## Excitation of light (photon) echo signals by a sequence of traveling and standing waves

V. A. Zuĭkov, V. V. Samartsev, and E. A. Turiyanskiĭ

Kazan' Physicotechnical Institute, USSR Academy of Sciences (Submitted 26 December 1980; resubmitted 15 April 1981) Zh. Eksp. Teor. Fiz. 81, 653–664 (August 1981)

In the special case of three-pulse action on a resonant medium (ruby,  ${}^{4}A_{2}-{}^{2}E(\overline{E})$  transition) it is shown that under certain conditions a sequence of optical traveling-wave pulses can be reversed in space at a certain instant of time by a standing wave. The parameters of the reversed responses of a resonant system (which have been named reversed echo signals) do not depend on the direction of action of the standing wave, and a much better signal/noise ratio can be obtained in the detection of the signal. This has made it possible to measure in ruby, for the first time, the temperature dependence of the optical echo-signal intensity and its attenuation curve in a zero magnetic field. The transverse relaxation times  $T_2$  in ruby (c = 0.05 wt.%) are determined at helium temperatures and for magnetic field of various intensities. The  ${}^2E(2A)-{}^2E(\overline{E})$  spontaneous transition time is also determined. It is found that the polarization of the reversed echo signals is the same as that of the traveling waves that cause, together with the standing wave, the emission of the reversed echo signals. The shape of the reversed echo signal is investigated under three-pulse action, is investigated for the first time, and it is shown that under certain conditions the shape can be reversed in time with respect to that of the pulses of the respective traveling waves.

PACS numbers: 78.20. - e, 42.65.Gv

Light (photon) echo<sup>1,2</sup> has by now found a number of applications in optical spectroscopy,<sup>3</sup> yielded for the hyperfine and superhyperfine line structures information masked by the inhomogeneous broadening, and yielded data on the irreversible-relaxation times and on the parameters of random processes. Photon-echo signals are quite intense (in ruby, for example, they reach several dozen watts at an exciting-pulse power 200-500 kW-Ref. 4), and can therefore find technical applications. These coherent responses promise to find use in the development of optical delay lines and of optical memory cells.<sup>5</sup> The indicated spectroscopic and technical applications of photon-echo signals can be greatly expanded by using the phenomenon of reversed photon echo,<sup>6</sup> which is the coherent optical response of a resonant system to two-pulse laser action if the first pulse is a traveling wave the second a standing wave. This echo signal is emitted in a direction opposite to that of the first pulse, and is independent of the direction of the action of the standing wave. If the wave vectors of the exciting laser pulses are noncollinear, the indicated spatial regularities in the generation of reversed photon echo uncover a possibility of controlling the propagation of optical pulsed signals, reminiscent of the Thomson-Quate effect well known from acoustoelectronics.7

The possible applications of reversed photon echo signals are greatly expanded by multiple excitation of the resonant system.

This paper is devoted to a theoretical and experimental investigation of the features of generation of reversed photon echo signals under three-pulse action. It is shown to generate these signals it is necessary that the first two laser pulses be traveling waves and the last pulse be a standing wave. Since the reversed photon echo signals are emitted in a direction opposite to that of the action of the traveling waves, and are independent of the direction of the standing wave, the photomultiplier is not subjected to the background scattered light of the excited pulses, so that the signal/noise ratio is increased. Making use of this advantage of reversed echo signals, the temperature dependence and the fall-off curves of ruby-emission intensity in a zero magnetic field are obtained for the first time ever. It is shown that the polarization of the reversed photon echo signals coincides with that of the corresponding traveling wave.

## 1. THEORY OF OPTICAL COHERENT RESPONSES UPON EXCITATION OF A RESONANT SYSTEM BY A MULIPULSE SEQUENCE OF TRAVELING AND STANDING WAVES

The object investigated in our experiments was ruby. Resonance conditions were obtained for the energy transition  ${}^{4}A_{2} - {}^{2}E(\overline{E})$ . It is known<sup>2</sup> that linearly polarized light propagating along the optical *c* axis of the crystal causes simultaneous equally probable transitions between the succeeding pairs of sublevels:

$${}^{4}A_{2}(M={}^{1}/_{2}) - {}^{2}E(M'={}^{1}/_{2})$$
 ( $\sigma^{-}$  transition)  
 ${}^{4}A_{2}(M={}^{-1}/_{2}) - {}^{2}E(M'={}^{-1}/_{2})$  ( $\sigma^{+}$  transition)

This reduces the problem of calculating the optical coherent responses in such a four-level system to the two-level problem, making it possible to consider the formation of the reversed photon echo signals separately for each transition.

