Anisotropic capture of 29-cm $^{-1}$ phonons in a ruby

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Using the capture of 29-cm $^{-1}$ phonons in an excited ruby, we observed experimentally the theoretically predicted[1] mechanism of radiation capture, which is connected with the anisotropy of the medium.

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The resonance capture of acoustic 29-cm $^{-1}$ phonons in optically pumped ruby Al_2O_3:Cr^{3+} is caused by multiple resonance scattering of phonons as a result of the $\bar{E} \rightleftarrows 2\bar{A}$ transition in the excited Cr^{3+} ions.[2] The resonance scattering of 29-cm $^{-1}$ phonons is elastic[3–5] and the phonons leave the material mainly by spatial diffusion and anharmonic decay.[2–5] The capture mechanism for the 29-cm $^{-1}$ phonons is basically different from the known Holstein–Biberman mechanism of resonance capture of phonons in a gas, in which, because of Doppler and collisional broadening of spectral lines, the phonon frequency changes significantly during scattering and the radiation is

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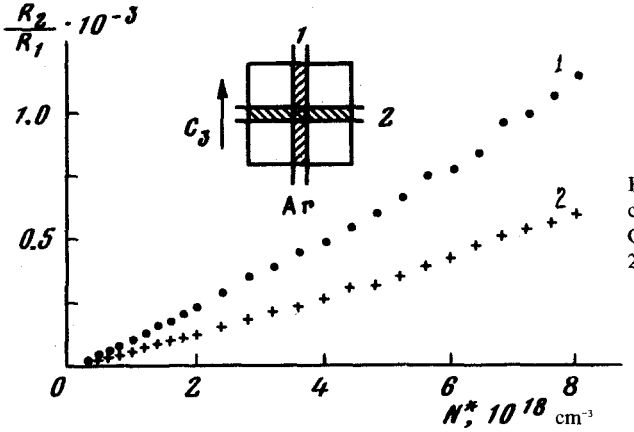

FIG. 1. Dependence of R_2/R_1 on concentration N^* of the excited Cr^{3+} ions: 1—exciting beam $\|C_3$; 2—exciting beam $\perp C_3$.

emitted from the bulk of the material, predominantly in the weakly absorbing wings of the line ("self-reversal").[6,7]

In this paper we use the 29-cm^{-1} phonons to report an experimentally observed new mechanism for release of the captured radiation, which is closely connected with the anisotropy of the system.[1,8]

Investigation of the phonon capture was carried out using oriented Al_2O_3:0.02 at% Cr single crystals at $T = 1.8$ K, according to the method described in Ref. 9. A steady-state pumping by an argon-ion laser ($\lambda = 5145$ Å) was used to excite the sample with a concentration N^* of Cr^{3+} ions in the metastable ^2E state. The sample had a strongly anisotropic "cylindrical" shape of a laser beam with the radius $r \approx 0.3$ mm and length ≈ 10 mm (Fig. 1). The relative intensity of the steady-state luminescence lines $R_2(2\bar{A} \rightarrow {}^4A_2)$ and $R_1(\bar{E} \rightarrow {}^4A_2)$ was measured. From the balance relations we obtain

$$\frac{R_2}{R_1} \approx \frac{I_{ph}}{I} \cdot \frac{T_1 \sigma}{\tau_R}, \qquad (1)$$

where I_{ph} is the rate of generation of the 29-cm^{-1} phonon in the sample, as a result of the $2\bar{A} \rightarrow \bar{E}$ transitions during the relaxation from the upper excited states of Cr^{3+}, I is the rate of excitation of the Cr^{3+} ions to the $^2E(\bar{E}, 2\bar{A})$ state (ratio $I_{ph}/I = 0.28$ [10]), $T_1 = 1.1$ nsec [10] is the lifetime of the $2\bar{A}$ level relative to the spontaneous single-phonon $2\bar{A} \rightarrow \bar{E}$ transition, τ_R (≈ 4 msec) is the time of the radiative decay of the $2\bar{A}$ and $\bar{E}$ levels with the emission of the R lines, σ is the phonon-capture factor, which is equal to the average number of scattering events of the 29-cm^{-1} phonons produced during their residence in the sample.

It was found that the capture factor σ in the extended cylindrical sample is anisotropic: its value depends on the orientation of the cylinder in the sample with respect to the crystallographic axis C_3. Figure 1 shows the dependence of R_2/R_1 on the concentration N^* of the metastable Cr^{3+} ions for two orientations of the pump laser beam in the crystal: $\|C_3$ and $\perp C_3$. For any fixed value of N^*, the R_2/R_1 ratio, i.e., the capture factor σ, is 1.5–2 times greater for the $\|C_3$ orientation than for the $\perp C_3$

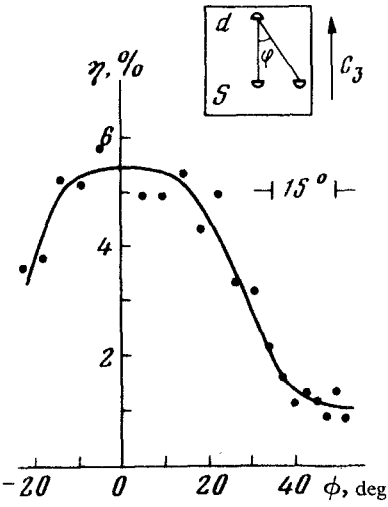

FIG. 2. Angular dependence of η. In both volumes $N^* = 2\times10^{17}$ cm^{-3}.

