CONTRIBUTION TO THE THEORY OF NONSTATIONARY PROCESS OF THE PHOTON-ECHO TYPE IN CRYSTALS WITH FERROMAGNETIC IMPURITIES

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A theory is constructed for nonstationary processes of the photon-echo type occurring in crystals with paramagnetic impurities after the passage through them of an arbitrary number of light pulses. The calculations are carried through to conclusion for the case of three exciting pulses of coherent radiation. The spatial synchronism, the phase relations, the intensity, energy, and attenuation of the coherent radiation produced in the medium are investigated. The results are discussed as applied to a ruby crystal.

INTRODUCTION. FORMULATION OF PROBLEM

THE photon-echo effect predicted in[1] promises to be an effective method of measuring the relaxation time both in solids[2-4] and in gases[5,6]. This method consists of successively passing through a resonant medium two exciting coherent pulses of light separated by a certain time interval τ_1. At the instant of time $2\tau_1$ after the passage of the first pulse, a coherent quantum state is produced in the medium and leads to the occurrence of a radiation signal-photon echo. From the attenuation of the intensity of the photon echo as a function of τ_1 it is possible to determine the relaxation time T_2 of the quantum coherence.

A characteristic feature of photon echo, unlike the widely known spin echo, is its spatial synchronism: it is necessary that the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ of the exciting pulses and the wave vector $\mathbf{k}$ of the photon echo satisfy the relation[3]

$$\mathbf{k} = 2\mathbf{k}_2 - \mathbf{k}_1. \qquad \text{(I)}$$

New effects of the photon-echo type appear when three and more light pulses act on a resonant medium. Under certain conditions, the so-called "stimulated" echo can arise (see Fig. 1, pulse with b = 2). The first experiments with such an excitation were performed on ruby[7,8], and the question of the spatial synchronism was considered by one of the authors[9].

In this paper we develop a theory of nonstationary processes of the photon-echo type in crystals with paramagnetic impurities, through which an arbitrary number of light pulses passes. The calculations are carried through to conclusion for the case of three exciting pulses of coherent radiation. We investigate the spatial synchronism, the phase relations, and the intensity and attenuation of the coherent radiation produced in the medium (see the summary table).

The theory of photon echo as applied to a gas has by now been thoroughly developed; namely, investigations were made of the polarization effects[10,11], and account was taken of the influence of the magnetic fields[12]. Besides taking into account the attenuation, the main difficulty lies in the fact that the energy levels of the atom have a degeneracy of one degree or another. At the same time, the spatial-synchronism condition (I) for such an isotropic medium, as a gas is satisfied only in the one simple case when all three vectors have the same direction, since they are equal in magnitude.

In anisotropic crystals with paramagnetic impurities, the spatial synchronism becomes more complicated, since the light beam breaks up into two rays, ordinary and extraordinary, having different phase velocities. However, the degree of degeneracy of the energy levels of the paramagnetic ions is small, and this makes it possible to simplify the problem considerably. It is known that the ground state of paramagnetic ions in a crystal is either nondegenerate (for an even number of electrons) or else is a Kramers doublet (for an odd number of electrons) (see, for example,[13]). In particular, in ruby with the electric vector parallel to the optical axis of the crystal (extraordinary wave), the following transitions are distinguished in the trivalent chromium ion[14]:

$$^4A_2(M_s = 1/2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = -1/2);\ ^4A_2(M_s = -1/2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = 1/2),$$

these are two independent two-level systems. A similar separation is valid also for light propagating along the optical axis of the crystal[3]. We shall therefore limit ourselves henceforth to a model of a nondegenerate two-level system. This makes it possible to use the spin formalism in such problems[15].

We assume further that the electromagnetic field is described classically, which is perfectly sufficient for the calculation of coherent radiation of impurity ions in a crystal. This radiation is concentrated in a very narrow interval of angles relative to the preferred direction, so that contribution of the coherent spontaneous radiation is very small[3].

We assume, finally, that in the absence of a field the level population remains practically unchanged during the time intervals in question, and the quantum coherence (the off-diagonal elements of the density matrix) attenuate exponentially with a time T_2. This is perhaps the crudest assumption, but this is precisely the assumption which makes it possible to carry through to conclusion the calculation of the attenuation of the light pulses that follow directly an arbitrary number of exciting pulses.

