Ultra-high-resolution echo spectroscopy in a strong magnetic field

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It is shown that in a strong magnetic field in which the Zeeman splitting of the resonant levels is much larger than the hyperfine splitting, the quantum beats of the three-level and two-pulse photon echoes have a very simple form and afford the possibility of determining independently in the same experiment the g factors of the levels, the hyperfine-splitting constants, and the irreversible-relaxation constants. On the basis of these quantum beats, a technique is proposed which provides ultrahigh resolution of nearby sublevels, which can lie either within or outside the Doppler width of the spectral line. This effect is interpreted physically as resulting from the interference of coherent waves emitted at individual atomic transitions between hyperfine sublevels in the Paschen-Back regime in the free decay of the induced atomic polarization. It is demonstrated that this effect is general in nature and can also be observed in other transient processes in a strong field **H** if ultrashort light pulses with specially chosen polarizations are used for the optical orientation of the atoms.

The specific rotation of the polarization and the quantum beats of the photon echo (PE) and three-level photon echo (TPE) in a gas have been considered previously[1–4] for the case of a weak magnetic field **H**, when the Zeeman splitting is linear in the field H (the Zeeman regime). In addition, the transition regime, in which the linear dependence on H breaks down, has been studied experimentally.[2,3] In both these regimes one can obtain clear spectroscopic information for atoms with nonzero nuclear spin only in special cases for which the exciting light pulses engage individual hyperfine sublevels of the resonant levels[4] or a single hyperfine sublevel of the lower level and two of the upper level.[2,3,5] If a larger number of hyperfine sublevels are engaged, then the specific rotation of the potential and quantum beats of the PE and TPE in a gas have a complex and disorderly form, in contrast to the case of atoms with zero nuclear spin, for which these effects under certain conditions have a regular nature.[1,4]

In the present paper we show that in a strong magnetic field **H**, when the Zeeman splitting again becomes linear in H and is larger than the hyperfine splitting (the Paschen-Back regime), the quantum beats of the PE and TPE in a gas, independently of the degeneracy of the levels, become orderly and in this way are fundamentally different from the chaotic beats of the PE and TPE in a weak **H** field in the presence of a hyperfine structure of the levels. The orderly nature of the quantum beats is due to the special form of the atomic energy in the Paschen-Back regime and requires a special choice of the polarizations of the exciting light pulses, corresponding to the selection rules for emission and absorption of a photon in a transverse **H** field. The orderliness of the quantum beats in a strong **H** field lies in the fact that the peaks of the intensity of the PE and TPE as a function of the time delay between the exciting pulses simultaneously undergo rapid harmonic oscillations, with a period which contains the g factors of the resonant levels, and slow oscillations, with a period that depends on the hyperfine splitting constants. The envelope through the maxima of the slow quantum beats decays exponentially, with an exponent containing the relaxation constant. In addition, at a fixed time delay the quantum beats of the PE and TPE as functions of H have a harmonic form, with a period which depends on the g factors of the resonant levels.

The extremely simple form of the quantum beats of the PE and TPE in a strong magnetic field in the presence of a hyperfine structure of the levels is due to the interference of the light waves emitted spontaneously in atomic transitions between hyperfine sublevels with fixed values of the electronic quantum numbers and different projections M_I of the nuclear spin **I** which satisfy the selection rule $\Delta M_i = 0$. Near the time of the maximum of the PE and TPE, these emitted waves are coherent, and their frequencies differ from one another by a small quantity which depends on the hyperfine splitting constants of the resonant levels. The contributions from individual pairs of hyperfine sublevels combine to form in the amplitude of the emitted wave a characteristic factor that takes into account the hyperfine structure of the resonant levels. As a result, a unique situation arises in which three different physical phenomena, viz., the precession of the induced atomic polarization and the magnetic moment of the excited atom around the direction of **H**, the hyperfine splitting of the levels, and the irreversible relaxation, introduce into the amplitude of the PE and TPE light waves separate and independent factors which permit independent experimental study of the g factors of the levels, the hyperfine splitting constants, and the irreversible-relaxation constants. The general physical significance of this effect is that it can also be manifested in other transient processes in which one is studying the coherent electromagnetic radiation of oriented atoms, due to the free decay of the induced atomic polarization in the presence of a strong magnetic field and of a hyperfine structure of the levels. In this connection we note that in the conventional methods of optical orientation[6–14] the sought for g factors, hyperfine splitting constants, and relaxation constants are contained the intensity of the emitted wave in the form of a definite combination, so

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that to determine one of these quantities requires knowledge of another. Another advantage of the proposed method is the exceptionally simple form of the quantum beats of the PE and TPE, which is independent of the gas temperature and the detuning from resonance and also of the intensity, shape, and spectral composition of the exciting pulses, which are usually poorly controlled in an experiment. The aforementioned universality of the quantum beats of the PE and TPE is a convincing incentive for new experimental studies.

1. BASIC EQUATIONS AND NOTATION

Let us consider a gas containing active atoms with nonzero nuclear spin. We assume that we are dealing with the common case in which the magnetic hyperfine interaction is large compared to the quadrupolar hyperfine interaction.[15] In a weak external magnetic field **H** the coupling between the electronic angular momentum **J** and the nuclear spin **I** is not broken, and so the energy of the atom is given by

$$\begin{aligned} E_{FM_F}&=E+\hbar\Delta_F+g_F\mu_0M_FH,\\ \Delta_F&=A[F(F+1)-J(J+1)-I(I+1)]/2\hbar,\\ g_F&=g[F(F+1)+J(J+1)-I(I+1)]/2F(F+1), \end{aligned} \tag{1}$$

where E is the energy of the atom at $\mathbf{H}=0$ in the absence of the hyperfine interaction, A is the magnetic hyperfine splitting constant, g is the gyromagnetic ratio (g factor), μ_0 is the Bohr magneton, and M_F is the projection of the total angular momentum $\mathbf{F}=\mathbf{J}+\mathbf{I}$ onto the quantization axis, which is directed along **H**. Beside the Zeeman regime (1), we shall also consider the Paschen-Back regime in a strong magnetic field, in which the coupling between the vectors **J** and **I** is broken, the Zeeman splitting $g\mu_0MH$ is considerably larger than the hyperfine splitting AMM_I, and the energy of the atom is of the form

$$E_{MM_I}=E+g\mu_0MH+AMM_I, \tag{2}$$

where M and M_I are the projections of the vectors **J** and **I** onto the quantization axis. The second term in (2) is large compared to the third but small compared to the fine-structure splitting of the level.

