu\mu'} = r_{\mu\mu'}(t - z/c). \end{aligned} \quad (6)$$

The matrix $r(t)$ is connected with its value $r(t_0)$ at the initial instant of time by the relation

$$r(t) = S(t - t_0) r(t_0) S^+(t - t_0).$$

The evolution operator $S(t - t_0)$ takes here in the matrix representation the form

$$\begin{aligned} S_{mm'}(t - t_0) &= A_{m+s} \delta_{mm'}, \quad S_{\mu\mu'}(t - t_0) = A_{\mu}^* \delta_{\mu\mu'}, \\ S_{\mu m}(t - t_0) &= i(-1)^{j_b - m} \frac{d_{ba}}{|d_{ba}|} B_{\mu} \delta_{\mu, m+s}, \\ S_{m\mu}(t - t_0) &= -S_{\mu m}^*(t - t_0), \\ A_{\lambda} &= \cos \frac{\Omega_{\lambda}(t - t_0)}{2} + i \frac{kv_z - \Delta}{\Omega_{\lambda}} \sin \frac{\Omega_{\lambda}(t - t_0)}{2}, \\ B_{\lambda} &= \frac{\chi_{\lambda}}{\Omega_{\lambda}} \sin \frac{\Omega_{\lambda}(t - t_0)}{2}, \quad \Omega_{\lambda} = [(kv_z - \Delta)^2 + \chi_{\lambda}^2]^{1/2}, \\ \chi_{\lambda} &= \frac{a|d_{ba}|}{\hbar} \begin{pmatrix} j_b & j_a & 1 \\ \lambda & s - \lambda & -s \end{pmatrix}, \end{aligned}$$

where d_{ba} is the reduced dipole moment of the atomic transition $j_a \rightarrow j_b$ (Ref. 8), $\Delta = \omega - \omega_{ba}$ is the detuning from resonance, and the parameter s takes on values 0, 1, and -1 respectively for linear and right- and left-circular polarizations of the pulse (1). The quantization axes are X and Z for linear and circular polarization, respectively.

The components of the density matrix ρ obtained in this manner for the instant of time $t = t_0 + \tau_p + z/c$, with allowance for the delay z/c , are the initial conditions for the solution of Eqs. (3) and (4) in the region $t_0 + \tau_p \leq t - z/c$, when the pulse (1) has already passed through $a = 0$ and the electromagnetic field produced by the polarized resonant medium in the presence of \mathbf{H} is substantial. The solution is obtained with the aid of Eq. (2) and manifests itself in the form of free optical induction or photon echo if the gas medium is successively excited by several light pulses (1).

2. INFLUENCE OF THE MAGNETIC-MOMENT PRECESSION ON THE TPE

To investigate the g factors of the levels belonging to a two-photon transition, we use the most informative TPE, which appears in the gas after the passage of three light pulses with specially chosen polarizations:

$$E_1 = I_x a_1 \cos(\omega t - kz + \Phi_1), \quad (7)$$

$$E_2 = \frac{1}{2} I^{(\sigma)} a_2 \exp[i(\bar{k}z - \bar{\omega}t - \Phi_2)] + \text{c.c.}, \quad (8)$$

$$E_3 = \frac{1}{2} I^{(\sigma)} a_3 \exp[i(\bar{k}z - \bar{\omega}t - \Phi_3)] + \text{c.c.}, \quad (9)$$

where the amplitudes a_1 , a_2 , and a_3 of the electric fields and the phase shifts Φ_1 , Φ_2 , and Φ_3 are real constants. The durations τ_1 , τ_2 , and τ_3 of the pulses (7)–(9) are shorter than the time intervals τ_{12} and τ_{23} between them. The pulse (7) enters the gas at the point $z = 0$ at $t = 0$. The frequencies $\omega = kc$ and $\bar{\omega} = \bar{k}c$ are close to the frequencies $\omega_{ba} = (E_b - E_a)/\hbar$ and $\omega_{cb} = (E_c - E_b)/\hbar$ of two adjacent atomic transitions $j_a \rightarrow j_b$ and $j_b \rightarrow j_c$, where j_a , j_b , and j_c are the total angular momenta of the atom in states with energies E_a , E_b , and E_c ($E_a < E_b < E_c$), while the lower level E_a is in the ground or an excited state.

The interaction of the pulse (7) with three-level atoms in the presence of a longitudinal magnetic field \mathbf{H} is described by expressions (2)–(6) with $a = a_1$ and $\Phi = \Phi_1$, and for pulses (8) and (9) it is necessary in Eqs. (2)–(6) to redesignate the amplitudes and phases, to make the substitutions $\omega \rightarrow \bar{\omega}$ and $k \rightarrow \bar{k}$, and to change the indices $a \rightarrow b$, $b \rightarrow c$, $m \rightarrow \mu$ for all the quantities. Here ν is the projection of the total angular momentum of an atom in a state with energy E_c and total angular momentum j_c .

