

Propagation and relaxation of rf acoustic phonons in thin crystal plates

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A phonon spectrometer with high spatial and temporal resolution has been used to study the propagation and relaxation of rf acoustic phonons in thin anthracene crystals. The nature of the propagation of the nonequilibrium phonons changes at a time ~ 40 ns after the excitation pulse.

Optical methods are widely used to excite and detect short-wave acoustic phonons, with frequencies 10^{11} – 10^{12} Hz. During the photoexcitation of crystals, rf acoustic phonons are excited in a time $\lesssim 10^{-10}$ s as a result of a radiationless relaxation of electronic excitations. These phonons begin to propagate out of the excitation region into the cold regions of the crystal. The propagation of the nonequilibrium phonons may be accompanied by a spontaneous coalescence or splitting of phonons, depending on the occupation numbers in their initial distribution and on the experimental geometry. The coalescence and splitting in turn play an important role in establishing the phonon propagation conditions in the crystal.^{1,2} In the present experiments we have used methods with a frequency selectivity and a high time resolution to detect the rf phonons. For the first time, we have been able to follow the relaxation of these phonons and the establishment of the propagation conditions for nonequilibrium phonons in thin crystal plates starting immediately after these phonons are excited.

In the experiments we use thin anthracene crystals ($d = 2$ – $15 \mu\text{m}$) with a textured [001] surface. The crystals are held in an optical helium constant-temperature bath at $T_0 = 4.5$ K. Short-wave phonons are produced by exciting the crystal surface with pulses from a nitrogen laser (pulse length $\tau_p \approx 4.5$ ns, repetition frequency of 25 Hz, and pump intensity $P = 10$ – 50 kW/cm^2). The laser light is absorbed in a layer $\sim 0.5 \mu\text{m}$ thick.

The appearance of nonequilibrium phonons in the bulk of the crystal under study is identified from a change in the absorption coefficient, $\kappa(\omega) - \kappa_0(\omega)$, in the frequency region near the bottom of the lowest exciton b band ($\omega_T = 25\,096 \text{ cm}^{-1}$). The difference $\kappa(\omega) - \kappa_0(\omega)$ is measured at various times after the pump pulse from the nitrogen laser, with the help of a tunable dye laser ($\tau_p \approx 3$ ns, output bandwidth $\Delta\omega \lesssim 1 \text{ cm}^{-1}$, light polarization $\mathbf{E} \parallel \mathbf{b}$ axis). The time delay of the pulses of the tunable laser with respect to the pump pulses can be varied over the range $t_D = 0$ – 150 ns by means of an optical delay line.

In anthracene crystals the polariton group velocity V_g is lower than the sound velocity u for the lower polariton branch at frequencies $\omega \gtrsim \omega_T$ (Refs. 3 and 4), so that one-phonon transitions involving the absorption of high-frequency acoustic phonons from polariton states below the bottom of the exciton band, into the region $\omega \gtrsim \omega_T$, are

allowed. Since the polariton state density increases sharply in this frequency region, and since there is a comparatively narrow ($\sim 3 \text{ cm}^{-1}$) peak in the state density at $\omega \approx \omega_T$, according to experimental data⁵ and calculations, the anti-Stokes scattering of polaritons with frequency ω is dominated by one-phonon transitions involving short-wave acoustic phonons of frequency $\Omega_a \approx \omega_T - \omega = \Delta$. Consequently, the increase in the absorption coefficient, $\kappa(\omega) - \kappa_0(\omega)$, can be said quite accurately to be proportional to the change (Δn) in the occupation numbers of phonons of frequency $\Omega_a \approx \Delta$. Accordingly, by varying the output frequency of the tunable laser and the delay time, one can determine Δn , the change in the occupation numbers, for nonequilibrium phonons of different frequencies at various times after the pump pulse. The net result is the development of a phonon spectrometer with a frequency resolution $\sim 3 \text{ cm}^{-1}$ and time resolution $\sim 3 \text{ ns}$, determined by the length of the laser pulse.