The propagation of the light pulse is described by the equations

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E}=\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\int \mathrm{Sp}(\rho\mathbf{d})\,d\mathbf{v}, \tag{3}$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla-\hat{\Gamma}\right)\rho=\frac{i}{\hbar}[\rho(\mathscr{H}+A\mathbf{JI}+V-\mathbf{Ed})-(\mathscr{H}+A\mathbf{JI}+V-\mathbf{Ed})\rho], \tag{4}$$

where **E** is the electric field of the pulse, ρ is the density matrix, c is the speed of light in vacuum, **v** is the thermal velocity of the atom, **d** is the dipole moment operator, $\mathscr{H}$ is the Hamiltonian of the atom in its center-of-mass system in the absence of external fields and the hyperfine interaction, $A\,\mathbf{J}\cdot\mathbf{I}$ is the magnetic hyperfine interaction operator, and V and $-\mathbf{E}\cdot\mathbf{d}$ are the interaction operators of the atom with the magnetic field **H** and electric field **E**. The term $\hat{\Gamma}\rho$ takes into account the additional pumping due to the Boltzmann distribution of the atoms over energy states, the departure of atoms from each level due to radiative decay and atomic collisions, and the arrival of atoms at the lower level due to spontaneous emission at the upper levels.

In the Paschen-Back regime (2) we solve Eq. (4) in the MM_I representation, which is constructed with the aid of the wave functions with quantum numbers $JIMM_I$ and energy E_{MM_I}. Then the term $\hat{\Gamma}\rho$ for the resonant states of the atom with indices a and b assumes the form

$$\begin{aligned} (\hat{\Gamma}\rho)_{M_aM_I,M'_aM'_I}&=\frac{(\gamma_aN_a-\gamma N_b)f(v)}{(2J_a+1)(2I+1)}\delta_{M_aM'_a}\delta_{M_IM'_I}\\ &\quad-\gamma_a\rho_{M_aM_I,M'_aM'_I}\\ &=+\frac{(2J_b+1)\gamma}{|d_{ba}|^2}\sum_{M_bM'_b}\mathbf{d}_{M_aM_b}\rho_{M_bM_I,M'_bM'_I}\mathbf{d}_{M'_bM'_a}, \end{aligned} \tag{5}$$

$$(\hat{\Gamma}\rho)_{M_bM_I,M'_bM'_I}=\frac{\gamma_bN_bf(v)}{(2J_b+1)(2I+1)}\delta_{M_bM'_b}\delta_{M_IM'_I}-\gamma_b\rho_{M_bM_I,M'_bM'_I}, \tag{6}$$

$$(\hat{\Gamma}\rho)_{M_bM_I,M_aM_I'}=-\gamma_{ba}\rho_{M_bM_I,M_aM_I'},$$
$$f(v)=(\pi^{1/2}u)^{-3}\exp(-v^2/u^2),\quad \gamma=4|d_{ba}|^2\omega_{ba}/3(2J_b+1)\hbar c^3. \tag{7}$$

Here N_a (N_b) is the total steady-state density of atoms at all the hyperfine sublevels of the level E_a (E_b) at $\mathbf{E}=\mathbf{H}=0$, γ is the probability of spontaneous emission of a photon $\hbar\omega_{ba}=E_b-E_a$ by an isolated atom, d_{ba} is the reduced dipole moment of the atomic transition $J_a\to J_b$, $f(v)$ is the Maxwellian distribution, u is the most probable speed of the atom, $\hbar\gamma_a$ ($\hbar\gamma_b$) is the width of each hyperfine sublevel of the given level E_a (E_b), and γ_{ba} is the half-width of the spectral line of the atomic transition between hyperfine sublevels of the upper level E_b and lower level E_a and is the same for all the hyperfine components. For the Zeeman regime (1) the good quantum numbers are $JIFM_F$ and the energy E_{FM_F}, and so we use the FM_F representation, for which the indices in Eqs. (5)–(7) are relabeled $M\to F(M'\to F')$ and $M_I\to M_F$ ($M'_I\to M_{F'}$) and the replacement $d_{M_aM_b}\to d_{F_a}M_{F_a}$, $F_bM_{F_b}$, with the subsequent summation over F_b and M_{F_b} (F'_b and $M_{F'_b}$), is made in Eq. (5).

At a sufficiently low gas pressure the main relaxation mechanism is radiative decay. If the gas pressure is not low, then γ_a, γ_b, and γ_{ba} take into account the spontaneous emission and atomic collisions. We henceforth consider an optically thin gaseous medium, and we solve Eq. (4) in the approximation of a given field **E**. We substitute ρ obtained in this way into (3) in order to find the influence of the gas back on the fixed field. Here it is assumed that the duration τ_n of the propagating light pulse is short:

$$\gamma_a\tau_n\ll1,\quad \gamma_{ba}\tau_n\ll1,\quad g_a\mu_0JH\tau_n\ll1\quad (g_{F_a}\mu_0F_aH\tau_n\ll1), \tag{8}$$

where the inequalities which are not shown are obtained by relabeling the indices a and b, and the index $n = 1, 2, 3$ is the number of the light pulse in the sequence when several pulses are sent through the gas in succession. Inequalities (8) allow us to neglect the irreversible relaxation and the magnetic field in the time interval in which the light pulse acts. When inequalities (8) hold, the evolution operator for Eq. (4) in the presence of hyperfine structure and a fixed **E** field is found by the method developed in Ref. 16. After the exciting pulse has acted, we must set $\mathbf{E} = 0$ in (4) and make a complete allowance for the irreversible relaxation and for the magnetic field. Prior to the passage of the light pulse, the gas atoms are in statistical equilibrium.

