PHOTON ECHOES IN DOUBLE RADIOOPTICAL RESONANCE

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The method of double radiooptical resonance is generalized to the case of coherent spontaneous radiation. The effect of photon echo is investigated in two limiting cases, those of strong and weak magnetic fields. In strong magnetic fields, the coherent response in the medium arises at different optical frequencies. Both components of the radiation experience modulation, depending on the interval between the exciting pulses of light and the amplitude of the radiofrequency field. In weak magnetic fields, there is a polarization dependence of the emerging coherent radiation. The results are valid for transitions in gases of the type $j = 1/2 \rightarrow j' = 1/2$ and in crystals with paramagnetic impurities of the ruby type.

IT is known that the photon-echo effect[1,2] is exceedingly sensitive to various kinds of hyperfine interactions. Measurement of the intensity of photon echo as a function of the interval τ between two exciting coherent pulses of light apparently makes it possible to extract information on the isotopic shifts of the excited levels[3], and can serve as a new method in spectroscopy of ultrahigh resolution[4]. Modulation of the echo signal in ruby was recently observed[5]. In this case the hyperfine interaction is equivalent to action of a certain radio frequency field, produced by the precessing magnetic moments of the crystal-matrix nuclei, on the ions of the paramagnetic impurities. It is obvious that such an alternating field has a complicated spatial and temporal structure, which in final analysis does not make it possible to solve this problem completely[1,6].

This raises the question of how the photon-echo effect changes when a stationary and homogeneous radiofrequency field is applied. In this case the field parameters are relatively simple and can be varied over a wide range at the observer's will. In particular, it is of interest to consider the problem of formation of photon echo in a medium in which the action of continuous radio-frequency pumping is effected under conditions of double radiooptical resonance, first investigated by Brossel and Kastler[7].

The method of radiooptical resonance has presently become widely known and is a reliable method of optical registration of EPR in excited states. It must be noted, however, that so far the action of the microwave field reduced only to a perturbation of incoherent spontaneous radiation. Therefore in our investigation the method of the radiooptical resonance has been extended to cover an essentially coherent process, namely the effect of photon echo. We have considered here the two most important limiting cases:

a) strong magnetic field, in which the following inequality is realized:

$$\Delta \ll d\tilde{E} / \hbar \ll |\delta\Omega|, \tag{1}$$

where $\Delta = 1/T_2^*$ is the inhomogeneous line width; $\delta\Omega = \Omega_2 - \Omega_1$, Ω_1, Ω_2 are quantities characterizing the Zeeman splitting of the levels of the ground and excited states; $\tilde{E}$ is the amplitude of the electric field of the light pulses and d is the reduced matrix element;

b) weak magnetic field, when the inequality

$$|\delta\Omega| \ll \Delta \ll d\tilde{E} / \hbar. \tag{2}$$

is satisfied. We note that the amplitude of the electric field of the light pulses was chosen to be sufficiently large[8]. With respect to the relaxation processes we assume that the damping constants for transitions to different Zeeman sublevels are the same. We assume also that the direction of the constant magnetic field coincides with the z axis—the quantization axis in the gas, or with the direction of the optical axis in the paramagnetic crystal.

Qualitatively, the main results can be obtained by means of the following physical reasoning. Both the optical and the radiofrequency electromagnetic fields are coherent. In case a), after the action of the first optical pulse, the three-level quantum system (see Fig. 1a) turns out to be in a superposition of the two upper states II and III. Such a system is a classical dipole radiator at the frequency ω. The continuous radio-frequency field mixes the states I and II, so that in this case the quantum system is a classical radiator at three frequencies ω, Ω, and $\omega + \Omega$[1)]. It is known that the amplitudes of the probabilities of observing the system in the states II and I vary periodically like $\cos(\lambda_\Omega t/2)$ and $\sin(\lambda_\Omega t/2)$ (see, for example,[9]), where λ_Ω depends on the amplitude of the alternating field. In our case, this will be exactly the law of variation of the amplitudes of the oscillations at the frequencies ω and $\omega + \Omega$. Thus, by the instant of action of the second optical pulse at $t = \tau$ the number of radiators at the frequencies ω and $\omega + \Omega$ will be proportional respectively to $\cos(\lambda_\Omega\tau/2)$ and $\sin(\lambda_\Omega\tau/2)$. The second optical pulse leads to "time reversal" for the dipole radiators at the same frequency ω[1], and will exert no noticeable influence on the radiators of the second optical frequency $\omega + \Omega$, for by virtue of the condition (1) the latter are away from resonance with the external light pulses.

