Theory of photon echo produced at resonant levels with hyperfine structure

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The intensity and polarization of a photon echo produced at resonant levels with hyperfine structure are found. The relations obtained are applicable to optically allowed transitions with arbitrary values of the electron angular momenta of the resonant levels. Principal attention is paid to transitions between the ground and excited states in atoms of the alkali metals, since it is precisely these atoms that have thus far been investigated experimentally. The polarization properties of a photon echo produced in an alkali-metal vapor are found to depend essentially on the structure of the resonant transition, i.e., on whether all or only one of the hyperfine components of each of the resonant levels participate in its production. This fact is important for the correct analysis of the experimental data when the purpose of the analysis is to extract information about the homogeneous width of the spectral line of an inhomogeneously broadened resonant transition. The pre-exponential factors determining the nonmonotonic dependence of the photon-echo signal on the time interval between the exciting pulses are computed. The results of the calculation are used to discuss the experiments.

The photon-echo method is at present one of the effective methods of nonlinear laser spectroscopy. The photon echo is used here to investigate both solid and gaseous media. In particular, an increasing number of experiments have over the past few years been devoted to the investigation of alkali-metal vapors (see, for example, Refs. 1–5). In all these experiments the spectral width of the exciting light pulses, was, as a rule, greater than, or of the order of, the hyperfine splitting of one or both of the resonant levels, so that the photon echo was produced on the entire set of transitions between the hyperfine components of the resonant levels.

Let us note that the photon echo produced at resonant levels which are a set of split sublevels has been theoretically investigated by Lambert *et al.*[6] They, however, neglect the polarization properties of the electromagnetic field. This limits substantially the region of applicability of the results, since the use of exciting pulses of different polarizations uncovers, as is well known, new capabilities of the photon-echo method and leads in a number of cases to a significant simplification of the experimental scheme. Let us emphasize that all the theoretical investigations of the polarization properties of the photon echo pertain to systems whose energy spectrum does not possess a hyperfine structure.

In the present paper we construct a consistent theory of the production of a photon echo in gaseous media at resonant levels with a hyperfine structure. As an example, we consider in detail the case of photon-echo production in alkali-metal vapors. The results of the calculation are used to discuss the experiments that have already been performed. It is shown that the theory developed allows us to extract more information from the experimental data.

1. THE BASIC EQUATIONS AND RELATIONS

As is well known (see, for example, Ref. 7), the interaction of the magnetic and quadrupole moments of a nucleus with the electron shell leads to the hyperfine splitting of levels with electron angular momenta J_a and J_b into a number of components each of which corresponds to a definite value of the total angular momentum F_a or F_b of the atom. Here $F_a = J_a + I, \ldots, |J_a - I|$ and $F_b = J_b + 1, \ldots, |J_b - 1|$, where I is the spin of the nucleus of the atom. Since the energy characterizing the interaction of the magnetic and quadrupole moments of the nucleus with the electron shell is always sufficiently small compared to the difference between the energies of the unperturbed levels, the splitting of each of the levels can be considered independently of the rest. In this approximation the quantities J, I, F, and M are good quantum numbers. Here M is the component of the total angular momentum of the atom along the axis of quantization. Thus, for the lower, a, and upper, b, resonance energy levels we have

$$E_{J_aIF_a}=E_a+\Delta E_{F_a}, \qquad E_{J_bIF_b}=E_b+\Delta E_{F_b}.$$

Here E_a and E_b are their energies when the hyperfine interaction is neglected and the quantities ΔE_{F_a} and ΔE_{F_b} depend on the quantum numbers F_a and F_b and are small compared to $E_b - E_a = \hbar\omega_0$.

To determine the intensity of the electric field of the photon echo, let us use the d'Alembert equation

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

and the quantum-mechanical equation for the density matrix $\hat{\rho}$ of the resonant atoms:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\hat{\mathscr{T}}\right)\hat{\rho}=\frac{i}{\hbar}[\hat{\rho}, \hat{H}_0-\hat{\mathbf{d}}\mathbf{E}]. \tag{2}$$

Here $\mathbf{E}$ is the electric field intensity in the medium, $\hat{H}_0$ is the Hamiltonian of the atom in its c.m.s., $\hat{\mathbf{d}}$ is the dipole-moment operator for the atom, and $\hat{\mathscr{T}}$ is the relaxation matrix that takes into account the radiative decay of the resonant levels, the elastic and inelastic collisions, as well as the radiative transition to the lower resonant level as a result of spontaneous emission at the upper level. The medium-polarization vector $\mathbf{P}$ in (1), which pertains to a group of atoms moving

