

# Characteristics of reversed photon echo resulting from nonsimultaneous four-wave interaction in ruby

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The results are reported of detailed theoretical and experimental investigations of reversed echo in ruby at 2°K. A prediction is made of an oscillatory dependence of the echo intensity on the degree of overlap of the opposite pump waves and on the intensity of the standing wave field. Reversal of the wavefront of the signal wave is produced by a reversing echo mirror with a correction for the additional aberration introduced into the wave.

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## INTRODUCTION

Transformation of the wavefronts of optical fields by the four-wave interaction method is currently the subject of extensive investigations in nonlinear optics and dynamic holography. A signal due to the four-wave simultaneous interaction was first demonstrated experimentally in the forward geometry<sup>1</sup> and in the geometry of complete wavefront reversal,<sup>2,3</sup> where the result of generation of a complex-conjugate wave was considered in terms of dynamic holography. The interaction signal appears simultaneously with the arrival of three coherent light pulses in a recording medium and its wave vector is  $k = k_3 + k_2 - k_1$ , where  $k_1$  is the vector of the wave being transformed, whereas  $k_2$  and  $k_3$  are the vectors of plane pump waves. This process is coherent and if the medium loses coherence rapidly, then all the three pump waves must be applied simultaneously.

However, under resonance conditions when the irreversible relaxation channels are suppressed, an atomic system may have a fairly long phase memory of the excitation coherence. Then, the condition of simultaneity of the action of all three waves need no longer be satisfied. Then, during the transient stage we can expect various coherent optical quantum phenomena such as the photon echo,<sup>4</sup> induced transient coherent population gratings in space,<sup>5</sup> optical nutation,<sup>6</sup> self-induced transparency,<sup>7</sup> etc. In particular, a complete analog of the simultaneous four-wave interaction is the three-pulse photon echo,<sup>8</sup> which is a coherent response of a medium to the action of three laser pulses arriving at different times and generally noncollinear. The ability of a medium with phase memory to "remember" the wave characteristics of optical pump waves can be used in dynamic holography. The feasibility of recording, reconstructing, and transforming optical wavefronts (complex conjugacy, doubling of fronts, etc.) in the photon echo geometry (echo holography) was considered theoretically in Ref. 9 and then in Ref. 10. The three-pulse nonsimultaneous excitation in the special case when the second and third pump waves are directed opposite to one another generates a polarization wave in the medium and this wave satisfies the phase matching condition<sup>11</sup> and it serves as a source of a coherent electromagnetic echo wave. This case of the opposite pump waves ( $k_3 = -k_2$ ) can ensure a complete wave reversal

of the wavefront in respect of the direction ( $k_3 = -k_1$ ) and time because the medium "remembers" not only the information on the fronts but also the order of arrival of the optical pulses.<sup>9</sup>

The first experimental observation of the three-pulse reversed photon echo was made in ruby<sup>12</sup> and then in sodium vapor.<sup>13</sup> Another special case of this geometry was considered theoretically in Ref. 14, where an analysis was made of the situation of a complete overlap of the opposite pump waves, i.e., in the case when the second and third pulses act simultaneously on a medium and are oppositely directed. In this case the pump field in the medium has a periodic spatial inhomogeneity (standing wave) and this alters the nature of the nonlinear interaction. The more general and more complex case when the opposite waves act on a medium nonsimultaneously but overlap partly has not yet been investigated. In this case after a time  $\tau > \delta$ , where  $\delta$  is the pulse duration, from the arrival of the first pulse a medium is excited first by a traveling wave (homogeneous excitation by the leading edge of the second pulse), then by a standing wave (inhomogeneous excitation by an interference field), and finally again by a traveling wave (homogeneous excitation by the trailing edge of the third pulse). We shall describe a detailed investigation of the influence of the degree of such overlap of the opposite pump waves on the characteristics of the three-pulse reversed photon echo.

