Relaxation of rf acoustic phonons

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The energy relaxation of rf acoustic phonons in thin anthracene crystals has been studied experimentally. A comparison with the results of model-based calculations has made it possible to estimate the parameters of the distribution of acoustic phonons generated during the relaxation of the electronic excitation and also the lifetime of the acoustic phonons relative to the anharmonic processes.

One of the more interesting and important problems in phonon physics is the relaxation of a nonequilibrium-phonon system in the course of the establishment of equilibrium temperature of the crystal. Early stages of such relaxation, accompanied by a slight excitation of the phonon system, where the spontaneous splitting of phonons is dominant, were observed experimentally by Happek *et al.*[1] and Baumgartner *et al.*[2] To describe the relaxation processes at high excitation levels, when the change in the temperature of the crystal may reach a level comparable to the initial temperature, $\Delta T = T_c - T_0 \cong T_0$, one must take into account the coalescence of rf acoustic phonons, in addition to their splitting.[3] We have measured the time evolution of the occupation numbers of rf nonequilibrium phonons in a crystal after the application of an intense laser pulse and we have made a quantitative comparison with the results of model-based calculations carried out in the "quasicontinuous"-phonon-spectrum approximation. We have used an optical method, with a frequency selectivity and high temporal resolution, proposed by Maksimov and Tartakovskiĭ,[4] to detect nonequilibrium acoustic phonons. This approach allowed us to track the change in the occupation numbers of phonons of various frequencies, beginning with the time immediately after the termination of the laser pulse. To eliminate the effect of phonon propagation on the phonon energy relaxation, we applied, as in Ref. 1, the method of uniform excitation of the crystal surface by a wide laser beam.

The experiment can be summarized as follows. The front surface (the developed ab plane) of anthracene single crystals of thickness $d = 2$–5 μm were placed in an optical helium constant-temperature bath held at $T_0 = 4.3$ K. The crystal surface was uniformly excited with pulses from a nitrogen TEA laser ($\omega = 28\,670$ cm^{-1}, pulse length $\tau_p \approx 0.5$ ns, repetition frequency of 100 Hz, maximum pump intensity of 100 kW/cm^2 with a sample). This laser was also used to excite the tunable dye laser with a lasing frequency ω near the bottom of the lowest exciton **b** band, $\omega_T = 25\,096$ cm^{-1} (half-width of the lasing band $\Delta\omega \approx 1$ cm^{-1}, pulse length $\tau_L \approx 0.3$ ns, polarization of light $\mathbf{E} \| \mathbf{b}$ axis). As was shown in Refs. 4 and 5, at the frequency $\omega \leqslant \omega_T$ the change in the absorption coefficient $\Delta k = k(\omega) - k_0(\omega)$ of anthracene crystals is porportional to the change $\Delta n(\Omega)$ in the occupation numbers of phonons with a frequency $\Omega = \omega_T - \omega$. We can thus obtain information on the relaxation of phonons of various energies by measuring the intensity of the tunable laser light which passes through the

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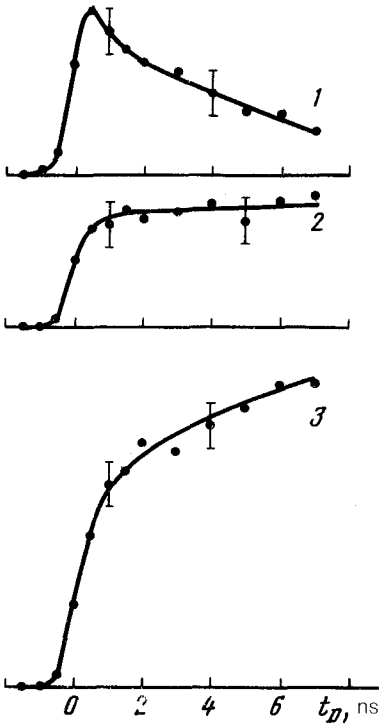

FIG. 1. Time evolution of the absorption coefficient $\Delta k(\omega)$ at several frequencies $\Omega = \omega_l - \omega$: 1—32; 2—25; 3—14 cm^{-1}. Excitation with the laser pulse occurs at the time $t_D = 0$.

crystal plate at different times t_D (the pulses from the tunable laser are delayed relative to the pulses from the N_2 laser with the help of an optical-delay line, $t_D = 2 - 10$ ns) at different lasing frequencies ω.