As a result, the matrix elements $r_{\nu\mu}(t)$ that contribute to the TPE at the frequency $\bar{\omega}$ in the region $t_3 + \tau_3 < t$ take the form

$$r_{\nu\mu}(t) = r_{\nu\mu}(t_3 + \tau_3) \exp\{[i(\Omega_b \mu - \Omega_c \nu + \bar{\Delta} - \bar{k}v_z) - \gamma_{cb}](t - t_3 - \tau_3)\}, \quad (10)$$

$$r_{\nu\mu}(t_3 + \tau_3) = \sum_{\mu'\nu'} S_{\nu\mu'}(\tau_3) r_{\mu'\nu'}(t_3) S_{\nu'\mu}^+(\tau_3), \quad (11)$$

$$r_{\mu\nu}(t_3) = r_{\mu\nu}(t_2 + \tau_2) \exp\{[i(\Omega_c \nu - \Omega_b \mu + \bar{k}v_z - \bar{\Delta}) - \gamma_{cb}]\tau_{23} + i(\Phi_2 - \Phi_3)\}, \quad (12)$$

$$r_{\mu\nu}(t_2 + \tau_2) = \sum_{\mu'\nu'} S_{\mu\nu'}(\tau_2) r_{\mu'\nu'}(t_2) S_{\nu'\mu}^+(\tau_2) + \sum_{\nu''\nu'} S_{\mu\nu''}(\tau_2) r_{\nu''\nu'}(t_2) S_{\nu'\nu}^+(\tau_2), \quad (13)$$

$$r_{\mu\mu'}(t_2) = r_{\mu\mu'}(\tau_1) \exp\{[i\Omega_b(\mu' - \mu) - \gamma_b]\tau_{12}\}$$

$$+ \frac{N_b f(v)}{2j_b + 1} \delta_{\mu\mu'} [1 - \exp(-\gamma_b \tau_{12})],$$

$$r_{\nu\nu'}(t_2) = \frac{N_c f(v)}{2j_c + 1} \delta_{\nu\nu'}, \quad (14)$$

$$r_{\mu\mu'}(\tau_1) = \sum_{\mu_1\mu_2} D_{\mu_1\mu_2}^{j_b+} \left(\frac{\pi}{2}, \frac{\pi}{2}, \pi \right) \times \left(\sum_{\mu_2\mu_3} S_{\mu_1\mu_2}(\tau_1) r_{\mu_2\mu_3}(0) S_{\mu_3\mu_4}^+(\tau_1) + \sum_{m m'} S_{\mu m}(\tau_1) r_{m m'}(0) S_{m' \mu}^+(\tau_1) \right) D_{\mu\mu'}^{j_b} \left(\frac{\pi}{2}, \frac{\pi}{2}, \pi \right), \quad (15)$$

$$r_{m m'}(0) = N_a f(v) \delta_{m m'} / (2j_a + 1), \quad r_{\mu\mu'}(0) = N_b f(v) \delta_{\mu\mu'} / (2j_b + 1),$$

$$r_{\mu\mu}(0) = r_{\nu\nu}(0) = 0,$$

where $t_1 = 0$, $t_2 = \tau_{12} + \tau_1$ and $t_3 = \tau_{12} + \tau_{23} + \tau_1 + \tau_2$ are the instants of time at which the pulses (7)–(9) enter the gas, N_c is the stationary density of the atoms on the level E_c in the absence of external fields, γ_{cb} is the damping constant of the optical-coherence matrix $\rho_{\mu\nu}$, and $D_{\mu\mu'}^{j_b}(\alpha, \beta, \gamma)$ is the Wigner function $D_{\mu\mu'}^{j_b}(\alpha, \beta, \gamma)$ at $\alpha = \beta = \pi/2$ and $\gamma = \pi$ (Ref. 8). In Eqs. (10)–(15) the quantization direction is along \mathbf{H} , and in (11) and (13) were retained only the terms essential for the formation of the TPE.

It can be seen from Eqs. (14) that in the time interval $\tau_1 \leq t \leq t_2$ the precession of the magnetic moments of the atoms in an excited state with energy E_b makes a large contribution to the phase, and this is reflected in final analysis in the polarization of the coherent radiation. Yet in the time intervals $t_2 + \tau_2 \leq t \leq t_3$ and $t_3 + \tau_3 \leq t$, according to (12) and (19), a significant role is played only by the precession of the induced atomic polarization; this precession is proportional to the density matrix elements that are not diagonal in the levels E_b and E_c . If $a_1 = 0$, we have in (13)

$$r_{\mu\mu'}(t_2) = N_b f(v) \delta_{\mu\mu'} / (2j_b + 1)$$

and Eq. (10) describes the formation of the usual two-pulse echo, to which only the precession of the induced atomic polarization contributes.