## THEORY

A calculation of the nonequilibrium polarization induced in a medium will be made by a quasiclassical method in which the field is regarded in the classical form but the behavior of the system is described using the formalism of the density matrix operator. In the interaction representation the equation of motion of this operator  $\hat{\rho}$  is

$$i\hbar \frac{\partial}{\partial t} \hat{\rho} = [\hat{\mathcal{H}}, \hat{\rho}], \quad (1)$$

where  $\hat{\mathcal{H}}$  is the Hamiltonian of the system of two-level atoms. The interaction with a coherent field of amplitude  $\mathcal{E}$  is described by

$$\hat{\mathcal{H}} = \sum_{j=1}^N \hbar \Delta_j \hat{R}_{j-} - \rho \mathcal{E} \sum_{j=1}^N [\hat{R}_{j+} \exp(ikr_j) + \hat{R}_{j-} \exp(-ikr_j)]. \quad (2)$$

In the interval between the pump pulses the Hamiltonian becomes

$$\hat{\mathcal{H}} = \sum_{j=1}^N \hbar \Delta_j \hat{R}_{3j} + \hat{\Gamma}, \quad (3)$$

where  $N$  is the number of active centers;  $2\pi\hbar$  is the Planck constant;  $\Delta_j = \omega_j - \omega$ ;  $\omega_j$  is the natural frequency of the  $j$ -th atom;  $\omega$  is the pump field frequency;  $p$  is the dipole moment of the transition;  $\hat{R}_{*j} = \hat{R}_{1j} \pm i\hat{R}_{2j}$ ;  $\hat{R}_{1j}$ ,  $\hat{R}_{2j}$ , and  $\hat{R}_{3j}$  are the transverse and longitudinal components of the Dicke operator<sup>15,16</sup>  $\hat{\Gamma}$  is the relaxation operator defined by

$$[\hat{\Gamma}, \hat{\rho}] = \begin{pmatrix} 0 & -\Gamma\rho_{12} \\ -\Gamma\rho_{21} & 0 \end{pmatrix};$$

$\Gamma$  is the half-width of the function representing a homogeneous broadening of the transition;  $\mathcal{E}_\mu$  is the electric field of the  $\mu$ -th pump pulse. This field can be represented in the form of rectangular pulses with  $\mu = 1, 2, 3$ , and 4 which are all plane waves:

- 1)  $0 \leq t \leq \delta_1$ ,  $E_1 = \mathcal{E}_1 \exp\{i(\omega t - \mathbf{k}_1 \mathbf{r})\} + \text{c.c.}$ ;
- 2)  $\delta_1 < t \leq \tau$ ,  $E = 0$ ;
- 3)  $\tau < t \leq \tau + \delta_2$ ,  $E_2 = \mathcal{E}_2 \exp\{i(\omega t - \mathbf{k}_2 \mathbf{r})\} + \text{c.c.}$ ;
- 4)  $\tau + \delta_2 < t \leq \tau + \delta_2 + \delta_3$ ,  $E_3 = \mathcal{E}_3 \cos \mathbf{k}_2 \mathbf{r} e^{i\omega t} + \text{c.c.}$ ;
- 5)  $\tau + \delta_2 + \delta_3 < t \leq \tau + \delta_2 + \delta_3 + \delta_4$ ,  $E_4 = \mathcal{E}_4 \exp\{i(\omega t + \mathbf{k}_2 \mathbf{r})\} + \text{c.c.}$ ;
- 6)  $t > \tau + \delta_2 + \delta_3 + \delta_4$ ,  $E = 0$ .

In these calculations we are assuming that the operator  $\hat{\rho}$  of a system of noninteracting two-level atoms has the usual form for optical transitions (before arrival of the pump fields):

$$\hat{\rho}(0) = 2^{-N} \prod_{j=1}^N (1 - 2\hat{R}_{3j}). \quad (5)$$

The solution of Eq. (1) for nondiagonal elements of the density matrix is

$$\hat{\rho}_{\text{int}}(t) = \hat{L}(t) \hat{\rho}(0) L^{-1}(t) \exp[-\Gamma t], \quad (6)$$

where  $\hat{L}(t)$  is the familiar unitary operator composed of the product of unitary operators corresponding to each time interval and taken in the interaction representation. Using the resultant expressions, which describe cophasal polarization waves responsible for the formation of radiation in the direction  $-\mathbf{k}_1$ , we then obtain from Eq. (6) the required polarization:

$$\mathcal{P}(t) = \text{Sp}(\hat{\rho}_{\text{int}} \hat{P}_{\text{int}}), \quad (7)$$

where  $\hat{P}_{\text{int}}$  is the operator of the dipole moment of the transition. It is interesting to consider the behavior of the polarization in a real situation when the pump wave pulses are symmetric and identical ( $\mathcal{E}_1 = \mathcal{E}_2 = \mathcal{E}_3 = \mathcal{E}_4$ ,  $\delta_2 = \delta_4$ ) and the laser frequency coincides exactly with the center of an inhomogeneous transition line ( $\omega = \omega_0$ ) described by the Gaussian function

$$g(\Delta) = \frac{N_0}{\Delta_p \pi^{1/2}} \exp\left[-\left(\frac{\Delta}{\Delta_p}\right)^2\right], \quad (8)$$

where  $N_0$  is the concentration of the atoms and  $\Delta_p$  is the line half-width.

After averaging over the distribution  $g(\Delta)$ , we find that Eqs. (6) and (7) yield the following expression for the polarization:

$$\begin{aligned} \mathcal{P}(\mathbf{r}, t-t') &= \frac{p}{2} \left\{ \int_{-\infty}^{\infty} B g(\Delta) \gamma \sin \theta_1 \left[ \cos \Delta(t-t') \right. \right. \\ &\quad \left. \left. + \frac{\Delta \sin \Delta(t-t')}{(\Delta^2 + \Omega^2)^{1/2}} \text{tg} \frac{\theta_1}{2} \right] d\Delta \right\} \exp[i(\omega_0 t + \mathbf{k} \cdot \mathbf{r})] + \text{c.c.}; \quad (9) \\ B &= \gamma^2 \sin^2 \frac{\theta_2}{2} [1 - J_0(2\theta_{03})] - \gamma^2 \sin \theta_2 \sin^2 \frac{\theta_2}{2} J_1(2\theta_{03}) \\ &\quad - 2\gamma \sin^2 \frac{\theta_2}{2} \cos^2 \frac{\theta_2}{2} J_2(2\theta_{03}) \left[ 1 - (1-\gamma) \text{tg}^2 \frac{\theta_2}{2} \right] \\ &\quad + \left[ 1 + (1-\gamma) \text{tg}^2 \frac{\theta_2}{2} \right] \left[ \gamma \sin^2 \frac{\theta_2}{2} J_0(2\theta_{03}) + 2\gamma^2 \sin \theta_2 \cos^2 \frac{\theta_2}{2} J_1(2\theta_{03}) \right] \\ &\quad + \left[ 1 + (1-\gamma) \text{tg}^2 \frac{\theta_2}{2} \right]^2 \cos^2 \frac{\theta_2}{2} [1 - J_0(2\theta_{03})], \\ \gamma &= \Omega^2 / (\Delta^2 + \Omega^2), \quad \theta_\mu = (\theta_{0\mu}^2 + \Delta^2 \delta_\mu^2)^{1/2}, \quad \theta_{0\mu} = \Omega \delta_\mu, \end{aligned}$$

$\Omega = p\hbar^{-1} \mathcal{E}$  is the Rabi frequency, and  $J_{0,1,2}(2\theta_{03})$  is a Bessel function of the first kind. Equation (9) may have a simple solution for certain usually made assumptions.

I. Very powerful and short pump pulses ( $\Omega \gg \Delta_p$  and  $\delta < 2\pi\Omega/\Delta_p^2$ ), when all the isochromates of an inhomogeneously broadened transition line are excited. In this case we have  $\gamma = 1$  and  $\theta_\mu = \theta_{0\mu}$ , and the amplitude of the polarization in Eq. (9) becomes

$$\begin{aligned} \mathcal{P}(t-t') &= 1/2 p \sin \theta_{01} \{1/2(1 + \cos^2 \theta_{02}) [1 - J_0(2\theta_{03})] - 1/2 \sin^2 \theta_{02} J_2(2\theta_{03}) \\ &\quad + \sin 2\theta_{02} J_1(2\theta_{03}) + \sin^2 \theta_{02} J_0(2\theta_{03})\} S(t-t'), \quad (10) \end{aligned}$$

where the function representing the shape of the echo signal is

$$S(t-t') = \frac{N_0}{\Delta_p \pi^{1/2}} \left[ 1 + \frac{\text{tg}(\theta_{01}/2) \Delta_p^2(t-t')}{2\Omega} \right] \exp\left\{-\left[\frac{\Delta_p(t-t')}{2}\right]^2\right\}, \quad (11)$$