2. INFLUENCE OF HYPERFINE STRUCTURE ON THE TPE IN A FIELD H

In order for the quantum beats of the TPE in a transverse magnetic field **H** to have the simplest form and the TPE to be easily distinguished from the other electromagnetic signals, the center frequencies and polarizations of the exciting pulses must be chosen as follows:

$$\mathbf{E}_1 = {}^1/_2\mathbf{l}^{(\sigma)}a_1 \exp[i(k_1 z-\omega_1 t-\varphi_1)]+\text{c.c.}, \quad \sigma=\pm 1, \tag{9}$$

$$\mathbf{E}_2 = {}^1/_2\mathbf{l}^{(0)}a_2 \exp[i(k_2 z-\omega_2 t-\varphi_2)]+\text{c.c.}, \tag{10}$$

$$\mathbf{E}_3 = {}^1/_2\mathbf{l}^{(0)}a_3 \exp[i(k_3 z-\omega_3 t-\varphi_3)]+\text{c.c.}, \quad \omega_3=\omega_2,$$
$$a_n=a_n(t'), \quad t'=t-t_n-z/c, \quad n=1, 2, 3. \tag{11}$$

Here the center frequencies $\omega_1 = k_1 c$ and $\omega_2 = k_2 c$ are close to the atomic frequencies $\omega_{ba} = (E_b - E_a)/\hbar$ and $\omega_{cb} = (E_c - E_b)/\hbar (E_a < E_b < E_c)$. The amplitude $a_n(t')$ is a slow function of time in the interval $0 \leqslant t' \leqslant \tau_n$. The light pulses enter the gas at the point $z = 0$ at the respective times $t_1 = 0$, $t_2 = \tau_{12} + \tau_1$, and $t_3 = \tau_{12} + \tau_{23} + \tau_1 + \tau_2$, where the time intervals τ_{12} and τ_{23} between the exciting pulses are arbitrary. The unit vectors for the linear $\mathbf{l}^{(0)}$ and circular $\mathbf{l}^{(\sigma)}$ polarizations are written

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

where $\mathbf{l}_x$ and $\mathbf{l}_y$ are the unit vectors of the Cartesian axes X and Y, and the vectors $\mathbf{l}_x$ and **H** are parallel. The exciting pulses (9)–(11) give rise to a TPE which in the lowest order of perturbation theory is a six-photon process, unlike the three-level echo of Ref. 17, which is based on a four-photon interaction.

For the sake of generality, we assume that with respect to the excited states of the atoms at the levels E_b and E_c, the magnetic field is strong and the Paschen-Back regime (2) obtains, whereas for the atom at the lower level E_a it is arbitrary, and either the Zeeman (1) or Paschen-Back regime (2) can obtain. In the Paschen-Back regime we have for Eq. (4)

$$[\rho(AJI+V)-(AJI+V)\rho]_{M_b M_I, M_b' M_I}$$
$$=(\hbar\Omega_b+A_b M_I)(M_b'-M_b)\rho_{M_b M_I, M_b' M_I},$$
$$[\rho(AJI+V)-(AJI+V)\rho]_{M_c M_I, M_b M_I}$$
$$=[(\hbar\Omega_b+A_b M_I)M_b-(\hbar\Omega_c+A_c M_I)M_c]\rho_{M_c M_I, M_b M_I},$$
$$\Omega_b=g_b\mu_0 H/\hbar, \quad \Omega_c=g_c\mu_0 H/\hbar,$$

where the quantization axis is chosen parallel to **H** and a perturbation theory is used which takes it into account that the hyperfine interaction energy is small. The omitted terms are smaller than the rest in order of magnitude by factors of $A_b I/\Omega_b$ and $A_c I/\Omega_c$. For the chosen polarizations of the exciting pulses (9)–(11), the structure of this operator provides a general picture of the quantum beats of the TPE. The beats assume an extremely simple form when the spectral width of the light pulses (9)–(11) is large compared to the hyperfine splitting of the resonant levels E_a, E_b, and E_c:

$$|\Delta_{F_a}| \ll 1/\tau_1, \quad |\Delta_{F_b}| \ll 1/\tau_1, \tag{12}$$

$$|\Delta_{F_b}| \ll 1/\tau_2(1/\tau_3), \quad |\Delta_{F_c}| \ll 1/\tau_2(1/\tau_3). \tag{13}$$

If the detuning from resonance is small, i.e.,

$$|\Delta_n|\tau_n \lesssim 1, \quad n=1, 2, \quad \Delta_1=\omega_1-\omega_{ba}, \quad \Delta_2=\omega_2-\omega_{cb},$$

then, by virtue of inequalities (12) and (13), all the hyperfine sublevels of each resonant level participate in the formation of the TPE. Since the atomic state with electronic angular momentum J_b is common to the two adjacent optically allowed transitions $J_a \to J_b$ and $J_b \to J_c$, it is for this level that the spectroscopic information contained in the TPE is most complete.