The data in Fig. 1 were obtained at the time of the pump pulse and 20 ns later. The excitation region on the crystal surface is a long, narrow strip with a transverse dimension $a \lesssim 20 \mu\text{m}$, oriented along the b axis. The light from the tunable laser, which is transmitted through the crystal plate, is detected by an optical system from a fixed region in the form of a narrow strip with a transverse dimension $< 10 \mu\text{m}$, oriented parallel to the excitation region. The spatial distribution of $\kappa(\omega) - \kappa_0(\omega) \sim \Delta n$ is measured by scanning the excitation spot over the crystal surface along the a axis (i.e., by varying l , the distance, between the points at which the phonons are excited and detected) at various times t_D . It can be seen from Fig. 1 that there are nonequilibrium phonons of essentially all frequencies at the initial time in the excitation region (in anthracene crystals, $\Omega_0 \approx 30 \text{ cm}^{-1}$ is the limiting frequency of the acoustic phonons). After the pump pulse, we observe a decrease in the occupation numbers of phonons with frequency $\Omega_a = 20\text{--}25 \text{ cm}^{-1}$. Their degradation is accompanied by an increase in the occupation numbers of phonons of lower energy and by a propagation of the latter into the cold regions of the crystal. According to the measurements carried out at time delays $t_D = 0\text{--}40 \text{ ns}$, phonons of frequency $\Omega_a = 20\text{--}25$

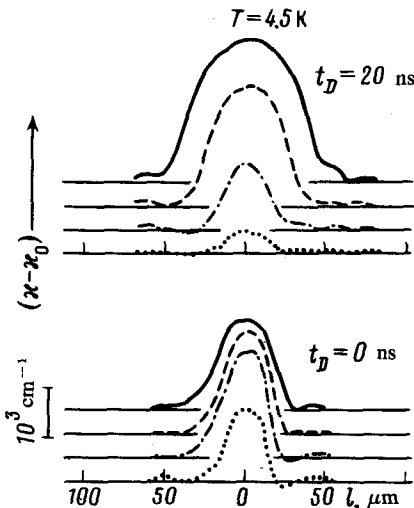


FIG. 1. Spatial change in the absorption coefficient during the pump pulse and 20 ns later at the following frequencies $\Delta = \omega_T - \omega$: Solid line— 10 cm^{-1} ; dashed line— 15 cm^{-1} ; dot-dashed line— 20 cm^{-1} ; dotted line— 25 cm^{-1} ($T_0 = 4.5 \text{ K}$, $d = 10 \mu\text{m}$, $P = 30 \text{ kW/cm}^2$).

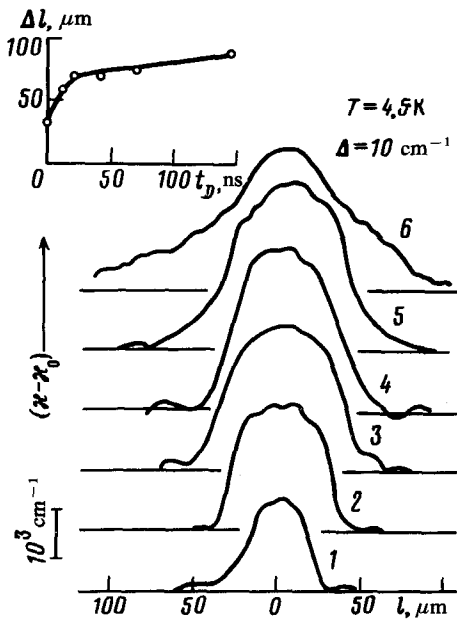


FIG. 2. Spatial change in the absorption coefficient ($\Delta = 10 \text{ cm}^{-1}$) for various delay times t_D : 1—0; 2—10; 3—20; 4—40; 5—68; 6—144 ns ($d = 10 \text{ }\mu\text{m}$, $P = 30 \text{ kW/cm}^2$).

cm^{-1} are essentially trapped in the excitation region. This trapping is apparently due to two factors: the short lifetime with respect to decay and scattering and the low group velocity of phonons of frequency $\Omega_a \approx \Omega_0$.