$0 \leq \theta_{03} \leq \Omega\delta$ ;  $\delta = \beta + \delta_3$  is the duration of the pump pulse;  $\beta$  is the delay of one opposite wave relative to the other (within the region of overlap of the opposite waves  $\beta < \delta$ ).

We shall first consider two limiting cases.

a) Complete overlap (only a standing wave is obtained), i.e.,  $\beta = 0$ ,  $\theta_{02} = 0$ . We can see from Eq. (10) that in this case the polarization reduces to the expression obtained in Ref. 14:

$$\mathcal{P}_0(t-t') = 1/2 p [1 - J_0(2\Omega\delta)] \sin \theta_{01} S(t-t'). \quad (12)$$

b) Complete nonoverlap of the opposite waves ( $\beta > \delta$ ). We then have  $\theta_{03} = 0$  and the polarization is described by

$$\mathcal{P}_c(t-t') = 1/2 p \sin \theta_{01} \sin^2 \theta_{02} S(t-t'), \quad (13)$$

i.e., by the well-known expression for the three-pulse echo.

c) We shall now deal with the more general case of partial time overlap of the opposite waves ( $0 < \beta < \delta$ ), when the echo polarization is the result of interference of several terms in Eq. (10). As  $\beta$  increases to  $\delta$ , the polarization generally decreases, i.e.,  $\mathcal{P}_0 > \mathcal{P}_c$ . However, the echo intensity does not fall monotonically but oscillates and the oscillation depth depends on the intensities of the opposite pump waves. Figure 1 shows this dependence calculated from Eq. (10) for different values of the areas under the pulses representing the opposite waves. The echo pulse profile is governed by the function  $S(t-t')$  and, as can be seen from Eq. (11),

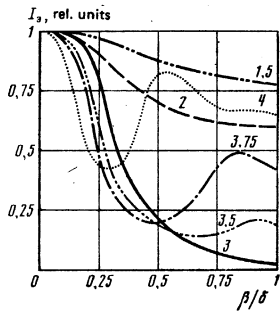


FIG. 1. Influence of the degree of the time overlap of the opposite pump waves on the intensity of the reversed echo for different values of  $\theta_2$  (theory).

it either has a symmetric Gaussian form with the dephasing parameter  $T_2^* = 2\Delta_p^{-1}$  on condition that  $\tan(\theta_{01}/2) \gg \Omega/2\Delta_p$  or it becomes asymmetric and its maximum is subject to an additional delay which is, for example, approximately  $\Omega^{-1}$  in the case when  $\theta_{01} = \pi/2$ .

II. We shall now consider an equally important case  $\Omega \ll \Delta_p$  which is frequently encountered in experiments. This case corresponds to partial excitation of the spectral transition line, when out of the whole ensemble of atoms only some isochromats are excited and they have detunings  $\Delta < \Delta_p$ . Then, in the case of the atoms satisfying the condition  $\Delta < \Delta_0 = (2\pi\Omega/\delta)^{1/2}$  the behavior of the polarization remains coherent but the spectral weight of the separate components is different because  $\gamma \neq 1$ . Under these assumptions we can, as before, assume that  $\theta_\mu = \theta_{0\mu}$  in Eq. (9) and this allows us to obtain an analytic solution

$$\mathcal{P}(t) = p \frac{\pi^{1/2} \Omega}{\Delta_p} (C + \eta D), \quad t \rightarrow t - t', \quad \eta = \begin{cases} +1, & t > 0 \\ -1, & t < 0 \end{cases} \quad (14)$$