By the instant of occurrence of the photon echo, the radio-frequency field acting after the second optical pulse again distributes the radiators over the frequen-

1) If the positions of the two lower levels are reversed, the combination frequency is $\omega - \Omega$.

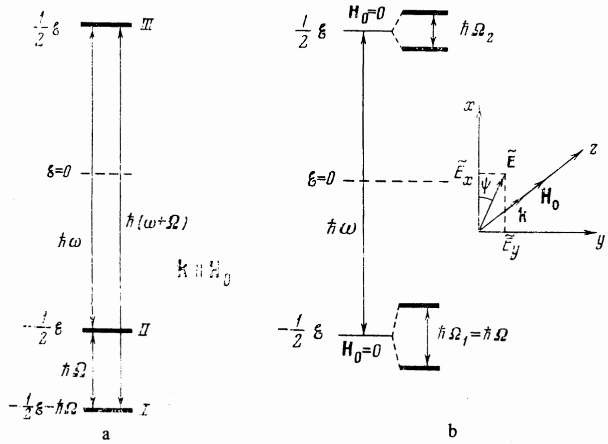

FIG. 1. Energy-level scheme for the realization of photon echo under conditions of double radiooptical resonance: a—in strong magnetic fields, b—in weak fields. $\tilde{E}$—amplitude of electric field of light wave; k—wave vector, ω—optical frequency, Ω—frequency of radio emission, H_0—constant magnetic field.

cies ω and $\omega + \Omega$ in the proportions indicated above, so that finally at $t = 2\tau$ their number will be proportional to $\cos^2(\lambda_\Omega \tau/2)$ and $\cos(\lambda_\Omega \tau/2)\sin(\lambda_\Omega \tau/2)$. Thus, both components of the photon echo will experience modulation, depending on the interval between the exciting pulses of light τ and the intensity of the radio-frequency pumping. We emphasize that if the equality $\lambda_\Omega \tau = \pi$ is satisfied, both components vanish simultaneously, and if $\lambda_\Omega \tau = 2\pi$, the combination-frequency component vanishes, whereas at the fundamental frequency the component is maximal in magnitude. The quantitative results are given in the form of expressions (17) and (18).

Similar reasoning applies also in case b). Here under the action of the continuous radio-frequency pumping there will be observed the same frequency variations of the optical field. But since the frequency shifts of the energy levels of the ground and the excited states in the magnetic field lie within the limits of the width of the optical line, the greater importance will be assumed not by the frequency dependence of the emerging coherent radiation, but by the polarization dependence. Thus, it can be shown that for the four-level system shown in Fig. 1b, the photon-echo polarization picture consists of two vectors, one of which is directed at an angle $2\psi_2 - \psi_1$ in Cartesian axes, and the direction of the other depends both on the magnitude of the constant magnetic field and on the interval τ between the exciting light pulses. When τ or the constant magnetic field is varied, the position of the second vector relative to the first changes, and as a result the intensity of the photon echo experiences oscillations. We note that in the absence of a radio-frequency field ($\lambda_\Omega = 0$ in formulas (27)—(29)), the photon-echo polarization vector is directed at an angle $2\psi_2 - \psi_1$. Thus, for the transition $j = 1/2 \leftrightarrow j' = 1/2$ there is no rotation of the plane of polarization of the photon echo at the instant $t = 2\tau$ in a constant magnetic field. The last result is valid for the type of transition considered by us, where the four-level system may be broken up into two independent two-level systems.

The rotation of the plane of polarization in an alternating nonresonant magnetic field was indicated in[10].

The calculation procedure of the present paper is based on an earlier paper by the authors[8], and we shall therefore confine ourselves henceforth only to the most general information concerning the character of the calculations, and will stop to discuss in detail only the new aspects in the calculation. The initial equation for the density matrix, just as in[8], is taken in the form

$$i\hbar\frac{\partial\hat{\rho}}{\partial t} = [\hat{\mathscr{H}}_0 + \hat{V}(t), \hat{\rho}] + i\hbar\left(\frac{\partial\hat{\rho}}{\partial t}\right)_\gamma, \tag{3}$$

where $\hat{\mathscr{H}}_0$ is the unperturbed Hamiltonian, but now with account taken of the constant magnetic field, $\hat{V}(t)$ is the interaction operator, and the term $i\hbar(\partial\hat{\rho}/\partial t)_\gamma$ describes the relaxation processes. The interaction with the light field will be considered in the dipole approximation, and the interaction with the radio-frequency field in the magnetic-dipole approximation; then

$$\hat{V}(t) = \hat{V}^\omega(t) + \hat{V}^\Omega(t),$$
$$\hat{V}^\omega(t) = -\hat{\mathbf{d}}\mathbf{E}_a(t), \qquad \hat{V}^\Omega(t) = -\mu\mathbf{H}^\Omega(t). \tag{4}$$