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with velocity $\mathbf{v}$, is connected with the density matrix $\hat{\rho}$ by the relation

$$\mathbf{P}=\mathrm{Sp}(\hat{\rho}\hat{\mathbf{d}}). \quad (3)$$

Below we shall dwell on only the relaxation of the optical-coherence matrix, since it is precisely this relaxation that leads to the decrease of the amplitude of the photon echo.[8] In the model of elastic depolarizing collisions (see, for example, Refs. 8 and 9), the relaxation of the various multipole moments of the resonant electron transition $b \to a$ occurs independently, and is described by the relaxation constants $\gamma^{(\varkappa)} - i\Delta^{(\varkappa)}$ $(|J_a - J_b| \leqslant \varkappa \leqslant J_a + J_b)$. Here $\gamma^{(\varkappa)} = (\gamma_a^{(0)} + \gamma_b^{(0)})/2 + \Gamma^{(\varkappa)}$, $\gamma_a^{(0)}$ and $\gamma_b^{(0)}$ are the reciprocal lifetimes of the levels a and b, as determined by the radiative decay and the inelastic gas-kinetic collisions, and the real quantities $\Gamma^{(\varkappa)}$ and $\Delta^{(\varkappa)}$ take account of the effect of the elastic depolarizing collisions.

In the case when the resonant levels possess a hyperfine structure, the depolarizing collisions lead to the transfer of coherence from one pair of the hyperfine components of the levels a and b to another. The probability per unit time of such processes is determined by the difference $\gamma^{(\varkappa)} - \gamma^{(1)}$ $(\varkappa \neq 1)$. We note that the explicit form of the collision integral describing the relaxation of a hyperfine super-multiplet has been written down and investigated by Rabane[9] in the model that assumes the severance of the hyperfine coupling in the interaction process.

The subsequent computations are carried out under the assumption that the following inequalities obtain:

$$|\gamma^{(\varkappa)}-\gamma^{(1)}|\tau<1, \quad |\Delta^{(\varkappa)}-\Delta^{(1)}|\tau<1. \quad (4)$$

Here τ is the time interval between the exciting light pulses. In this approximation the relaxation of the optical-coherence matrix is described by the expression

$$\langle F_aM_a|\hat{\mathscr{T}}\rho|F_bM_b\rangle=(\gamma^{(1)}-i\Delta^{(1)})\langle F_aM_a|\hat{\rho}|F_bM_b\rangle. \quad (5)$$

Here $\gamma^{(1)}$ is the homogeneous halfwidth of the spectral line and $\Delta^{(1)}$ is the line shift due to the elastic depolarizing collisions. Let us emphasize that, in the case of transitions accompanied by changes $J_b = 1/2 \to J_a = 1/2$ and $J_b = 3/2 \to J_a = 1/2$ in the total electron angular momentum, the relaxation characteristics $\gamma^{(\varkappa)}$ and $\Delta^{(\varkappa)}$ respectively coincide at all possible values of $\varkappa$ with $\gamma^{(1)}$ and $\Delta^{(1)}$ (see, for example, Ref. 9). Thus, the inequalities (4) and the expression (5) are valid for these transitions at any value of τ, and it is precisely at such transitions that all the published experiments on the photon echo in alkali-metal vapors were performed.

Let exciting light pulses linearly polarized in different planes be incident on the boundary $z = 0$ of a gaseous medium respectively at the moments of time $t = 0$ and $t = \tau + T_1$ and propagate in the positive direction of the Z axis. Let us write the intensities of the electric fields of the exciting pulses in the form

$$\mathbf{E}_1=e^{(1)}(\mathbf{l}_x\cos\psi+\mathbf{l}_y\sin\psi)g_1(\zeta)\exp[i(\omega t-kz+\Phi_1)]+\text{c.c.}, \quad 0\leqslant\zeta\leqslant T_1, \quad (6)$$

$$\mathbf{E}_2=e^{(2)}\mathbf{l}_x g_2(\zeta-\tau-T_1)\exp[i(\omega t-kz+\Phi_2)]+\text{c.c.}, \quad \tau+T_1\leqslant\zeta\leqslant\tau+T_1+T_2. \quad (7)$$

Here the amplitudes $e^{(1)}$ and $e^{(2)}$, as well as the phase shifts Φ_1 and Φ_2, are constants; T_1 and T_2 are the durations of the exciting pulses; ω is the carrier frequency of these pulses; $\zeta = t - z/c$; $\mathbf{l}_x$ and $\mathbf{l}_y$ are the unit vectors of the corresponding Cartesian axes; and the functions g_1 and g_2 describe the shape of the exciting pulses, with

$$\frac{1}{T_l}\int_0^{T_l} g_l(s)\,ds=1, \quad l=1,2.$$

The spectral widths δ_1 and δ_2 of the exciting pulses are determined by the form of the functions g_1 and g_2. Thus, for rectangular exciting pulses (for which $g_1 = g_2 = 1$), we have $\delta_{1,2} = 1/T_{1,2}$. As follows from (6) and (7), the X axis is oriented along the polarization vector of the second exciting pulse, and the polarization vector of the first exciting pulse forms an angle ψ with the polarization vector of the second pulse.