1. FUNDAMENTAL EQUATIONS

The state of the impurity ion in the crystal is described by a density matrix $\hat{\rho}(t)$ satisfying the equation

$$i\hbar \frac{\partial \hat{\rho}}{\partial t} = [\hat{H}_0 + \hat{V}, \hat{\rho}] + i\hbar \left(\frac{\partial \hat{\rho}}{\partial t}\right)_\gamma . \quad (1.1)$$

The unperturbed Hamiltonian for a two-level system is

$$\hat{H}_0 = -\hbar\omega_{10}\hat{S}_z, \quad (1.2)$$

where $\hbar\omega_{10}$ is the energy interval between the resonant states $|1\rangle$ and $|0\rangle$, and $\hat{S}_Z$ is the operator of the fictitious spin $S = 1/2$. For interaction with an electric field $\mathbf{E}$ we have, in the dipole approximation,

$$\hat{V} = -\hat{\mathbf{d}}\mathbf{E}, \quad (1.3)$$

where

$$\hat{\mathbf{d}} = \mathbf{d}_{01}\hat{S}_+ + \mathbf{d}_{10}\hat{S}_-, \quad \hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y. \quad (1.4)$$

The term $i\hbar(\partial\hat{\rho}/\partial t)_\gamma$ describes the relaxation processes; under the assumptions made here it takes the form

$$\left(\frac{\partial \hat{\rho}}{\partial t}\right)_\gamma = -\gamma(\rho_{01}\hat{S}_+ + \rho_{10}\hat{S}_-), \quad \gamma = \frac{1}{T_2}. \quad (1.5)$$

At the initial instant of time the ion is in the ground state, and therefore the initial condition for Eq. (1.1) is written in the form

$$\hat{\rho}^{(0)} = 1/2 \cdot \hat{1} + \hat{S}_z. \quad (1.6)$$

Assume that a series of coherent light pulses whose electric field $\mathbf{E}$ is represented by the expression

$$\mathbf{E}_a(\mathbf{R}_j, t) = \mathrm{Re}\,\{\vec{\mathscr{E}}_a(\mathbf{R}_j, t)\exp[i\mathbf{k}_a\mathbf{R}_j - i\omega t + i\varphi_a]\} \quad (1.7)$$

passes through the crystal in sequence. Here $\mathbf{R}_j$ is the coordinate of the center of gravity of the j-th ion in the crystal, $\vec{\mathscr{E}}_a$ is the slowly-varying amplitude of the field (a real quantity); $\mathbf{k}_a$ is the wave vector and ω is the field frequency and is close to the transition frequency ω_{10} in the ion; φ_a is an arbitrary initial phase; a = 1, 2, 3... numbers the sequence of pulses. The field of the light pulses will be assumed fixed and consequently the concentration of the impurity ions will be assumed quite small (see the criterion (4.7)), so that the electromagnetic field produced in the medium is much weaker than the external field.

In fixed fields (1.7), the solution of (1.1) makes it possible to calculate the induced dielectric polarization of the medium at the point $\mathbf{R}$ by means of the formula

$$\mathbf{P} = \sum_j \mathrm{Sp}\,\{\hat{\rho}(\mathbf{R}_j)\hat{\mathbf{d}}\}\,\delta(\mathbf{R} - \mathbf{R}_j), \quad (1.8)$$

where the summation is over all the impurity ions.

We note that the time T_2 describes the homogeneous broadening of the levels of the ion, whereas the deviation of the transition frequency ω_{10} from a certain mean value ω_0 leads to inhomogeneous broadening of the levels. If $g(\omega_{10} - \omega_0)$ is the distribution function of the transition frequencies of the ions, then the polarization (1.8) can be rewritten in the form

$$\mathbf{P} = N_0 \int d\omega_{10}\, g(\omega_{10} - \omega_0)\,\mathrm{Sp}\,\{\hat{\rho}(\mathbf{R})\hat{\mathbf{d}}\}, \quad (1.9)$$

where N_0 is the impurity concentration and the function $g(\omega)$ is normalized to unity. With the aid of (1.9) we can solve Maxwell's equations for a nonmagnetic medium:

$$\mathrm{rot\,rot}\,\mathbf{E} + \frac{\partial^2}{\partial t^2}\frac{1}{c^2}(\mathbf{E} + 4\pi\mathbf{P}) = 0, \quad \mathrm{div}(\mathbf{E} + 4\pi\mathbf{P}) = 0, \quad (1.10)$$

and for a definite geometry we can calculate the field and the intensity of the coherent radiation resulting from the action of the incident light.