The index a in (4) numbers the sequence of light pulses and assumes values a = 1 and 2. We write the electromagnetic field in the form

$$\mathbf{E}_a(\mathbf{R}_j, t) = \mathrm{Re}\,\{\tilde{\mathbf{E}}_a(\mathbf{R}_j, t)\exp[i(\mathbf{k}\mathbf{R}_j - \omega t + \Phi_a)]\} \tag{5}$$

for the optical pulses and

$$\mathbf{H}^\Omega(t) = \mathrm{Re}\,\{\mathbf{H}_0^\Omega \exp[-i(\Omega t + \Phi_H)]\}, \quad \mathbf{H}_0^\Omega \perp \mathbf{H}_0. \tag{6}$$

for the radio-frequency pumping.

In expressions (5) and (6), $\tilde{\mathbf{E}}_a(\mathbf{R}_j, t)$ are slow amplitudes of the optical pulses, $\mathbf{R}_j$ is the coordinate of the j-th radiator, $k = \omega n(\omega)/c$ is the wave vector of the incident coherent radiation, Φ_a and Φ_H are phases, $\mathbf{H}_0^\Omega$ is the amplitude of the radio-frequency field of frequency Ω, and $\mathbf{H}_0$ is the constant magnetic field. In writing down (6) it was assumed that the wavelength of the radio emission greatly exceeds the dimensions of the medium, i.e., $L\Omega n(\Omega)c^{-1} \ll 1$ (L—dimensions of the medium, $n(\Omega)$—refractive index, c—velocity of light in vacuum).

During the time of action of the optical pulses, we assume that their duration δ_a is much smaller than the characteristic relaxation time of the "quantum coherence" of the medium γ^{-1} ($\delta_a \ll \gamma^{-1}$), and the amplitudes of the light field are much larger than the amplitudes of the radio-frequency field ($V^\omega \gg V_{ij}^\Omega$; i, j—indices of the states). Then Eq. (3) takes the form

$$i\hbar\frac{\partial\hat{\rho}}{\partial t} = [\hat{\mathscr{H}}_0 + \hat{V}_a^\omega, \hat{\rho}]. \tag{7}$$

We shall solve this equation in the given-field approximation, for which it is necessary to satisfy the condition

$$2\pi\omega L N_0 d^2 T_2^* / \hbar v\varepsilon \ll 1, \tag{8}$$

where N_0 is the concentration of the radiators and ϵ is the dielectric constant of the medium. We shall henceforth confine ourselves to calculation of the polarization of the medium, an expression for which can be written as follows:

$$\mathbf{P}(\mathbf{R}, t) = N_0 \int d\mathscr{E} g(\mathscr{E} - \hbar\omega)\mathrm{Sp}\{\hat{\rho}(\mathbf{R}, t)\hat{\mathbf{d}}\}. \tag{9}$$

In formula (9), $g(\mathscr{E})$ is the energy-level distribution function, and the integration is carried out over all possible values of the energy levels.

Let us consider for simplicity a certain system (either a crystal with paramagnetic impurities or a gas) having a small degree of degeneracy in the absence of a magnetic field. Assume that it is equal, say, to two for both the ground and excited states. The conditions of strong and weak constant magnetic fields means that in the former case we should work with a three-level system and in the latter case with a four-level system. We choose them as shown in Fig. 1. To make the succeeding expressions compact, we shall use the expansion of the three-row matrices in terms of the matrices of the effective spin S = 1 (see, for example,[11], and expand the four-row matrices in the Dirac γ matrices[12]. Let us discuss each case separately.