At zero time $\zeta = t - z/c = 0$, prior to the arrival of the first exciting pulse at the point z of the gaseous medium, the density matrix of the resonant atoms has the form

$$\hat{\rho}(\zeta=0)=f(\mathbf{v})\sum(n_a|F_aM_a\rangle\langle M_aF_a|+n_b|F_bM_b\rangle\langle M_bF_b|). \quad (8)$$

Here the summation is over all the possible values of F_a, F_b, M_a, and M_b; n_b and n_a are the population densities of the Zeeman sublevels of the hyperfine components of the resonant levels b and a at $\zeta \leqslant 0$; $f(\mathbf{v})$ describes the Maxwellian velocity ($\mathbf{v}$) distribution of the atoms.

The system (1)–(3) with the initial condition (8) was solved in the given-field approximation (6) and (7), i.e., without allowance for the reaction of the resonant medium on the field of the exciting pulses. This approximation is usually entirely admissible for experiments on the photon echo in gases. Further, as is usually done in the investigation of the photon echo in gases, we have assumed that the durations T_l $(l = 1, 2)$ of the exciting pulses are short compared to the irreversible relaxation times and the time interval τ between the pulses. Furthermore, a necessary condition for the observation of the photon echo in gases is inhomogeneity of the broadening of the resonant spectral line and shortness of the reversible Doppler relaxation time T_0 compared with the time interval τ between the exciting pulses. Here T_0 is connected with the mean thermal velocity u of the resonant atoms by the relation $T_0 = 1/ku$, where $k = \omega/c$ is the modulus of the wave vector of the exciting pulses.

The problem of finding the intensity of the electric field of the photon echo has been solved for exciting pulses of small area,[8] which are more suitable for the polarization spectroscopy of gaseous media. The calculations carried out yield for the intensity of the electric field of the photon echo the expression

$$\mathbf{E}^e=-\frac{\pi}{\hbar^3}\omega\frac{L}{c}|d|^4e^{(1)}T_1e^{(2)2}T_2^2N_0I_e\mathbf{e}^e \times\exp[i(\omega t-kz+2\Phi_2-\Phi_1)-i\Delta\omega(t'-\tau)-\gamma^{(1)}(t'+\tau)]+\text{c.c.}, \quad (9)$$

where $t' = t - \tau - T_1 - T_2 - z/c$, $\Delta\omega = \omega - \omega_0 - \Delta^{(1)}$, L is the length of the gaseous medium, $N_0 = n_b - n_a$, and d is the reduced matrix element of the dipole-moment operator for the transition $b \to a$. Further, the real quantity I_e, which characterizes the shape of the photon-echo pulse, is given by the relation

$$I_e = \int a_1(v)[a_2^*(v)]^2 \exp[ikv(t'-\tau-{}^1/_2T_1)]f(v)\,dv, \quad (10)$$

where

$$a_l(v) = \frac{1}{T_l}\int_{-T_l/2}^{T_l/2} g_l(s+T_l/2)\exp(ikvs)\,ds, \qquad l=1,2,$$

and $f(v)$ is the Maxwellian distribution over the components v of the velocity of the atoms along the Z axis. Finally, the vector $\mathbf{e}^e$, which characterizes the polarization properties of the photon echo, has the form

$$\mathbf{e}^e = {}^2/_3(c_0+2c_2)\cos\psi\mathbf{l}_x + (c_2-c_1)\sin\psi\mathbf{l}_y, \quad (11)$$

where

$$c_\varkappa = \sum(-1)^{F_b'-F_b}\begin{Bmatrix}\varkappa & 1 & 1\\ F_b & F_a & F_a'\end{Bmatrix}\begin{Bmatrix}\varkappa & 1 & 1\\ F_b' & F_a & F_a'\end{Bmatrix} G_{F_bF_a} \times G_{F_b'F_a}G_{F_b'F_a'}G_{F_bF_a'}\exp[i(\Delta_{F_bF_a'}t' - \Delta_{F_b'F_a'}\tau)]. \quad (12)$$

Here the quantities $G_{F_aF_b}$, which are proportional to the coefficients that interrelate the matrix elements of the dipole moment operator in the F, M and J, M_J representations, are given by

$$G_{F_bF_a} = [(2F_a+1)(2F_b+1)]^{1/2}\begin{Bmatrix} I & F_a & J_a\\ 1 & J_b & F_b\end{Bmatrix},$$

where the $6j$ symbols are designated in the usual manner, while

$$\Delta_{F_iF_j'} = (\Delta E_{F_i} - \Delta E_{F_j'})/\hbar, \quad i,j=a,b.$$

The summation in (12) is over all the values of the total angular momenta of the hyperfine components of the resonant-levels that participate in the production of the photon echo.