The results of measurements at several frequencies Ω are shown in Fig. 1. We see that the kinetics of variation of the phonon occupation numbers depends essentially on the frequency of the phonons. For phonons with a frequency $\Omega \approx 32$ cm^{-1} (the limiting frequency of the acoustic phonons in anthracene crystals is $\Omega_0 \approx 45$ cm^{-1}), for example, the occupation numbers decrease, for phonons of frequency $\Omega \approx 25$ cm^{-1} the occupation numbers remain virtually constant over a time $t_D \leqslant 10$ ns, and for phonons with $\Omega \approx 14$ cm^{-1} the occupation numbers, in contrast, increase. We also see that at zero time the energy distribution of acoustic phonons, which are produced in the crystal as a result of fast, nonradiative processes in the course of the relaxation of the electronic excitations, differs markedly from the equilibrium distribution.

The results presented here allow us to conclude that pulsed laser pumping leads to a generation of acoustic phonons over a broad frequency range. We also conclude that at zero time the principal excitation energy is concentrated in the rf region of the phonon spectrum. The occupation numbers of the rf phonons, $\Omega > 25$ cm^{-1}, decrease in the course of the relaxation, while the occupation numbers of phonons of the lower frequencies, $\Omega < 25$ cm^{-1}, increase. The total energy of the phonon distribution is

conserved, since adiabatic conditions are rigorously maintained in the experiment up to a time $t_D \approx 1$ μs.

To describe theoretically the relaxation of a system of nonequilibrium acoustic phonons under conditions corresponding to those of the experiment, we have numerically simulated the kinetics of the change in the occupation numbers of phonons of various frequencies. The kinetic equation for the occupation numbers of phonons of various frequencies, which was written in accordance with Ref. 6 in the model of one longitudinal and two transverse, degenerate, dispersion-free, isotropic branches, was solved numerically on a uniformly spaced grid: the frequency interval $0 - \Omega_0$ was partitioned into N levels, where $N = 32$. The matrix elements of the anharmonic three-phonon processes were assumed to be proportional to the product of the moduli of the phonon wave vectors:

$$|f(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)|^2 \propto |\mathbf{k}_1| \cdot |\mathbf{k}_2| \cdot |\mathbf{k}_3|. \tag{1}$$

We took into account all three-phonon processes of the type $LA \rightarrow TA + TA$ and $LA \rightarrow LA + TA$, which satisfy the energy and quasimomentum conservation laws. The model was checked for the correspondence of the lifetime of LA phonons relative to