CASE OF STRONG MAGNETIC FIELD

The action of a continuous radio-frequency pump in a medium leads to the occurrence of magnetization and to a change in the population of the resonant energy levels. Upon saturation, the populations become equalized, and the magnetization tends to zero. This means that the off-diagonal elements of the density matrix, which describes this two-level system, become much smaller than the diagonal ones. Indeed, the standard procedure of calculating stationary problems in the theory of magnetic resonance (see, for example,[13]) shows that this condition is valid when the following inequality is satisfied:

$$\frac{1}{\varkappa}\,\mathrm{th}\left(\frac{\hbar\Omega}{2kT}\right)\frac{|\mu_{21}H_0^\Omega|T_2}{\hbar}\ll 1\mp\frac{1}{\varkappa}\,\mathrm{th}\left(\frac{\hbar\Omega}{2kT}\right),$$

$$\varkappa=1+T_1T_2\hbar^{-2}|\mu_{21}H_0^\Omega|^2,$$

where μ_{21} is the matrix element of the operator of the magnetic moment for the transition between the energy levels II and I, H_0^Ω is the amplitude of the alternating radio-frequency field (6), T is the temperature, and T_1 and T_2 are the times of longitudinal and transverse relaxation. The minus (plus) sign in the right side of the inequality pertains to the case when the optical excitation is realized from the level II (from the level I). We see that when $\kappa \gg 1$ (saturation), the inequality is satisfied in the entire temperature interval. When $\kappa \sim 1$ it is violated in the region of very low temperatures. Therefore at the instant of action of the first optical pulse the initial condition for Eq. (3) can be written in the form

$$\hat{\rho}^{(0)}=\alpha+\frac{(1-\alpha)}{2}\hat{S}_z+\frac{(1-3\alpha)}{2}\hat{S}_z^2, \tag{10}$$

where α characterizes the relative population of level II (with allowance for the radio-frequency field) and $\hat{S}_z$ is the matrix of the effective spin S = 1.

The procedure for solving Eq. (7) in strong electromagnetic fields ($d\tilde{E}_a T_2^* h^{-1} \gg 1$) is described in[8], and we therefore present only the final result:

$$\hat{\rho}(t)=e^{-i\hat{\Delta}(t-t_a)}e^{-i\hat{D}}\hat{\rho}(t_a)e^{i\hat{D}}e^{i\hat{\Delta}(t-t_a)}, \tag{11}$$

where

$$\hat{\Delta}=\frac{\omega}{2}(\hat{S}_z^2-\hat{S}_z-1),$$

$$\hat{D}=-\frac{\Omega}{2}(t-t_a)\hat{S}_z(\hat{S}_z+1)+\frac{A}{2\sqrt{2}}\{(\hat{S}_+-[\hat{S}_+,\hat{S}_z]^+)+\text{h.c.}\},$$

$$A=-\frac{\theta_a}{2}\exp[-i(\mathbf{kR}_j+\Phi_a)+i\omega t_a],$$

$$\theta_a=\frac{d^\omega}{\hbar}\int_{t_a}^{t}E(\mathbf{R},t')dt'. \tag{12}$$

The expression for the exponential operator in (11) can be obtained in the standard manner. To this end it is necessary to find a unitary transformation that reduces the matrix $\hat{D}$ to diagonal form, and then find $\exp(\pm i\hat{D})$ with the aid of the similarity transformation. As a result we obtain

$$e^{i\hat{D}}=\cos\frac{\theta_a}{2}+\frac{1}{2}\left\{\exp[-i\Omega(t-t_a)]-\cos\frac{\theta_a}{2}\right\}(\hat{S}_z+\hat{S}_z^2)+i\frac{\sin(\theta_a/2)}{\theta_a}\left\{\frac{A}{\sqrt{2}}(\hat{S}_+-[\hat{S}_+,\hat{S}_z]^+)+\text{h.c.}\right\}. \tag{13}$$

For the instant of time between the optical pulses, we write Eq. (3) only for those components of the density matrix which contribute to the coherent radiation at the optical frequencies:

$$i\hbar\dot{\rho}_{13}=-i\hbar\gamma\rho_{13}-(\mathscr{E}+\hbar\Omega)\rho_{13}+V_{12}\rho_{23},$$
$$i\hbar\dot{\rho}_{23}=-i\hbar\gamma\rho_{23}-\mathscr{E}\rho_{23}+V_{21}\rho_{13}, \tag{14}$$

where $1/\gamma$ is the time of irreversible relaxation. The solution of this system entails no difficulty[2)]:

$$\left\|\begin{matrix}\rho_{13}(t)\\ \rho_{23}(t)\end{matrix}\right\| = e^{\gamma}\left\|\begin{matrix} e^{i\Omega(t-t_0)}\cos\frac{\lambda_\Omega(t-t_0)}{2}; & -2i\frac{F_{21}^*e^{i\Omega t}}{\hbar\lambda_\Omega}\sin\frac{\lambda_\Omega(t-t_0)}{2}\\ \cos\frac{\lambda_\Omega(t-t_0)}{2}; & -2i\frac{F_{21}e^{-i\Omega t_0}}{\hbar\lambda_\Omega}\sin\frac{\lambda_\Omega(t-t_0)}{2}\end{matrix}\right\| \times\left\|\begin{matrix}\rho_{13}(t_0)\\ \rho_{23}(t_0)\end{matrix}\right\|, \tag{15}$$

where

$$e^{\gamma}=\exp(-\gamma+i\mathscr{E}\hbar^{-1})(t-t_0),\quad F_{21}=\mu_{21}H_0^\Omega/2,\quad \lambda_\Omega=2|F_{21}|/\hbar.$$

The obtained solutions (11) and (15) now make it possible to calculate the density matrix, and consequently also the polarization produced in the medium after the passage of two coherent light pulses.

We take the origin at the boundary of the medium and assume that the first light pulse reaches it at the instant of time t = 0. We assume that the wave vectors of the exciting pulses are equal, i.e., $\mathbf{k}_1 = \mathbf{k}_2$, and that the propagation direction coincides with the z axis. For radiators with arbitrary coordinate $\mathbf{R}_j$, the instants t_a of turning on the optical field will be

$$t_1=\frac{kz_j}{\omega},\quad t_2=\frac{kz_j}{\omega}+\tau+\delta_1, \tag{16}$$

where δ_a is the duration of the pulses and τ is the interval between them. Substituting expressions (10) and (13) in (11), we obtain the density matrix after the first pulse. Further, retaining the terms responsible for the photon echo we obtain, taking the solution of the system (14) into account, the density matrix at the start of action of the second pulse, and so on up to the instant of time $t > t_2 + \delta_2$. Substituting the obtained values of $\hat{\rho}(t)$ in (9)

2)The solution of (14) was obtained under the assumption $\Omega \gg \lambda_\Omega$, τ^{-1}.

and integrating with respect to the energy, we obtain the final expression for the polarization:

$$\mathbf{P} = \mathbf{P}^{\omega} + \mathbf{P}^{\omega+\Omega},$$
$$\mathbf{P}^{\omega} = \mathrm{Re}\,\{\mathbf{P}^{\omega}\exp[i(kz-\omega t+\Phi^{\omega})]\}, \tag{17}$$
$$\mathbf{P}^{\omega+\Omega} = \mathrm{Re}\,\{\tilde{\mathbf{P}}^{\omega+\Omega}\exp[i(kz-\omega t-\Omega t+\Phi^{\omega+\Omega})]\},$$

$$\tilde{\mathbf{P}}^{\omega} = \alpha N_0 \mathbf{d}^{\omega} fg\left(t-\frac{z}{v}-t_m\right)\cos\frac{\lambda_\Omega\tau}{2}\cos\frac{\lambda_\Omega}{2}\left(t-\frac{z}{v}-\tau-\delta_1-\delta_2\right)$$
$$\times\exp\left[-\gamma\left(t-\frac{z}{v}-\delta_1-\delta_2\right)\right], \tag{18}$$
$$\tilde{\mathbf{P}}^{\omega+\Omega} = \alpha N_0 \mathbf{d}^{\omega+\Omega} fg\left(t-\frac{z}{v}-t_m\right)\cos\frac{\lambda_\Omega\tau}{2}$$
$$\times\sin\frac{\lambda_\Omega}{2}\left(t-\frac{z}{v}-\tau-\delta_1-\delta_2\right)\exp\left[-\gamma\left(t-\frac{z}{v}-\delta_1-\delta_2\right)\right].$$

In expressions (18), f is a factor that depends on the "rotation angles" of the dipole-moment vector $\mathbf{d}^{\omega}$ in the electric fields of the light waves, $f = \sin\theta_1\sin^2(\theta_2/2)$; Φ^{ω}, $\Phi^{\omega+\Omega}$—oscillation phases, $\Phi^{\omega} = 2\Phi_2 - \Phi_1 - \pi/2$, $\Phi^{\omega+\Omega} = 2\Phi_2 - \Phi_1 + \Phi_H$; g(t)—correlation function connected with the distribution function $g(\mathscr{E})$ by the usual Fourier transformation; v—velocity of propagation of light in the medium; $t_m = 2\tau + \delta_1 + \delta_2$—instant when the photon echo is maximal.