In the general case the photon echo (9)–(12) is elliptically polarized, and propagates along the Z axis with carrier frequency ω.

As follows from (10), when the photon echo is produced on a spectral line that is narrow relative to both exciting pulses (i.e., for which $T_0 \gg T_l$, $l=1, 2$), the peak of the echo pulse occurs at the instant $t = 2\tau + z/c$, and the duration of the pulse is of the order of T_0. When the photon echo is produced on a spectral line that is broad relative to both exciting pulses ($T_0 \ll T_l$), the peak of the echo pulse is shifted relative to the instant $t = 2\tau + z/c$ by a period of the order of the total duration of the exciting pulses, while the duration of the pulse turns out to be of the order of the longer of the two exciting-pulse durations.

Let us emphasize that the polarization properties of the photon echo (9)–(12) do not depend on either the areas or the shapes of the exciting pulses, nor do they depend on the type of spectral line (wide or narrow) on which the echo is produced. The polarization dependence of the photon echo (9)–(12) turned out to be simple and suitable for the processing of experimental data.

2. DISCUSSION OF THE RESULTS

An important role is played in the production of the photon echo at resonant levels with hyperfine structures by the relations between the hyperfine splittings of the resonant levels, the Doppler width of the spectral line, and the spectral width of the exciting pulses. In the present paper we consider the following three possible cases: $F_b \to F_a$, $J_b \to F_a$, and $J_b \to J_a$.

1. The case $F_b \to F_a$ occurs when the photon echo is produced on only one pair of hyperfine components of the resonant levels characterized by the total-angular-momentum values F_b and F_a. It is realized when the hyperfine splittings of both the upper and lower resonant levels are large compared with the Doppler width of the spectral line and the spectral width of the exciting light pulses, and the carrier frequency ω of the exciting pulses is close to the $F_b \to F_a$ transition frequency, i.e., $|\Delta\omega - \Delta_{F_bF_a}| < \delta_l$ ($l = 1, 2$). It is assumed that the differences in the frequencies of the transitions between the various components of the hyperfine structures of the upper and lower resonant levels are large compared with the spectral width of the exciting pulses.

In the case under consideration only the term with $F_a' = F_a$ and $F_b' = F_b$ remains in the sum (12), and the photon echo is linearly polarized irrespective of the value of the parameter $|\Delta\omega - \Delta_{F_bF_a}|\tau$. For the angle φ_e between photon-echo polarization vector and the polarization vector of the second exciting pulse, which is determined from the equation

$$\operatorname{tg}\varphi_e = e_y^e / e_x^e,$$

we have

$$\operatorname{tg}\varphi_e = {}^3/_2(a_2-a_1)(a_0+2a_2)^{-1}\operatorname{tg}\psi, \quad (13)$$

where

$$a_\varkappa = \begin{Bmatrix}\varkappa & 1 & 1\\ F_b & F_a & F_a\end{Bmatrix}^2. \quad (14)$$

As follows from (13) and (14),

$$\operatorname{tg}\varphi_e = (F-1)(F+2)(3F^2+3F-1)^{-1}\operatorname{tg}\psi \quad (15)$$

for the $F \to F$ transitions and

$$\operatorname{tg}\varphi_e = -2F(F+2)(4F^2+8F+5)^{-1}\operatorname{tg}\psi \quad (16)$$

for the $F \leftrightarrow F+1$ transitions. Thus, for the $F \to F$ transitions the photon-echo polarization vector can have one of three orientations: it can lie outside the angle ψ ($F = 1/2$), or coincide with the polarization vector of the second exciting pulse ($F = 1$), or lie within the angle ψ. The polarization vector of the photon echo produced at the transitions $F \leftrightarrow F+1$ either coincides with the polarization vector of the second exciting pulse ($F = 0$), or lies outside the angle ψ. Let us note that, in the case of relatively small values of F ($F < 10$), Eqs. (15) and (16) can be used to determine F from the experimentally determined relation $\varphi_e = \varphi_e(\psi)$. Consequently, in the $F_b \to F_a$ case the formulas (15) and (16) can be used to identify the resonant transition at which the photon echo is produced.

2. The second possible case $J_b \to F_a$ is realized when the photon echo is produced at transitions between one hyperfine component of the lower, and all the hyperfine compo-

nents of the upper resonant level that are allowed by the selection rules. It occurs when the hyperfine splittings of the lower resonant level are large compared with the Doppler width of the spectral line and the spectral width of the exciting pulses, while the hyperfine splittings of the upper resonant level are small compared with the spectral width of the exciting pulses. Here it is assumed that the frequencies of the transitions between the given component of the hyperfine structure of the lower resonant level and all the components of the hyperfine structure of the upper are sufficiently close to the carrier frequency ω of the exciting pulses, while the frequencies of the transitions between each of the remaining components of the hyperfine structure of the lower level and all the components of the hyperfine structure of the upper level are sufficiently far from the carrier frequency of the exciting pulses. Notice that the present case can correspond fully to the experimental situation, since the hyperfine splitting of the ground state of atoms is significantly greater than the hyperfine splitting of the excited states,[7,10] and it is precisely on such a system of resonant levels that the photon echo is produced in experiments on alkali-metal vapors.

