$K_1(\mathrm{Ar})+K_{-1}(\mathrm{Ar})=(8\pm4)\cdot10^3\ \mathrm{sec}^{-1}\ \mathrm{Torr}^{-1}$.

The constants K_2, K_0, K_1, and K_{-1} can be determined with the aid of the principle of detailed balancing. Since the vibrational state ν_3 of CH_3F is nondegenerate, ν_6 is doubly degenerate, and ν_4 of CH_4 is triply degenerate, we have at room temperature

$$K_0=0.87K_{-0};\quad K_2=0.82K_{-2};\quad K_1=1.05K_{-1}.$$

The measured values of the vibrational energy-transfer constants K_{-0} and K_{-2} correspond approximately to 160 and 45 gas-kinetic collisions whereas, according to Earl *et al.*, [2] the number of collisions for transfer between the isotopic molecules $^{12}CH_3F$ and $^{13}CH_3F$ is about 6. It is, therefore, much easier, in fact, to excite selectively CH_3F in the CH_3F-CH_4 system than in the $^{12}CH_3F-^{13}CH_3F$ system.

In conclusion, the authors express their gratitude to A. A. Savenko who participated in the experiments.

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Translated by S. Chomet

Self-induced transparency and photon echo in lithium aluminate

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(Submitted July 15, 1976)
Zh. Eksp. Teor. Fiz. **72**, 1702–1709 (May 1977)

Self-induced transparency and photon echo were investigated in a lithium aluminate crystal activated with trivalent chromium ions. The dynamics of the deformation and self-contraction of the laser beam in this crystal was investigated. A study of the character of the bleaching of the medium has made it possible to determine the electric dipole moment of the resonant transition. An experimental study of the character of the decrease of the photon-echo intensity with increasing time between the pulses has made it possible to find the values of the times of the transverse irreversible relaxation of the system in a constant magnetic field and without a field. The concentration dependence of the photon-echo intensity has a quadratic character, so that superradiance model can be used for a description of this phenomenon.

PACS numbers: 42.65.Gv, 42.60.He, 78.20.−e

1. INTRODUCTION

The phenomena of photon echo (PE)[1,2] and self-induced transparency (SIT)[3] have by now found various scientific and technical applications. The most thoroughly studied were the formation of the PE and SIT effects in gases (see, e.g., [6,7]), although they were first observed in ruby. [2,3] The reason lies in the technical difficulty of performing PE and SIT experiments in solid-state samples, which must be at liquid-helium temperatures to suppress rapid relaxation processes. The performance of such experiments, however is promising because in relatively small solid volumes it is possible to obtain a high concentration of active centers. Such investigations are needed also because so close a packing of the active centers in crystals and different (compared with gases) mechanisms of inhomogeneous broadening of the spectral lines exert appreciable influences on the course of these processes and on their parameters. The symmetry of the real environment of the active center influences the PE and SIT to a considerable degree. All this makes experiments on the observation of PE and SIT in solids an urgent problem.

The present paper is devoted to an investigation of the conditions and singularities of the onset of PE and SIT in a lithium aluminate crystal (ordered phase of $LiAl_5O_8+Al_2O_3$ [8]) activated by trivalent chromium ions.

To observe the PE and STP effects it is necessary that the time of resonant interaction of the field with the medium be shorter than the time T_2 of the transverse irreversible relaxation. In this case the nonequilibrium electric polarization of the resonance medium, which is a nonlinear function of the intensity amplitude and of the phase of the exciting waves, preserves the information on the characteristics of the coherent excitation that has been effected at preceding instants of time.