Thus, the coherent radiation arises immediately at two frequencies ω and $\omega + \Omega$. The polarization is determined by the structure of the matrix elements $\mathbf{d}^{\omega}$ and $\mathbf{d}^{\omega+\Omega}$ and, in final analysis, depends on the type of the transitions under consideration. We note that when the wavelength of the radio-frequency field becomes comparable with the dimensions of the medium or smaller than these dimensions, then to observe the radiation of frequency $\omega + \Omega$ it is necessary also to satisfy the condition of spatial synchronism: $\mathbf{k}(\omega+\Omega) = \mathbf{k}(\omega) + \mathbf{k}(\Omega)$.

CASE OF WEAK MAGNETIC FIELD

In this limiting case it is necessary to choose as the working system the four-level system shown in Fig. 1b. We shall assume that the optical pulses are linearly polarized and that the polarization planes form angles ψ_a with the x axis. We choose as the initial structure the structure of matrix elements of the dipole moment for Cr ions in Al_2O_3. Such a choice does not limit the generality of the results and is useful because the condition $|\delta\Omega| \ll \Delta$ can be satisfied not only in weak magnetic fields, when the Zeeman splitting is much smaller than the line width ($\Omega_{1,2} \ll \Delta$), but also in the opposite case, when the condition $\Omega_{1,2} \gg \Delta$ is satisfied[5]. The final results will be valid also for the transition $j = 1/2 \leftrightarrow j' = 1/2$ in the gas atom, with Ω_2 replaced by $-\Omega_2$. The condition $|\delta\Omega| \ll \Delta$ then goes over automatically into the condition $\Omega_{1,2} \ll \Delta$.

The matrix elements of the operator d in ruby can be obtained by the method of Sugano and Tanabe[14]. For light propagating along the optical axis, the dipole-moment operator can be represented in the following compact form:

$$\hat{\mathbf{d}} = d(\hat{\gamma}^3\mathbf{i} - i\hat{\gamma}^0\mathbf{j})\hat{\gamma}^5, \tag{19}$$

where d is the reduced matrix element; $\hat{\gamma}^0$, $\hat{\gamma}^3$, and $\hat{\gamma}^5$ are Dirac matrices[12]; i and j are unit vectors in the directions of x and y. The initial conditions for the fundamental equation (3) are now written in the form

$$\hat{\rho}^{(0)} = {}^1\!/_4(\hat{1} - \hat{\gamma}^5). \tag{20}$$

The solution of equation (7) during the time of action of the optical pulses using the approximation (8) will be described also by formula (11) with $\hat{\Delta}$ and $\hat{D}$ replaced with the new quantities

$$\hat{\Delta} = {}^1\!/_2\omega\hat{\gamma}^5, \tag{21}$$

$$\hat{D} = (\cos\psi_a\hat{\gamma}^3 - i\sin\psi_a\hat{\gamma}^0)\left(\frac{A-A^*}{2} + \frac{A-A^*}{2}\hat{\gamma}^5\right) \tag{22}$$

In expression (22), A takes on the value (12). For the exponential operator $\exp(i\hat{D})$ we can obtain similarly

$$e^{i\hat{D}} = \cos\frac{\theta_a}{2}\hat{1} + \theta_a^{-1}\sin\frac{\theta_a}{2}$$
$$\times(i\cos\psi_a\hat{\gamma}^3 + \sin\psi_a\hat{\gamma}^0)[A - A^* + (A + A^*)\hat{\gamma}^5]. \tag{23}$$

In the intervals between the optical pulses, solving Eq. (3) for the components $\rho(t)$ that contribute to the coherent radiation, we obtain[3)]

$$\rho_{jk}(t) = e_{jk}^{\gamma}\left[\rho_{jk}(t_0)\cos\frac{\lambda_\Omega(t-t_0)}{2} - i\rho_{j+1,k}(t_0)\frac{2F_{21}{}^*e^{i\Omega_1 t_0}}{\hbar\lambda_\Omega}\sin\frac{\lambda_\Omega(t-t_0)}{2}\right]$$
$$j = 1,\quad k = 3, 4; \tag{24}$$

$$\rho_{jk}(t) = e_{jk}^{\gamma}\left[\rho_{jk}(t_0)\cos\frac{\lambda_\Omega(t-t_0)}{2} - i\rho_{j-1,k}(t_0)\frac{2F_{21}e^{-i\Omega_1 t_0}}{\hbar\lambda_\Omega}\sin\frac{\lambda_\Omega(t-t_0)}{2}\right]$$
$$j = 2;\quad k = 3, 4, \tag{25}$$

where

$$e_{jk}^{\gamma} = \exp\{[+i\hbar^{-1}(\mathscr{E}_k - \mathscr{E}_j) - \gamma](t - t_0)\}. \tag{26}$$