The intensity of the electric field of the photon echo is then again given by Eqs. (9)–(12), in which the summation in (12) must now be performed over all possible values of F_b and F_b' for $F_a'=F_a$. For sufficiently small distances between the components of the hyperfine structure of the upper resonant level, i.e., distances such that

$$\max\{|\Delta_{F_bF_b'}|\}\tau<1, \tag{17}$$

the photon echo is linearly polarized. The angle φ_e between photon-echo polarization vector and the polarization vector of the second exciting pulse can be determined from Eq. (13), in which $a_\varkappa$ now has the form

$$a_\varkappa=\begin{Bmatrix} \varkappa & 1 & 1 \\ J_b & J_a & J_a \end{Bmatrix}^2 \begin{Bmatrix} \varkappa & J_a & J_a \\ I & F_a & F_a \end{Bmatrix}^2. \tag{18}$$

In the case of greater spacing between the components of the hyperfine structure of the upper resonant level, such that

$$\max\{|\Delta_{F_bF_b'}|\}\tau\gtrsim1, \tag{19}$$

the photon-echo polarization is, in general, elliptical. The echo intensity as a function of the time interval τ between the exciting pulses takes the form of beats superimposed on a damping proportional to $\exp(-4\gamma^{(1)}\tau)$. This effect limits the applicability of the traditional procedure for processing experimental data, in which the entire τ dependence is assumed to be connected with the factor $\exp(-4\gamma^{(1)}\tau)$. Apparently, it is precisely because of this that researchers usually limit themselves in experiments on the photon echo in alkali-metal vapors to the determination of the quantity $d\gamma^{(1)}/dp$ (where p is the pressure of the gas) rather than $\gamma^{(1)}$ itself. The latter would require knowledge of the pre-exponential factors, which also depend on τ. With the aid of the formulas obtained in the present paper we can compute these pre-exponential factors for any system and, consequently, use them to process experimental data.

Notice that, as follows from the results under certain conditions the photon-echo pulse contains modulation oscillations and oscillations of the ratio of the axes of the polarization ellipse.

To illustrate the indicated relationships, let us consider as an example the $3P_{1/2}\to3S_{1/2}$ transition in ^{23}Na, a transition widely used in photon-echo experiments (see, for example, Refs. 4 and 5). The spin of the nucleus of the ^{23}Na atom is $I=3/2$. The intercomponent spacing is $\Delta_a=1.77$ GHz for the $F_a=2$ and $F_a=1$ doublet of the $3S_{1/2}$ level and $\Delta_b=0.188$ GHz for the $F_b=2$ and $F_b=1$ doublet of the $3P_{1/2}$ level.[10] If the photon echo is produced at transitions between one component of the lower $3S_{1/2}$ level and both components of the upper $3P_{1/2}$ level, then from (11) and (12) we obtain for the nonzero components of the vector $\mathbf{e}^e$ when

$$\hbar(\Delta\omega-{}^1/_2\Delta_b)=\Delta E_{F_b=1}-\Delta E_{F_a}$$

the expressions

$$e_x^e={}^1/_{72}\cos\psi\,\{3\cos[{}^1/_2\Delta_b(t'+\tau)]+9\cos[{}^1/_2\Delta_b(t'-\tau)]+8i\sin[{}^1/_2\Delta_b(t'-\tau)]\}\exp[i\Delta\omega(t'-\tau)], \tag{20}$$

$$e_y^e=-{}^1/_{72}\sin\psi\{3\cos[{}^1/_2\Delta_b(t'-\tau)]-\cos[{}^1/_2\Delta_b(t'+\tau)]+3i\sin[{}^1/_2\Delta_b(t'-\tau)]\}\exp[i\Delta\omega(t'-\tau)] \tag{21}$$

in the case of the $3P_{1/2}\to F_a=1$ transition and

$$e_x^e={}^1/_{72}\cos\psi\{3\cos[{}^1/_2\Delta_b(t'+\tau)]+17\cos[{}^1/_2\Delta_b(t'-\tau)]\}\times\exp[i\Delta\omega(t'-\tau)], \tag{22}$$

$$e_y^e=-{}^1/_{72}\sin\psi\{9\cos[{}^1/_2\Delta_b(t'+\tau)]+\cos[{}^1/_2\Delta_b(t'-\tau)]-5i\sin[{}^1/_2\Delta_b(t'-\tau)]\}\exp[i\Delta\omega(t'-\tau)] \tag{23}$$

in the case of the $3P_{1/2}\to F_a=2$ transition.

