Effect of short heat pulses on coherently excited optical centers in ruby

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A change caused in the amplitude of the coherent emission of the system of Cr^{3+} impurity centers in Al_2O_3 by nonequilibrium terahertz-frequency phonons has been observed. A change has also been observed in the intensity of the optical echo in ruby when a pulsed magnetic field was applied before the lasing pulses. In this case the echo modulation cannot be described by the known mechanisms involving the effect of an rf field on an impurity spin system.

As part of the research on the physics of terahertz-frequency phonons, a significant number of studies, beginning with the experiments by Renk and Deisenhofer,[3] have been devoted to the optical detection of nonequilibrium acoustic phonons. In most experiments of this sort, optical pumping puts an impurity in an excited state, and the appearance of nonequilibrium terahertz phonons in the sample causes a redistribution of the luminescence intensity in the various spectral lines. In particular, the absorption of 29-cm^{-1} phonons by excited Cr^{3+} ions in ruby at liquid-helium temperature gives rise to a luminescence in the R_2 line, which is associated with a radiative relaxation of the $2\overline{A}(^2E)$ state. The primary relaxation mechanism is not the $2\overline{A}(^2E) \rightarrow {}^4A_2$ transition to the ground state but a return to the $\overline{E}(^2E)$ state accompanied by the emission of a 29-cm^{-1} phonon. As a result, it is necessary to detect the R_2-line luminescence by a photon-counting method.[4]

Experiments of this sort concern incoherent states; changes in phase relations upon transitions have no effect on the signal which is detected. It would be natural to suggest that in phase-sensitive experiments, such as optical-echo experiments, the existence of a relaxation mechanism involving a loss of phase would lead to a significant change in the detectable signal. One could work from the change in the intensity of the optical-echo signal to learn about the number of excited Cr^{3+} ions which are sent into the $2\overline{A}(^2E)$ state by the phonon pulse. It would be virtually impossible to estimate this number from the change in the R_1 luminescence in the nanosecond range, since the time scale of the relaxation between the components of the ^2E doublet is less than 10^{-9} s.

In this letter we are reporting a study of how nanosecond current pulses applied to a thin-film heater on the sample affect the magnitude of the optical echo in ruby with Cr^{3+} concentrations of 0.6% and 0.9%. The measurements were carried out under inverse-echo conditions at $T_0 = 2$ K in a static magnetic field $H = 120$ Oe, directed parallel to the C_3 axis. The echo was excited by pulses (with a length $\tau = 10$

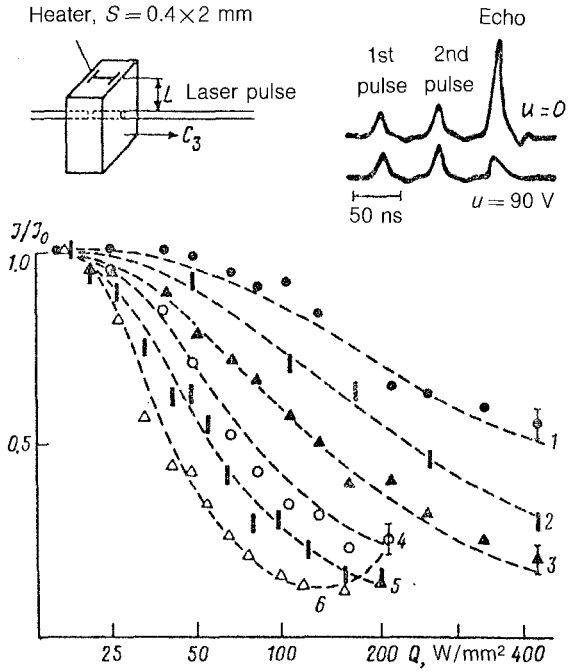

FIG. 1. Relative intensity of the optical echo in ruby with a Cr^{3+} concentration of 0.9% (curves 1–3) or 0.6% (curves 4–6) versus the power supplied to the heater, Q. 1, 4—$L = 1$ mm; 2, 5—$L = 0.75$ mm; 3, 6—$L = 0.5$ mm. The current pulse is applied at the same time as the first laser pulse. Curves 1–3 were recorded with the help of a heater with dimensions of 0.4×1 mm. Some typical oscilloscope traces of the observed signals are shown at the top.