From now on the procedure is perfectly analogous to the preceding case and when multiplying the Dirac matrices it is necessary to use their corresponding properties[12]. Retaining in the density matrix $\hat{\rho}(t)$ the terms responsible for the photon-echo effect, we obtain the following expression for the polarization:

$$\mathbf{P}(z,t) = \mathrm{Re}\left\{\tilde{P}\left(t-\frac{z}{v}\right)\exp(ikz - i\omega t + i\Phi_e)\right.$$
$$\times\mathbf{l}_1(t)\cos\frac{\lambda_\Omega\tau}{2}\cos\frac{\lambda_\Omega}{2}\left(t-\frac{z}{v}-\tau-\delta_1-\delta_2\right)$$
$$\left.+\mathbf{l}_2(t)\sin\frac{\lambda_\Omega\tau}{2}\sin\frac{\lambda_\Omega}{2}\left(t-\frac{z}{v}-\tau-\delta_1-\delta_2\right)\right\}, \tag{27}$$

where $\tilde{P}(t - z/v)$ is the slow polarization amplitude, equal to

$$N_0 dfg\left(t-\frac{z}{v}-t_m\right)\exp\left[-\gamma\left(t-\frac{z}{v}-\delta_1-\delta_2\right)\right\}.$$

We see that it is maximal at the instant of time $t_m = 2\tau + \delta_1 + \delta_2$; $\Phi_e = 2\Phi_2 - \Phi_1 - \pi/2$ is the phase of the oscillations and $\mathbf{l}_1$ and $\mathbf{l}_2$ are unit vectors:

$$\mathbf{l}_b = \mathbf{i}\cos\varphi_b + \mathbf{j}\sin\varphi_b,\quad b = 1, 2; \tag{28}$$

$$\varphi_1(t) = 2\psi_2 - \psi_1 + \frac{(\Omega_1-\Omega_2)}{2}\left(t-\frac{z}{v}-t_m\right),$$
$$\varphi_2(t) = \psi_1 + \frac{\Omega_1}{2}\left(t-\frac{z}{v}-\delta_1+\delta_2\right) - \frac{\Omega_2}{2}\left(t-\frac{z}{v}-\delta_1-\delta_2\right). \tag{29}$$

Thus, expressions (17), (18), and (27)–(29) describe the photon-echo polarization in double radiooptical resonance in the two limiting cases of strong and weak

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3)The solution of Eqs. (24)–(26) is valid if $|\Omega_2 - \Omega_1| \gg \lambda_\Omega, \tau^{-1}$.

constant magnetic fields. In the former case the coherent response in the medium arises at two optical frequencies ω and $\omega + \Omega$. The intensity of each of the components depends on the "rotation angle" of the dipole moment by the radio frequency field. When the photon echo vanishes at the fundamental frequency, the component at the combination frequency also vanishes. When $\lambda_\Omega \tau = \pi/2$ the echo signal at the combination frequency is maximal in magnitude. This can be used for spectral separation of the photon-echo signal from the external exciting pulses of the coherent light, which apparently is of importance for measurements of relatively short relaxation times γ^{-1} of the "quantum coherence." In addition, in strong radio-frequency fields when the field line width is of the same order as the optical line width ($\lambda_\Omega \sim \Delta$), after the response time T_2^* the frequency of the oscillations may change, leading to oscillations of the intensity in time.