As follows from (20)–(23), in the general case the polarization of the echo is elliptical. If the spectral line on which the echo is produced is narrow, and $\Delta_b\gtrsim ku$, modulation oscillations with frequency Δ_b occur inside the echo pulse, as well as oscillations of the ratio of the axes of the polarization ellipse. If $\Delta_b<ku$, or if the spectral line is broad, these oscillations do not occur, and the echo is linearly polarized. We have then from (20)–(23)

$$e_x^e={}^1/_{72}\cos\psi(9+3\cos\Delta_b\tau),\quad e_y^e=-{}^1/_{72}\sin\psi(3-\cos\Delta_b\tau) \tag{24}$$

for the $3P_{1/2}\to F_a=1$ transition, and at $3P_{1/2}\to F_a=2$ we have

$$e_x^e={}^1/_{72}\cos\psi(17+3\cos\Delta_b\tau),\quad e_y^e=-{}^1/_{72}\sin\psi(1+9\cos\Delta_b\tau). \tag{25}$$

It follows from (24) and (25) that beats with frequency $2\Delta_b$ appear in the echo intensity as a function of the period τ and that the angle φ_e between the echo polarization vector and the polarization vector of the second exciting pulse depends on τ in the following manner:

$$\operatorname{tg}\varphi_e=-{}^1/_3(3-\cos\Delta_b\tau)(3+\cos\Delta_b\tau)^{-1}\operatorname{tg}\psi \tag{26}$$

in the case of the $3P_{1/2}\to F_a=1$ transition and

$$\operatorname{tg}\varphi_e=-(1+9\cos\Delta_b\tau)(17+3\cos\Delta_b\tau)^{-1}\operatorname{tg}\psi \tag{27}$$

in the case of the $3P_{1/2}\to F_a=2$ transition. Thus, for the $3P_{1/2}\to F_a=1$ transition the echo polarization vector always lies outside the angle ψ between the polarization vectors of the exciting pulses, while for the $3P_{1/2}\to F_a=2$ transition it can lie either outside or inside the angle ψ, depending on τ. Finally, for $\Delta_b\tau<1$, i.e., in the case when the inequality (17) holds, Eqs. (26) and (27) assume the simple form

$$\operatorname{tg}\varphi_e=-{}^1/_6\operatorname{tg}\psi \qquad (28)$$

in the case of the $3P_{1/2}\to F_a=1$ transition and

$$\operatorname{tg}\varphi_e=-{}^1/_2\operatorname{tg}\psi \qquad (29)$$

in the case of the $3P_{1/2}\to F_a=2$ transition. We emphasize that the formulas (28) and (29) can also be easily obtained directly from (13) and (18).

3. The third possible case $J_b\to J_a$ occurs when all the hyperfine components of both the lower and the upper resonant level participate in the production of the echo. This case is realized when the hyperfine splittings of the resonance levels are small compared with the spectral width of the exciting pulses and the carrier frequency ω of the exciting pulses is close to the resonant transition frequency ω_0 if the hyperfine interaction is neglected, i.e., when $|\Delta\omega|<\delta_l$ ($l=1,2$). Then the intensity of the electric field of the photon echo is again given by (9)–(12), in which the summation is, however, over all the possible values of F_a, F'_a, F_b, and F'_b. For sufficiently small intercomponent spacing in the hyperfine structure of the lower resonant level, such that

$$\max\{|\Delta_{F_aF_a'}|\}\tau<1, \qquad (30)$$

the photon echo in the $J_b\to J_a$ case is linearly polarized. The angle φ_e between the photon-echo polarization vector and the polarization vector of the second exciting pulse can be determined from Eq. (13), in which $a_\varkappa$ now has the form

$$a_\varkappa=\begin{Bmatrix}\varkappa & 1 & 1\\ J_b & J_a & J_a\end{Bmatrix}^2. \qquad (31)$$

We emphasize that (13) and (31) coincide with the corresponding formulas of Ref. 8, which are derived without allowance for the hyperfine structure of the resonance levels. Thus, when the photon echo is produced at transitions between all the hyperfine components of the lower resonant level and all the hyperfine components of the upper, the effect of the hyperfine structure of the resonant levels on the polarization properties of the photon echo is no way manifested when the inequalities (4) and (30) are satisfied. Notice that the formulas (13) and (31) can be used to identify the type of resonance transition ($J\to J$ or $J\rightleftarrows J+1$) when $J\gg1$. Such an identification is carried out in Ref. 8 for the experimental data obtained by Alimpiev and Karlov[11] and Vasilenko and Rubtsova[12] through direct processing of the data with the aid of the formulas obtained in that paper.

