Propagation of 29-cm^{-1} phonons in ruby under conditions of resonant scattering, and estimate of their lifetimes

A. P. Abramov, I. N. Abramova, I. Ya. Gerlovin, and I. K. Razumova

S. I. Vavilov State Optical Institute
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It is demonstrated experimentally that there is no "bottleneck" effect in experiments on optical detection of 29-cm^{-1} phonons in ruby. It is concluded that at 2 K the lifetime of the 29-cm^{-1} phonons exceeds 2.7 μsec.

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Experiments on optical detection of 29-cm^{-1} phonons, carried out on ruby crystals,[1–4] have led to a simple and lucid interpretation of the phenomena that occur at high concentrations of excited Cr^{3+} ions. When the excited chromium ion absorbs a phonon, the upper component of the state $^2E\ \bar{E} \rightarrow 2\bar{A}$ transition is populated. The presence of phonons is revealed by the luminescence from the state $2\bar{A}$ (R_2 line) at 2 K. Since the main relaxation channel of the $2\bar{A}$ state is the return to the $\bar{E}$ state with emission of a 29-cm^{-1} phonon, the excited chromium ions, while making it possible to detect 29-cm^{-1} phonons, serve simultaneously as resonant-scattering centers for these phonons. This scattering comes into play at excited-ion concentrations $C > 10^{14}$ cm^{-3}, when the ballistic flight of the phonons through the excited volume gives way to diffuse motion.[1] As a result, with increasing C, the fall-off time (τ) of the R_2-line luminescence observed following a short-duration injection of thermal phonons into the crystal increases. It was observed[2] that the increase of τ with increasing excitation density is subject to saturation; this was interpreted as the result of complete "dragging" of the phonons in the illuminated volume adjacent to the heater. The limiting damping time corresponding to these conditions (1.5 μsec) was ascribed to the proper phonon lifetime due to their spontaneous decay.

Subsequent experiments have shown that τ continues to increase slowly with further increase of the excitation density, without experiencing saturation. This growth was attributed in[3] to a "bottleneck" effect wherein the measured lifetime consists of the phonon lifetime ($\tau_p = 1.4$ μsec) and the total time that the chromium ions remain in the $2\bar{A}$ state during the intervals between the phonon absorption and emission. If at $C \approx 10^{16}$ cm^{-3} the phonon experiences during its lifetime approximately 10^3 reabsorptions, then the total time increases to 2.5 μsec.

The interpretation presented in[3], which is based on the analysis of only the trailing edge of the luminescence pulse rather than on the entire pulse shape, is not sufficiently well founded. In fact, if the damping is due to spontaneous decay of the phonon, then it should begin immediately after the end of the thermal pulse, the duration of which is 0.1 μsec. At the same time, as follows from the data given in[3], under strong excitation conditions the detected pulse has almost a flat top with dura-

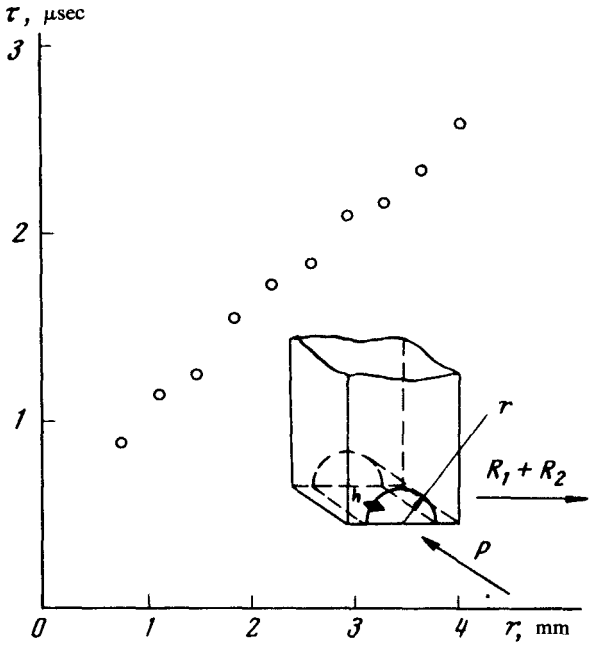

FIG. 1.

tion up to 1 μsec, and only then does exponential damping set in with a time constant 2.5 μsec. This pulse shape suggests that the decrease of the number of phonons is due to their departure from the excited volume, while the duration of the flat top is determined by the minimal time of phonon propagation from the heater to the boundary.

