Inelastic collision of terahertz acoustic phonons in ruby crystals

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An inelastic interaction of a focused beam of 0.87-THz transverse ballistic phonons with higher frequency ($\overline{\omega} \geq 2$ THz) phonons has been observed in a ruby at $T = 1.8$ K by means of a technique for the optical detection of nonequilibrium acoustic phonons.

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Three-phonon anharmonic interactions of acoustic oscillations play an important role in many physical processes in crystals (heat conductivity, sound attenuation, etc.). Direct observation of these interactions has been accomplished for 10^6- to 10^9-Hz ultrasonic waves (see, for example, Ref. 1). For acoustic phonons of the terahertz (10^{12} Hz) band the experiment is limited by the fruitless attempt to ob-

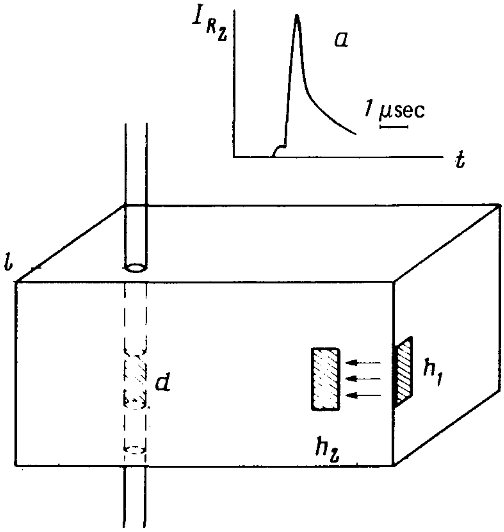

FIG. 1.

serve the interaction of thermal pulses in crystals.[2] In our work we have observed for the first time the inelastic interaction of terahertz acoustic phonons by optical detection of acoustic phonons[3] that propagate ballistically in a ruby under phonon focusing conditions.[4,5] We have observed processes in which 0.87-THz (29 cm^{-1}) transverse phonons adhere to acoustic phonons of much higher frequencies $\bar{\omega} \gtrsim 70$ cm^{-1}.

In our experiments (Fig. 1) a 14 X 7.5 X 6-mm rectangular $Al_2O_3:0.02\% \ Cr^{3+}$ single crystal was immersed in liquid helium at $T = 1.8$ K. Near the edge of one end we deposited a 3 X 1-mm thin-film thermal-pulse injector h_1, which was heated by current pulses (duration $\Delta t = 200$ nsec and repetition frequency $f = 50$ kHz). A steady-state excited volume "d" was established at the opposite end of the sample by using an Ar laser (0.3-mm beam diameter and 50-mW power). This volume served as a luminescent resonant detector of phonons with a 0.87-THz frequency (29 cm^{-1}).[3] The long edge of the sample parallel to the h_1-d line was oriented along the crystallographic direction in the σ_v symmetry plane at a 38° angle to the trigonal axis C_3. It corresponds to the direction of acute-angle focusing ($\approx 1°$) of fast, transverse (*FTA*) phonons.[4,5] Therefore, the 29-cm^{-1} *FTA* phonons, which are injected from h_1 and which propagate ballistically along the h_1-d line, are the focused phonons; they account for the main contribution to the narrow and intense pulse of R_2 luminescence $I(t)$ induced in "d"[4,5] –see Fig. 1a. There is a second, 3 X 1-mm, thin-film injector of thermal pulses h_2 ($\Delta t = 200$ nsec and $f = 50$ kHz) on the sample face adjacent to h_1-d. It was assumed that the inelastic interaction of phonons injected from h_2 with the focused 29-cm^{-1} *FTA* phonons will remove the latter from the ballistic beam, which is propagating along the h_1-d line, and reduce the intensity of the beam that reaches "d". The effect was investigated by analyzing the amplitude I of the R_2-luminescence pulse $I(t)$ using a differential method of digital synchronous detection with a storage, which made it possible to take into account the phonons from h_2 arriving at "d".

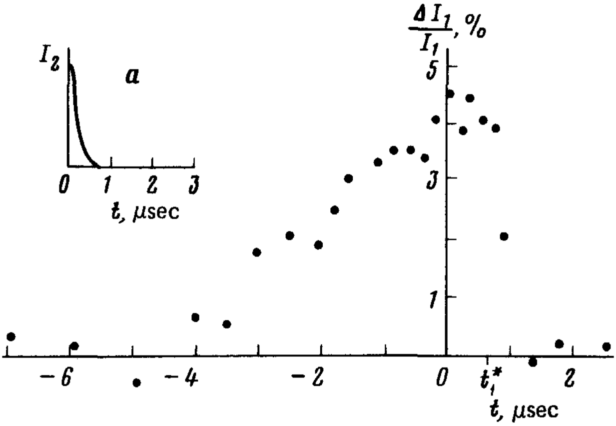

FIG. 2.