Assume that a two-level system of particles is subjected to resonant action by n (n = 1, 2, 3, ...) laser pulses. The electric field of the exciting pulse at the location **r** of the *j*-th particle can be represented in the form of a superposition of f (f = 1, 2, 3...) simultaneously acting linearly polarized traveling waves:

$$\mathbf{E}_{n}(\mathbf{r}_{j};t) = \sum_{j} \mathbf{e}_{j} E_{oj} \cos\left(\omega t - \mathbf{k}_{j} \mathbf{r}_{j} + \varphi_{oj}\right), \tag{1}$$

where  $\omega$  is the carrier frequency of the pulse,  $e_f$ , and

 $\mathbf{E}_{of}$  are the polarization unit vector and the amplitude of the electric field of the *f*-th wave,  $\mathbf{k}_f$  and  $\varphi_{of}$  are the wave vector and the initial phase of the *f*-th wave. The density matrix of the system at the instant of time *t* can be calculated in the interaction representation from the equation

$$\rho^{*}(t) = L \rho_{\circ} L^{-1}, \qquad (2)$$

where *L* is the system-evolution operator;  $\rho_0$  is the equilibrium density matrix and can be written in the formalism of the energy spin  $R = \frac{1}{2}$  (Ref. 8) in the form

$$\rho_0 = \rho(t=0) = \frac{1}{2^N} \prod_{j=1}^N (1-2R_{j_2}), \qquad (3)$$

N is the number of active centers, and  $R_{j3}$  is the longitudinal component of the energy spin.

Following Ref. 2, it can be shown that the system evolution operator can be written here in the form

$$L = \exp\left[-i\sum_{j}\Delta\omega_{j}(t-t_{n}-\Delta t_{n})R_{j3}\right]\exp\left[i\sum_{j}b_{nj}R_{j+}+b_{nj}R_{j-}\right]$$
$$\times \exp\left[-i\sum_{j}\Delta\omega_{j}\tau_{n-1}R_{j3}\right]\exp\left[i\sum_{j}b_{n-1,j}R_{j+}+b_{n-1,j}R_{j-}\right]$$
$$\dots \exp\left[-i\sum_{j}\Delta\omega_{j}\tau_{i}R_{j3}\right]\exp\left[i\sum_{j}b_{1j}R_{j+}+b_{1j}R_{j-}\right], \qquad (4)$$

where  $\Delta \omega_j = \omega_{0j} - \omega$ ;  $\omega_{0j}$  is the frequency of the resonant transition of the *j*-th particle;  $\omega$  is the carrier frequency of the exciting pulses, assumed to be the same for all pulses;  $R_{j\pm} = R_{1j} \pm iR_{2j}$ ;  $R_{1j}$  and  $R_{2j}$  are the transverse components of the energy spin of the *j*-th particle;  $t_n$  is the instant of application of the *n*-th exciting pulse;  $\tau_{\eta}$  is the time interval between the  $\eta$ -th and the  $(\eta + 1)$ st pulses (Fig. 1a);  $b_{\eta j}$  is the parameter of the interaction of the *j*-th particle with the  $\eta$ -th exciting pulse. If the pulse is a traveling wave, this parameter is of the form<sup>2</sup>

$$\frac{i}{2\theta_{\eta}} \exp\left[i(\mathbf{k}_{\eta}\mathbf{r}_{j}-\boldsymbol{\varphi}_{tr\eta})\right], \tag{5}$$

while in the case of a standing wave

$$\theta_{\eta} \cos \left( \mathbf{k}_{\eta} \mathbf{r}_{j} - \varphi_{st\eta} \right),$$
 (6)

where  $\varphi_{tr \eta}$  and  $\varphi_{st\eta}$  are the initial phases of the traveling and standing waves, respectively.

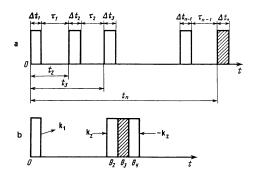


FIG. 1. Order of excitation of a resonant system by a sequence of traveling waves and a standing wave. The shaded rectangles are the standing waves, and the remaining signals are the traveling waves: a) case of n-pulse action; b) case of three-pulse action, with two of the pulses propagating counter to each other and overlapping partially.

The parameter  $\theta_{\eta}$  is the "area" of the pulse, which has in the case of a rectangular pulse the form

$$\theta_{\eta} = \frac{d}{\hbar} E_{o\eta} \Delta t_{\eta} , \qquad (7)$$

where  $E_{0\eta}$  is the amplitude of the electric field intensity of the  $\eta$ -th exciting pulse; d is the modulus of the electric dipole moment of the transition between the considered levels;  $\Delta t_{\eta}$  is the duration of the exciting pulse.