In the present experiment the duration of the exciting pulses, Δt was 10–18 nsec, and the time τ between

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pulses was varied in the range 20–120 nsec. The R_1 line of the Cr^{3+} ion spectrum was excited in the investigated crystal, which was maintained at a temperature 1.7–2.2 K, by ruby-laser radiation of wavelength $\lambda \approx 6935$ Å, the laser radiation falling in the wing of the R_1 absorption line.[9] Estimates of the times of the transverse irreversible relaxation due to the electric and magnetic dipole-dipole interactions (in the absence of a constant magnetic field) leads to values > 25 nsec. The hyperfine and super-hyperfine interactions are characterized by longer relaxation times. Relaxation rates estimated at not more than 10^5 sec^{-1} were obtained for: a) the Orbach process $1/\tau_{Orb} \propto \Delta^3 \exp\{\Delta/k_B T\}$ and b) the Raman process $1/\tau_{Ram} \propto \Delta^{-4} T^9$ (where k_B is the Boltzmann constant, T is the absolute temperature of the sample, and Δ is the splitting between some resonant sublevel and the nearest nonresonant sublevel. Thus, the conditions for coherent excitation were satisfied in this experiment. It is well known[1,2] that the intensity of the PE decreases exponentially like $\exp[-4\tau/T_2]$ with increasing τ. The apparatus used in the present experiment made it possible to record PE signals also at $\tau > T_2$ (up to values $\tau \approx 4T_2$).

2. APPARATUS AND MEASUREMENT PROCEDURE

The experimental setup used to observe the SIT process in lithium aluminate activated with Cr^{3+} ions is shown in Fig. 1. The working sample 11 was placed in the helium bath of an optical cryostat and was kept at a temperature 1.7–2.2 K. The Cr^{3+} ion concentration in the SIT experiment was 0.03 wt.%. In this case a laser beam of 0.16 cm diameter traversed a path of 0.97 cm in the sample. Preliminary investigations of the optical properties of the samples have shown (data obtained by M. N. Baranov) that it has a preferred threefold axis (which we shall call the **C** axis), and variations of the polarization of the light propagating along the **C** axis produce no changes in the absorption spectrum. In our experimental observation of the SIT we investigated the singularities of the transmission and deformation of the pulse of a ruby laser (with passive Q switching) in the direction of the **C** axis of the lithium-aluminate crystal. The active element of this laser was at a temperature ≈ 78 K. The laser pulse duration was 15–18 nsec.

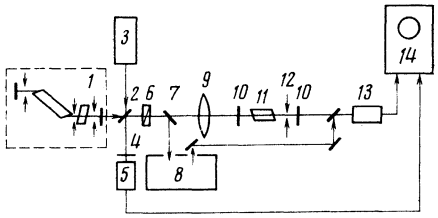

FIG. 1. Block diagram of the experimental setup for the observation of the effect of self-induced transparency: 1—ruby laser, 2—adjusting beam-splitting plate, 3—adjusting He–Ne laser (LG-36), 4—light filters, 5—high-speed photomultiplier (ÉLU-FT), 6—Glan prism, 7—beam-splitting plate, 8—optical delay line, 9—focusing lens, 10—neutral light filters, 11—investigated sample, 12—diaphragm, 13—high-speed photomultiplier (ÉLU-FT), 14—time-interval meter (I2–7).

In the experiments aimed at observing the PE in lithium aluminate, the Cr^{3+} ion concentration was 0.013, 0.018, and 0.03 wt.% (at a sample length 0.325 cm). The experimental setup used to observe PE is described in detail in[4]. In place of a passive shutter, we used a Kerr cell with two crossed Glan prism for active Q switching. An additional positive lens was placed behind the optical cryostat, and an optical shutter was placed in its focus to eliminate the light scattered by the optical elements and the sample. The shutter was synchronized with the laser radiation and opened at the instant of arrival of the PE signal. The duration of the exciting pulses was 10–12 nsec. The signals were detected with a high-speed ELU-FT photoreceiver with resolution not worse than 2.7 nsec. It was possible to apply to the setup a constant magnetic field $\mathbf{H}_0 \| \mathbf{C}$ (produced by Helmholtz coils) of not more than 200 G.