For greater intercomponent spacing in the hyperfine structure of the lower resonant level, such that

$$\max\{|\Delta_{F_aF_a'}|\}\tau\geqslant1, \qquad (32)$$

the photon-echo polarization is, in the general case, elliptical. The echo intensity as a function of the time interval τ between the exciting light pulses contains beats. Under certain conditions there also occur modulation oscillations inside the photon-echo pulse and oscillations of the ratio of the axes of the polarization ellipse.

To illustrate these assertions, let us again consider the $3P_{1/2}\to3S_{1/2}$ transition in ^{23}Na. If the photon echo is produced at the transitions between both components of the hyperfine structure of the upper $3P_{1/2}$, and both components of the hyperfine structure of the lower $3S_{1/2}$, resonant level, we obtain from (11) and (12) for the nonzero components of the vector $\mathbf{e}^e$ in the case

$$\hbar(\Delta\omega+{}^1/_2\Delta_a-{}^1/_2\Delta_b)=\Delta E_{F_b=1}-\Delta E_{F_a=1}, \qquad (33)$$

when the echo is produced on a narrow spectral line and the conditions $\Delta_b\tau<1$ and $\Delta_a\gtrsim ku$ are fulfilled, the expressions

$$e_x^e={}^1/_9\cos\psi\{4\cos[{}^1/_2\Delta_a(t'-\tau)]-i\sin[{}^1/_2\Delta_a(t'-\tau)]\}\times\exp[i\Delta\omega(t'-\tau)], \qquad (34)$$

$$e_y^e=-{}^1/_{18}\sin\psi\{5\cos[{}^1/_2\Delta_a(t'+\tau)]+3\cos[{}^1/_2\Delta_a(t'-\tau)]-2i\sin[{}^1/_2\Delta_a(t'-\tau)]\}\exp[i\Delta\omega(t'-\tau)]. \qquad (35)$$

As follows from (34) and (35), in the present case there occur inside the photon-echo pulse modulation oscillations and oscillations of the ratio of the axes of the polarization ellipse with frequency Δ_a. We emphasize that the conditions under which the formulas (34) and (35) are valid correspond to the parameters of the experiment of Nakatsuka *et al.*,[5] in which modulation oscillations with frequency Δ_a were observed inside the photon-echo pulse.

Let us now discuss the instant $t'=\tau$ corresponding to the time of the photon-echo intensity peak when the echo is produced on a narrow spectral line and of the entire photon-echo pulse when it is produced on a broad spectral line. In the case of the $3P_{1/2}\to3S_{1/2}$ transition in ^{23}Na we obtain for the nonzero components of the vector $\mathbf{e}^e$ from (11) and (12) the expressions

$$e_x^e={}^1/_{36}\cos\psi(13+3\cos\Delta_a\tau+3\cos\Delta_b\tau-3\cos\Delta_a\tau\cos\Delta_b\tau), \qquad (36)$$

$$e_y^e=-{}^1/_{18}\sin\psi(1+2\cos\Delta_a\tau+2\cos\Delta_b\tau+3\cos\Delta_a\tau\cos\Delta_b\tau). \qquad (37)$$

Thus, the photon echo (9), (10), (36), and (37) is linearly polarized, and its intensity beats with frequency $2\Delta_a$, $2\Delta_b$, $\Delta_a-\Delta_b$, or $\Delta_a+\Delta_b$, depending on the time interval τ. The angle φ_e between the echo polarization vector and that of the second exciting pulse is τ dependent, and can lie either within or outside the angle ψ between the polarization vectors of the exciting pulses. When $\Delta_a\tau<1$, i.e., when the condition (30) is fulfilled, the angle φ_e always lies outside the angle ψ, since, as follows from (36) and (37),

$$\operatorname{tg}\varphi_e=-\operatorname{tg}\psi. \qquad (38)$$

Notice that the formula (38) can easily be derived from (13) and (31).

3. POSSIBILITY OF EXPERIMENTAL IDENTIFICATION OF THE STRUCTURE OF RESONANT TRANSITIONS

The photon-echo polarization properties found in the present paper enable us to determine which of the considered cases of photon-echo production at resonant levels possessing hyperfine structure is realized in a given experiment. We emphasize that the simplest of the formulas obtained correspond to the case (30) in which the condition (17) is automatically fulfilled as well. Therefore we should, in setting up experiments aimed at determining the structure of resonant transitions by the photon-echo method, take the corresponding limitations into consideration. Notice that even when the inequality (30) is violated the formulas obtained in the paper still enable us to determine the structure of the resonant transition.