We consider now a specific case when the first m (m = 1, 2, 3, ..., n-1) pulses are traveling and the last (n-th) is a standing wave Fig. 1(a). Calculations using Eqs. (1)-(7) shows that the corresponding part of the density matrix, which contains terms with wave vectors directed opposite to the wave vectors of the first m exciting pulses, is given by

$$\rho^{*(-\mathbf{k}_m)} = \frac{1}{2^N} \prod_j [1 - (M_j^{(-\mathbf{k}_m)} + M_j^{+(-\mathbf{k}_m)})]; \qquad (8)$$

$$\boldsymbol{M}_{j}^{(-\mathbf{k}_{m})} = \frac{i}{2} R_{j+} F \exp \left\{ i \left[ -\mathbf{k}_{m} \mathbf{r}_{j} - \Delta \omega_{j} \left( t - 2t_{n} + t_{m} \right) - \varphi_{0} \right] \right\}, \tag{9}$$

$$F = \left(\prod_{p < m} \cos \theta_p\right) \sin \theta_m \left(\prod_{m < q \leq n-1} \cos^2 \frac{\theta_q}{2}\right) \cdot [1 - J_0(2\theta_n)], \quad (10)$$

where  $\mathbf{k}_m$  is the wave vector of the *m*-th (m = 1, 2, ..., n - 1) exciting pulse;  $t_m$  is the instant of time of application of the *m*-th pulse);  $t_m$  is the instant of application of the standing-wave pulse;  $\varphi_0 = -\varphi_{0m} + \varphi'_{0n} + \varphi''_{0n}$ , with  $\varphi_{0m}$  the initial phase of the *m*-th traveling waves, and  $\varphi'_{0n}$  and  $\varphi''_{0n}$  the initial phases of the two waves that form the standing wave (the last exciting pulse);  $\theta_{p(q)}$  is the area of the p(q)-th traveling-wave pulse (p < m;  $m < q \leq n-1$ );  $J_0$  is a bessel function of the first kind and zero order.

Knowledge of the density matrix of the system makes it possible to calculate the macroscopic electric dipole moment for the transitions  $\sigma^*$  and  $\sigma^-$ , and then also the electric field of the dipole emission of the system (assuming that the electric field of the exciting pulses is polarized in a plane perpendicular or nearly perpendicular to the optical axis of the crystal). As a result, the reversed photon echo emission field acquires at the instant t in the direction  $-\mathbf{k}_m$  the form

$$\mathbf{E}_{m}(t) = \frac{\omega^{2} d}{c^{2} R_{0}} F \mathbf{x} \sum_{j} \sin \left[ \omega t + \Delta \omega_{j} (t - 2t_{n} + t_{m}) + (\mathbf{k} + \mathbf{k}_{m}) \mathbf{r}_{j} - \mathbf{k} \mathbf{R}_{0} + \varphi_{0} \right],$$
(11)

where x is the unit vector of the laboratory frame in the X-axis direction, with the Z axis directed along the optical axis of the crystal;  $R_0$  is the distance from the origin of the laboratory frame to the observation point; d is the modulus of the electric dipole moment for the transitions  $\sigma^*$  and  $\sigma^-$ .

Expression (11) is substantially different from zero for each concrete m in the case  $\mathbf{k} = -\mathbf{k}_m$ . Thus, when a sample is excited in the indicated sequence there are formed in it n-1 reversed photon echo signals, which propagate in directions opposite to the action of the first n-1 exciting pulses at the instants of time  $t^{(m)} = 2t_n$  $-t_m$ . As a result it is possible to generate in this excitation regime n-1 reversed photon echo signals at various instants of time in directions opposite to the propagation direction of the corresponding traveling waves. This circumstance seems quite important in problems dealing with the control of the motion of the light beam. We shall dwell now on several particular cases.

1. n=2, with the first pulse a traveling wave and the second a standing wave. In this case there is a reversed photo echo signal in the  $-k_1$  direction generated simultaneously with the primary photon echo (PPE) (emitted in the direction  $k_{PPE} = 2k_2 - k_1$ , where  $k_1$  and  $k_2$  are the wave vectors of the first and second exciting pulses), in agreement with the results of the theoretical paper.<sup>10</sup> The amplitude of the reversed photon echo emission field is proportional to

 $F = \sin \theta_1 [1 - J_0(2\theta_2)].$ 

It is well known that the results obtained by the method of evolution operators agree with the results obtained by solving equations of the Bloch type.<sup>11</sup> In the Blochequations procedure the echo amplitude turns out to be proportional to the relaxation factor, which in our case is written in the form  $\exp\{-2\tau_1/T_2\}$ , where  $\tau_1$  is the time interval between the exciting pulses and  $T_2$  is the time of irreversible transverse relaxation.