Experimentally it is easy to extract from the exciting pulses (9)–(11) the projection of the electric field of the TPE onto the Y axis:

$$E_{1y}=\varepsilon_1(t-z/c)\exp[i(k_2 z-\omega_2 t-\varphi_e)]+\text{c.c.}, \tag{14}$$

where $\varphi_e = 2\varphi_3 - \varphi_2$ and the amplitude $\varepsilon_1(t)$ is given by the expression

$$\varepsilon_1(t) = -\frac{i}{2^{1/2}}\frac{\varepsilon_{01}}{2I+1}$$
$$\times\sum_{M_I}\int f(v)\exp[i(\Delta_2-\mathbf{k}_2\mathbf{v})(t-t_e)-\gamma_{cb}(t-\tau_{12})$$
$$-\gamma_b\tau_{12}]\{W_1^{(\sigma)}\exp[-i(\Omega_b+A_b M_I\hbar^{-1})\tau_{12}$$
$$-i(\Omega_b+\Omega_c+(A_b+A_c)M_I\hbar^{-1})\tau_{23}]$$
$$+W_{-1}^{(\sigma)}\exp[i(\Omega_b+A_b M_I\hbar^{-1})\tau_{12}$$
$$+i(\Omega_b+\Omega_c+(A_b+A_c)M_I\hbar^{-1})\tau_{23}]\}dv,$$

$$W_q^{(\sigma)} = \sum_{\varkappa=1}^{2J_b} Q_\varkappa^{(\sigma)} R_{\varkappa q}^{(0)}(t-t_e), \quad q=\pm 1, \quad \sigma=\pm 1, \quad J_b>0,$$

$$\varepsilon_{01}=2\pi k_2 L|d_{cb}|N_{ab}, \quad N_{ab}=N_a/(2J_a+1)-N_b/(2J_b+1), \tag{15}$$

$$Q_\varkappa^{(\sigma)} = \frac{1}{N_{ab}}\sum_\mu (-1)^{J_b-\mu}\begin{pmatrix} J_b & J_b & \varkappa \\ \mu & -\mu & 0\end{pmatrix}\left[|B_{1\mu}^{(\sigma)}|^2\frac{N_a}{2J_a+1}\right.$$
$$\left.+|A_{1\mu}^{(\sigma)}|^2\frac{N_b}{2J_b+1}\right],$$

$$R_{\varkappa q}^{(0)}(t)=(2\varkappa+1)\,d_{0q}^{\varkappa}\left(\frac{\pi}{2}\right)\sum_{\nu\mu}(-1)^{J_b-\mu}\begin{pmatrix} J_c & J_b & 1\\ \nu & -\mu & -q\end{pmatrix}$$

$$\times\begin{pmatrix} J_b & J_b & \varkappa\\ \nu & -\mu & -q\end{pmatrix}A_{2\nu}^{(0)}B_{2,\mu+\sigma}^{(0)*}B_{3\nu}^{(0)}B_{3\mu}^{(0)}$$

$$\times\exp\{i[(\Omega_b+A_bM_I/\hbar)\mu-(\Omega_c+A_cM_I/\hbar)\nu]t\},$$

$$\Omega_a=g_a\mu_0H/\hbar,\quad t_e=\tau_{12}+2\tau_{23}+\tau_1+\tau_2+\tau_3.$$

Here L is the length of the optical path of the exciting pulses in the gas, d_{cb} is the reduced dipole moment of the atomic transition $J_b\to J_c$, $\mathbf{k}_2$ is the wave vector of light pulse (10), and the values 1 and -1 of the index σ refer to right- and left-polarized exciting pulses, respectively. The quantity $d_{0q}^{\varkappa}(\beta)$ is the Wigner D function $D_{pq}^{\varkappa}(\alpha,\beta,\gamma)$ for $p=\alpha=\gamma=0$. For the $3j$ symbols and the other quantities we use the notation of Ref. 18.

If the light pulse with polarization $\mathbf{l}^{(s)}$, real amplitude $a_n(t)$, center frequency $\omega_n=k_nc$, and detuning $\Delta_n=\omega_n-\omega_{ba}$ has a wide spectral composition $k_nu+|\Delta_n|\ll 1/\tau_n$, then one should use the expression

$$A_{n\mu}^{(s)}=\cos\left(\frac{1}{2}\Lambda_\mu^{(s)}\int_0^{\tau_n}a_n(t)\,dt\right),$$

$$B_{n\mu}^{(s)}=\sin\left(\frac{1}{2}\Lambda_\mu^{(s)}\int_0^{\tau_n}a_n(t)\,dt\right),$$

$$\Lambda_\mu^{(s)}=\frac{|d_{ba}|}{\hbar}\begin{pmatrix} J_b & J_a & 1\\ \mu & s-\mu & -s\end{pmatrix},\qquad s=0,\pm1,\ n=1,2,3.$$

In the case of a square light pulse with constant amplitude a_n and an arbitrary value of the parameter $(k_nu+|\Delta_n|)\tau_n$ we have

$$A_{n\mu}^{(s)}=\cos\frac{\Omega_{n\mu}^{(s)}\tau_n}{2}+i\frac{\mathbf{k}_n\mathbf{v}-\Delta_n}{\Omega_{n\mu}^{(s)}}\sin\frac{\Omega_{n\mu}^{(s)}\tau_n}{2},$$

$$B_{n\mu}^{(s)}=\frac{a_n\Lambda_\mu^{(s)}}{\Omega_{n\mu}^{(s)}}\sin\frac{\Omega_{n\mu}^{(s)}\tau_n}{2},$$

$$\Omega_{n\mu}^{(s)}=[(\mathbf{k}_n\mathbf{v}-\Delta_n)^2+a_n^2(\Lambda_\mu^{(s)})^2]^{1/2}.$$