Using (19) with the substitution $t \rightarrow t - z/c$ we obtain the final expression for the electric field E_e of the considered TPE:

$$E_e = \varepsilon(t - z/c) \exp[i(\bar{k}z - \bar{\omega}t - \Phi_e)] + \text{c.c.}, \quad (16)$$

where $\Phi_e = 2\Phi_3 - \Phi_2$, and the amplitude $\varepsilon(t)$ takes the form

$$\varepsilon(t) = \varepsilon_0 \int d\nu f(v) \{ I^{(1)} W_1^{(\sigma)} \exp[i(\sigma - 1)(\Omega_b \tau_{12} + (\Omega_b + \Omega_c) \tau_{23})] + I^{(-1)} W_{-1}^{(\sigma)} \exp[i(\sigma + 1)(\Omega_b \tau_{12} + (\Omega_b + \Omega_c) \tau_{23})] \} \times \exp[i(\bar{\Delta} - \bar{k}v_z)(t - t_e) - \gamma_{cb}(t - \tau_{12})],$$

$$\varepsilon_0 = 2\pi \bar{k}L |d_{cb}| N_0, \quad N_0 = N_a / (2j_a + 1) - N_b / (2j_b + 1),$$

$$W_q^{(\sigma)} = \sum_x Q_x R_{xq}^{(\sigma)}, \quad q = \pm 1, \quad \sigma = \pm 1,$$

$$Q_x = \sum_{\mu} \frac{(-1)^{j_b - \mu}}{N_0} \begin{pmatrix} j_b & j_b & \kappa \\ \mu & -\mu & 0 \end{pmatrix} \left[\left(\frac{N_a (B_{1\mu}^{(0)})^2}{2j_a + 1} + \frac{N_b (|A_{1\mu}^{(0)}|^2 - 1)}{2j_b + 1} \right) e^{-\gamma_b \tau_{12}} + \frac{N_b}{2j_b + 1} - \frac{N_c}{2j_c + 1} \right]$$

$$R_{\nu q}^{(\sigma)} = (2\kappa + 1) \sum_{\nu} (-1)^{j_b - \mu} \begin{pmatrix} j_c & j_b & 1 \\ \nu & -\mu & -q \end{pmatrix} \times \begin{pmatrix} j_b & j_b & \kappa \\ \nu - \sigma & -\mu & \sigma - q \end{pmatrix} \bar{A}_{2\nu}^{(\sigma)} \bar{B}_{2, \mu + \sigma}^{(\sigma)}$$

$$\times \bar{B}_{3\nu}^{(\sigma)} \bar{B}_{3, \mu + \sigma}^{(\sigma)} d_{0, \sigma - q}^{\kappa} \left(\frac{\pi}{2} \right) \exp[i(\Omega_b \mu - \Omega_c \nu)(t - t_e)],$$

$$A_{n\nu}^{(\sigma)} = \cos \frac{\Omega_{n\nu}^{(\sigma)} \tau_n}{2} + i \frac{k\nu_z - \Delta}{\Omega_{n\nu}^{(\sigma)}} \sin \frac{\Omega_{n\nu}^{(\sigma)} \tau_n}{2},$$

$$B_{n\nu}^{(\sigma)} = \frac{\chi_{n\nu}^{(\sigma)}}{\Omega_{n\nu}^{(\sigma)}} \sin \frac{\Omega_{n\nu}^{(\sigma)} \tau_n}{2},$$

$$\Omega_{n\nu}^{(\sigma)} = [(k\nu_z - \Delta)^2 + \chi_{n\nu}^{(\sigma)2}]^{1/2}, \quad \chi_{n\nu}^{(\sigma)} = \frac{a_n |d_{ba}|}{\hbar} \begin{pmatrix} j_b & j_a & 1 \\ \nu & s - \nu & -s \end{pmatrix}$$

$$n = 1, 2, 3, \quad s = 0, \pm 1, \quad \Omega_c = g_c \mu_0 H / \hbar,$$

$$t_e = \tau_{12} + 2\tau_{23} + \tau_1 + \tau_2 + \tau_3, \quad \bar{\Delta} = \bar{\omega} - \omega_{cb}. \quad (17)$$

Here L is the length of the gas volume and $d_{qp}^{\kappa}(\beta)$ stands for the Wigner function $D_{qp}^{\kappa}(\alpha, \beta, \gamma)$ at $\alpha = \gamma = 0$ (Ref. 8). The quantities $\bar{A}_{n\nu}^{(\sigma)}$ and $\bar{B}_{n\nu}^{(\sigma)}$ are obtained from $A_{n\nu}^{(\sigma)}$ and $B_{n\nu}^{(\sigma)}$ by making the substitutions $\omega \rightarrow \bar{\omega}$, $k \rightarrow \bar{k}$, $\omega_{ba} \rightarrow \omega_{cb}$, $j_a \rightarrow j_b$, $j_b \rightarrow j_c$ and $d_{ba} \rightarrow d_{cb}$, where d_{cb} is the reduced dipole moment of the atomic transition $j_b \rightarrow j_c$. The indices $\sigma = 1$ and $\sigma = -1$ pertain to right- and left-circularly polarized exciting pulses, respectively.