We note that under conditions of partial overlap of the two traveling waves that make up the standing wave there can also be formed an echo signal in the  $-k_1$  direction, since this case is intermediate between the two- and three-pulse actions (if  $k_2$  is antiparallel to  $k_3$ —Ref. 12). Calculation shows that the amplitude of the radiated field is in this case proportional to

$$\sin \theta_{i} \left\{ \frac{1}{2} \left[ 1 - J_{0}(2\theta_{3}) \right] \left[ 1 + \cos \theta_{2} \cos \theta_{4} \right] \right.$$

$$\left. + \frac{1}{2} J_{2}(2\theta_{3}) \left[ \cos \theta_{2} \cos \theta_{4} - 1 \right] + J_{0}(2\theta_{3}) \sin \theta_{2} \sin \theta_{4} \right\},$$

$$(12)$$

where  $J_1$  and  $J_2$  are Bessel functions of the first kind and of first and second order, respectively. The areas of the corresponding pulses and their overlap sections are shown in Fig. 1b. The region of overlap of the second and third pulses corresponds to a standing wave  $(\theta_3)$  whose action, together with the traveling wave, leads to the formation of the reversed photon echo. If  $\theta_2 = \theta_4 = 0$ , Eq. (12) corresponds to the expression for the case n = 2. At  $\theta_3 = 0$ , Eq. (12) goes over into the expression for the stimulated photon echo signal in the three-pulse case.<sup>12</sup>

2. n = 3. The first two pulses are traveling waves in the directions  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , and the third is a standing wave. Calculation shows that at the instants of time  $2(\tau_1 + \tau_2)$  and  $\tau_1 + 2\tau_2$ , where  $\tau_1$  and  $\tau_2$  are the time intervals between the first and second and between the second and third exciting pulses, the system emits reversed photon echo signals in the directions  $-\mathbf{k}_1$  and  $-\mathbf{k}_2$ , respectively, with the reversed photon echo signal in the  $-\mathbf{k}_1$  direction proportional to

 $\sin\theta_1\cos^2\frac{\theta_2}{2}[1-J_0(2\theta_3)],$ 

and in the  $-\mathbf{k}_2$  direction to

 $\cos\theta_1\sin\theta_2[1-J_0(2\theta_3)].$ 

In addition to the indicated reversed photon echo signals, the resonant medium will emit the usual primary photon echo and stimulated photon echo signals. Moreover, calculation similar to the foregoing, shows that the primary and simulated photon echo signals can become reversed in this excitation regime. To this end, the resonant medium is acted upon by a standing-wave pulse immediately after the generation of the primary and stimulated photon echo signals due to the action of two or three traveling waves. As a result, at the instants  $2\tau_2$  and  $\tau_2 + 2\tau_3$  respectively the system will emit in the directions  $k_1 - 2k_2$  and  $k_1 - k_2 - k_3$  reversed photon echo signals due to the action of the primary and stimulated photon echo signals and of the standing wave. The radiation field in the direction  $k_1 - 2k_2$  is proportional to

$$\sin\theta_1\sin^2\frac{\theta_2}{2}\left[1-J_0(2\theta_3)\right],$$

and in the direction  $\mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3$  to

 $\sin \theta_1 \sin \theta_2 \sin \theta_3 [1 - J_0 (2\theta_1)].$ 

In particular the reversed primary photon echo can be regarded as an inverted variant of the reconstructed echo signal.<sup>12+13</sup> We consider now the question of the photon-echo signal polarization when the first m (m = 1, 2,...,n-1) exciting pulses are traveling waves, and the last pulse is a standing wave (questions connected with the polarization of the photon echo by standing waves only are considered in detail in Ref. 14).

Let each of the exciting pulses be linearly polarized at an angle  $\psi_m$  to the axis (which lies in the plane perpendicular to the crystal optical axis). In a calculation similar to that in Ref. 2, the amplitude of the pulse electric field intensity is expressed in terms of the projections on the axes X and Y. The calculation leads to the following expression for the field of the reversed photon echo signal due to the action of the *m*-th pulse and of the standing wave:

$$\mathbf{E}_{m} = \frac{\omega^{2} d}{c^{2} R_{0}} F(\mathbf{x} \cos \psi_{m} + \mathbf{y} \sin \psi_{m})$$

$$\times \sum_{j} \sin \left[ \omega t + \Delta \omega_{j} (t - 2t_{n} + t_{m}) + (\mathbf{k} + \mathbf{k}_{m}) \mathbf{r}_{j} - \mathbf{k} \mathbf{R}_{0} + \varphi_{0} \right], \qquad (13)$$

where F is determined from (10). It follows from (13) that the polarization of the reversed photon echo signal coincides with the polarization of the corresponding traveling wave<sup>1</sup> (whose action together with that of the standing wave generated the reversed photon echo). We note that in the case when the exciting pulses act at appreciable angles to the optical axis, it becomes necessary to take into account the presence of an ordinary and an extraordinary wave in the crystal.<sup>16</sup>