$$C = J_0[\Omega(\delta^2 - t^2)^{1/2}] \{ [1 - J_0(2\theta_{03})] + 2 \sin \theta_{01} \sin \theta_{02} J_1(2\theta_{03}) e^{-\alpha|t|} + 2^{1/2} \sin \theta_{01} \sin^2(\theta_{02}/2) [3J_0(2\theta_{03}) - 1 - \cos \theta_{02} J_2(2\theta_{03})] \}$$

$$\times [1 + 2^{1/2} \Omega |t|] \exp(-2^{1/2} \Omega |t|) - 2 \sin \theta_{01} \sin \theta_{02} \sin^2 \frac{\theta_{03}}{2} J_1(2\theta_{03}) (1 + \Omega |t|) e^{-\alpha|t|} + 5 \cdot 2^{-1/2} \sin \theta_{01} \sin^4 \frac{\theta_{03}}{2} [1 - 5J_0(2\theta_{03}) + 2J_2(2\theta_{03})] \times [1 + 2^{1/2} \Omega t + 0, 2(2^{1/2} \Omega t)^2] \exp(-2^{1/2} \Omega |t|),$$

$$D = \sin \theta_{01} \{ [1 - J_0(2\theta_{03})] e^{-\alpha|t|} + 2^{1/2} \sin \theta_{02} J_1(2\theta_{03}) \Omega |t| \exp(-2^{1/2} \Omega |t|) + [3J_0(2\theta_{03}) - 1 - \cos \theta_{02} J_2(2\theta_{03})] \sin^2(\theta_{02}/2) \Omega |t| e^{-\alpha|t|} - 2^{1/2} \sin \theta_{02} \sin^2(\theta_{02}/2) J_1(2\theta_{03}) \Omega |t| (1 + 2^{1/2} \Omega |t|) \exp(-2^{1/2} \Omega |t|) + 1/2 \sin^4(\theta_{02}/2) \Omega |t| (1 + \Omega |t|) [1 - 5J_0(2\theta_{03}) + 2J_2(2\theta_{03})] e^{-\alpha|t|} \}.$$

This expression can be analyzed quite simply in the two limiting cases of: a) complete overlap of the opposite waves ( $\beta = 0$ ,  $\theta_{02} = 0$ ,  $\theta_{03} = \Omega\delta$ ); b) consecutive action of the opposite waves ( $\beta > \delta$ ). Figure 2 shows examples of the shape of the polarization pulses calculated from Eq. (14) for these two cases in the approximation of small areas under the pulses ( $\sin \theta_{0\mu} \approx \theta_{0\mu}$ ). We can see that the nature of phase matching depends strongly on the

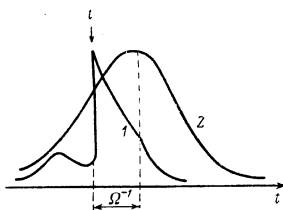


FIG. 2. Profile of the echo pulses: 1)  $\beta = 0$ ; 2)  $\beta > \delta$ .

delay  $\beta$ . In the first case (curve 1) the echo duration and its leading edge are much shorter than in the second case (curve 2) and there is no additional shift of the echo maximum. In the second case the echo pulse is more symmetric and it is delayed by about  $\Omega^{-1}$ .

## EXPERIMENTS

The reversed photon three-pulse echo was investigated using apparatus described earlier.<sup>12, 17</sup> The only modification was that in the present experiments a beam III was formed independently using an additional OLZ-2 optical delay line, which made it possible to compensate accurately the path difference between opposite beams II and III. The resonant medium was a ruby single crystal with a  $\text{Cr}^{3+}$  concentration of  $\sim 0.03$  at.%. The sample was a cube with a 10-mm edge and it was cooled to 2°K in an optical cryostat in order to increase the duration of the phase memory of the medium. A resonance at the  ${}^4A_2(\pm\frac{3}{2}) - {}^2E(\bar{E})$  transition in ruby ( $\lambda = 6934 \text{ \AA}$  at 2°K) was excited by a ruby laser which was cooled to 77°K and generated the second component of the same  ${}^4A_2(\pm\frac{3}{2}) - {}^2E(\bar{E})$  doublet at  $\lambda = 6933.97 \text{ \AA}$ . Single-pulse emission was ensured by active Q switching of the resonator cavity, which made it possible to suppress the stray optical coupling between the resonator elements and generate pulses of 10 nsec duration and about 1 MW power. The sample was pumped approximately along the optic axis of the crystal, and the polarizations of all the beams were perpendicular to the plane of their incidence on the sample. The angle between the beams I and II was 5°, which ensured a satisfactory spatial decoupling between the pump and echo signals. A weaker longitudinal static magnetic field (0–250 Oe) was applied to the sample by Helmholtz coils and it lifted partly the degeneracy of the ground state  ${}^4A_2$ , so that the signal echo became stronger. The coherent echo response was detected with wide-band ELU-FT photomultipliers and a Tektronix-466 oscilloscope.