The three-level photon echo (16) is produced at the instant of time $t = t_e$ at the frequency $\bar{\omega}$, and is in the general case elliptically polarized. The magnetic field rotates the axes of the TPE polarization ellipse about the Z axis clockwise through an angle $\Omega_b \tau_{12} + (\Omega_b + \Omega_c) \tau_{23}$ when viewed along \mathbf{H} . The physical causes of this rotation are the precessions, about the direction of the vector \mathbf{H} , of the magnetic moment of the excited atom and of the induced atomic polarization. The first precession yields the term $\Omega_b \tau_{12}$ and the second $(\Omega_b + \Omega_c) \tau_{23}$.

The radiation intensity $I^{(x)}$ corresponding to the x -projection of the electric field (16) is described, after averaging over the period $2\pi/\bar{\omega}$, by the formula $I^{(x)} = c|\mathcal{E}_x(t)|^2/2\pi$ at $t = t_0$ or

$$I^{(x)} = \{1 - b \cos [2\Omega_b \tau_{12} + 2(\Omega_b + \Omega_c) \tau_{23} + 2\alpha]\} I_0 \exp(-4\gamma_b \tau_{23}), \quad (18)$$

where the slow functions are taken at the instant of time $t = t_0$ of the maximum of the TPE, and we use the notation

$$I_0 = c\mathcal{E}_0^2 (|\bar{W}_1^{(\sigma)}|^2 + |\bar{W}_{-1}^{(\sigma)}|^2) / 4\pi, \quad (19)$$

$$b = 2|\bar{W}_1^{(\sigma)} \bar{W}_{-1}^{(\sigma)}| / (|\bar{W}_1^{(\sigma)}|^2 + |\bar{W}_{-1}^{(\sigma)}|^2), \quad (20)$$

$$\alpha = (\varphi_1^{(\sigma)} - \varphi_{-1}^{(\sigma)}) / 2, \quad (21)$$

where a superior bar denotes statistical averaging

$$\bar{W}_q^{(\sigma)} = \int d\nu f(\nu) W_q^{(\sigma)} = |\bar{W}_q^{(\sigma)}| \exp(-i\varphi_q^{(\sigma)}), \quad (22)$$

and Eq. (22) serves as the definition of the phase $\varphi_q^{(\sigma)}$ at $q = \pm 1$.

If the common level E_b lies above the other two, $E_a < E_c < E_b$, or below them, $E_b < E_a < E_c$, ($E_b < E_c < E_a$), the basic formulas (18)–(22) remain valid provided the substitutions $\omega_{ba} \rightarrow |\omega_{ba}|$ and $\omega_{cb} \rightarrow |\omega_{cb}|$ are made.

Since the quantum beats (18) are due to the specific rotation of the TPE polarization in the magnetic field, the intensity $I^{(y)}$ corresponding to the y -projection of the field (16) is described by Eq. (18) with a plus sign in front of the amplitude b , and the quantities in it are the same (19)–(21).

The quantum beats (18) can be used to determine the g factors of the levels E_b and E_c , inasmuch as under the usual conditions $N_b \ll N_a$ and $N_c \ll N_a$ the quantities (20) and (21) are independent of τ_{12} and τ_{23} , and $I_0 \sim \exp(-2\gamma_b \tau_{12})$. By investigating a set of echo signals at different values of τ_{12} and with the other parameters fixed, we easily obtain a plot of the oscillations (18), whose frequency is proportional to g_b if $0 < \Omega_b \tau_{12} \lesssim 2\pi$. The modulation depth is quite large here, as can be seen from the numerically calculated values of b shown in Fig. 1. This makes it possible to determine experimentally the g factor g_b for any degeneracy multiplicity of the level E_b , with the exception of the case $j_b + j_c < 3$. If $j_b + j_c < 3$, we have $b = 0$ and the TPE polarization becomes circular as in the case of the exciting pulses (8) and (9). As $b \rightarrow 1$ the polarization ellipse is drawn out into a segment making an angle $(\Omega_b \tau_{12} + (\Omega_b + \Omega_c) \tau_{23} + \alpha + \pi/2)$ with the