2. SOLUTION OF THE EQUATION FOR THE DENSITY MATRIX

1. Case of Strong External Field

The solution of Eq. (1.1) for specified external fields is sought in the approximation $\delta_a \ll T_2$ (δ_a is the pulse duration). Then the relaxation term (1.5) can be neglected during the time of action of the pulse. We introduce the operator

$$\hat{\Delta} = -\omega\hat{S}_z, \quad (2.1)$$

and subject the sought density matrix $\hat{\rho}(t)$ to the unitary transformation

$$\tilde{\hat{\rho}}(t) = e^{i\hat{\Delta}(t-t_a)}\hat{\rho}(t)\,e^{-i\hat{\Delta}(t-t_a)}, \quad (2.2)$$

where t_a are the instants when the external field is turned on. Taking (2.2) into account, we obtain for the equation

$$i\hbar \frac{\partial \tilde{\hat{\rho}}}{\partial t} = [\hat{\sigma} + \tilde{\hat{V}}, \tilde{\hat{\rho}}], \quad (2.3)$$

where

$$\hat{\sigma} = \hat{H}_0 - \hbar\hat{\Delta}, \quad \tilde{\hat{V}} = e^{i\hat{\Delta}(t-t_a)}\hat{V}(t)\,e^{-i\hat{\Delta}(t-t_a)}. \quad (2.4)$$

After averaging[1] in Eq. (2.3) over the time interval $(\mathscr{E}_a d/\hbar)^{-1} \gg t \gg \omega^{-1}$, we obtain

$$i\hbar \frac{\partial \tilde{\hat{\rho}}}{\partial t} = [\hat{\sigma} + \hat{U}, \tilde{\hat{\rho}}], \quad (2.5)$$

where

$$\hat{U} = -1/2\,\mathbf{d}_{01}\vec{\mathscr{E}}_a \exp(-i\mathbf{k}_a\mathbf{R}_j - i\varphi_a + i\omega t_a)\,\hat{S}_+ - 1/2\,\mathbf{d}_{10}\vec{\mathscr{E}}_a \exp(i\mathbf{k}_a\mathbf{R}_j + i\varphi_a - i\omega t_a)\,\hat{S}_-.$$

In strong electric fields, when the inequality $\mathscr{E}_a d \gg \hbar/T_2^*$ is satisfied (T_2^* characterizes the inhomogeneous broadening, which is the largest in this case), Eq. (2.5) is replaced by

$$i\hbar \frac{\partial \tilde{\hat{\rho}}}{\partial t} = [\hat{U}, \tilde{\hat{\rho}}]. \quad (2.6)$$

The solution of (2.6) can be expressed by the formula

$$\tilde{\hat{\rho}}(t) = e^{-i\hat{F}_a\Theta_a}\hat{\rho}(t_a)\,e^{i\hat{F}_a\Theta_a}, \quad (2.7)$$

where

$$\Theta_a = \frac{d}{\hbar}\int_{t_a}^{t} \mathscr{E}_a(t')\,dt', \quad (2.8)$$

$$\hat{F}_a = -1/2\exp(-i\mathbf{k}_a\mathbf{R}_j - i\varphi_a + i\omega t_a)\hat{S}_+ - 1/2\exp(i\mathbf{k}_a\mathbf{R}_j + i\varphi_a - i\omega t_a)\hat{S}_-. \quad (2.9)$$

Finally, the density matrix in a strong field is given by the expression

$$\hat{\rho}(t) = e^{-i\hat{\Delta}(t-t_a)}\,e^{-i\hat{F}_a\Theta_a}\hat{\rho}(t_a)\,e^{i\hat{F}_a\Theta_a}\,e^{i\hat{\Delta}(t-t_a)}. \quad (2.10)$$

2. Without External Field

In the absence of an external field, Eq. (1.1) simplifies to

[1] Such an averaging means that we neglect the rapidly-varying part of V(t).

$$i\hbar\frac{\partial\hat{\rho}}{\partial t}=[\hat{H}_0,\hat{\rho}]+i\hbar\left(\frac{\partial\hat{\rho}}{\partial t}\right)_\gamma. \qquad (2.11)$$

A solution of (2.11) that allows for relaxation in the form (1.5) is

$$\hat{\rho}(t)=e^{-i\hat{H}_0(t-t_0)/\hbar}\hat{\rho}(t_0)\,e^{i\hat{H}_0(t-t_0)/\hbar}\varkappa(t-t_0)$$
$$\varkappa(t-t_0)=\begin{cases}1 & i=k,\\ e^{-\gamma(t-t_0)} & i\neq k.\end{cases} \qquad (2.12)$$