FIG. 2. Oscillograms of the reversed photon echo signals in ruby  $[{}^{4}A_{2} - {}^{2}E(E)]$  in the case of three-pulse excitation (the third pulse from the left is the standing wave). The first two from the right are the reversed photon echo signals;  $\tau_{1} = 24$  nsec;  $\tau_{2} = 36$  nsec;  $H_{0} = 400$  Oe; markers—10 nsec.

Since the exciting pulses are quite long  $(\Delta t_{\eta} = 10 \text{ nsec}, where \eta = 1, 2, ..., n)$ , it is important to take into account the dephasing of the electric dipoles during the time of the exciting pulses, and not only in the interval between the pulses.<sup>4</sup> Under conditions when  $T_2^* < \Delta t < T_1$ , and  $T_2^* < \Delta t < T_2$ , where  $T_1$  is the longitudinal relaxation time and  $T_2$  and  $T_2^*$  are the times of the transverse irreversible and reversible relaxations, the evolution operator of the system takes the form

$$L = \exp\left[-i\sum_{j}\Delta\omega_{j}(t-t_{n}-\Delta t_{n})R_{js}\right]\exp\left[i\sum_{j}a_{nj}R_{js} + b_{nj}R_{j+}+b_{nj}R_{j-}\right] \dots \exp\left[-i\sum_{j}\Delta\omega_{j}\tau_{i}R_{js}\right]$$
$$\times \exp\left[i\sum_{j}a_{ij}R_{js}+b_{ij}R_{j+}+b_{ij}R_{j-}\right], \qquad (14)$$

where  $a_{\eta j} = -\Delta \omega_j \Delta t_{\eta}$   $(\eta = 1, 2, ..., n)$  and the expressions for  $b_{\eta j}$  were written our earlier.

The following recurrence relations are useful in the calculation of the density matrix of the system from Eq. (2):

$$\exp [i(a_{vv}R_{j_{3}}+b_{v_{i}}R_{j_{i+}}+b_{vi}\cdot R_{j_{-}})]R_{j_{3}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}R_{j_{i+}}+b_{vi}\cdot R_{j_{i-}})] = A_{vv}R_{j_{3}}+B_{vv}R_{j_{i+}}+B_{vi}\cdot R_{j_{-}},$$

$$\exp [i(a_{vv}R_{j_{3}}+b_{vv}R_{j_{i+}}+b_{vi}\cdot R_{j_{-}})]R_{j_{1}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}R_{j_{1}}+b_{vi}\cdot R_{j_{-}})] = C_{vv}R_{j_{3}}+D_{vv}R_{j_{1}}+E_{vv}R_{j_{-}},$$

$$\exp [i(a_{vv}R_{j_{3}}+b_{vi}R_{j_{+}}+b_{vi}\cdot R_{j_{-}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}R_{j_{2}}+b_{vv}\cdot R_{j_{-}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{3}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{-}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{3}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{-}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{3}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{-}})]R_{j_{-}} \exp [-i(a_{vv}R_{j_{3}} + b_{vv}\cdot R_{j_{3}})]R_{j_{-}} \exp [-$$

$$+b_{\eta j}R_{j+}+b_{\eta j}R_{j-})]=C_{\eta j}R_{j3}+E_{\eta j}R_{j+}+D_{\eta j}R_{j-},$$

where

$$A_{\eta j} = \frac{1}{\Phi_{\eta j}^{2}} (a_{\eta}^{2} + 4b_{\eta} b_{\eta j}^{2} \cos \Phi_{\eta j}),$$

$$B_{\eta j} = b_{\eta j} \left[ \frac{1}{\Phi_{\eta}^{2}} (1 - \cos \Phi_{\eta j}) a_{\eta j} - i \frac{\sin \Phi_{\eta j}}{\Phi_{\eta j}} \right],$$

$$C_{\eta j} = 2b_{\eta j} \cdot \left[ \frac{1}{\Phi_{\eta j}^{2}} (1 - \cos \Phi_{\eta j}) a_{\eta j} - i \frac{\sin \Phi_{\eta j}}{\Phi_{\eta j}} \right],$$

$$D_{\eta j} = \frac{1}{\Phi_{\eta j}^{2}} \left[ a_{\eta j}^{2} \cos \Phi_{\eta j} + 2b_{\eta j} b_{\eta j}^{2} (1 + \cos \Phi_{\eta j}) \right] + i a_{\eta j} \frac{\sin \Phi_{\eta j}}{\Phi_{\eta j}},$$
(16)