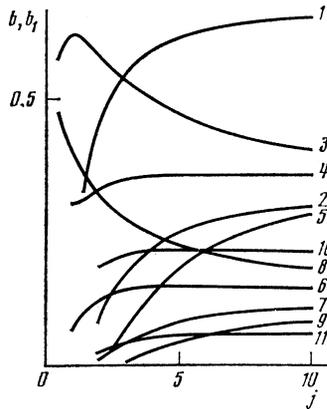


FIG. 1. Amplitudes b and b_1 of the quantum beats (18) and (28) as functions of the angular momentum j . Curves 1–11 pertain to the atomic transitions $j_a \rightarrow j_b \rightarrow j_c$ and $j_a \rightarrow j_b$, respectively, of the following types $j \rightarrow j - j$; $j \rightarrow j - j - 1$; $j \rightarrow j + 1 - j + 1$; $j \rightarrow j - j + 1$; $j \rightarrow j - 1 - j - 1$; $j \rightarrow j + 1 - j$; $j \rightarrow j - 1 - j$; $j \rightarrow j + 1 - j + 2$; $j \rightarrow j - 1 - j - 2$; $j \rightarrow j$ and $j \rightarrow j + 1(j + 1 - j)$. It is assumed that $\Delta = \bar{\Delta} = 0$, $N_c < N_b \ll N_a$ and $k\nu\tau_n \ll 1$ ($n = 1, 2, 3$). Furthermore, $\Theta_1^{(0)} = \Theta_3^{(1)} = \pi$ and $\Theta_2^{(1)} = \pi/2$ for TPE and $\Theta_1^{(0)} = \pi/2$ and $\Theta_2^{(1)} = \pi$ for two-level photon echo. The relative arrangement of curves 1–11 is the criterion for the identification of the atomic transitions.

X axis. The angles are positive when measured from X to Y.

Similarly, by varying τ_{23} under the conditions $0 < (\Omega_b + \Omega_c)\tau_{23} \lesssim 2\pi$ and $\tau_{12} = \text{const}$ we can determine from experiment the sum $\Omega_b + \Omega_c$ which contains the sought g factor g_c of the level E_c belonging to the dipole-forbidden atomic transition $j_a \rightarrow j_c$.

If the values of (20) and (21) are not to change noticeably in response to echo signals with different values of τ_{12} and τ_{23} , the fluctuations δa_n of the amplitudes of the exciting pulses (7)–(9) must be small:

$$\frac{\partial \Theta_n^{(s)}}{\partial a_n} \delta a_n \ll 1, \quad n=1, 2, 3, \quad (23)$$

where $\Theta_n^{(s)} = \tau_n |\chi_{nv}^{(s)}|_{\text{max}}$ is the area of the n -th exciting pulse, whose polarization is characterized by the parameter s , while $|\chi_{nv}^{(s)}|_{\text{max}}$ is the maximum value of the quantity $|\chi_{nv}^{(s)}|$ as a function of ν . In the case of linear ($s=0$) and circular ($s = \pm 1$) polarizations of these pulses we have from Ref. 9:

$$\Theta_n^{(0)} = \beta_n [j/(j+1)(2j+1)]^{1/2}$$

for the atomic transition $j \rightarrow j$,

$$\Theta_n^{(0)} = \beta_n [(j+1)/(2j+1)(2j+3)]^{1/2}$$

for $j \rightarrow j+1, j+1 \rightarrow j, j=0, 1, \dots$

$$\Theta_n^{(0)} = \beta_n [4(j+1)]^{-1/2}$$

for $j \rightarrow j+1, j+1 \rightarrow j, j - \text{half-integer}$,

$$\Theta_n^{(\pm 1)} = \beta_n (4j+2)^{-1/2} \quad \text{for } j \rightarrow j, j=0, 1, \dots$$

$$\Theta_n^{(\pm 1)} = \beta_n [(2j+1)/8j(j+1)]^{1/2}$$

for $j \rightarrow j, j - \text{half-integer}$,

$$\Theta_n^{(\pm 1)} = \beta_n (2j+3)^{-1/2} \quad \text{for } j \rightarrow j+1, j+1 \rightarrow j,$$

$$\beta_1 = a_1 \tau_1 |d_{ba}| \hbar^{-1}, \quad \beta_n = a_n \tau_n |d_{cb}| \hbar^{-1} \quad \text{at } n=2, 3.$$

Violation of inequality (23) affects only the amplitude (20) and the phase (21), but not the oscillation frequency (18). This attests to the reliability of the experimental determination of the g factors by the proposed method. Moreover, for small areas of exciting pulses, $\Theta_n^{(s)} \ll 1$, we have $\alpha = 0$ or $\alpha = \pi/2$, depending on the type of the atomic transition, while the amplitude (20) does not depend on a_n at all.