Thus, the obtained solutions (2.10) and (2.12) enable us to calculate the density matrix, and consequently also the induced polarization of the medium (1.9) for an arbitrary number of consecutively acting strong-field light pulses. Indeed, at the initial instant of time the state of each ion is determined by expression (1.6); after the action of the first pulse this state is described by a density matrix (2.10), in which $\hat{\rho}(t_a)=\hat{\rho}^{(0)}$; before the action of the second pulse this state is expressed by formula (2.12), in which $\hat{\rho}(t_0)$ should be replaced by the matrix (2.10) with allowance for the statement made above, and so on.

We consider below the case of three light pulses.

3. INDUCED POLARIZATION

Assume that the crystal is acted upon successively by three light pulses at the instants t_1, t_2, and t_3; the pulses have respective durations δ_1, δ_2, and δ_3 (see Fig. 1). We place the origin at the center of gravity of any particular ion and assume that for this ion $t_1 = 0$. Then for ions with an arbitrary coordinate $\mathbf{R}_j$ we have

$$t_1=\mathbf{k}_1\mathbf{R}_j/\omega,\quad t_2=\mathbf{k}_2\mathbf{R}_j/\omega+\tau_1+\delta_1,\ t_3=\mathbf{k}_3\mathbf{R}_j/\omega+\tau_1+\tau_2+\delta_1+\delta_2, \qquad (3.1)$$

where τ_1 and τ_2 are the distances between pulses.

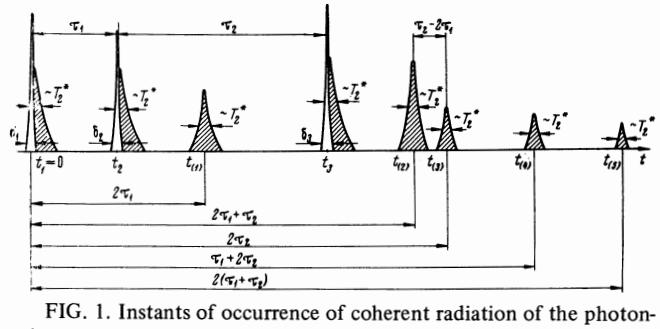

FIG. 1. Instants of occurrence of coherent radiation of the photon-echo type upon excitation by three coherent pulses of light: $\mathcal{E}_a$–slowly varying amplitude of the external field; t_a–instance when the field is turned on; $t_{(b)}$–instance of echo occurrence; τ_1 and τ_2–distances between the exciting pulses; shaded–coherent radiation of the crystal.

Following the procedure described in Sec. 2, we find the density matrix $\hat{\rho}(t)$, after which it is easy to calculate the average dipole moment induced in the j-th ion. The induced dipole moment then contains eight terms, which determine the coherent radiation of the medium. Three pulses of the medium occur simultaneously with the external ones, and since their intensity is small compared with the incident pulses, they are not considered here. The remaining five are responsible for the formation of the signals of the photon-echo type, and are the only ones considered from now on. To simplify the formulas that follow, we assume that the pulse durations δ_a are small compared with the intervals τ_a between them, and let furthermore $\tau_2 > \tau_1$ (the case $\tau_2 < \tau_1$ is considered in Sec. 5). With respect to the light pulses, we shall assume also that they are linearly polarized along the optical axis of the crystal and propagate perpendicular to it. Then $k_a = \omega n_e(\omega)/c$, where $n_e(\omega)$ is the refractive index of the extraordinary wave. Substituting the obtained dipole moment in (1.9), after integration with respect to the frequencies, we obtain the following final expression for the polarization:

$$P(\mathbf{R},t)=N_0 d\sum_{b=1}^{5}\mathrm{Re}\left\{\mathcal{P}_b(\mathbf{R},t)\exp\left[i\frac{\omega_0}{\omega}\mathbf{k}_{(b)}\mathbf{R}-i\omega_0 t\right]\right\}, \qquad (3.2)$$

$$\mathcal{P}_b(\mathbf{R},t)=f_b g\left(t-\frac{\mathbf{k}_{(b)}\mathbf{R}}{\omega}-t_{(b)}\right)\cdot$$
$$\times\exp\left[-\gamma\left(t-\frac{\mathbf{k}_{(b)}\mathbf{R}}{\omega}-T_{(b)}(\mathbf{R})+i\Delta\omega t_{(b)}+i\Phi_{(b)}\right)\right], \qquad (3.3)$$

where g(t) is the autocorrelation function connected with the distribution function $g(\omega)$ by means of a Fourier transformation, $\Delta\omega = \omega_0 - \omega$, and d is the reduced matrix element. The time in expressions (3.2) and (3.3) is $t > t_3(R)$ for all the terms with the exception of b = 1, for which $t > t_2(R)$.