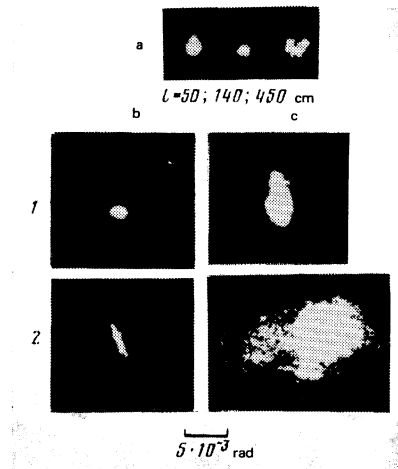


FIG. 3. Angular divergence of the beam after reflection by: a), b) reversing echo mirrors; c) plane mirror located in the plane of the sample; 1) without a phase plate; 2) with a phase plate.

The beam divergence was minimized by employing a confocal configuration with a 4 m base in both delay lines. A system of conjugate lenses with  $f=50$  cm (Ref. 12) made it possible, on the one hand, to increase the intensity of the pump field in the sample and, on the other, to combine automatically the caustics of all the beams in the same region. Moreover, because of the small divergence of the laser beam, the opposite beams II and III obtained in this geometry had approximately complex-conjugate wavefronts,<sup>12</sup> which made it possible to reduce the distortions of the reversed echo wavefront representing the complex conjugate to the wavefront of a beam I. In fact, the reversed echo beam IV had the reverse divergence relative to the first beam. This is demonstrated in Fig. 3a, which shows photographs of the transverse cross section of the echo beam at different distances  $l$  from the sample. Away from the sample the echo beam converged to an angle approximately equal to the divergence of the pump beam ( $\sim 8'$ ). At a distance  $l$  equal to the path from the exit mirror of the laser to the sample, the echo beam cross section was identical with the cross section at the exit from the laser. For greater values of  $l$  the echo beam cross section increased again. The quality of the wavefront reversal was checked by introducing deliberate aberrations into the signal beam I by inserting an etched phase plate directly in front of the sample.

Figures 3b and 3c show examples of compensation of the aberrations in the reversed echo signal. Clearly, in the case when the signal wave I had a wider spatial frequency spectrum (in the presence of the phase plate), the echo reproduced more accurately the pump beam geometry. This was clearly associated with the more favorable mixing of the spatial frequencies, a consequence of which was a more homogeneous excitation of the sample. Information on the wavefront of the pump beam I was reproduced accurately in the echo wave and this made it possible to regard the system as a reversing echo mirror. The wavefront reversal occurred due to a characteristic time reversal, i.e., at those points in the sample where the vibrations of the dipoles were advanced in phase during the application of the first nonplanar wave, and exactly the same phase lag appeared after the action of the pulse III. In accordance with the general principle of holography, this should produce a pseudoscopic image of the object. This can be illustrated clearly by the simple example of collinear three-pulse excitation (Fig. 4a) on the assumption that the signal wave I is characterized by a small phase

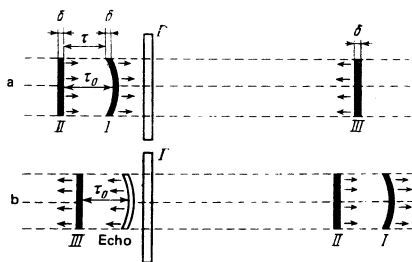


FIG. 4. Reversed echo of a wavefront in the three-pulse pumping case: a) before pumping; b) after echo response.