It is important to note that the magnetic field H does not enter in the quantities (19)–(21). Therefore each of the g factors g_b and g_c can be easily determined also by another method that may turn out to be experimentally preferable. A set of measurements of the intensity (18) at different values of H and at fixed τ_{12} and τ_{23} (and next τ'_{12} and τ'_{23}) leads to the system of algebraic equations

$$g_b(\tau_{12} + \tau_{23}) + g_c \tau_{23} = c_1, \quad g_b(\tau'_{12} + \tau'_{23}) + g_c \tau'_{23} = c_2$$

for the sought g_b and g_c , where the numerical values of c_1 and c_2 are obtained from experiment. For the typical situation $g_b \sim g_c \sim 1$, $\tau_n \sim 10^{-9}$ and $\tau_{12} \sim \tau_{23} \approx 10^{-8}$ see the order of magnitude of H ranges from 1 to 10 Oe.

The formation of the TPE (17) is due to the Doppler mechanism of reversible relaxation. The Doppler shifts $k \cdot v$ of the resonant frequency for atoms moving with different velocities v are not commensurate, and this leads in the course of time to total dephasing of the individual radiators and to a monotonic damping of the coherent-radiation intensity. The radiation is also influenced by the scatter of the resonant levels due to Zeeman splitting. However, the radiator phase shifts, due to the Zeeman scatter of the levels, make up an enumerable set of multiples. This leads to periodic oscillations of the intensity $I(t) = c|\epsilon(t)|^2/2\pi$ of the coherent radiation, which attenuate with time only in the case of simultaneous action of the reversible Doppler relaxation or when irreversible relaxation processes come into play. These intensity fluctuations, as functions of the time t , will also be referred to as quantum beats, since they are a consequence of quantization of the angular momentum in the field H .

In a weak magnetic field $j_b |\Omega_b - \Omega_c| \ll \bar{k}u$ the quantum beats as functions of t do not manifest themselves, owing to Doppler relaxation. But in a strong magnetic field

$$\bar{k}u < j_b |\Omega_b - \Omega_c| \ll 1/\tau_n, \quad n=1, 2, 3, \quad (24)$$

the quantum beats of the TPE intensity as a function of t take the form of a large dome-shaped peak and a number of lateral small peaks that diminish gradually. This picture is repeated with a period $2\pi/|\Omega_b - \Omega_c|$, so that the TPE intensity profile breaks up into a number of peaks located under the envelope that describes the echo signal at $H = 0$ (Fig. 2). By dividing the distance between the principal peaks by the peak width at $2/3$ its height we can determine, from the experimental plot of the TPE intensity, the angular momentum j_b at any multiplicity of level degeneracy. Numerical calculations have shown that the result of such a division differ from the true value $2j_b$ at $1 < j_b < 10^2$ by several percent if the areas under the exciting pulses lie in the interval $0 < \Theta_1^{(0)} \leq \pi$ at $\Theta_1^{(0)} = 2\Theta_2^{(\pm 1)} = \Theta_3^{(\pm 1)}$. The modulus $|g_b - g_c|$ of the difference of the g factors is calculated at the

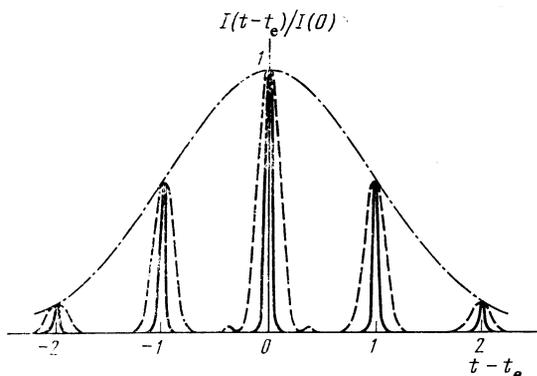


FIG. 2. Quantum beats of TPE vs time. The solid and dashed curves pertain to angular momenta j_b equal to 10 and $5/2$, respectively. The dash-dot envelope describes the profile of the echo signal at $H = 0$. It is assumed that $j_a = j \rightarrow j_b = j \rightarrow j_c = j$, $\Delta = \bar{\Delta} = 0$, $\Theta_1^{(0)} = \Theta_3^{(1)} = \pi$, $\Theta_2^{(1)} = \pi/2$, $\gamma_{cb}\tau_{23} \ll 1$, $\gamma_b\tau_{12} \ll 1$, $N_c < N_b \ll N_a$ and $|\Omega_b - \Omega_c| \ll 1/\tau_n$, where $n = 1, 2, 3, \dots$. Unity on the abscissa axis corresponds to $2\pi|\Omega_b - \Omega_c|^{-1}$. The ordinate is $I(t) = c|\epsilon(t)|^2/2\pi$ in relative units.

same time.