ns) from a low-temperature ruby laser with passive Q switching. The angle between the first laser pulse and the second was 2°; the delay between the first and second pulses was $\tau_{12} = 57$ ns. Current pulses with an amplitude $0 \leqslant U \leqslant 100$ V and a length $\tau_h = 17$ ns were generated by an avalanche-transistor source and applied to the heater, a resistance $R = 50\ \Omega$, in synchronization with the laser pulses.[5] Where necessary, the current pulse could be applied before the first laser pulse, which would be sent through an auxiliary optical delay line in this case. When a lens with a focal length $F = 50$ cm was used, the optical echo was formed over the entire thickness ($d = 5$ mm) of the sample, in a filament about 300 μm in diameter. The region of optical excitation could be moved through the sample with the help of a micrometer and could be set within 50 μm.

It was observed that the application of a current pulse to the heater causes a sharp decrease in the intensity of the optical echo (Fig. 1). The nonequilibrium phonons emitted by the heater excite $E(^2E) \rightarrow 2\overline{A}(^2E)$ transitions when they reach the optically excited volume. These transitions disrupt the phase of the coherent state. It can be shown that the intensity of the echo in this case is given by

$$J = J_0 \cos^2 \sqrt{n(\hbar\omega/k_B T_e A(c, L, \tau_h)}, \qquad (1)$$

where n is the phonon distribution function, and T_e is the electron temperature in the heater film. The experimental results (the dashed lines in Fig. 1) can be described well by expression (1) in the Perrin–Budd model[6] with appropriate choices of the quantity $A(c,L,\tau)$ (a function of the impurity concentration c), L (the distance between the heater and the excited volume), and τ_h (the length of the current pulse at the heater).

Further experiments showed, however, that phonons are not the only entities playing an important role in the decay of the echo intensity. As the distance L is increased in experiments in which the current pulse and the first laser pulse are applied at the same time, not even the ballistic LA phonons, which have the highest propagation velocity, manage to reach the excited volume by the time ($t = 2\tau_{12}$) of echo formation, starting at $L_{\max} = 2\tau_{12}V_1$ ($V_1 = 11.1$ km/s is the velocity of the longitudinal-polarization phonons). Nevertheless, as curve *1* in Fig. 2 shows, the condition $J/J_0 < 1$ holds even at $L > L_{\max}$. It was suggested that a pulse of a weak transverse magnetic field, $H_h = i/2\pi L \simeq 4$ Oe, which arises as the current i flows through the heater, plays an important role in the decay of the echo signal. There were two ways to test this suggestion.

First, the phonon component of the current pulse could be suppressed. For this purpose, the heater was placed on a separate substrate at a distance $L' \simeq 0.4$ mm from the ruby sample. In this case, with $L = L' + 0.5$ mm and $U = 90$ V, we observed an echo smaller by a factor of 6 or 7 (Fig. 3), despite the fact that an effect of the thermal phonons was completely ruled out.

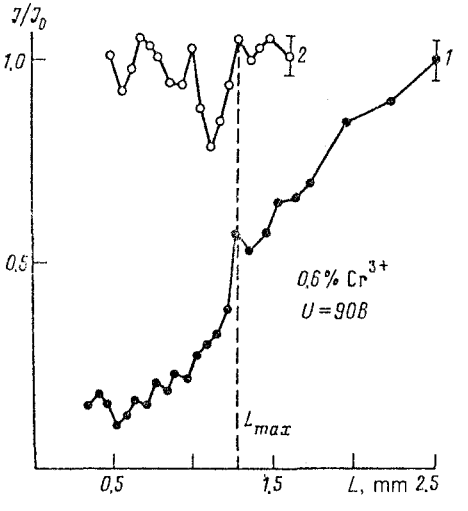

FIG. 2. Plot of J/J_0 in ruby versus the distance to the heater, L. 1—The heater is applied to the sample as a strip with dimensions of 0.4×2 mm; 2—the heater is in the form of a meandering pattern. The current pulse is applied at the same time as the first laser pulse.