 $E_{\eta j} = 2b_{\eta j} \cdot \frac{(1 - \cos \Phi_{\eta j})}{\Phi_{\eta j}^2}$ 

and the parameter  $\Phi_{\eta j}$  for the traveling wave is

$$D_{\eta j} = (a_{\eta j}^2 + \theta_{\eta j}^2)^{\frac{1}{2}},$$

đ

and for the standing wave

$$\Phi_{\eta j} = [a_{\eta j}^2 + 4\theta_{\eta j}^2 \cos^2(\mathbf{k}_{\eta}\mathbf{r}_j - \varphi_{c\eta})]^{\frac{1}{2}}$$

Calculation of the density matrix in the considered case (when the first m (m = 1, 2, ..., n - 1) pulses are traveling waves and the *n*-th is a standing wave) leads to expression (8), where

$$M_{j}^{(-\mathbf{k}_{m})} = 2R_{j+} \left(\prod_{p < m} A_{pj}\right) B_{mj}^{*} \left(\prod_{m < q < n-1} D_{qj}^{*}\right) E_{nj}^{*}$$

$$\times \exp\left[-i\Delta\omega_{j}\left(t - 2t_{n} + t_{m} - \sum_{q=1}^{n} \Delta t_{q}\right)\right].$$
(17)

In the limiting case  $a_{\eta j} = 0$  (the subscript  $\eta$  runs through the numbers of all the exciting pulses), expression (17) goes over into (9). In particular, in the case n = 2, when the first pulse is a traveling wave and the second a standing one, the calculation leads to the following expression for the reversed photon echo in the -k, direction:

$$M_{j} = \frac{1}{2} R_{j+} \theta_{ij} \left[ \frac{a_{ij}}{\Phi_{ij}^{2}} \left( 1 - \cos \Phi_{ij} \right) + i \frac{\sin \Phi_{ij}}{\Phi_{ij}} \right] \\ \times \left\{ \left[ 1 - J_{0} (2\theta_{2}) \right] + \sum_{u=1}^{\infty} \sum_{v=1}^{u} (-1)^{u} \frac{C_{u}^{\sigma-4} C_{2v}}{\left[ 2 (u+1) \right]!} a_{2j}^{2(u-v+1)} \theta_{2j}^{2v} \right\} \\ \times \exp \left\{ i \left[ -\mathbf{k}_{i} \mathbf{r}_{j} - \Delta \omega_{j} (t - 2\tau_{1} - \Delta t_{1} - \Delta t_{2}) - \varphi_{0} \right] \right\},$$
(18)

where  $C_{u}^{v}$  is the number of combinations of u taken v at a time.

Thus, just as in the short-pulse case, the reversed photon echo signal turns out to be independent of the direction of the action of the standing wave. Allowance for the reversible relaxation during the time of action of the pulses (as well as between the pulses) does not prevent generation of the reversed photon echo signal, and only deforms the shape of this signal.

## 2. RESULTS OF EXPERIMENTAL INVESTIGATION OF REVERSED PHOTON ECHO SIGNALS FOLLOWING RESONANT EXCITATION OF A SYSTEM BY A SEQUENCE OF TRAVELING AND STANDING WAVES

The experiments were performed with the setup described in Ref. 17. The density of the Cr<sup>3+</sup> ions in the investigated ruby crystal was 0.05 wt.%. The sample temperature ranged from 1.7 to 2.2 K. A constant magnetic field  $H_0 \le 1500$  Oe was applied to the sample in a direction parallel to c and caused an increase in the intensity of the reversed photon echo signals. The exciting laser pulses were separated in time. The reversed photon echo signals were investigated under three-pulse excitation. The oscillograms of these signals in ruby are shown in this case in Fig. 2. The signals were emitted in a direction opposite to that of the corresponding traveling waves and were independent of the direction of the action of the standing waves. Under conditions when the direction of the action of the standing wave was close to the propagation direction of the traveling waves, the reversed photon echo signals were generated simultaneously with the corresponding primary photon echo signals. In three-pulse excitation, for example, they were observed simultaneously with the primary photon echo signals that were emitted at

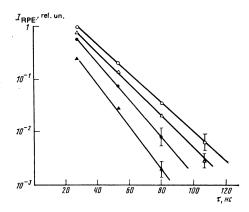


FIG. 3. Relative intensity of reversed photon echo (RPE) in ruby (under two-pulse excitation) vs. the time interval  $\tau_1$  between the pulses, in a constant magnetic field  $H_0 = 460$  (O), 77 ( $\Delta$ ), 40 ( $\bullet$ ), and 0 Oe ( $\blacktriangle$ ).