Gas atoms usually have $ku < 10^9 \text{ sec}^{-1}$, $j_b \sim 10$, and $|g_b - g_c| \sim 1$, so that the inequality (24) is easily satisfied if pulses (7)–(9) with picosecond durations $\tau_n < 10^{-11} \text{ sec}$ are used in a magnetic field $H \sim 10^2 \text{ Oe}$. In the region (24), after substituting $a_n \tau_n \rightarrow \int a_n(t) dt$, Eqs. (16)–(23) are valid for pulses (7)–(9) with arbitrary profile. The proposed method for the determination of the angular momenta is not inferior to the known methods,^{10,11} especially in the region $j_b \gg 1$.

3. INVESTIGATION OF ADJACENT ATOMIC TRANSITION

To determine the g factor g_a and the angular momentum j_a of the level E_a of an adjacent transition $j_a \rightarrow j_b$ at any degeneracy multiplicity we must use the two-pulse photon echo produced by irradiating the gas with two pulses with linear (7) and circular

$$E_2 = \frac{1}{2} I^{(0)} a_2 \exp [i(kz - \omega t - \Phi_2)] + \text{c.c.} \quad (25)$$

polarizations, separated by a time interval τ . In this case the electric field of the two-pulse photon echo takes the form

$$E_{1e} = e_1(t - z/c) \exp [i(kz - \omega t - 2\Phi_2 + \Phi_1)] + \text{c.c.}, \\ e_1(t) = e_{01} \int d\nu f(\nu) \{ I^{(1)} U_1 \exp [i(\sigma - 1)(\Omega_a + \Omega_b)\tau] \} \quad (26)$$

$$+ I^{(-1)} U_{-1}^{(\sigma)} \exp [i(\sigma + 1)(\Omega_a + \Omega_b)\tau] \exp [i(\Delta - kv_a)(t - t_{1e}) - \gamma_{ba} t], \\ e_{01} = 2\pi kL |d_{ba}| N_0, \quad t_{1e} = 2\tau + \tau_1 + \tau_2,$$

$$U_q^{(\sigma)} = \sum_x G_x P_{xq}^{(\sigma)}, \quad G_x = \sum_{\mu} \begin{pmatrix} j_b & j_a & \kappa \\ \mu & -\mu & 0 \end{pmatrix} A_{1\mu}^{(\sigma)} B_{1\mu}^{(\sigma)},$$

$$P_{xq}^{(\sigma)} = (-1)^\sigma (2\kappa + 1) \sum_{\mu m} \begin{pmatrix} j_b & j_a & 1 \\ \mu & -m & -q \end{pmatrix} \begin{pmatrix} j_b & j_a & \kappa \\ m + \sigma & \sigma - \mu & q - 2\sigma \end{pmatrix}$$

$$\times B_{2\mu}^{(\sigma)} B_{2, m + \sigma}^{(\sigma)} d_{2\sigma - q, 0}^{x*} \left(\frac{\pi}{2} \right) \exp [i(\Omega_a m - \Omega_b \mu)(t - t_{1e})],$$

$$q = \pm 1, \quad \sigma = \pm 1. \quad (27)$$

In contrast to (18), the quantum beats of the two-pulse photon echo as a function of H or τ are connected only with the precession of the induced atomic polarization, and take the form

$$I_1^{(\sigma)} = \{1 - b_1 \cos [2(\Omega_a + \Omega_b)\tau + 2\alpha_1]\} I_{01} \exp(-4\gamma_{ba}\tau), \quad (28)$$

where the quantities I_{01} , b_1 , and α_1 are obtained from (19)–(21) by replacing $W_q^{(\sigma)}$ by $U_q^{(\sigma)}$ at $t = t_{1e}$ and $q = \pm 1$. Since I_{01} , b_1 , and α_1 do not depend on H , the quantum beats (28) are useful for the determination of the g -factor sum $g_a + g_b$ by varying H , so that g_a can be calculated if g_b is known. For this purpose it is possible also to vary τ under the condition $\gamma_{ba}\tau \ll 1$. The modulation depth in (28) is large in a wide range of the parameters it contains (Fig. 1), and for atomic transitions with $j_a + j_b < 3$ we have $b_1 = 0$. At another choice of the polarizations of the exciting pulses, the quantum beats of the two-pulse photon echo in two-level systems are complicated in form and do not permit a determination of $g_a + g_b$, as can be seen from the investigation results.^{1-3,12}