Another case of considerable interest is that of a light pulse of small area $\theta_n^{(s)}$, with a real or complex amplitude $a_n(t)$; in accordance with Ref. 19, the area of the pulse is defined as

$$\theta_n^{(s)}=\max_\mu|\Lambda_\mu^{(s)}|\int_0^{\tau_n}|a_n(t)|\,dt.$$

The values of $\theta_n^{(s)}$ for linear ($s=0$) and circular ($s=\pm1$) polarization of the light pulse and for different atomic transitions $J_a\to J_b$ are given by

$$\theta_n^{(0)}=\beta_n[J/(J+1)(2J+1)]^{1/2}\quad\text{for } J\to J,$$

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

$$\text{for } J\leftrightarrow J+1,\ J=0,1,\ldots,$$

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

$$\text{for } J\leftrightarrow J+1 \text{ c with half-integral } J$$

$$\theta_n^{(\pm1)}=\beta_n(4J+2)^{-1/2}\quad\text{for } J\to J,\quad J=0,1,\ldots,$$

$$\theta_n^{(\pm1)}=\beta_n[(2J+1)/8J(J+1)]^{1/2}$$

$$\text{for } J\to J \text{ c with half-integral } J$$

$$\theta_n^{(\pm1)}=\beta_n(2J+3)^{-1/2}\quad\text{for } J\leftrightarrow J+1,$$

$$\beta_n=\hbar^{-1}|d_{ba}|\int_0^{\tau_n}|a_n(t)|\,dt,\qquad n=1,2,\ldots.$$

For a small area $\theta_n^{(s)}\ll 1$ we get

$$A_{n\mu}^{(s)}=\exp[i(\mathbf{k}_n\mathbf{v}-\Delta_n)\tau_n/2],$$

$$B_{n\mu}^{(s)}=\frac{1}{2}\Lambda_\mu^{(s)}\int_0^{\tau_n}a_n(t')\exp\left[i(\mathbf{k}_n\mathbf{v}-\Delta_n)\left(t'-\frac{\tau_n}{2}\right)\right]dt',$$

where the parameter $(k_nu+|\Delta_n|)\tau_n$ is arbitrary and the function $a_n(t')$ takes on complex values in the presence of phase modulation of the pulses (9)–(11).

The factor $B_{1\mu}^{(\sigma)}$ in amplitude (15) is equal to the quantity $B_{n\mu}^{(s)}$ given above for $n=1$ and $s=\sigma$, while the factors $A_{2\mu}^{(0)}$, $B_{2\mu}^{(0)}$ and $B_{3\mu}^{(0)}$ are obtained from $A_{n\mu}^{(s)}$ and $B_{n\mu}^{(s)}$ with the replacement $\omega_{ba}\to\omega_{cb}$, $J_a\to J_b$, $J_b\to J_c$, $d_{ba}\to d_{cb}$, $s=0$ and $n=2$ (3).

According to (15), the maximum of the TPE, reached at the time $t=t_e$. The term $(\Omega_b+A_bM_I/\hbar)\tau_{12}=g_b\mu_0H_{bI}\tau_{12}\hbar^{-1}$ in the phase in (15) describes the precession of the magnetic moment of the atom in the time interval $\tau_1\leqslant t\leqslant\tau_1+\tau_{12}$ in a total magnetic field $H_{bI}\mathbf{l}_x=\mathbf{H}+\mathbf{l}_xA_bM_I/g_b\mu_0$, where the second term represents the contribution from the hyperfine interaction and $\mathbf{l}_x$ is the unit vector along the quantization axis ($\mathbf{l}_x\|\mathbf{H}$). The other term in the phase, $[\Omega_b+\Omega_c+(A_b+A_c)M_I/\hbar]\tau_{23}$, describes the precession of the induced atomic polarization. The amplitude in (15) contains a contribution only from the excited states of the atom at the levels E_b and E_c. It is precisely for this reason that the Paschen-Back regime is used for these excited states, whereas for the lower level the magnetic field is arbitrary. In these excited states the coupling between the electronic angular momentum and the nuclear spin is broken, and each of these quantities is conserved separately, precessing around $\mathbf{H}$. Because of the independence of the electronic and spin states and the selection rule $\Delta M_I=0$, the amplitude in (15) contains contributions solely from the atomic transitions between hyperfine sublevels with the same index M_I. Since the radiative transitions between hyperfine sublevels with the same M_I and fixed electronic quantum numbers occur independently, their contributions to the amplitude in (15) are added with allowance for the statistical weight of the spin state for all M_I in the interval $-I\leqslant M_I\leqslant I$. The coherence of the emitted waves is maximum near the time $t=t_e$, when the TPE is maximum. At this time a characteristic factor

$$\sin\{(2I+1)[A_b\tau_{12}+(A_b+A_c)\tau_{23}]/2\hbar\}/$$
$$\sin\{[A_b\tau_{12}+(A_b+A_c)\tau_{23}]/2\hbar\},$$

which takes into account the hyperfine structure of the levels, is separated in the amplitude (15). The irreversible relaxation and the precession of the induced atomic polarization and of the magnetic moment of the atom about **H** are taken into account in the amplitude in (15) by other independent factors.