As a result, it was established that the thermal pulses from h_2 reduce by several percent the amplitude of the R_2-luminescence pulse, which was induced by the 29-cm^{-1} *FTA* phonons emitted from h_1. This is a direct indication of the scattering of the ballistic 29-cm^{-1} phonons by phonons injected from h_2. The relative attenuation $\Delta I/I$ of the amplitude of the R_2 pulse depends on the time delay t between the moments t_1 and t_2 of injection of the thermal pulses at h_1 and h_2, respectively ($t = t_2 - t_1$). Figure 2 shows the $\Delta I/I(t)$ dependence, where it is assumed that $t_1 = 0$, and it also shows the moment t_1^* when the 29-cm^{-1} ballistic phonons fly past the heater h_2 ($t_1^* = h_2 - h_1/v_{FTA}$, where the distance between the heaters is $h_2 - h_1 = 3$ mm). For delays $t > t_1^*$, i.e., when the thermal pulses are injected from h_2 after the passage of 29-cm^{-1} ballistic *FTA* phonons, the beam attenuation effect disappears. This shows that the phonon-interaction region is located at the h_2 level. The value $\Delta I/I$ decreases very slowly with $|t|$ for "negative" delays $t < t_1^* = 0$, which correspond to the injection of thermal pulses from h_2 long before the injection of the 29-cm^{-1} phonons from h_1. This shows that the "cloud" of phonons injected from h_2, which are very effective in scattering the 29-cm^{-1} ballistic phonons, attenuates very slowly (with $\tau \approx 2$ μsec). This phonon cloud is localized in the immediate vicinity of h_2, which is evident from the absence of the effect of thermal injection from h_2 on the ballistic beam of 29-cm^{-1} phonons after their trajectory h_1-d has been moved (by shifting the location of d) from h_2 a distance $l \approx 0.3$ mm.

From the attenuation kinetics we can obtain a lower estimate of the average frequency $\bar{\omega}$ of the phonons injected from h_2, which interact very efficiently with the 29-cm^{-1} *FTA* phonons. As shown in Ref. 6, because of the strong frequency dependence ($\sim \omega^4$) of the cross section for elastic scattering of phonons by impurities and defects, the phonons of different frequencies injected from the heater are segregated in the crystal. The low-frequency phonons quickly (ballistically) enter the bulk of the crystal, while the high-frequency phonons are delayed in the surface layer near the heater, which they slowly leave through diffusion (and through anharmonic decay). The R_2 luminescence was used for a direct measurement of the behavior of the concentration near the heater h_2 of the 29-cm^{-1} phonons injected from it (Fig. 2a). It corresponds to the ballistic removal of phonons from h_2 in a time much shorter than the attenuation time $\tau = 2$ μsec of the $\bar{\omega}$ phonons. Thus the slow kinetics of attenuation of the $\bar{\omega}$ phonons, like their localization near h_2, make it possible to allege that these phonons have a frequency much higher than $\omega_0 = 29$ cm^{-1} ($\bar{\omega} \gg 29$ cm^{-1}).

The interaction of phonons ($\omega_0 = 29$ cm^{-1}) with higher frequency phonons ($\bar{\omega} > \omega_0$) apparently is similar to the three-phonon processes $T+L \to L$ (Ref. 7) and the $T+T \to L$ process: the TA phonon ω_0 adheres to the LA (TA) phonon $\bar{\omega}$ with the formation of an LA phonon with a frequency $\bar{\omega} + \omega_0$. The cross section of these processes[1,2] is $\sim \omega_0 \bar{\omega} (\bar{\omega} + \omega_0)^3 \sim \bar{\omega}^4$; this increases the role of high-frequency phonons in the scattering of the 29-cm^{-1} FTA phonons. For a Planck spectrum of injected phonons the spectral dependence of the extinction coefficient of 29-cm^{-1} phonons, as they fly past h_2, has the form $K(\omega) \sim \omega^6 [\exp(h\omega/kT) - 1]^{-1}$. The maximum of $K(\omega)$ is located at $h\omega_{\max} \approx 6\,kT$; this gives $\omega_{\max} \approx 65$ cm^{-1} when h_2 is heated by $T \approx 15$ K. It can be seen that ω_{max} is located at the frequencies >29 cm^{-1}. The number of injected high-frequency phonons can be higher because of a non-Planckian shape of the spectrum of thermal pulses.[8]

Using a known time for the transit of the 29-cm^{-1} FTA phonons past the heater h_2 and the value (~5%) of the ballistic-beam attenuation, we estimate the fusion time $\tilde{\tau}$ of the FTA phonons with the high-frequency phonons, which are injected from h_2, to be $\tilde{\tau} \approx 3$ μsec. Consequently, the anharmonic lifetime of the 29-cm^{-1} phonons near the thermal injector is determined not only by their decay but also by their fusion; this is manifested, in particular, in the experiments with resonant capture of 29-cm^{-1} phonons.[3]

The observed attenuation time $\tau \approx 2$ μsec of a screening phonon cloud is anomalously long if it is compared directly with the times for diffusive or decompositional removal of high-frequency phonons of a constant frequency from the heater h_2. Taking into account the strong ($\sim \omega^4$) frequency dependence of the phonon-fusion cross section, we can assume that the spectral redistribution of the phonons injected from h_2 exerts over a time a strong influence on the behavior of $\Delta I/I(t)$ and the value of τ.

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