To verify this assumption, we have investigated in the present study the dependence of the kinetics of the detected phonon pulse on the dimensions of the illuminated volume at a constant concentration of the excited centers. The geometry of sample excitation is shown in the inset of Fig. 1 (h–heater, P–pump, R_1+R_2–luminescence). We used in the experiment a thermal-pulse technique similar to that described in[1-3]. The measurements were made at a temperature 2 K. To separate the R_2 line we used a DFS-12 monochromator, and to register the kinetics we used a multichannel AI-256-6 analyzer with "time-amplitude" conversion. The power density of the excited light incident on the sample corresponded to an excited-ion concentration 1.2×10^{16} cm^{-3} (i.e., close to the largest of those attained in[3]) and remained constant when the light-spot radius (r) was varied. It must be emphasized that to exclude effects connected with possible diffusion redistribution of the phonon density in the volume we registered in this experiment light from the entire excited volume of the ruby, and not from some segment of the ruby. Figure 1 shows a plot of the damping time against the dimension of the excited volume. The observed monotonic increase of τ in the entire range of r denotes that the damping is due to departure of the phonons from the volume, and not from their decay inside the volume. Starting from the volume dimensions, the measured values of τ, and the known speed of sound in ruby ($v=5\times10^5$ cm/sec), it can be shown that in the diffusion approximation the number of reabsorptions (n) experienced by the phonons prior to its departure from the volume does not exceed twenty.

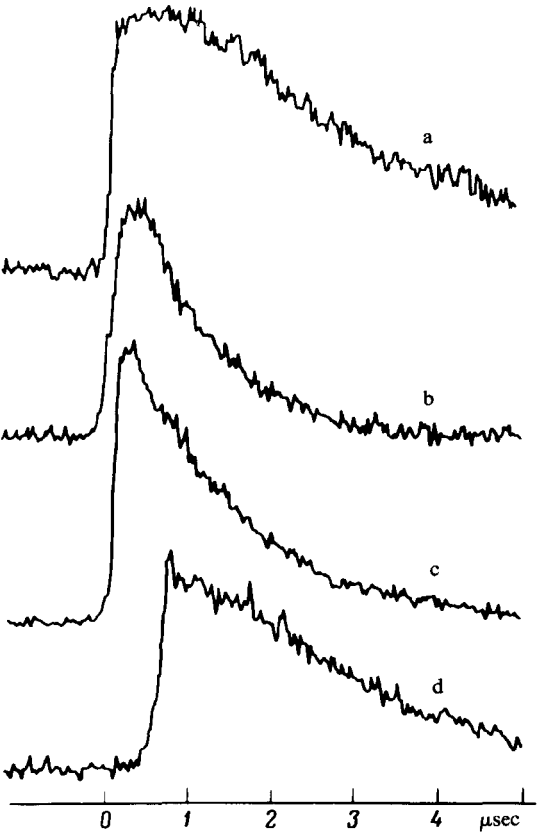

FIG. 2.

A detailed interpretation of the obtained dependence of τ on r is made complicated both by the inaccuracy of the diffusion model when the number of reabsorptions is small and by the fact that the kinetics of the damping of the phonon momentum is not elementary. As seen from Figs. 2(a) and 2(b), which show the results of measurements carried out under conditions of strong and weak excitations, the detected pulse has a rather complicated shape even in the case of ballistic propagation of the phonons (Fig. 2b). This pulse shape is apparently connected with the conditions of anisotropic propagation of the phonons in the crystal[4] through a semicylindrical excited volume. Allowance for the foregoing considerations, however, can only decrease the value of $\bar{n}$, which is in any case much lower than the value necessary to observe the "bottleneck" effect.

Direct proof of the absence of the bottleneck effect is provided by the results, shown in Figs. 2(c) and 2(d), of measurements of the kinetics of the emission of individual sections of the excited volume, namely, in the region adjacent to the heater (2c) and in the peripheral part of the volume (2d). It is seen from the figures that the pulses have equally steep leading fronts, and the front of the pulse registered in the peripheral part is shifted by an amount corresponding to the time of the ballistic flight of the phonons from the heater. Thus, even at high excitation densities, the ballistic

phonons reach the boundary of the detecting volume, i.e., their mean free path is comparable with the value of r. Naturally, under these conditions the departure of the phonon from the volume cannot be accompanied by a large number of reabsorptions.

The obtained data force us to change the opinions advanced in[3] concerning the mechanism of propagation of the resonant phonons and concerning their estimated lifetimes. It can be concluded that under the real conditions of the experiments on optical detection of phonons, no bottleneck effect is observed in ruby: the lifetime of the $2\bar{A}$ level does not make a substantial contribution to the results of the kinetic measurements. Since the measured damping times are determined only by the time that the phonons remain in the detecting volume, the lowest estimated lifetime of the 29-cm^{-1} phonons in ruby should be taken to be the largest of the measured damping times, i.e., $\tau_p > 2.7 \times 10^{-6}$ sec.

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