At $t=t_e$ the intensity $I_1(t)=c|\varepsilon_1(t)|^2/2\pi$ corresponding to electric field (14) is of the form

$$I_1(t_e)=I_{01}\exp(-2\gamma_b\tau_{12}-4\gamma_{cb}\tau_{23})\times\cos^2[\Omega_b\tau_{12}+(\Omega_b+\Omega_c)\tau_{23}+\alpha_1]\times\frac{\sin^2\{(2I+1)[A_b\tau_{12}+(A_b+A_c)\tau_{23}]/2\hbar\}}{(2I+1)^2\sin^2\{[A_b\tau_{12}+(A_b+A_c)\tau_{23}]/2\hbar\}}. \quad (16)$$

$$I_{01}=c\varepsilon_{01}^2|\overline{W}_1^{(\sigma)}|^2/\pi, \quad \sigma=\pm1,$$

$$\overline{W}_q^{(\sigma)}=\int f(v)W_q^{(\sigma)}dv=|\overline{W}_q^{(\sigma)}|\exp(-i\xi_q^{(\sigma)}), \quad q=\pm1,$$

$$\alpha_1=(\xi_1^{(\sigma)}-\xi_{-1}^{(\sigma)})/2, \quad |\overline{W}_1^{(\sigma)}|=|\overline{W}_{-1}^{(\sigma)}|, \quad (17)$$

where formula (17) defines the phase and modulus of the complex quantity $\overline{W}_q^{(\sigma)}$ which arises after the statistical averaging of $W_q^{(\sigma)}$ at $t=t_e$. If the common level E_b lies above ($E_a<E_c<E_b$) or below ($E_b<E_a<E_c$) the other two levels, then in the preceding formulas (14)–(17) one should make the replacement $\omega_{cb}\to\omega_{bc}$ ($\gamma_{cb}\to\gamma_{bc}$) or $\omega_{ba}\to\omega_{ab}$, respectively, for $\gamma\ll\Omega_a$. In the case $E_a<E_b<E_c$ and $\gamma\gtrsim\Omega_a$, the quantities I_{01} and α_1 have other expressions and depend on H, while Eq. (16) is valid for $\gamma_a\tau_{12}>1$. We stress that the separation in (16) of the characteristic factor that takes the influence of the hyperfine structure into account occurs only in front of the intensity corresponding to the projection of the electric field of the TPE onto the physically preferred axis Y, which is perpendicular to the vectors $\mathbf{H}, \mathbf{l}^{(0)}$, and $\mathbf{k}_{1,2,3}$. This factor stems from the interference of the coherent waves emitted at the atomic transitions $E_{M_bM_I}\to E_{M_cM_I}$ between individual pairs of hyperfine sublevels of the upper E_c and lower E_b levels, with fixed M_c and M_b but different M_I.

Intensity (16) as a function of the delay time $\tau_{12}(\tau_{23})$ and of the modulus of the magnetic field strength H undergoes oscillations (quantum beats) which have an exceptionally simple form independent of the degeneracy and the number of components in the hyperfine structure of the levels. Since the quantities I_{01} and α_1 do not contain τ_{12}, τ_{23}, and H, the quantum beats (16) are universal, independent of the shape, detuning, and spectral composition of the exciting pulses, which are subject to certain fluctuations in an experiment. Furthermore, the Doppler width of the spectral line and the distance between hyperfine and Zeeman components are not related by any kind of condition, so that one can study the distance between hyperfine sublevels located both inside and outside the Doppler width of the spectral line. The only requirement is that the atomic transition $J_b\to J_c$ be inhomogeneously broadened: $k_2u>\gamma_{cb}$.

If intensity (16) is considered as a function of τ_{12} or τ_{23} at fixed **H**, then the quantum beats will be rapid harmonic oscillations with a frequency of $2\Omega_b$ or $2(\Omega_b+\Omega_c)$, respectively, which are superposed on slow oscillations with a frequency of $|A_b|\hbar^{-1}$ or $|A_b+A_c|\hbar^{-1}$, respectively. The rapid harmonic oscillations contain information on the g factors of the resonant levels. Meanwhile, the intensity averaged over the rapid harmonic oscillations, $\langle I_1(t_e)\rangle$, will execute slow, damped oscillations from which one can determine the hyperfine splitting constants. It fact, by making a series of measurements of the averaged intensity $\langle I_1(t_e)\rangle$ with different values of τ_{12} at fixed τ_{23} and **H**, one can easily determine the quantity $|A_b|$ for the common level E_b of the two adjacent atomic transitions (Fig. 1). Here the envelope through the maxima of the slow oscillations is described by the exponential function $\exp(-2\gamma_b\tau_{12})$, making it possible to find γ_b.

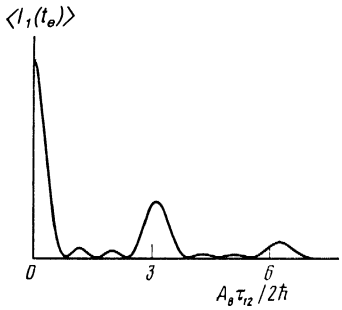

FIG. 1. Averaged intensity of the TPE in arbitrary units. We have considered the transitions $3S_{1/2}\to3P_{1/2}\to4S_{1/2}$ for sodium vapor $_{11}{}^{23}$Na, with nuclear spin $I=3/2$. We have set $A_a/\hbar=5.56\cdot10^9$ sec^{-1}, $A_b/\hbar=0.59\cdot20^9$ sec^{-1}, $A_c/\hbar=1.26\cdot10^9$ sec^{-1} ($A_b+A_c)\tau_{23}/2\hbar=n\pi$ (n is an integer), $\gamma_b=0.6\cdot10^8$ sec^{-1}, $\gamma_{cb}=0.37\cdot10^8$ sec^{-1}, $H\gtrsim10^3$ Oe, and $k_2u\tau_{23}>1$.

Analogously, by varying τ_{23} at fixed τ_{12} and **H**, we find $|A_b+A_c|$, and the envelope for these oscillations is proportional to $\exp(-4\gamma_{cb}\tau_{23})$ and yields the relaxation constant γ_{cb}. For experimental determination of the g factors, one can also use an alternative method in which intensity (16) is studied as a function of H at fixed values of τ_{12} and τ_{23}. In all the cases mentioned, the inequality $k_2u\tau_{23}>1$ must hold in order to eliminate the contribution of the optical free induction arising after the end of the three pulses (9)–(11).