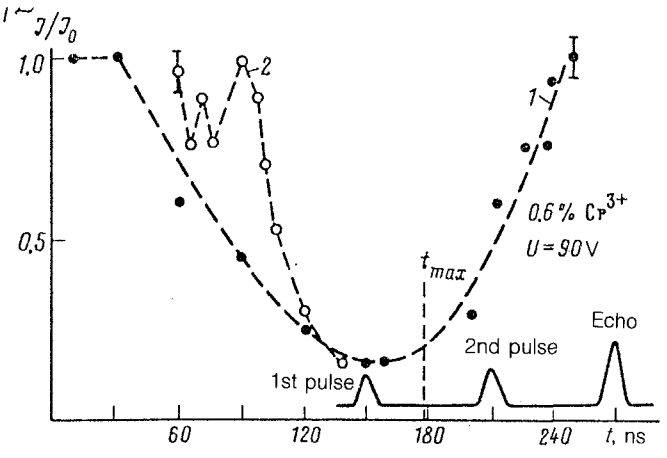

FIG. 3. Optical-echo signal versus the time at which the current pulse is applied to the heater. 1: The heater is sputtered on the sample. Here t_{max} is the latest time at which LA phonons manage to reach the excited volume by the time at which the echo is formed. 2: The heater is separated from the sample by a layer of helium with a thickness $L' = 0.4$ mm.

Second, the magnetic component of the current pulse could be suppressed or at least substantially reduced. For this purpose, a heater in the form of a meandering pattern, cut from strips 0.4 mm wide with a gap of 0.08 mm, was placed on the sample. The results are shown by curve 2 in Fig. 2.

Both curves in Fig. 2 show that the arrival of the ballistic LA phonons at the excited volume causes a significant decrease in the echo signal. Since the intensity of the echo signal is proportional to the square of the population of the excited level, N^2, we can thus estimate the relative change which occurs in the population of the $\overline{E}(^2E)$ level as a result of the phonon pulse: $\Delta N/N \simeq 0.15$. We recall that in the usual experiments this value would be masked by the short relaxation time of the $2\overline{A}(^2E)$ level.

The measurements showed that in a zero external magnetic field the quantity J/J_0 is comparable to the signal modulation amplitude in a field $H \simeq 120$ Oe. In those cases (Fig. 3) in which the magnetic pulse is applied after the first light pulse ($t_1 < t_h < t_1 + 2\tau_{12}$), the modulation of the echo amplitude is described by[1,2]

$$J = J_0\{1 - 2\sin^2\vartheta\cos^2\Omega_2(2\tau_{12} + t_1 - t_h)\}\exp(-2\gamma\tau_{12}), \qquad (2)$$

where $\vartheta = \mu_e H_h \tau_h/\hbar$ is the area under the field pulse, $\Omega_2 \simeq \omega_c$ is the shift of the precession phase velocities of the electric dipoles of the coherently excited impurities, ω_c is the superhyperfine interaction of the Cr^{3+} ions with the Al^{27} nuclei, in frequency units, and γ is the lifetime of the optically excited coherent state.

At $t_h < t_1$ the echo modulation is described empirically by a similar expression with $\Omega_1 \neq \Omega_2$. From the data in Fig. 3 we can find the modulation frequencies: $\Omega_1 = 1.7 \times 10^7$ rad/s and $\Omega_2 = 3.1 \times 10^7$ rad/s. From the change in J/J_0 as a function

of the pulsed magnetic field (Fig. 1), we find the constant of the interaction ($\mu_{\text{eff}} H_h$) of the field with the impurity spin system. In frequency units, it turns out to be $\omega_{\text{int}} \simeq 7 \times 10^7$ rad/s. This result is on the order of the energy of the interaction of the field with the electron spin system of the Cr^{3+} impurity, $\mu_{\text{eff}} \simeq \mu_e$.

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