the instants  $\tau_1 + 2\tau_2$  and  $2(\tau_1 + \tau_2)$  in the directions  $2k_3$  $-\mathbf{k}_2$  and  $2\mathbf{k}_3 - \mathbf{k}_1$  (where  $\mathbf{k}_3$  is the wave vector of that standing-wave-generating traveling wave whose propagation direction was close to that of the preceding traveling wave). We call attention to the fact that with increasing intensity of the first pulse, the theory calls for the first reversed photon echo to decrease and for the second to increase. Conversely, with increasing intensity of the second traveling wave the intensity of the first reversed photon echo should increase (albeit not as much as in the preceding case), and that of the second should decrease. By varying the intensities of the traveling waves it is possible to transfer energy from one reversed photon echo to the other. This is precisely the behavior of the reversed photon echo signal which we observed in our experiments with increasing intensity of the corresponding traveling waves.

We note that we have experimentally registered (at the noise level) rather weak signals of reversed primary and stimulated photon echos.

Since a much better signal/noise ratio is reached in the detection of reversed photon echo signals, it seemed natural to use these signals under situations in which the procedure of the primary and stimulated echo turns out to be inapplicable because of the low sensitivity. Such a situation is realized in ruby, for example, in a zero magnetic field, when the very detection of primary and stimulated photon echo signals is a praiseworthy accomplishment. Figure 3 shows the intensity fall-off curves of one of the reversed photon echo signals both in a zero magnetic field and in magnetic field of varying intensity. We determined from these curves, accurate to  $\pm 12\%$ , the times  $T_2$  of transverse irreversible relaxation in ruby (c = 0.05wt.%) at a temperature 2.2 K, namely

 $H_0$ , Oe: 0 40 77 460  $T_2$ , nsec 23.4 68 91 107

We investigated the temperature dependence of the reversed photon echo intensity in a zero magnetic field (Fig. 4). At temperatures lower than 5 K the relaxation due to the interaction between the electrons of the chromium ions with the aluminum nuclei predominates. At temperatures higher than 5 K the Orbach relaxation with participation of the  ${}^{2}E(2\overline{A})$  state begins to prevail. The relaxation factor of the reversed photon echo signal can be written in the form  $\exp(-2t_{e}/\tau_{e})$ , where  $t_{e}$  is the

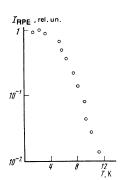


FIG. 4. Temperature dependence of the reversed photon echo intensity in ruby in a zero magnetic field;  $\tau_1 = 52$  nsec.

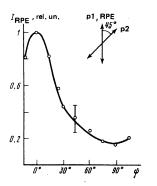


FIG. 5. Relative intensity of reversed photon echo intensity in ruby  $[{}^{4}A_{2}-{}^{2}E(\bar{E})]$  vs. the angle at which the polarization of this signal is fixed;  $H_{0}$ =460 Oe. The arrows in the right-hand corner indicate the directions of the polarization vectors of the corresponding pulses and of the reversed photon echo signal.

instant of observation of the echo-signal maximum and  $\tau_e$  is the lifetime of the echo signal. Using the results of Geschwind,<sup>18</sup> we have at  $T > 5K \tau_e = \frac{1}{4} \tau_0 \exp(\Delta/k_B T)$ , where  $\tau_0$  is the lifetime of the spontaneous transition from the state  ${}^2E(2A)$  to the state  ${}^2E(\overline{E})$ ,  $\Delta = 29 \text{ cm}^{-1}$ ,  $k_B$  is the Boltzmann constant, and T is the absolute temperature of the sample. Analysis of the temperature dependence of the reversed photon echo yielded the estimate  $\tau_0 = 10^{-9}$  sec.

We have also investigated experimentally the dependence of the reversed photon echo intensity on the angle at which the polarization of this signal is fixed (Fig. 5). If the polarization vectors of the traveling and standing waves are mutually perpendicular, a similar dependence was plotted in Ref. 6. It follows from these relations that the reversed photon echo polarization coincides with the polarization of that traveling wave whose action together with the standing wave led to the generation of the reversed photon echo; this agrees with the conclusions of the theory developed.