3. PHOTON ECHO IN A STRONG H FIELD

An ultrahigh resolution of the sublevels of the lower level E_a is expediently achieved with the aid of the PE formed in a gas after passage of two exciting pulses (9) and (10) with identical center frequencies ω_1 and ω_2, $\omega_1=\omega_2=\omega(\mathbf{k}_1=\mathbf{k}_2=\mathbf{k})$, which are close to the atomic frequency ω_{ba} of the investigated atomic transition $J_a\to J_b$. The other characteristics of these light pulses remain as before. Suppose the magnetic field $\mathbf{H}\|\mathbf{l}_x$ is strong and the Paschen-Back regime (2) obtains for the lower and upper levels of the atom. The projection of the electric field of the PE onto the Y as is of the form

$$E_y=\varepsilon(t-z/c)\exp[i(kz-\omega t-\varphi_{1e})]+\text{c.c.}, \quad (18)$$

$$\varepsilon(t)=-\frac{i}{2^{1/2}}\frac{\varepsilon_0}{2I+1}\times\sum_{M_I}\int f(v)\exp[i(\Delta-\mathbf{kV})(t-t_{1e})-\gamma_{ba}t]$$

$$\times\{V_1^{(\sigma)}\exp[-i(\Omega_a+\Omega_b+(A_a+A_b)M_I\hbar^{-1})\tau]-(-1)^\sigma V_{-1}^{(\sigma)}$$

$$\times\exp[i(\Omega_a+\Omega_b+(A_a+A_b)M_I\hbar^{-1})\tau]\}d\nu,$$

$$V_q^{(\sigma)}=\sum_\varkappa G_\varkappa^{(\sigma)}T_{\varkappa q}(t-t_{13}),\quad q=\pm1,\quad \sigma=\pm1,$$

$$G_\varkappa^{(\sigma)}=\sum_\mu\begin{pmatrix}J_b & J_a & \varkappa\\ \mu & \sigma-\mu & -\sigma\end{pmatrix}A_{1\mu}^{(\sigma)}B_{1\mu}^{(\sigma)*}, \tag{19}$$

$$T_{\varkappa q}(t)=(2\varkappa+1)d^\varkappa_{\sigma,-q}\left(\frac{\pi}{2}\right)\sum_{\nu\mu}\begin{pmatrix}J_a & J_b & 1\\ \nu & -\mu & q\end{pmatrix}$$

$$\times\begin{pmatrix}J_b & J_a & \varkappa\\ \nu & -\mu & q\end{pmatrix}B_{2\nu}^{(0)}B_{2\mu}^{(0)}\exp\{i[(\Omega_a+A_aM_I\hbar^{-1})\nu$$

$$-(\Omega_b+A_bM_I\hbar^{-1})\mu]t\},$$

$$\varepsilon_0=2\pi kL|d_{ba}|N_{ab},\quad \varphi_{1e}=2\varphi_2-\varphi_1,\quad t_{1e}=2\tau_{12}+\tau_1+\tau_2.$$

We see that the transverse magnetic field **H** causes a precession only of the induced atomic polarization, leading to quantum beats and to a rotation and change of the axes of the polarization ellipse of the PE. Here the projection of the electric field of the PE onto the physically preferred axis Y has the simplest form. As in the case of the TPE, the contributions to amplitude (19) of individual pairs of hyperfine sublevels, corresponding to the selection rule $\Delta M_I=0$, add up as independent. The interference of the waves emitted at individual pairs of atomic transitions between hyperfine sublevels $E_{M_bM_I}$ and $E_{M_aM_I}$ with fixed M_a and M_b but different M_I leads to the characteristic factor

$$\sin^2[(2I+1)(A_a+A_b)\tau_{12}/2\hbar]/\sin^2[(A_a+A_b)\tau_{12}/2\hbar],$$

which reflects the influence of the hyperfine structure of the levels. The precession of the reduced atomic polarization and the irreversible relaxation are taken into account in the amplitude in (19) by other independent factors. The intensity $I(t)=c|\varepsilon(t)|^2/2\pi$ corresponding to the projection of the electric field (18) at the time $t=t_{1e}$ of the PE maximum is given by the formula

$$I(t_{1e})=I_0\exp(-4\gamma_{ba}\tau_{12})\cos^2[(\Omega_a+\Omega_b)\tau_{12}+\alpha]$$

$$\times\frac{\sin^2[(2I+1)(A_a+A_b)\tau_{12}/2\hbar]}{(2I+1)^2\sin^2[(A_a+A_b)\tau_{12}/2\hbar]},$$

$$I_0=c\varepsilon_0^2|\bar V_1^{(\sigma)}|^2/\pi, \tag{20}$$

$$\bar V_q^{(\sigma)}=\int f(v)G_\varkappa^{(\sigma)}T_{\varkappa q}(0)\,d\mathbf{v}=|\bar V_q^{(\sigma)}|\exp(-i\alpha_q^{(\sigma)}),\quad q=\pm1,$$

$$\alpha=(\alpha_1^{(\sigma)}-\alpha_{-1}^{(\sigma)})/2,\quad |\bar V_1^{(\sigma)}|=|\bar V_{-1}^{(\sigma)}|.$$

Because I_0 and α are independent of τ_{12} and H, a study of the rapid harmonic oscillations of intensity (20) as a function of τ_{12} (or H) for fixed **H** (or τ_{12}) makes it possible to determine $|g_a+g_b|$. As a function of τ_{12}, the intensity averaged over these rapid oscillations, $\langle I(t_{1e})\rangle$, executes slow oscillations with a period $|A_a+A_b|\hbar^{-1}$, permitting one to find $|A_a+A_b|$ (Fig. 2). From the values found for $|g_a+g_b|$ and $|A_a+A_b|$, one can form an opinion about $|g_a|$ and $|A_a|$, since $|g_b|$ and $|A_b|$ are known from the previous study of the TPE. We note that the envelope through the maxima of the slow quantum beats is damped by the exponential law $\exp(-4\gamma_{ba}\tau_{12})$, which contains information on the relaxation constant γ_{ba}. The variation of τ_{12} should be done in observance of the condition $ku\tau_{12}>1$, which rules out the contribution of the optical free induction after the end of the two exciting pulses. The possibility of experimental measurement of the desired quantities with the aid of dye lasers can be seen from the graphs in Figs. 1 and 2, which were constructed for actual parameter values taken from published sources.[15]