The values of the quantities $t_{(b)}$, $T_{(b)}$, $\mathbf{k}_{(b)}$, and $\Phi_{(b)}$ are summarized in the table. The data in the table coincide with the results obtained earlier by one of the authors[9] by another method, which is suitable for the calculation of coherent radiation produced under the influence of an arbitrary number of light pulses of finite duration without allowance for the attenuation processes. The same results are obtained also by calculation of the coherent radiation by the Dicke method, used by Kopvillem[16] to investigate "stimulated light echo." In[16] there are discussed also certain features of incoherent spontaneous radiation, which we do not

Parameters of polarization of a medium excited by three coherent light pulses

b	$t_{(b)}$	$\mathbf{k}_{(b)}$	f_b	$T_{(b)}(\mathbf{R})$	$\tau_{(b)}$	$\Phi_{(b)}$
1	$2\tau_1$	$2\mathbf{k}_2-\mathbf{k}_1$	$\sin\theta_1\sin^3(\theta_2/2)$	$2(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}/\omega$	$2\tau_1$	$2\varphi_2-\varphi_1-\pi/2$
2	$2\tau_1+\tau_2$	$\mathbf{k}_3+\mathbf{k}_2-\mathbf{k}_1$	$^1/_2\sin\theta_1\sin\theta_2\sin\theta_3$	$\tau_2+2(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}/\omega$	$2\tau_1$	$\varphi_3+\varphi_2-\varphi_1-\pi/2$
3	$2\tau_2$	$2\mathbf{k}_3-2\mathbf{k}_2+\mathbf{k}_1$	$\sin\theta_1\sin^2(\theta_2/2)\sin^2(\theta_3/2)$	$2(\mathbf{k}_2-\mathbf{k}_3)\mathbf{R}/\omega$	$2\tau_2$	$2\varphi_3-2\varphi_2++\varphi_1+\pi/2$
4	$\tau_1+2\tau_2$	$2\mathbf{k}_3-\mathbf{k}_2$	$\cos\theta_1\sin\theta_2\sin^2(\theta_3/2)$	$\tau_1+2(\mathbf{k}_2-\mathbf{k}_3)\mathbf{R}/\omega$	$2\tau_2$	$2\varphi_3-\varphi_2-\pi/2$
5	$2(\tau_1+\tau_2)$	$2\mathbf{k}_3-\mathbf{k}_1$	$\sin\theta_1\cos^2(\theta_2/2)\sin^2(\theta_3/2)$	$2(\mathbf{k}_1-\mathbf{k}_3)\mathbf{R}/\omega$	$2(\tau_1+\tau_2)$	$2\varphi_3-\varphi_1-\pi/2$

Note. Here b–index of coherent pulse of the medium; $\mathbf{k}_{(b)}$–wave vector; $t_{(b)}$–instants of time at which the pulses are maximal; f_b–factor determining the intensity; $\Phi_{(b)}$–phases; $\tau_{(b)}$–attenuation parameters (see the text, formula (3.2)).

second and third pulses, and also for the echo from the first and third pulses.

However, simultaneous observation of all five pulses of the coherent radiation of the crystal can be realized only if the condition for the spatial synchronism is satisfied for all the vectors $\mathbf{k}_{(b)}$. It is easy to see that the case of total spatial synchronism is realized for the vectors $\mathbf{k}_{(b)}$ and $\mathbf{k}_{(a)}$ lying on a single straight line, with

$$\mathbf{k}_{(b)} = \mathbf{k}_a, \quad b = 1 \div 5. \tag{5.1}$$

The condition (5.1), incidentally, can be satisfied also for small angles between the vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$. The general picture of the occurrence of the photon echo for this case is shown in Fig. 1. Such a sequence of pulses is observed if $\tau_2 > 2\tau_1$. On the other hand, if $\tau_1 < \tau_2 < 2\tau_1$, then the pulses with b = 2 and b = 3 "interchange places," that is, first to come is the pulse with b = 3 and then already with b = 2. At $\tau_2 = 2\tau_1$ both echo signals merge into one, and interference is possible.

