Investigation of the mechanism of broadening of resonance lines in ruby by the photon echo method

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From an analysis of the experimental dependence of the photon echo on the polarization, contour, and propagation directions of the exciting pulses, on the intensity of the constant magnetic field, and on the time τ between the pulses, it is found that the decisive contribution to the damping of the echo in the ruby, at a Cr^{3+} ion concentration ~ 0.1 at.%, is made by magnetic dipole-dipole interactions. It is established that this damping is determined by the exponential $-\exp(-A\sqrt{\tau})$, where A is a damping constant corresponding to the magnetic dipole-dipole interactions.

The photon echo method[1,2] is effective both in investigations of the nature of the broadening of the resonance line and the characteristic relaxation time,[3–5] in the analysis of the dynamics of generation of optical pulses, and in the study of their waveform and spectral composition. This is made possible by the fact that the optical quantum system consists of a set of quasi-independent "spin packets," which interact in different manners with each of the exciting pulses. Therefore the anomalies in the waveform and in the instants of the appearance of the coherent responses are connected to a large degree with the extent to which the sequence of exciting pulses recombines these elementary acts of ion-photon interaction. Calculation shows that in the case of nonresonant and multifrequency excitation of coherent responses, time shifts should be observed, together with waveform distortions and changes in the characteristics of the photon echo. When the pulse duration is decreased (and consequently their spectra are broadened), these phenonema become more and more manifest, as is confirmed by our experiments.

The photon-echo signal is a coherent spontaneous optical response of a resonant medium to a two-pulse laser excitation. The duration Δt of the exciting pulses and the time τ between them should be shorter than all the irreversible relaxation times. Figure 1 shows the oscillograms of the signals in ruby, which were obtained by us experimentally. As seen from Fig. 1a, the maximum of the photon-echo signal takes place at the instant of time 2τ. The direction of the wave vector $\mathbf{k}_e$ of this signal satisfies the spatial-synchronism condition $\mathbf{k}_e = 2\mathbf{k}_2 - \mathbf{k}_1$, where $\mathbf{k}_\eta$ $(\eta = 1, 2)$ are the wave vectors of the exciting pulses. In a number of cases, however, the instant of the appearance of the maximum of the echo was shifted from the instant 2τ by 4–5 nsec. In addition, a correlation was observed between the waveform of the echo and the contours of the exciting pulses. The oscillograms of such signals (we shall call them "anomalous") are shown in Fig. 2.

The working sample, in the form of a plate (with sides 1×1 cm and thickness 0.07 cm) of Al_2O_3 crystal

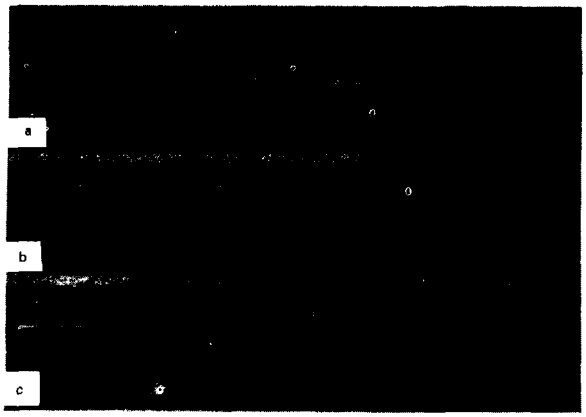

FIG. 1. Oscillograms of observed photon-echo signals in ruby; τ - 44.5 nsec; H $\parallel C_0$, where C_0 is the optical axis in ruby; $H = 200$ Oe: a—two exciting laser pulses and photon-echo signal (third on the left); b—action of first pulse only; there is no echo; c—action of second pulse only; there is no echo.

width Cr^{3+} iron concentration ~0.1 at.% was at liquid-helium temperature. The excitation was produced by ruby-laser radiation (the active element of the laser was kept at liquid-nitrogen temperature) at a wavelength $\lambda \approx 6935$ Å. Under these conditions, the frequency of degeneration of the ruby laser (which operated in an active Q-switching regime) and the frequency of the transition ${}^4A_2(M=\pm\frac{1}{2}) \longrightarrow {}^2E(\bar{E})$ in the investigated sample coincide. The pulse duration was 7—10 nsec, and the time between pulses was varied between 40 and 120 nsec. The light signals were received with the aid of a high-speed ÉLU-FT photoreceiver with a resolution not worse than 2.7 nsec, the output signal of which was fed to a time interval meter I2-7 (sweep ~250 nsec). The constant magnetic field H was produced by Helmholtz coils.

The experimentally obtained plots of the echo intensity (I) against τ and against the angle ϕ between the analyzer used to record the echoes and the polarization vectors l_1 and l_2 ($l_1 \parallel l_2$) of the exciting pulses are shown in Fig. 3.