An investigation of the shapes of the reversed photon echo signals and of its correlation with the shapes of the corresponding traveling-wave pulses has shown that if the intensity of the traveling waves is low and that of the standing wave high, the shape of the reversed photon echo signals in the inverse of the shape of the corresponding traveling wave pulses. This is clearly seen



FIG. 6. Oscillogram illustrating the effect of the correlation of the shapes of the reversed photon echo signals in ruby with the shapes of the corresponding exciting pulses. The power of the third exciting pulse ( $\approx 300 \text{ kW}$ ) is one order of magnitude higher than that of the first two pulses;  $\tau_1 = 24 \text{ nsec}$ ,  $\tau_2 = 35 \text{ nsec}$ ,  $H_0 = 460 \text{ Oe}$ .

from the oscillograms of Fig. 6. The conditions under which the shapes of the echo signals correlate with the shapes of the pulses were discussed in Ref. 5.

## CONCLUSION

It was shown above that a sequence of laser pulses (which are traveling waves) can be reversed in space with the aid of a standing wave. Since the reversal effect does not depend on the direction of the action of the standing wave, it follows that the generation of the sequence of reversed photon echo signals can be effected in practically any arbitrarily chosen section of the sample. This permits spectroscopic probing of various sections of the sample. A sequence of reversed photon echo signals with decreasing amplitude can then be used to determine the relaxation time  $T_2$  at definite intervals between the pulses (without plotting the dependences of the reversed photon echo intensity on the various values of these intervals). In this respect it recalls the reversed variant of the Carr-Purcell technique.

The effect of reversal of laser signals in the spinecho procedure can be used in technology, since it permits the motion of these signals at specified instants of time in a previously chosen volume of a resonant medium.

In conclusion, the authors thank Professor A.I. Alekseev for a discussion of the polarization of two-pulse reversed photon echo, as well as Professor E.A. Manykin for a discussion of the shapes of the echo signals.

 $^{1)}\mathrm{A}$  similar result was first obtained in the case n=2 by Alekseev and Basharov.  $^{15}$ 

- <sup>1</sup>U. Kh. Kopvillem and V. R. Nagibarov, Fiz. Met. Metallov. **15**, 313 (1963).
- <sup>2</sup>I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966).
- <sup>3</sup>V. V. Samartsev, Zh. Prikl. Spektrosk. 30, 581 (1979).
- <sup>4</sup>V. V. Samartsev, R. G. Usmanov, G. M. Ershov, and B. Sh. Khamidullin, Zh. Eksp. Teor. Fiz. 74, 1979 (1978) [Sov. Phys. JETP 47, 1030 (1978)].
- <sup>5</sup>V. A. Zuikov, V. V. Samartsev, and R. G. Usmanov, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 293 (1980) [JETP Lett. **32**, 270 (1980)].
- <sup>6</sup>V. A. Zuikov, V. V. Samartsev, and R. G. Usmanov, *ibid*. **31**, 654 (1980) [**31**, 617 (1980)].
- <sup>7</sup>R. H. Thomson and C. F. Quate, J. Appl. Phys. 42, 907 (1971).
- <sup>8</sup>R. H. Dicke, Phys. Rev. 93, 99 (1954).
- <sup>9</sup>R. Kachru, T. W. Mossberg, E. Whittaker, and S. R. Hartmann, Opt. Commun. **31**, 223 (1979).
- <sup>10</sup>N. S. Shiren, Appl. Phys. Lett. 33, 299 (1978).
- <sup>11</sup>G. M. Ershov and U. Kh. Kopvillem, Zh. Eksp. Teor. Fiz. **72**, 130 (1977) [*sic*].
- <sup>12</sup>S. M. Zakharov, E. A. Manykin, and E. V. Onishchenko, *ibid.* 59, 1307 (1979) [Sov. Phys. JETP 32, 714 (1971)].
- <sup>13</sup>E. A. Manykin, Pis'ma Zh. Eksp. Teor. Fiz. 7, 345 (1968) [JETP Lett. 7, 269 (1968)].
- <sup>14</sup>A. I. Alekseev, A. M. Basharov, and V. I. Belobrodov, Zh. Eksp. Teor. Fiz. **79**, 787 (1980) [Sov. Phys. JETP **52**, 401 (1980)].
- <sup>15</sup>A. N. Alekseev and A. M. Basharov, Kvant. Elektron. (Moscow) 8, 182 (1981) [Sov. J. Quantum Electron. 11, 101 (1981)].
- <sup>16</sup>S. M. Zakharov and E. A. Manykin, Opt. Spektrosk. 33, 966 (1972).
- <sup>17</sup>V. A. Zuikov, V. V. Samartsev, and R. G. Usmanov, Zh. Tekh. Fiz. **49**, 2272 (1979) [Sov. Phys. Tech. Phys. **24**, 1260 (1979).
- <sup>18</sup>S. Geschwind, transl. in: Sverkhtonkie vzaimodeľsviya v tverdykh telakh (Hyperfine Interactions in Solids), Mir, 1979, pp. 103-162.

Translated by J. G. Adashko