The operating ability of the setup was tested with a ruby single crystal with a resonance on the energy transition $^4A_2 \rightarrow 2E(\bar{E})$. The test revealed all the singularities typical of the PE and SIT phenomena.[1–3] In the investigation of the SIT, the ruby control sample with Cr^{3+} concentration 0.05 wt.% and length 2.1 cm was cut at a Brewster angle to the optical axis **C**, along which the laser pulses propagated.

3. SELF-INDUCED TRANSPARENCY IN LITHIUM ALUMINATE ACTIVATED WITH Cr^{3+} IONS

An important parameter of a laser pulse in a resonant medium is the "area" $\theta(z, t)$ of the pulse, defined as follows:

$$\theta(z,t) = \hbar^{-1} p \int_{-\infty}^{t} \mathscr{E}_0(z,t')\,dt', \tag{1}$$

where p is the modulus of the electric dipole moment of the transition between the resonant sublevels; $\mathscr{E}_0(z, t)$ is the amplitude of the electric field intensity of the light wave at the instant of time t' at the point z in the case of propagation along the **z** axis ($\mathbf{z} \| \mathbf{k}$); **k** is the wave vector of the pulse. We shall henceforth take θ_0 to mean the "area" of the pulse at the entrance to the sample ($z = 0$).

Self-induced transparency constitutes optical bleaching of a resonant medium under the influence of a laser pulse in the case when $\theta_0 > \pi$ and $\Delta t < T_2$. The energy absorbed by an ensemble of particles from the leading front of the pulse is re-radiated in an induced manner by the medium during the rest of the pulse. This causes deformation of the pulse and its delay in the sample. The re-radiation ("reaction") of the resonant medium in the course of passage of the laser pulse must be taken into account if

$$\alpha_r l > 1, \tag{2}$$

where α_r is the coefficient of resonant absorption ($\alpha_r \approx 3$ cm^{-1} and ≈ 5 cm^{-1} for lithium aluminate and ruby, respectively) and l is the sample length.

The reaction of the medium can be mathematically calculated by solving the simultaneous system of Max-

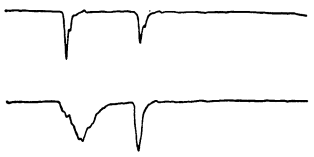

Fig. 2. Oscillograms illustrating the self-contraction (top) and lengthening (bottom) of a pulse in ruby. Right—control pulse, left—pulse at exit from the sample.

well's differential equations and the kinetic equations.[3] Calculation has shown that under these conditions the group velocity of the laser pulse in the medium is determined by the expression[3,10]

$$v=c[1+{}^1/_2\alpha_r\Delta t_r c]^{-1}, \tag{3}$$

where c is the phase velocity of the light in the medium; $1/\Delta t_r$ is the spectral width of the pulse. The pulse delay time in the sample t_d is equal to

$$t_d\approx{}^1/_2\alpha_r\Delta t_r l. \tag{4}$$

To observe the SIT it is necessary that the diffraction losses be small. This occurs if the laser-beam diameter exceeds $2(\lambda l)^{1/2}$, where l is the wavelength of the light.

The area of the pulse at the exit from the sample can be obtained from the formula[3]

$$\theta=2\,\mathrm{arctg}\left\{\mathrm{tg}\frac{\theta_0}{2}\exp[-\alpha_r l/2]\right\}. \tag{5}$$

The SIT phenomenon is accompanied by formation of a pulse with area $\theta=2\pi$ in the resonant medium (this is called a 2π pulse). The intensity and duration of each produced 2π pulse can be different, depending on the pulse area θ_0 at the entrance. If $\pi<\theta_0<2\pi$, then the area of the pulse increases after passing through the resonant medium, and its intensity decreases, thus