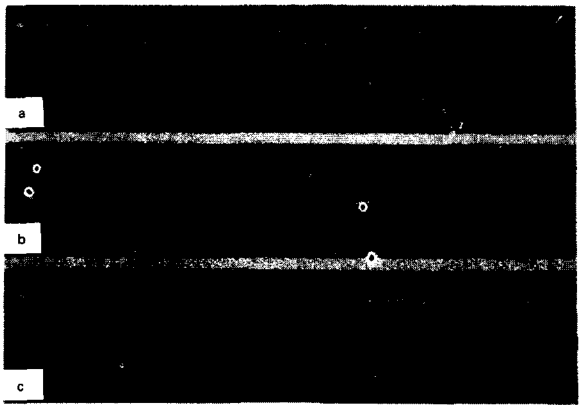

FIG. 2. Oscillograms of "anomalous" photon-echo signals: a—photon-echo signal with non-Gaussian contour; $\tau = 44.5$ nsec; H $\parallel C_0$ and $H = 65$ Oe (the fourth signal on the left can be easily eliminated by passing the first two exciting pulses through diaphragms); b—the same, $\tau = 82$ nsec, H $\parallel C_0$, and $H = 210$ Oe; c—shift of the instant of appearance of the photon-echo signal; $\tau = 82$ nsec, H $\parallel C_0$, and $H = 210$ Oe.

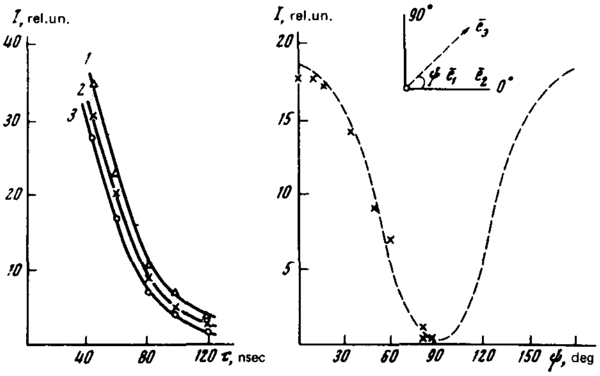

FIG. 3. Dependences of the photon-echo intensity on τ and ψ: a) 1—in field $H = 200$ Oe; 2—$H = 250$ Oe; 3—$H = 177$ Oe, b) ψ is the angle between the analyzer that records the echo and the linear polarization of the first and second pulses; $l_1 \parallel l_2$; l_e is the echo polarization vector.

Theoretical investigations of the damping mechanisms in ruby[6] have shown that at a Cr^{3+} concentration ~0.1 at.% the following can predominate among them: 1) electric dipole-dipole interactions of the Cr^{3+} ion; 2) magnetic dipole-dipole interactions of the Cr^{3+} ion; 3) electron-nuclear $Cr^{3+} \longleftrightarrow Al^{27}$ interactions.

Estimates show that by virtue of the small population of the excited electronic levels (as a result of which the "flip-flop" processes are "frozen") the relaxation due to the mechanism 1 will not be shorter than 5×10^{-7} sec. The remaining two mechanisms influence the echo formation differently. When mechanism 2 predominates, the decrease of the echo intensity with increasing τ should have a monotonic character. When mechanism 3 predominates, owing to the hyperfine interaction, a transverse magnetization component will be induced at the Al^{27} nuclei, and this component will modulate, in the course of precession, the splitting of the working electron levels; as a net result this modulates the $I(\tau)$ dependence.[4] Under the conditions of our experiment the $I(\tau)$ curve has, as seen from Fig. 3a, a monotonic character, so that we can conclude that the decisive contribution to the damping of the echo is made by magnetic dipole-dipole interactions.

The investigation of the experimental $I(\tau)$ curve has shown that the decrease of the intensity with increasing τ is determined by the function $\sim \exp(-A\sqrt{\tau})$, where the decrement A is equal to 1.7×10^4 sec$^{-1/2}$. Theoretical calculations of this component with allowance for the magnetic dipole-dipole interactions between the Cr^{3+} ions, using the procedure of[6], yield $A \approx (4-6)\times10^5\sqrt{C}$, where C is the chromium ion concentration. Since C is of the order of 10^{-3} in the investigated sample, it appears that the magnetic dipole-dipole interactions explain the character of the fall-off of the $I(\tau)$ curve. This conclusion is confirmed also by Anderson's theory,[7] within the framework of which the "square-root" layer is attributed to the contribution made to the relaxation by the dipole-dipole interactions. In particular, in the case of quadrupole-quadrupole interaction, the decrease in accordance with this theory should be given by $\sim \exp[-B\tau^{3/8}]$, where B is the decrement.[5,7] Thus, the character of the fall-off can serve as a criterion for the

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predominance of a definite type of interaction. The polarization curve $I(\psi)$ (Fig. 3b) obtained by us, unlike in[2], has in the case $l_1 \parallel l_2$ a minimum for the values $\psi = n(\pi/2)$, where $n = 1, 3, 5, \ldots$. As is well known,[2] when the angle between l_1 and l_2 is 45°, the behavior of the polarization curve has a similar character. This behavior of the echo polarization, as follows from the theory,[8] is determined by the type of the transition ($J = \frac{1}{2} \longleftrightarrow J' = \frac{1}{2}$).

An analysis of the conditions for the appearance of distortions in the wave form of the echo and of a shift of its maximum allows us to suggest that inasmuch as the inhomogeneous width in ruby is $\Delta\nu^* \sim 10^9$ Hz, it follows that pulses with $\Delta t \sim 10$ nsec will excite not the entire inhomogeneously-broadened line, but only a group of spin packets. The waveform of the echo will then be sensitive to which part of the resonance-line contour is "cut out" by the pulse and depends on the waveform of the pulses. The formulas of[9] underestimate the shift of the echo maximum in comparison with the data obtained in our experiment (4—5 nsec).

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