In the latter case, the continuous radio-frequency pumping gives rise to a unique polarization dependence of the emerging coherent radiation. In weak magnetic fields at the instant of time $t_m = 2\tau + \delta_1 + \delta_2$, i.e., when this is maximal, the orientation of the vector l_1 does not depend on the constant magnetic field, whereas the vector l_2 forms an angle $\varphi_2 = \psi_1 + (\Omega_1 - \Omega_2)\tau + \Omega_1\delta_2$ with the x axis. The absolute values of the polarization vectors, as follows from (27)—(29), are determined by the intensity of the radio-frequency field λ_Ω and by the time interval τ between the optical pulses. When the equation $\lambda_\Omega \tau = \pi$ is satisfied, the photon-echo polarization plane experiences uniform rotation about the direction of the constant magnetic field depending on τ and δ_2. This can serve as a means of a sufficiently exact measurement of the g factors of the ground and excited states.

It is interesting to note that the case of a strong magnetic field can be realized in a gas of Cs atoms, and also in ruby in the absence of any magnetic field at all. In Cs, for example, continuous radio-frequency pumping can be "turned on" between the components of the hyperfine structure of the ground state $6^2S_{1/2}(F = 3) \leftrightarrow 6^2S_{1/2}(F = 4)$. The frequency of this transition is 9.2 GHz, i.e., it lies in the microwave band. The optical transitions can be excited either at the level $6^2P_{1/2}(F = 3)$, or at the level $6^2P_{1/2}(F = 4)$. We note that in Cs each of these levels is multiply degenerate, and therefore to obtain strict quantitative results it is necessary to solve the problem with allowance for the degeneracy. In ruby, the ground level $^4A_2$ of the trivalent chromium ions is split under the influence of the trigonal field and the spin-orbit interaction into two doublets (Fig. 2) (Ω = 11.4 GHz). The structure of the levels $^4A_2$ and $^2E(\overline{E})$ in ruby is also in accord with the case (a) considered by us. The singularity lies only in the fact that all the levels are doubly degenerate, and the degeneracy is lifted only by the magnetic field. However, for linearly-polarized optical pulses, and also for a linearly-polarized radio-frequency field, the six-level system $\{^4A_2, {}^2E(\overline{E})\}$ can be broken up into two independent three-level systems (Fig. 2), each of which interacts with circularly-polarized waves of a definite type. Thus, if the frequency of the exciting optical pulses is close to the frequency of the transitions $^4A_2(M_S = \pm 1/2) \leftrightarrow {}^2E(\overline{E})(\pm 1/2, u_\pm)$, then the right-hand wave causes the

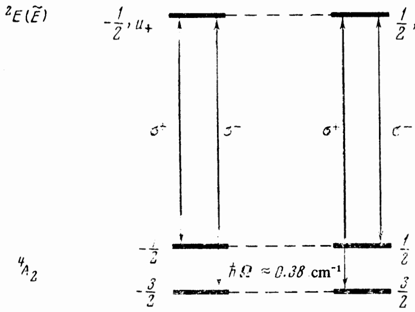

FIG. 2. Energy level scheme of the Cr^{3+} ion in ruby in the absence of a constant magnetic field. σ^+—transition caused by a light wave with right-hand circular polarization, σ^-—transition caused by a light wave with left-hand circular polarization.

transition $\sigma^+ {}^4A_2(M_S = -1/2) \leftrightarrow {}^2E(\overline{E})(-1/2, u_+)$, and the left-hand wave the transition $\sigma^- {}^4A_2(M_S = 1/2) \leftrightarrow {}^2E(\overline{E})(+1/2, u_-)$. Analogously, the right-hand wave of the radio-frequency field causes the transition $^4A_2(M_S = 1/2) \leftrightarrow {}^4A_2(M_S' = 3/2)$, and the left-hand wave the transition $^4A_2(M_S = -1/2) \leftrightarrow {}^4A_2(M_S' = -3/2)$. As a result, the photon-echo radiation at the frequency $\omega + \Omega$, emerging from the crystal, turns out to be likewise linearly polarized, and the position of the plane of polarization will be determined by the polarizations of both the light pulses and the radio-frequency field.

Let us estimate the amplitudes of the radio-frequency electromagnetic field needed in order to be able to observe the effects described above. From (17), (18), and (24)—(29) we see that it is necessary to satisfy the condition $\mu H_0^\Omega \hbar^{-1}\tau \sim 1$. For average values $\tau \sim 100$ nsec and $\mu \sim \mu_B = 10^{-20}$ we obtain $H_0^\Omega \sim 1$ G.

In conclusion, we note that the results described by formulas (27)—(29) are also valid in the limiting case when the frequency shift of the optical lines in the magnetic field $\delta\Omega_{12}$ is of the same order as the line width, i.e., $\delta\Omega_{12} \sim \Delta$.

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