($\sigma^{\parallel} > \sigma^{\perp}$) orientation. This result is also confirmed by direct measurements of the capture time of phonons in the cylindrical volume by using the pulsed method.[2] Thus, the experiment indicates the presence of a certain specific "anisotropic" channel for the escape of the 29-cm^{-1} phonons from the excited volume. The channel is effective in that geometry of the experiment in which the small volume (cylinder radius r) is directed along the C_3 axis.

To interpret this effect, we proceed from the concepts[1] which predict theoretically the possibility of this kind of anisotropic effect. We attribute the observed effect to microanisotropy of phonon interaction with chromium ions[1,1] under the conditions of phonon mode mixing during scattering. In fact, the phonon wave vector $\mathbf{q}$ and polarization $\mathbf{u}$ change as a result of preserving the phonon frequency during scattering. The transverse (TA) and longitudinal (LA) phonons have different lengths of the free path $\bar{l}$ with respect to the resonance scattering by the Cr^{3+} ions (^2E). Moreover, the LA phonons with $\mathbf{q}\|C_3$ generally do not interact with the $\bar{E}\rightleftarrows 2\bar{A}$ transition (which is forbidden by the selection rules) and become absorbed as $\mathbf{q}$ deviates from C_3 and increases with increasing angle $\theta = \sphericalangle\mathbf{q}C_3$.[1,1] Consequently, the "axial" LA phonons with the direction $\mathbf{q}$ near C_3, which are generated as a result of scattering, may emerge from the excited volume without reabsorption. Evidently, the critical angle of free escape is θ_{max}, such that $\bar{l}(\theta_{max}) \approx L_{\parallel}$, where $L_{\parallel}$ is the length of the volume along C_3. In a cylindrical volume with the axis $\perp$ to C_3, $L_{\parallel}$ is small and equal to $r \approx 0.3$ mm, whereas the angle of taper for the free escape of phonons θ_{max} is sufficiently large to provide effective phonon escape. However, when the cylinder is $\|C_3$, $L_{\parallel} \approx 10$ mm, the angle θ_{max} is very small, and the escape channel for the axial phonons is almost blocked. This was observed in the experiment (Fig. 1) where the phonon-capture factor in the first case ($\sigma_{\perp}$) was noticeably smaller than in the second ($\sigma_{\parallel}$).

Our interpretation was confirmed by direct observation of the "axial" emission of captured phonons from the excited volume. By splitting the laser beam into two separate beams in the crystal, we produced two parallel excited cylindrical volumes spaced

the region of small energy densities at the frequency 947.98 cm^{-1}, the dependence of the absorbed energy E on the density of the irradiation energy Φ has the form $E \sim \Phi^{0.5}$ which indicates that the single-photon absorption near the Q branch of the main 0–1 transition is the dominant contribution. For the peak at the frequency 944.46 cm^{-1} this dependence has the form $E \sim \Phi^{1.65}$ in the entire range of investigation. This peak, is apparently connected with the Q branch of the two-photon transition to one of the sublevels of the vibrational state $v = 2$. The frequency position of this peak and the data on the frequency of the $3\nu_3 SF_6$ harmonic[6,7] lead us to conclude that the anharmonic splitting in SF_6 is large.

It follows from Eq. (1) that the observed increase of R_2/R_1 may be attributable only to the increase of phonon generation I_{ph}, i.e., additional entry (ΔI_{ph}) into the "d" volume of phonons emitted by the "s" volume: $\eta = \Delta I_{ph}/I^0_{ph}$, where I^0_{ph} is the generation of phonons in the "d" volume due to the $2\bar{A} \rightarrow \bar{E}$ transitions. The absence of any effect of the "s" volume on the capture factor σ (on N^*) in the "d" volume was verified specially. Thus, there is an emission of phonons from the "s" volume, which is concentrated predominantly in the directions close to C_3. The focused phonon emission is also confirmed by the relatively weak experimental dependence of η on the spacing between the "s" and "d" volumes.

In conclusion, we emphasize that the observed mechanism of "anisotropic" capture is analogous to the Holstein–Biberman capture mechanism.[1] In both cases, as a result of multiple scattering, the radiation is converted to weakly absorbing modes which are emitted from the sample without reabsorption. If the Holstein–Biberman model represents conversion of the spectral radiation frequency, then our model represents transformation of the polarization and of the wave vector of (phonon) radiation.

[1)]The emission of phonons, which were delayed as a result of capture in the excited volume, was observed in Ref. 11.

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