The intensities of the coherent-radiation pulses appearing in the crystal depend on the "rotation angles" θ of the dipole moment by the electric field of the light wave (see the values of f_b in the table). The usual photon echo is maximal, as expected, if the first and second pulses are respectively $\pi/2$ and π pulses. The "stimulated echo" is maximal when all three pulses are $\pi/2$ pulses. We note that the intensity of the echo from the second and third pulses depends on the "angle of rotation" by the first pulse. Thus, for example, if the first pulse is a $\pi/2$ pulse, then there will be no photon echo with b = 4 at all. Conversely, the echo is maximal if the first pulse is a π pulse, the second a $\pi/2$ pulse, and the third a π pulse. In exactly the same manner, the intensity of the echo from the first and third pulses (b = 5) depends on θ_2. There will be no pulse with b = 5 if the second pulse is a π pulse.

In the case $\tau_2 < \tau_1$ the usual photon echo occurs already after the third pulse, and therefore the pulse with b = 3 will be missing, and only the photon echo from each pair of pulses and the "stimulated echo" remain. In addition, the factor f_1, which determines the intensity of the ordinary photon echo, will contain a function of the angle θ_3:

$$f_1 = \sin\theta_1 \sin^2\frac{\theta_2}{2}\cos^2\frac{\theta_3}{2}. \tag{5.2}$$

We see thus that the optimal conditions for the observation of certain pulses of coherent spontaneous radiation are not the same as for others.

At arbitrary angles between the vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$, the condition of spatial synchronism for the coherent state of the ions and the radiation field is not satisfied, and therefore the intensity decreases sharply by a factor $(L/\lambda)^2$. However, the spatial synchronism for the "stimulated echo" (b = 2) and the pulse with b = 3 can be reconstructed at certain angles between $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ (see Fig. 2). Thus, the synchronism of the "stimulated echo" is realized for the directions

$$1)\ \mathbf{k}_{(2)} = \mathbf{k}_3, \quad \text{if} \quad \mathbf{k}_1 = \mathbf{k}_2, \tag{5.3}$$

In case 1) the ordinary echo will be observed, and in case 2) echo from the first and third pulses will be observed in the direction $\mathbf{k}_1$. For the pulse with b = 3 we have

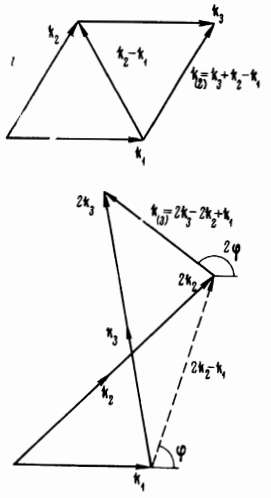

FIG. 2. Vector diagram illustrating the directions of the spatial synchronism for "stimulated echo" (a) and a pulse with b = 3(b).

$$\begin{aligned} &2)\ \mathbf{k}_{(2)} = \mathbf{k}_2, \quad \text{if} \quad \mathbf{k}_1 = \mathbf{k}_3, \\ &3)\ \mathbf{k}_{(2)} = -\mathbf{k}_1, \quad \text{if} \quad \mathbf{k}_2 = -\mathbf{k}_3. \end{aligned} \tag{5.4}$$

If case 1) of (5.4) is realized, there will be produced in the direction $\mathbf{k}_2$ also an echo from the second and third pulses, while in case 3) of (5.3) or 2) of (5.4), only pulses with b = 2 and b = 3 respectively are realized.

Let us estimate the obtained expressions for the ions Cr^{3+} in Al_3O_3: $T_2^* = 10^{-10}$ sec, corresponding to a line width 0.1 cm^{-1} at T = 4.2°K and $d \sim 10^{-20}$ $g^{1/2}\,cm^{-1/2}\,sec^{-1}$; we then have for the electric field $\mathscr{E}_a \gg \hbar/T_2^* d \sim 3 \times 10^5$ V/cm. The pulse duration for such fields should be of the order of $\delta_a \sim \pi\hbar/\mathscr{E}_a d \ll 3 \times 10^{-10}$ sec. In order for the effect of multiple photon echo not to appear at fields 3×10^6 V/cm and at durations $\delta_a \sim 10^{-11}$ sec it is necessary to choose the ion concentration and the sample length such that $LN_0 \ll \mathscr{E}_a \lambda \delta_a \sqrt{\epsilon_{\parallel}}/4\pi^2 d T_2^* \sim 3 \times 10^{17}\ cm^{-2}$.