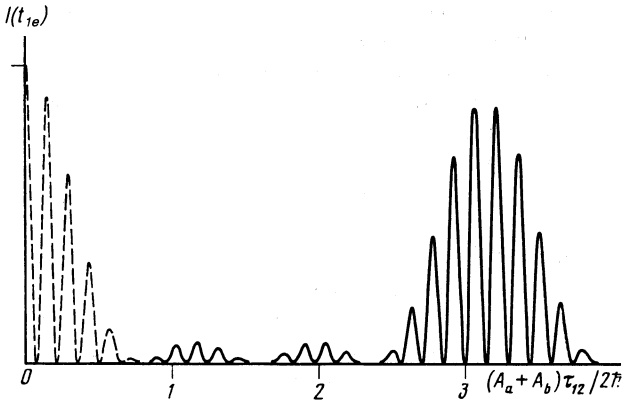

FIG. 2. Intensity of the PE in arbitrary units. We have considered the transition $3S_{1/2}\to3P_{1/2}$ for $_{11}{}^{23}$Na vapor. We have set $H=3.97\cdot10^3$ Oe, $g_a=2$, $g_b=0.665$, $\gamma_{ba}=0.3\cdot10^8$ sec^{-1}. The dashed part of the curve represents the region in which the optical induction can influence the PE.

4. DISCUSSION

Owing to the Paschen-Back regime, the results obtained above remain in force in the presence of depolarizing elastic collisions of the atoms. In a strong magnetic field (2) there is a breaking of the hyperfine coupling, and the electronic and nuclear subsystems therefore behave independently in an individual collision event. The relaxation of the electronic subsystem is described by the standard model of elastic atomic collisions,[9,10,20,21] according to which the damping of the multipole moments of the atom is characterized by the electronic relaxation constants $\gamma_b^{(\varkappa)}$ for $0\leqslant\varkappa\leqslant2J_b$ and $\gamma_{cb}^{(\varkappa)}$ for $|J_c-J_b|\leqslant\varkappa\leqslant J_c+J_b$ ($\gamma_{ba}^{(\varkappa)}$ for $|J_b-J_a|\leqslant\varkappa\leqslant J_b+J_a$). For different indices $\varkappa$ they vary by 10–40% in the case of $\gamma_b^{(\varkappa)}$ and by 10–30% for $\gamma_{cb}^{(\varkappa)}$ and $\gamma_{ba}^{(\varkappa)}$. Consequently, in the presence of elastic atomic collisions the formulas given above remain valid, after the substitutions $\gamma_b\to\gamma_b^{(1)}$, $\gamma_{cb}\to\gamma_{cb}^{(1)}$ and $\gamma_{ba}\to\gamma_{ba}^{(1)}$, in the time interval

$$|\gamma_b^{(1)}-\gamma_b^{(\varkappa)}|t\ll1,\quad |\gamma_{cb}^{(1)}-\gamma_{cb}^{(\varkappa)}|t\ll1,\quad |\gamma_{ba}^{(1)}-\gamma_{ba}^{(\varkappa)}|t\ll1,$$

here, for angular momenta J_a, J_b, and J_c which assume values from the set of numbers 0,0,1 (0,1,1) and 1/2, 1/2, 3/2, the relations $\gamma_{cb}^{(\varkappa)}=\gamma_{cb}^{(1)}$ and $\gamma_{ba}^{(\varkappa)}=\gamma_{ba}^{(1)}$ are satisfied,[9] and therefore the second and third inequalities drop out.

The method proposed above for determining the optical characteristics is based on the high senstivity of a polarized

atom to the effect of a magnetic field **H** and to the occurence of relaxation processes which depolarize the initial orientation of the atom. In this sense it is analogous to the various other methods of optical orientation using the broadening of the Hanle signal[6,7] the depolarization of the fluorescence,[8–10] level crossings,[11,12] the rf-optical double resonance,[12,13] and the quantum beats in the spontaneous emission.[14] However, the conventional methods[6–14] apply when one is studying the damping of the precession of the magnetic moment in a weak **H** field and the behavior of the density matrix of an atom at an individual level E_b on the basis of incoherent spontaneous emission, which enables one to find γ_b ($\gamma_b^{(x)}$) for a known value of the g factor g_b, and also to obtain certain information regarding the hyperfine splitting constant A_b. Here the most complete information on these quantities can evidently be obtained in the case of incoherent spontaneous emission during fluorescence in a strong magnetic field,[22] but this problem requires a separate treatment. Meanwhile, in the method proposed here, which uses coherent radiation, one studies the precession of the induced atomic polarization in a strong **H** field in the Paschen-Back regime and the relaxation of the optical coherence, which is described by a density matrix which is nondiagonal in the levels E_c and E_b or E_b and E_a, making it possible to determine independently the g factors of the levels, the hyperfine splitting constants, and the relaxation constants γ_{cb} ($\gamma_{cb}^{(x)}$) or γ_{ba} ($\gamma_{ba}^{(x)}$). In certain cases one can also determine γ_b ($\gamma_b^{(x)}$), when the precession of the magnetic moment contributes to the intensity of the coherent radiation. Thus the approach proposed above can be regarded as a method of optical orientation using coherent radiation in a strong magnetic field.

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