perturbation of the wavefront, and that the opposite waves II and III are plane. The echo is generated in such a way that the time lag in the axial part of the beam by  $\tau_0 - \tau$  results in reversal of the wavefront relative to the initial direction of propagation of the echo wave (Fig. 4b). It is worth recalling that in the general case described by Eq. (9) the moment of phase matching depends on many conditions, for example, there is a shift of the echo in the case of pumping by long pulses when it is no longer possible to neglect dephasing of the dipoles during the action of the pump pulse. This may result in distortion of the wavefront of the complex-conjugate replica of the signal wave if the intensity of the signal wave has a strongly inhomogeneous spatial distribution, so that in some parts of the sample there may be an additional induced dephasing of dipoles (local lag and advance). Distortions of the wavefront can be ignored in the case of excitation by short pulses ( $\delta < 2\pi\Omega/\Delta_p^2$ ) discussed above.

When the condition  $\Omega \gg \Delta_p$  is also satisfied, the wavefront distortions are again slight because the delay or advance of the echo is the same at all points in the sample. We observed a shift of the echo maximum depending on the excitation conditions. For example, in the  $\beta > \delta$  case (Fig. 5), i.e., when all the three pump pulses (the first two correspond to 1 and the third one

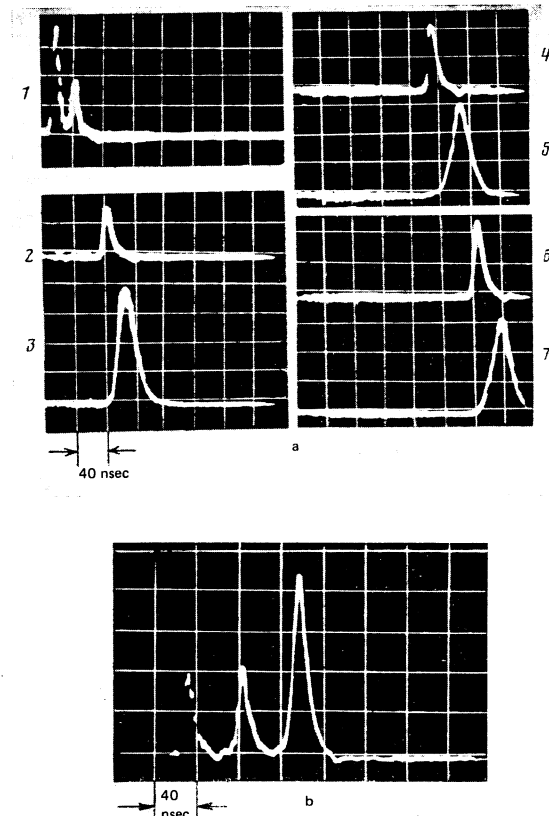


FIG. 5. a) Three-pulse echo obtained for different delays of the opposite pump waves: 1) pump pulses I and II for  $\tau = 27$  nsec; 2), 4), 6) pulse III,  $\beta = 50, 150, 200$  nsec; 3), 5), 7) reversed echo (pulse IV). b) Autocollimation photon echo in the direction  $-k_1$  for  $\tau = 54$  nsec (the first two pulses are the pump waves and the third one is the echo).

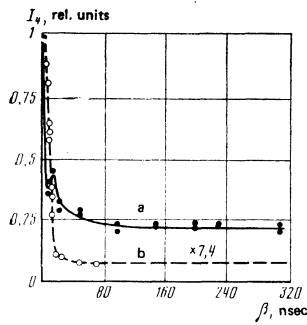


FIG. 6. Intensity of the four-wave interaction signal in the case of pump pulses noncoincident in time: a)  $\tau = 27$  sec; b)  $\tau = 0$ .