Figure 2 shows data on the spatial distribution of nonequilibrium phonons of frequency $\Omega_a \approx 10 \text{ cm}^{-1}$ at various times after the pump pulse. It follows from these results that the "spreading" of the phonon packet initially ($t_D = 0\text{--}20 \text{ ns}$) occurs at a velocity $\approx 10^5 \text{ cm/s}$, close to the sound velocity in anthracene crystals [$u = (1\text{--}4) \times 10^5 \text{ cm/s}$, depending on the polarization and propagation direction^{6,7}]; i.e., the phonons propagate in a quasiballistic manner over a distance $L \approx 20 \text{ }\mu\text{m}$. At later times, the propagation of the phonons slows down markedly (see the inset in Fig. 2; Δl is the width at half-maximum of the spatial distribution of phonons of frequency $\Omega_a \approx 10 \text{ cm}^{-1}$), demonstrating a decrease in the mean free path of the phonons at $t_D > 20 \text{ ns}$, at least to $l_u < L \approx 20 \text{ }\mu\text{m}$. An estimate of the mean free path of the phonons with respect to phonon-phonon scattering with flip, involving thermal (equilibrium) phonons, at a crystal temperature $T_0 = 4.5 \text{ K}$, $l_u^0 \approx l_0 \exp\{\Omega_0/T\}$, where $l_0 \approx 0.01 \text{ }\mu\text{m}$, yields $l_u^0 \approx 140 \text{ }\mu\text{m}$. A significant decrease in the mean free path, $l_u \ll l_u^0$, could be caused only by an increase in the occupation numbers of phonons with energies near the Debye energy ($\Omega_a \approx \Omega_0$) in the volume of the crystal occupied by nonequilibrium phonons. As follows from the data in Fig. 1, phonons of frequency $\Omega_a \approx \Omega_0$, which arise in the crystal during the pump pulse, are localized in the excitation region. Consequently, an increase in the occupation numbers of phonons with energies near the Debye energy in parts of the crystal far from the excitation region can be achieved only as a result of the coalescence of lower-frequency nonequilibrium phonons. An increase in the occupation numbers on the high-frequency tail of the phonon distribution, with the result that there is a change in the propagation regime

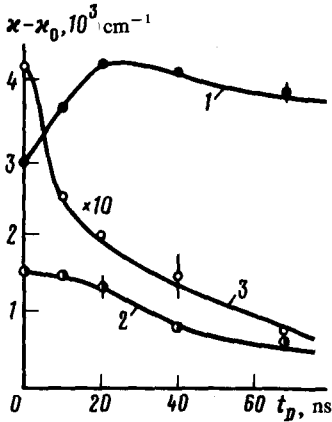


FIG. 3. Time evolution of the absorption coefficient at several frequencies Δ : 1—10; 2—15; 3—20 cm^{-1} (uniform excitation of the crystal surface at $P = 8 \text{ kW/cm}^2$, with $d = 12 \text{ }\mu\text{m}$).

for the nonequilibrium phonons, would naturally be linked with the establishment of a temperature $T > T_0$ in the crystal. Estimates show¹⁾ that under these experimental conditions the temperature can reach $T \approx 9 \text{ K}$; at this temperature we would have $l_u \approx 1 \text{ }\mu\text{m}$. The observed transport of energy from the excitation region into the cold regions of the crystal is reminiscent of the mechanism of nonlocal thermal conductivity proposed in Ref. 8.

In the case of uniform excitation of the entire crystal surface, the conditions for a plane experimental geometry hold, so that the propagation of phonons along the surface of the sample can be ignored. It follows from the data in Fig. 3 that just after the excitation (at $t_D < 20 \text{ ns}$) the changes in the occupation numbers of the phonons of different frequencies are quite different. For example, the occupation numbers of high-frequency phonons with $\Omega_a \approx 20 \text{ cm}^{-1}$ decrease sharply, because of their fast spontaneous decay. For lower-frequency phonons with $\Omega_a \approx \Omega_0/3 \approx 10 \text{ cm}^{-1}$, the occupation numbers increase because the decay of the phonons of a given frequency is slower than the rate at which they are generated by the decay of high-energy phonons. For phonons of intermediate frequency, $\Omega_a \approx 15 \text{ cm}^{-1}$, the occupation numbers stabilize, demonstrating that the excitation and spontaneous decay of phonons of a given frequency do not cancel out at $t_D \leq 20 \text{ ns}$. Later (at $t_D \geq 40 \text{ ns}$), the occupation numbers of phonons with $\Omega_a = 10\text{--}20 \text{ cm}^{-1}$ fall off smoothly, and they tend toward their equilibrium values as the temperature $T > T_0$ is established. The complex nature of the kinetics may also be due to the existence of different spontaneous-decay times for phonons of the quasilongitudinal and quasitransverse branches.⁹

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¹⁾The crystal temperature can be estimated from the relation

$$\frac{\pi^2(T^4 - T_0^4)}{10(\hbar\bar{u})^3} \approx P\tau_p \frac{a}{d\Delta l},$$

where $\bar{u} \approx 2 \times 10^5 \text{ cm/s}$ is the average sound velocity in the anthracene crystal, under the assumption that a region with a transverse dimension $\approx \Delta l$ is heated uniformly.

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