A. A. Kaplyanskii. *Optical Studies of High-Frequency (10^{12}Hz) Acoustic Phonons in Crystals.* The traditional methods of ultrasonic acoustics and Mandel'shtam-Brillouin scattering can be used to study acoustic vibrations in crystals up to frequencies of 10^{10}–10^{11} Hz, i.e., on a small initial segment of the Brillouin zone. Fundamental interest attaches to study of higher-frequency acoustic vibrations in the terahertz range (10^{12} Hz). Optical methods for detection and generation of terahertz acoustic vibrations (phonons) have been developed in recent years.

Figure 1(a) illustrates the principle of optical detection of terahertz phonons[1] as exemplified by a crystal with impurity centers the excited (radiative) state of which has a pair of nearby electronic sublevels 1 and 2; the interval Δ between them is in the tens of cm^{-1} (terahertz range). The crystal is at low (helium) temperature. Steady illumination creats a small, ~1 mm^3, volume "*d*" in the specimen with centers on the excited sublevel 1 ($\Delta \gg kT$); "*d*" radiates a fluorescence line $1 \to 0$. Thermal pulses[2] that propagate through the crystal are injected into the specimen by pulsed heating of metallic film "h" on the surface. When they reach volume "*d*," the phonons with $\hbar\omega = \Delta$ in the continuous phonon spectrum of the pulses induce $1 \to 2$ electronic transitions. A flash of radiation in the line $2 \to 0$ then appears in the luminescence spectrum of "*d*." The excited volume "*d*" therefore serves as a narrow-band resonant detector for nonequilibrium phonons with $\hbar\omega = \Delta$.

In ruby $Al_2O_3:Cr^{3+}$, on which most experiments have been performed, the levels 1 and 2 are components of a metastable state $\bar{E}$, $2\bar{A}$ with $\Delta = 29$ cm^{-1} (0.87 THz). In a tunable optical phonon detector, levels 1 and 2 are sublevels arising from the splitting of a degenerate electron state of a center in an external field, and Δ may vary. An example is a $CaF_2:Eu^{2+}$ "piezospectroscopic" detector, where doublet splitting of the Γ_8^+ radiative level by the excited $4f^6 5d$ configuration of Eu^{2+} on uniaxial elastic deformation of the crystal makes it possible to detect phonons with $\omega = 0$ to 2.5 THz.[3,4] These two-level systems are suitable not only for optical detection, but also for optical generation of terahertz-band monochromatic phonons [Fig. 1(b)]. On optical excitation to higher states, levels 1 and 2 are populated and $2 \to 1$ transitions create nonequilibrium phonons with $\hbar\omega = \Delta$ in the lattice.[5,6]

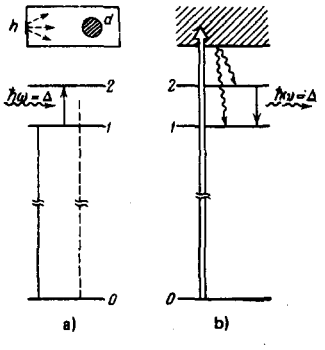

Рис. 1.

Several important properties of THz-band acoustic phonons have been studied on the basis of the above detection and generation schemes.

1. Resonant interaction of phonons with impurities. Optical detection of heat pulses propagating in crystals under ballistic conditions makes it possible to investigate the interaction of two-level impurities ($1 \rightleftarrows 2$ transitions) with individual acoustic-phonon modes. Strong mode dependence (anisotropy) has been observed for the absorption cross sections of ballistic LA and TA phonons with $\hbar\omega = 29$ cm^{-1} in $Al_2O_3:Cr^{3+}$[7] and with $\hbar\omega \approx 10$ cm^{-1} in uniaxially compressed $CaF_2:Eu^{2+}$.[4] This dependence was interpreted in terms of the symmetry selection rules of quadrupole-type for single-phonon transitions under dynamic deformation in an acoustic wave.[4,7]

2. Resonant dragging of phonons. Multiple resonant scattering of 29 cm^{-1} phonons by $\bar{E} \rightleftarrows 2\bar{A}$ transitions results in phonon dragging in the excited volume in $Al_2O_3:Cr^{3+}$.[1,8] It is manifested in lengthening of the R_2 fluorescence flash induced by heat pulses with an increase in the concentration N^* of the excited Cr^{3+} ions in the detecting volume "*d*" (Fig. 2). The specific mechanism of phonon dragging in Al_2O_3 involves the "radiative" nature of phonon-line broadening, which is determined for practical purposes by the $2\bar{A} \to \bar{E}$ decay with emission of a phonon. As a result, the $E \rightleftarrows 2A$ scattering event is purely elastic, and the phonons can escape from the volume only by spatial diffusion toward its boundaries.[9,10] An additional "anisotropic" channel for spatial escape of photons has also been detected[8,11] in association with scattering of phonons into weakly absorbed modes that exist as a result of the anisotropy of the resonant absorption crosssection of 19 cm^{-1} phonons.

3. Phonon lifetime. The idea behind optical experiments to determine the anharmonic lifetimes of terahertz phonons is to investigate the decay of luminescence from the upper sublevel 2 (see Fig. 1) under "phonon-bottleneck" conditions. Under these conditions, the relative population of levels 2 and 1 is determined by phonons with $\hbar\omega = \Delta$, and the kinetics of the $2 \to 0$ luminescence by the time τ of escape of phonons from the volume [the phonons are injected into the volume

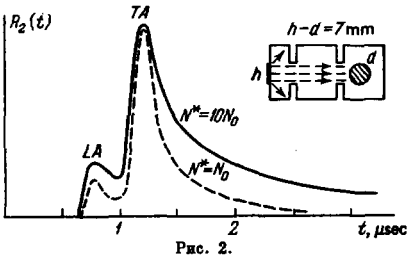

Рис. 2.

either by heat pulses or by optical generation; see Fig. 1(b)]. The time of spatial (diffusion) escape of phonons in resonant scattering (29 cm^{-1} phonons in $Al_2O_3:Cr^{3+}$ [1,8]) or in nonresonant Rayleigh scattering by structure defects (experiments with $CaF_2:Eu^{2+}$ [12]) can be made quite large. In this case, τ is determined by the intrinsic lifetime τ_p of the phonons, averaged over the modes. The measurements gave $\bar{\tau}_p \approx 2$ μsec for phonons with $\hbar\omega = 1$ THz in Al_2O_3 and $\bar{\tau}_p \approx 2 \cdot 10^{-7}$ sec for phonons with $\hbar\omega \approx 2$ THz in CaF_2.[12]

4. Experiments with ballistic phonons. Optical detection of thermal pulses in Al_2O_3 at various distances $h-d$ [see Fig. 1(a)] made it possible to measure the decay time of 21 cm^{-1} phonons.[13] In the propagation of LA phonons along the C_2 axis, the mean free path is $l = 21$ mm, and free path time is $\tau = 1.9$ μsec; this time gives the lower limit for the anharmonic lifetime τ_p for a single mode ($\tau_p \geq \tau$). Optical detection of ballistic 29 cm^{-1} phonons in angle scanning of the phonon flight direction $h-d$ led to direct observation of a phonon-focusing effect due to the elastic anisotropy of Al_2O_3.[14,15] Sharp concentration (focusing) of the ballistic-phonon flux into an extremely narrow (less than 70) angle range is observed.

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