It should be noted that if (5.1) is satisfied then our results are valid for Cr^{3+} ions in Al_2O_3 not only for light that is linearly polarized along the optical axis of the crystal. All the foregoing applies also in this case for light propagating along the optical axis. For a wave circularly polarized in the right-hand direction there are separated in "pure form" the independent transitions

$$\begin{aligned} &1)\ \mathbf{k}_{(3)} = \mathbf{k}_1, \quad \text{if} \quad \mathbf{k}_3 = \mathbf{k}_2, \\ &2)\ (\widehat{\mathbf{k}_{(3)}, \mathbf{k}_1}) = 2\varphi, \quad \text{if} \quad \varphi = (\widehat{\mathbf{k}_1, 2\mathbf{k}_2 - \mathbf{k}_1}). \end{aligned}$$

and for a left-hand circularly polarized wave we get the transition

$$\begin{aligned} &{}^4A_2(M_s = {}^3/_2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = {}^1/_2), \\ &{}^4A_2(M_s = -{}^1/_2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = -{}^1/_2), \end{aligned}$$

The authors are most grateful to A. M. Afanas'ev Yu. A. Bykovskiĭ and V. M. Galitskiĭ for help in the work and for a useful discussion.

$$\begin{aligned} &{}^4A_2(M_s = {}^1/_2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = {}^1/_2), \\ &{}^4A_2(M_s = -{}^3/_2) \leftrightarrow {}^2E(\bar{E})\,(M_{s'} = -{}^1/_2). \end{aligned}$$

[1] U. Kh. Kopvillem and V. R. Nagibarov, FMM 15, 313 (1963).

consider here because it is small compared with the coherent radiation.

4. FIELD AND INTENSITY OF COHERENT RADIATION

To find the electromagnetic field it is necessary to solve Maxwell's equations (1.10) with allowance for the polarization (3.2) and for the definite geometry of the crystal. Let the crystal be a layer of thickness L, let its surfaces be normal to the vector $\mathbf{k}_{(b)}$, and let the direction of the z axis be parallel to the vector $\mathbf{k}_{(b)}$. The amplitude of the macroscopic polarization $\mathcal{P}_b$ is a slow function of the point t and of the distance z, so that the electric field will also be sought in the form

$$E(z,t)=\sum_{b=1}^{5}\mathrm{Re}\{\mathcal{E}_b(z,t)\exp[ik_{(b)}z-i\omega_0 t]\},\qquad(4.1)$$

where $\mathcal{E}_b(z,t)$ is a slowly varying function of z and t compared with the wavelength of light and the period of its oscillation, $k_{(b)}=\omega_0/v$, and $v=c/n_e(\omega_0)$. Substituting (3.2) and (4.1) in (1.10), we obtain for the slowly-varying amplitude of the field the following equation:

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}\right)\mathcal{E}_b(z,t)=i\frac{2\pi\omega_0}{n_e^2}\mathcal{P}_b(z,t).\qquad(4.2)$$

After changing over to new variables

$$\tau=t-z/v,\quad \xi=z\qquad(4.3)$$

Eq. (4.2) takes the form

$$v\frac{\partial}{\partial\xi}\mathcal{E}_b(\xi,\tau)=i\frac{2\pi\omega_0}{n_e^2}\mathcal{P}_b(\xi,\tau).\qquad(4.4)$$

If

$$L\ll v\tau\qquad(4.5)$$

the values of $T_{(b)}(R)$ in (3.3) can be chosen at the point R = 0, and then we write the solution of (4.4) in the form

$$\mathcal{E}_b\left(t-\frac{z}{v}\right)=i\frac{2\pi\omega_0 z}{v\varepsilon_\parallel}\mathcal{P}_b\left(t-\frac{z}{v}\right),\qquad(4.6)$$

where $\epsilon_\parallel=n_e^2(\omega_0)$, and $0\le z\le L$. When (4.5) is satisfied, we have at all points of the crystal $t_2>t_1$ and $t_3>t_2$, and consequently the external pulses do not overlap in the crystal.