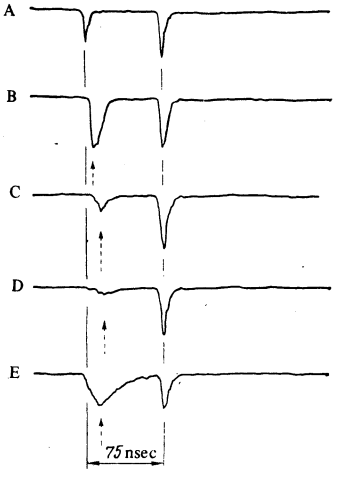

FIG. 3. Oscillograms demonstrating the process of formation of 2π pulse in ruby. The time between the pulse at the exit from the sample (left) and the control pulse (right) is 75 nsec at $\theta_0 \ll\pi$. A) $t_d=0$; B) $t_d=7$ nsec; C) $t_d=14.5$ nsec, D) $t_d=17.5$ nsec, E) $t_d=13.8$ nsec.

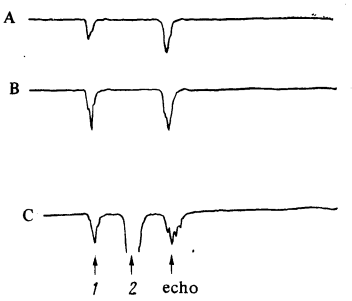

FIG. 4. A) Oscillogram showing the behavior of the laser pulse in lithium aluminate in the case $\theta_0<\pi$. B) Bleaching and self-contraction of laser pulse in lithium aluminate in the case $3\pi>\theta_0>2\pi$. C) Oscillogram of PE (first pulse on the right) in lithium luminate; 1,2—exciting pulses; $\tau=39$ nsec.

leading to an increase of the pulse duration at the exit from the medium (Fig. 2, bottom). In the case when $3\pi>\theta_0>2\pi$, the pulse area decreases and the intensity increases. As a result we have self-contraction of the pulse in the resonant medium (Fig. 2, top). If $\theta_0<\pi$, then the laser pulse is absorbed in the resonant regions in accord with the well known Beer's law.

The process of formation of a 2π pulse in a ruby sample is demonstrated by the oscillograms of Fig. 3, where the pulses at the exit from the sample (the signals on the left) are compared with control pulses that have passed through a delay line. Therefore, in the time scale (the time increases from left to right), the pulses at the exit lie ahead of the pulses at the entrance into the sample.

To determine the pulse delay time in the medium and to monitor the pulse waveform, the intensity of the pulse at the exit from the sample was matched, with the aid of calibrated neutral light filters, to the intensity of the control pulse. Oscillogram A of Fig. 3 corresponds to $\theta_0\ll\pi$, while oscillograms B–E illustrate the deformation and delay of the pulse in the ruby laser as the value of θ_0 is increased to $\approx\pi$.

The ruby sample was replaced with a lithium aluminate crystal activated with Cr^{3+} ions. Oscillograms A and B of Fig. 4 correspond to the cases of the passage of laser pulses with $\theta_0<\pi$ and $3\pi>\theta_0>2\pi$ in a lithium aluminate crystal. In the former case the pulse attenuates in accord with Beer's law, and in the latter case we have SIT accompanied by self-contraction of the laser pulse for 2–3 nsec, in agreement with the predictions of the theory.[3]

Figure 5 shows the experimentally obtained plot of

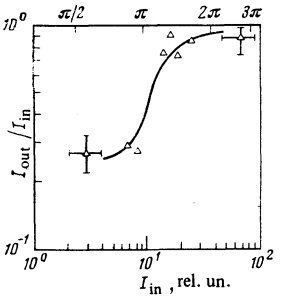

FIG. 5. Experimental dependence of I_{out}/I_{in} on I_{in} for a lithium aluminate crystal activated with Cr^{3+} ions.