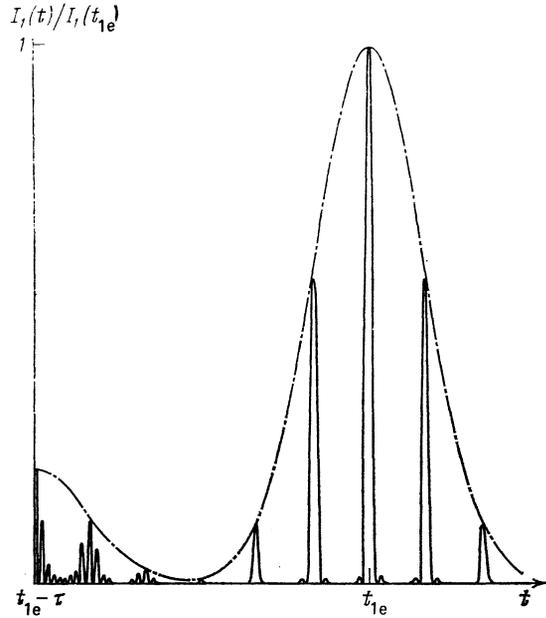


FIG. 3. Quantum beats of FOI and of two-pulse photon echo in a strong field \mathbf{H} (solid curve). The dash-dot curve describes the coherent radiation at $\mathbf{H} = 0$. It is assumed that $j_a = j - j_b = j$, $j = 5$, $\Delta = 0$, $\Theta_1^{(0)} = \pi/2$, $\Theta_2^{(1)} = \pi$, $ku\tau = 2\pi$, $6ku = |\Omega_a - \Omega_b|$ and $\gamma_{ba}\tau \ll 1$. The left-hand side shows the damping of the free optical induction on whose basis the two-pulse photon echo is produced at the instant $t = t_{1e}$.

We have left out of (26) the terms that describe at $\tau + \tau_1 + \tau_2 \leq t$ the free optical induction (FOI), which attenuates by the instant the two-pulse photon echo is produced, owing to the reversible Doppler relaxation. The contribution of these terms to the intensity $I_1(t) = c|\mathcal{E}_1(t)|^2/2\pi$ of the coherent radiation in the region (24) is shown in Fig. 3. The Zeeman splitting of the resonant levels in a strong field \mathbf{H} breaks up the two-pulse photon echo into a set of individual peaks with approximate width $\pi/j_a |\Omega_a - \Omega_b|$ at $2/3$ height, and with a distance $2\pi/|\Omega_a - \Omega_b|$ between them if $2\Theta_1^{(0)} = \Theta_2^{(\pm 1)}$ and $0 < \Theta_2^{(\pm 1)} \leq \pi$. This enables us to calculate j_a and $|\Omega_a - \Omega_b|$. Yet the FOI structure consists of one principal peak and two side lobes, and is repeated with a period $2\pi/|\Omega_a - \Omega_b|$.

4. CONCLUSION

To produce a TPE with an aim at observing the effect (18) one can use also a linearly polarized pulse (7) propagating in a direction opposite to that of the two other circularly polarized ones. In this case the amplitude of the produced TPE is described by Eq. (17) with the substitution $k_{uz} \rightarrow -k_{uz}$ in the quantities $A_{1\mu}^{(0)}$ and $B_{1\mu}^{(0)}$.

In addition to those considered above, there are also other TPE versions, some of which were used in experiments¹³ at $\mathbf{H} = 0$ to study the relaxation of optically forbidden transitions. From among them, only stimulated TPE makes it possible to determine the g factor of the common level E_b as a result of precession of the magnetic moments of the excited atoms. The others are not suitable for the determination of the g factor of each level separately, since they lead in the presence of a field \mathbf{H} to harmonic quantum beats with a frequency proportional to the sum of the g factors, or

else lead to a more complicated dependence of the echo-signal intensity on the values of τ_{12} , τ_{23} , and H .

When the delay times τ_{12} , τ_{23} , and τ are comparable with or exceed the characteristic time of the irreversible relaxation, the quantities (18) and (28) decrease exponentially as functions of τ_{12} , τ_{23} , and τ , and this makes it possible to determine γ_{ba} , γ_{cb} , and γ_b in experiment. Yet the amplitudes b at $N_c < N_b \ll N_a$ and b_1 , as well as the phases α and α_1 , are not altered by the irreversible relaxation characterized by the constants γ_{ba} , γ_{cb} , and γ_b . Moreover, the amplitudes and phase of the quantum beats (18) and (28) are not subject to relaxation also in the case when the model of elastic depolarizing atomic collisions^{7,14} is applicable, provided that exciting pulses with small areas are used. This means that the proposed procedure for determining g factors can be used also in the presence of irreversible relaxation, without the need for exact knowledge of the relaxation parameters. At the same time, in methods in which level crossing⁵ and the Hanle effect⁶ are used, the g factor of the excited level enters only in the combination g_b/γ_b with the decay constant γ_b of the excited state, so that to determine one of these quantities in experiment we must know the other. In addition, the methods of Refs. 5 and 6 cannot be used to determine the g factor of the ground level E_a and of the excited E_c belonging to a forbidden transition.

We have neglected above the Faraday rotation. If it is appreciable, the dependence of the radiation intensities (18) and (38) on τ_{12} , τ_{23} , and τ remains unchanged, but terms representing the Faraday angle of the wave-polarization rotation must be added to α and α_1 . The character of the quantum beats on Figs. 2 and 3 likewise remains unchanged.

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