From (4.6) we obtain the condition for neglecting the reaction of the medium on the external field:

$$T_2^*\omega_0 zN_0 d/v\varepsilon_\parallel\delta_a\ll\mathcal{E}_a,\quad d\mathcal{E}_a\delta_a/\hbar\sim 1,\qquad(4.7)$$

since $\mathcal{P}_b\sim N_0 d$. For the radiation flux density we obtain

$$I(z,t)=\frac{v}{8\pi}\mathcal{E}_b\mathcal{E}_b^*=\frac{\pi(\omega_0 zN_0 d)^2}{2v\varepsilon_\parallel^2}f_b^2 g^2\left(t-\frac{z}{v}-t_{(b)}\right)\cdot$$
$$\times\exp\left\{-2\gamma\left(t-\frac{z}{v}-T_{(b)}(0)\right)\right\}.\qquad(4.8)$$

The values of the energy flux density will be maximal at $t-z/v=t_{(b)}$ and equal to

$$I_b^{max}=\frac{\pi(\omega_0 zN_0 d)^2}{2v\varepsilon_\parallel^2}f_b^2 e^{-2\gamma\tau_{(b)}}.\qquad(4.9)$$

It is now easy to calculate the energy of the coherent pulse by means of the formula

$$W=S\int_{-\infty}^{\infty}dt\,I(z,t),\qquad(4.10)$$

where S is the cross section of the beam or the transverse dimensions of the plate. In the case when the inhomogeneous broadening is dominating ($T_2^*\ll T_2$), the correlation function has a δ-like character, so that after substituting (4.8) in (4.10) we obtain

$$W(z)=\beta S^2\frac{(\omega_0 zN_0 d)^2}{v\varepsilon_\parallel^2}T_2^* f_b^2\exp\{-2\gamma(t_{(b)}-T_{(b)}(0))\},\qquad(4.11)$$

where $\beta=\pi/2$ for a Lorentz line shape and $\beta=\pi\sqrt{\pi}$ for a Gaussian shape.

5. DISCUSSION OF RESULTS

In Sec. 3 we obtain a general expression for the macroscopic polarization (3.2) produced in a crystal when three coherent light pulses pass through it. Whereas excitation of the medium by two pulses causes the coherent "response" to appear at the instant of time $2\tau_1$, where τ_1 is the interval between the pulses, in the case of three light pulses the picture becomes much more complicated. In the general case the number of coherent "responses" of the medium is equal to 5. Indeed, the term with b = 1 in (3.2) describes the usual photon echo, which appears earlier than the third pulse, since $\tau_2>\tau_1$. The polarization responsible for the appearance of the photon echo is characterized by the wave vector $(2\mathbf{k}_2-\mathbf{k}_1)\omega_0/\omega$ and attenuates like $\exp(-\tau_{(1)}/T_2)=\exp(-2\tau_1/T_2)$. We emphasize that the radiation occurs at a frequency ω_0 that differs from the frequency of the external incident pulses.

At the instant of time $2\tau_1+\tau_2$ there appears in the medium polarization with a wave vector $(\mathbf{k}_3+\mathbf{k}_2-\mathbf{k}_1)\omega_0/\omega$ (b = 2), the attenuation of which does not depend on the interval τ_2 between the second and third pulses. This is the "stimulated echo." Such an unusual result is connected with the fact that under the influence of the second pulse a change takes place in the relaxation time of the quantum coherence of the medium. In fact, under ordinary conditions the coherent state relaxes with a characteristic transverse-relaxation time T_2, something we took into account as the attenuation of the near-diagonal element of the density matrix. The second light pulse transfers part of the "phase memory" of the medium to the diagonal density-matrix elements, in other words, to a population that attenuates with the longitudinal relaxation time T_1 of the medium. In paramagnetic crystals the inequality $T_1\gg T_2$ is usually satisfied, and therefore the damping of the "stimulated echo" will be significant only in the intervals between the second and third pulses, which are comparable with T_1.

The term with b = 3 describes the coherent state of the matter, stored by the usual photon echo and "realized" by the third light pulse. This attenuation is determined both by the interval between the ordinary photon echo and the third pulse, and by the attenuation of the photon echo itself $\tau_{(3)}=2(\tau_2-\tau_1)+2\tau_1=2\tau_2$. It is interesting to note that at the instant of time $2\tau_1$ there is essentially no external strong field at all in the crystal (we have assumed that the field produced by the medium is much weaker than the external field). Nonetheless the crystal is in this case in the superradiating state produced when the first two pulses of light passed. This coherent state is then "realized" by the third pulse at the instant of time $2\tau_2$.

Analogously, the terms with b = 4 and b = 5 in (3.2) are responsible for the photon echo from the

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