to 2, 4, and 6 for  $\beta = 50, 150$ , and  $200$  nsec, respectively) are separated on the time axis, we observed—in addition to  $\tau$ —an echo delay of approximately 6 nsec. The echo profile was then fairly symmetric. This was in reasonable agreement with the calculations (Fig. 2). In the case of complete overlap of the opposite waves ( $\beta = 0$ ) there was no additional delay and the echo pulse became shorter. This appeared particularly clearly in the case of pumping by weak pulses (Fig. 5b). The echo pulse in Fig. 5b was obtained in the case when the pump pulse III was not applied at all and the opposite pump wave was formed by reflection of the beam II from the rear surface of the sample. The echo was then observed only in the case of exact autocollimation of the beam II and it corresponded to practically complete overlap of the opposite waves ( $\beta = 0$ ). The dependence of the echo intensity on  $\beta$  in the case of complete separation of all the three pump pulses on the time axis ( $\beta > \delta$ ) was fairly simple (Fig. 6) since the fall of the intensity of this induced echo was governed by the longitudinal relaxation with a time constant  $T_1$  ( $\sim 10^{-3}$  sec).

A similar dependence was observed for  $\beta > \delta$  also in the case of simultaneous four-wave interaction, i.e., in the  $\tau = 0$  case (Fig. 6). For the time intervals  $\beta > \delta$  this signal could be regarded as a result of diffraction of the beam III by a grating of populations formed by the simultaneous action of two noncollinear beams.<sup>1</sup> The ef-

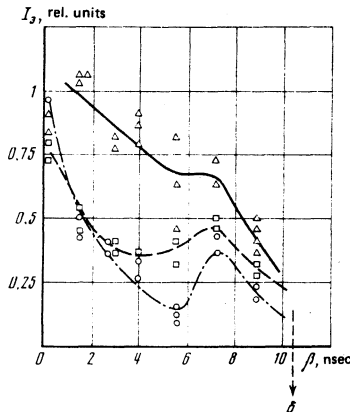


FIG. 7. Influence of the degree of the time overlap of the opposite pump waves on the echo intensity (experimental results):  $\square$   $\theta_{02}$ ;  $\Delta$   $0.71\theta_{02}$ ;  $\circ$   $0.53\theta_{02}$ .

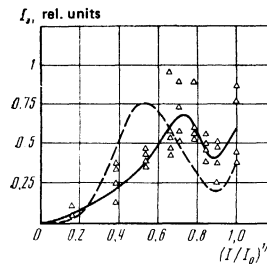


FIG. 8. Dependence of the echo intensity for  $\beta = 0$  on the intensity of the field of a standing pump wave. The dashed curve is the theory for the  $\Omega \gg \Delta_p$  case; the continuous curve is experimental.

fective lifetime of such a grating was governed by the decay time of the level populations and by the diffusion of the active centers.

In the region of overlap of the opposite pump waves ( $\beta < \delta$ ) the intensity of the reversed signal increased and the behavior of the echo parameters became much more complex. This is demonstrated by the results in Fig. 7, which shows the dependences  $I_e(\beta)$  obtained for ruby using opposite pump waves of different intensities  $I$ . Here, depending on the degree of overlap of the opposite waves, there were oscillations of the echo intensity  $I_e(\beta)$  and the oscillation amplitude changed as a result of an increase in the pump power. This was in qualitative agreement with the behavior of the echo calculated in the approximation of a strong field (Fig. 1). The somewhat unsatisfactory quantitative match was due to the fact that the strong field condition  $\Omega \gg \Delta_p$  was not satisfied in the experiments, i.e., only part of the inhomogeneously broadened line of the  ${}^4A_2(\pm\frac{1}{2}) - {}^2E(\bar{E})$  transition was excited. This was indicated by the above comparison of the results in Figs. 5a and 5b with those in Fig. 2. The agreement between the experiment and calculation considered in the strong field approximation should improve on increase in the field intensities of the opposite waves.

This was indeed observed at high pump intensities. The dependence of the reverse echo signal on the standing wave intensity (in the  $\beta = 0$  case) had the form shown in Fig. 8. An estimate of the Rabi frequency from the echo shift (see Fig. 5) and from the minimum of the  $I_e$  curve gave  $\sim 2 \times 10^8$  Hz, which was much less than the half-width of the  $R_1$  absorption line of the sample, amounting to  $\sim 3 \times 10^9$  Hz under our conditions.

Consequently, the nature of the nonlinearity of the interference field in the case of partial time overlap of the opposite pump waves gave rise to fundamentally new features of the behavior of the parameters of the reversed photon echo.

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