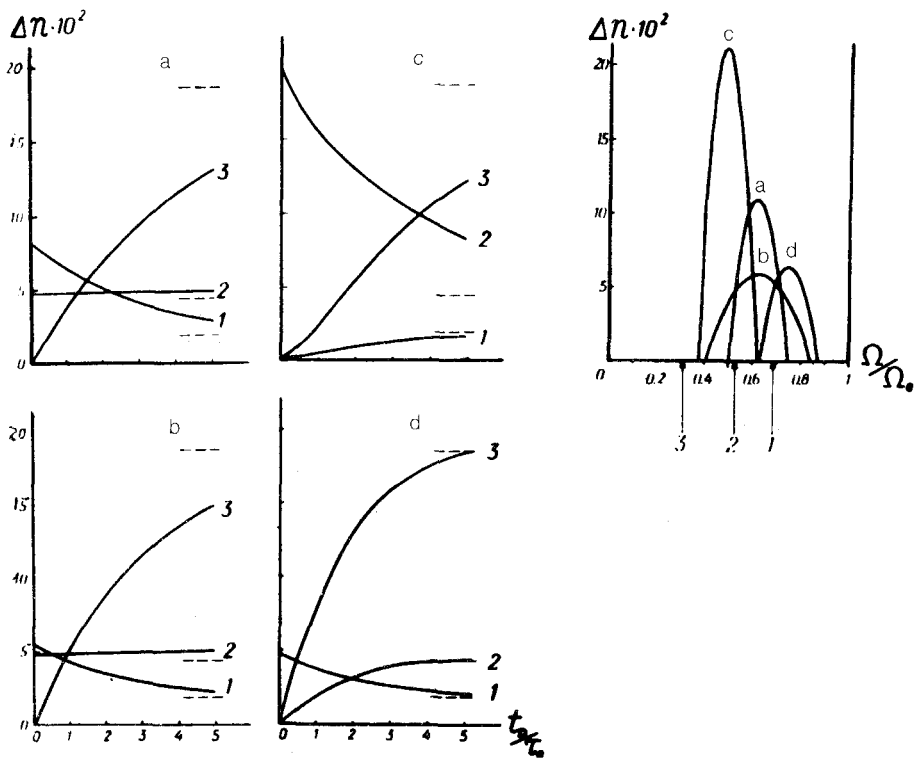

FIG. 2. Calculated time evolution of the occupation numbers of phonons of various frequencies Ω: 1—32; 2—25; 3—14 cm^{-1} at various initial distributions a–d, illustrated in the inset at $t_D = 0$. The dashed lines denote the finite values of $\Delta n(\Omega)$ upon establishment of the finite temperature $T_c = 11.5$ K.

the decay processes, $\tau_{decay}(\Omega) \propto (\Omega/\Omega_0)^{-5}$, at low perturbations. One of the variable parameters of the calculation was the coefficient in relation (1). It is more useful to represent this parameter as a change in the lifetime of the Debye phonons, $\tau_0 = \tau_{decay}(\Omega_0)$. Comparing the calculations for various values of τ_0 with experiment, we found, primarily on the basis of the behavior of phonons with $\Omega \approx 32$ cm^{-1}, that the best agreement is obtained at $\tau_0 \approx 1$ ns.

As the second adjustable parameter we chose the spectrum of acoustic phonons which were produced from optical phonons, i.e., the initial energy distribution of the acoustic phonons. We changed the position of the maximum of this spectrum, Ω_{max}, and its half-width, $\Delta\Omega$. The total energy of the phonon system, which was assumed to be constant, corresponded to the finite temperature $T_c \approx 11.5$ K, consistent with the experimental conditions. We see from several calculated curves in Fig. 2 that the nature of the relaxation depends essentially on the parameters Ω_{max} and $\Delta\Omega$ of the initial distribution. The best agreement with the experiment occurs at $\Omega_{max} = 0.6\ \Omega_0$ and $\Delta\Omega \lesssim \Omega_0/4$. The position $\Omega_{max} \approx 28$ cm^{-1} agrees with the frequency $\Omega_{opt}^{min} = 49$ cm^{-1} of the lowest optical phonon in the anthracene crystal and with the results of Ref. 7, where it is shown that primarily acoustic phonons with half the frequency are produced as a result of optical phonon decay.

In contrast with the "generation" model[8] with the set of levels Ω_0, $\Omega_0/2$, $\Omega_0/4$, etc., a system of phonon states uniformly spaced in frequency is more compatible with the continuous phonon spectrum. This system makes it possible to study the behavior of phonons with intermediate frequencies $\Omega_0/2 < \Omega < \Omega_0$ and therefore to quantitatively compare the calculations with experiment, whereas the generation model, because of its crude frequency separation, puts the experimental kinetic dependences essentially in the range of a single phonon generation. The low-frequency phonon relaxation shows the strongest disagreement with the results of the generation model, because in the model with a quasicontinuous phonon spectrum these phonons are produced in the first decay, while in the generation model they are produced only after several successive decays.

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