As an example, let us again consider the $3P_{1/2}\to 3S_{1/2}$ transition in ^{23}Na in the case $\Delta_a \tau < 1$. If the photon echo is produced at the transtions between both components of the hyperfine structure of the lower resonant level and both components of the hyperfine structure of the upper, the angle φ_e can be found from Eq. (38). Further, if the photon echo is produced at transitions between one hyperfine component of the lower resonant level, and one hyperfine component of the upper, we find from (15) and (16) that

$$\operatorname{tg} \varphi_e = 0, \quad (39)$$

for the $F_b = 1 \to F_a = 1$ transition,

$$\operatorname{tg} \varphi_e = {}^4/_{17} \operatorname{tg} \psi, \quad (40)$$

for the $F_b = 2 \to F_a = 2$ transition, and

$$\operatorname{tg} \varphi_e = -{}^6/_{17} \operatorname{tg} \psi \quad (41)$$

for the $F_b = 2 \to F_a = 1$ and $F_b = 1 \to F_a = 2$ transitions.

Finally, if the photon echo is produced at transitions between one hyperfine component of the lower $3S_{1/2}$ level and both hyperfine components of the upper $3P_{1/2}$ level, the angle φ_e can be found from Eqs. (28) and (29).

Thus, the formulas (28), (29), and (38)–(41) allow us to determine the structure of the transition at which the photon echo is produced in a given experiment in the case of the $3P_{1/2}\to 3S_{1/2}$ transition in ^{23}Na, since they lead to entirely different polarization properties of the photon echo. Let us emphasize that the range of validity of (28), (29), and (39)–(41) is broader than $\Delta_a \tau < 1$.

Formulas similar to (28), (29), and (38)–(41) can be written down for other transitions in sodium, as well as for the transitions between the energy levels of other atoms possessing hyperfine structure.

In conclusion, let us note that the entire analysis in the present paper has been performed in the limit, most suitable for the purposes of the polarization spectroscopy of gaseous media, of small areas of the exciting light pulses. As the areas of the exciting pulses are increased, the polarization properties of the echo begin to depend generally speaking, on these areas and on the shape of the exciting pulses. The formulas for the intensity of the electric field of the photon echo are then, even in the case of rectangular exciting pulses, too unwieldy and unsuitable for use in the processing of experimental data. But is should be emphasized that, in the case of the transitions $0 \leftrightarrow 1$, $1/2 \to 1/2$, and $1/2 \leftrightarrow 3/2$, which are accompanied by a change in the electron angular momentum, the polarization properties of the echo are described by the formulas (11) and (12) for arbitrary areas of the exciting pulses. Thus, in the case when the photon echo is produced at the transitions $J_b = 1/2 \to J_a = 1/2$ and $J_b = 3/2 \to J_a = 1/2$, which are accompanied by a change in the electron angular momentum, and at which the photon echo is produced in alkali-metal vapors, the intensity of the electric field of the photon echo is, if it is produced on a narrow spectral line, given by the formulas (9), (11), and (12), in which the quantity I_e has the form

$$I_e = \frac{4}{\theta_1 \theta_2^2} \sin\theta_1 \sin^2\frac{\theta_2}{2} \exp\left[-\frac{1}{4}(ku)^2(t'-\tau)^2\right],$$

where the areas of the exciting pulses are given by the relation

$$\theta_l = \sqrt{2}|d|e^{(l)}T_l/(\hbar\sqrt{3}), \quad l=1, 2.$$

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[1]T. Baer and I. D. Abella, Phys. Lett. A **59**, 371 (1976).
[2]T. Baer and I. D. Abella, Phys. Rev. A **16**, 2093 (1977).
[3]S. Aoki, Phys. Rev. A **20**, 2013 (1979).
[4]R. Kachru, T. W. Mossberg, and S. R. Hartmann, J. Phys. B **13**, L363 (1980).
[5]H. Nakatsuka, M. Fujita, and M. Matsuoka, Opt. Commun. **36**, 234 (1981).
[6]L. Q. Lambert, A. Compaan, and I. D. Abella, Phys. Rev. A **4**, 2022 (1971).
[7]I. I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov (Introduction to the Theory of Atomic Spectra), Nauka, Moscow 1977 (Eng. Transl., Pergamon, Oxford, 1973).
[8]I. V. Evseev and V. M. Ermachenko. Zh. Eksp. Teor. Fiz. **76**, 1538 (1979) [Sov. Phys. JETP **49**, 780 (1979)].
[9]V. N. Rebane, Dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of the Physico-Mathematical Sciences, Leningrad State University, Leningrad, 1980.
[10]E. Arimondo, M. Inguscio, and P. Violino, Rev. Mod. Phys. **49**, 31 (1977).
[11]S. S. Alimpiev and N. V. Karlov, Zh. Eksp. Teor. Fiz. **63**, 482 (1972) [Sov. Phys. JETP **36**, 255 (1973)].
[12]L. S. Vasilenko and N. N. Rubtsova, in: Lazernye sistemy (Laser Systems), Inst. Theor. Phys., Siberian Branch of the USSR Academy Sciences, Novosibirsk, 1982, p. 143.

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