I_{out}/I_{in} against the values of I_{in} (where I_{out} and I_{in} are the intensities of the laser pulses at the entrance and exit of the sample), thus demonstrating the bleaching of the resonance medium in the case of SIT. It is known[3,7] that the inflection point I_{infl} of this curve corresponds to a pulse, so that we can use the formula $p \approx \pi\hbar c^{1/2}/5I_{infl}^{1/2}\Delta t$ to estimate the electric dipole moment of the resonant transition: $p \approx 3\times10^{-21}$ cgs esu. The observed pulse delay in the sample for the SIT in the lithium aluminate crystal was 5 nsec. The experimental values of t_d are smaller than the theoretical values obtained from (5), both for lithium aluminate and for ruby. This may be due to the noncentral character of the excitation of the inhomogeneously broadened lines.[11]

4. PHOTON ECHO IN LITHIUM ALUMINATE

Photon echo is a coherent optical response of a resonant system to the action of two laser pulses applied with a time delay τ between them. This signal is generated at the instant 2τ in a direction satisfying the following spatial synchronism condition:

$$\mathbf{k}=2\mathbf{k}_2-\mathbf{k}_1,$$

where $\mathbf{k}$, $\mathbf{k}_1$, and $\mathbf{k}_2$ are respectively the wave vectors of the echo and of the first and second pulses. An oscillogram of a PE signal observed in lithium aluminate is shown in Fig. 4C.

The conditions for the realization of the present experiment correspond to the case of excitation, by long laser pulses, of a system of particles with resonance on a spectral line with large inhomogeneous broadening ($\Delta t > T_2^*$, where T_2^* is the time of the transverse reversible relaxation). The PE theory developed for this case[12-14] shows that the total intensity of this coherent response can be calculated from the following formula:

$$I=I_0N^2\frac{\lambda^2}{4S}\sin^2\theta_1\sin^4\frac{\theta_2}{2}\exp\left\{-4\frac{\tau}{T_2}\right\}AB, \qquad (6)$$

where N is the number of active particles; I_0 is the intensity of the spontaneous emission of an individual particle; θ_1 and θ_2 are the areas of the first and second exciting pulses; S is the area of the cross section of the laser beam; A is a factor that describes the influence of various random processes such as thermal oscillations of the environment of the paramagnetic center and

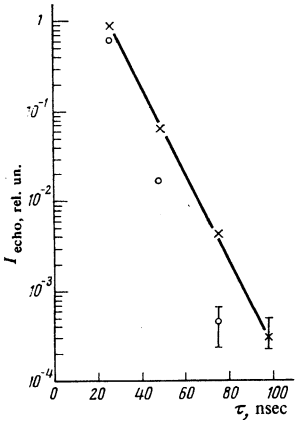

FIG. 6. PE intensity in lithium aluminate vs time τ between pulses: ×—H = 160 G, ○—H = 0.

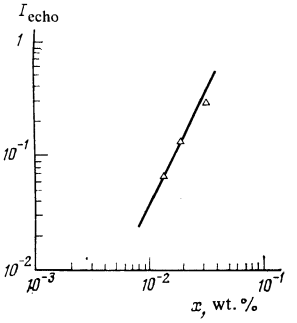

FIG. 7. PE intensity I_{echo} in lithium aluminate vs the concentration x of the Cr^{3+} ions.

rotational reorientation of the particles[15] ($A \sim 1$ at liquid-helium temperatures); $B=\frac{9}{4}(T_2^*/\Delta t)^2$. The presence of the factor B in (6) is due to the fact that in our case only a group of "spin packets" was excited[12] rather than the entire inhomogeneously broadened line. Bearing in mind that for lithium aluminate activated with Cr^{3+} ions we have $T_2^* \sim 10^{-11}$ sec, the presence of the factor B explains the low intensity of the echo in comparison with the intensity of the exciting pulses.

In the present experiment we plotted the PE intensity I against the time. The measurements were made both in an external magnetic field $H_0 = 160$ G and without a field (Fig. 6). Since $I \sim \exp\{-4\tau/T_2\}$, we determined from the experimental $I(\tau)$ plots the values of the time of irreversible relaxation, viz., $T_2 = 26.8$ nsec ($H_0 = 0$) and $T_2 = 35.5$ nsec ($H_0 = 160$ G).

In addition, we investigated the intensity of the PE as a function of the intensity of the constant magnetic field when the direction of the echo polarization vector coincided with the direction of the exciting-pulse polarization. It was observed that the influence of an external magnetic field on the PE intensity in lithium aluminate is much smaller than in ruby, apparently because of the larger local magnetic field H_L ($H_L \approx 12$ G in ruby[2]).

It follows from (6) that the intensity of the PE is proportional to the square of the number of active centers. Following Dicke,[16] such signals are customarily called superradiance. Figure 7 shows the concentration dependence of the PE intensity in lithium aluminate activated with Cr^{3+} ions. The triangles correspond to the experimental point and the solid line is the theoretical plot ($I \sim N^2$). In the quantitative measurements, all the sample parameters were kept constant, except the concentration of the Cr^{3+} ions. The plot obtained at Cr^{3+} ion concentrations 0.013–0.03 wt.% allows us to state that the superradiance formalism can be used to describe the PE.

The authors thank I. Kh. Khadyev for help with the experiments, and E. F. Kustov and M. N. Baranov for supplying the crystals.

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Theory of hysteresis reflection and refraction of light by a boundary of a nonlinear medium

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(Submitted July 29, 1976)
Zh. Eksp. Teor. Fiz. **72**, 1710–1726 (May 1977)

A theory is constructed for the reflection and refraction of light from the boundary of a semi-infinite nonlinear medium whose refractive index depends on the light intensity. It is shown that when the incidence angle or the intensity of the incident light is varied, hysteresis jumps should be observed from the transmission regime to the regime of total internal reflection (TIR) and back. At small nonlinearity, the necessary condition for the existence of the hysteresis effect is closeness of the linear refractive indices of both media (linear and nonlinear) and smallness of the glancing angles; all the observed effects should in this case be independent of the polarization of the incident field. At a negative nonlinearity, the phenomenon is due to the ambiguity of the transmission regime, while at positive nonlinearity it is due to the ambiguity of the TIR. At a definite light intensity, complete transparentization of the boundary can take place for all the incident angles in the region of stability of the transmission regime; in this case jumps take place from total reflection to total transmission and vice versa.

PACS numbers: 42.65.Bp

INTRODUCTION

1. In a preceding paper[1] I reported the possibility of observing new effects in the case of almost glancing incidence of light from a linear medium onto the boundary of a nonlinear medium whose permittivity ε_1 depends on the field intensity and is close to the permittivity ε_0 of the linear medium. The principal effect was that when the glancing angle or the incident-field intensity were varied, strong hysteresis jumps should be observed in the refractive index and in the reflection coefficient, from the nonlinear reflection regime ("transmission regime") to the regime of nonlinear total internal reflection (TIR), and back. A theory of this phenomenon is constructed in the present paper.

The Snell formulas that follow from the generalized boundary conditions (§ 1) for the refraction angle and the Fresnel formulas for the amplitudes of the fields become coupled to one another by virtue of the nonlinearity of the refracting medium, and this leads to a single self-consistent equation for the nonlinear refraction (§ 2). In the case of TIR, a surface wave propagates inside the refractive medium along the boundary, and is likewise described by a nonlinear wave equation (§ 3). The self-consistency of these equations leads, under definite conditions, to the appearance of severally physically realizable solutions (states), some of which are unstable and this in fact is the reason for the hysteresis. The transmission regime becomes ambiguous at negative nonlinearity ($\Delta\varepsilon_{nl} < 0$, § 4), and the TIR regime at psoitive nonlinearity ($\Delta\varepsilon_{nl} > 0$, § 5). At a certain field intensity, total nonlinear transparentization of the boundary takes place at all incidence angles (§ 2), and the hysteresis jumps take place between the states of the total transmission and the total reflection, while the boundary operates as an ideal "optical flip-flop" (§§ 4, 5).

2. One of the main premises for the observation of the indicated effect is the matching of the optical densi-

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