The spectrum of a phonon hot spot in fluorite crystals

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The frequency dependences of the occupation numbers of nonequilibrium phonons, produced by pulsed means in a fluorite crystal cooled to 2 K, with the help of a thin-film metal heater, have been studied experimentally. It was found that on using heat pulses with energies greater than 10 μJ, an equilibrium heated local region—a hot spot—is formed in the subsurface layer of the crystal. Numerical estimates and the results of additional experiments showed that the establishment of local equilibrium is facilitated by the return of phonons by diffusive motion to the metal film (the heater), which plays the part of an effective transformer of phonon frequencies.

In experiments on the propagation of heat pulses in cooled crystals, ballistic (i.e., without scattering) motion is generally observed at relatively small (of the order of μJ/mm^2) thermal energy densities.[1] An increase in energy is usually accompanied by an increase in the duration of the signal recorded, which indicates the appearance of diffusive motion of the nonequilibrium phonons forming the heat pulse. It is assumed that the reason for the increase in the duration is the concentration in a small volume of strongly scattered high-frequency phonons, the decay of which leads to the formation in the crystal of a superheated region (a so-called hot spot[2]), which acts as a secondary source of phonon emission. According to theoretical calculations,[3] the establishment of local thermal equilibrium is possible if certain initial conditions in the hot spot are fulfilled, and is characterized by a temperature appreciably higher than that of the crystal as a whole, and the equilibrium is preserved even for diffusive spreading of the heated region. In the present work we report the results of an experimental investigation of the dynamics of a hot spot and the spatial and temporal transformation of the spectrum of nonequilibrium phonons on generating heat pulses by a metal film heater.

The material studied was a fluorite crystal activated by Eu^{2+} ions. The method of fluorescence detection with piezospectroscopic frequency retuning[4] was used for phonon recording. The principle of the method consists in recording the luminescence from the raised sublevel of the excited state of the impurity ion, which arises on the appearance of nonequilibrium phonons in the cooled crystal. In the present case the orbital doublet Γ_8^+ of the $4f5d$ configuration of the divalent europium ion, split by the action of uniaxial compression, served as the luminescence state. The magnitude of the splitting Δ, which provides the frequency ν of the resonant-detected phonons could be changed in a controlled way by varying the strength of the compression ($\Delta = \nu/c$, where c is the velocity of light). The method of fluorescence detection has the appreciable advantage that it enables the absolute values of the occupation numbers $n(\nu)$ of resonance phonons at a given point of the crystal to be determined from the magnitude of the ratio of the populations N_2/N_1 of the upper and lower sublevels of the doublet. This advantage is most clearly seen when recording the spatially isotropic diffusive phonon flow, when the ratio N_2/N_1 is exactly equal to $n(\nu)/[n(\nu)+1]$, while for small occupation numbers it is simply $n(\nu)$. The ratio of the numbers N_2/N_1 is determined experimentally by measuring the intensity of the luminescence lines from the upper and lower sublevels of the excited state. Normalization, the necessity of which is determined by the difference in the polarization of the lines of the doublet, was carried out by measuring the temperature behavior of the intensities of the corresponding lines in the luminescence spectrum on stationary heating of the crystal, and extrapolation of the results obtained to infinite values of the temperature, at which the populations of the sublevels equalize.

The experiments were carried out at a temperature of 2 K on a crystal immersed in liquid helium. The specimen was a rectangular parallelepiped with dimensions 3×4×5 mm, cut out of a fluorite crystal containing about 0.001% of divalent europium ions. Phonons were generated by using a thin (thickness ≈ 100 nm) constantan film, deposited on the side face of the specimen, heated by electric pulses. The heat pulses generated were of 0.1 μsec duration and the maximum energy in a pulse was 20 μJ. The method for recording the signal was the same as described before.[5] The dependence of the amplitude and shape of the recorded signals on the experimental conditions was studied: the energy of the electrical pulse supplied to the heater and the heater-detector distance. The magnitude of the piezo-splitting Δ was varied within the limits 4–30 cm^{-1}.

At moderate energies in the heat pulse, the signal clearly contained two compartments: a rapid one with duration of the order of 0.1 μs, produced by the arrival of ballistic phonons at the detector, and a slow one, the existence of which is associated with phonons travelling diffusively. The frequency dependences of the occupation numbers of nonequilibrium phonons, or more exactly, of the relative populations of the detector levels, recorded at the moment of the ballistic signal, are shown in Fig. 1, curve 1, and after 1.5 μs (when only the slow component remains in the signal) in curve 2. As can be seen in the figure, the transition from the ballistic regime for motion to diffusion is accompanied by transformations of the phonon spectum—the occupation numbers at small frequencies increase and those at high frequencies decrease. This result is a direct experimental confirmation of the existence of processes for decay of high-frequency phonons into phonons of lower frequency.

On analyzing the experimental results, attention is drawn to the fact that on the chosen semilogarithmic plot the points on the graphs lie well on straight lines, i.e., the

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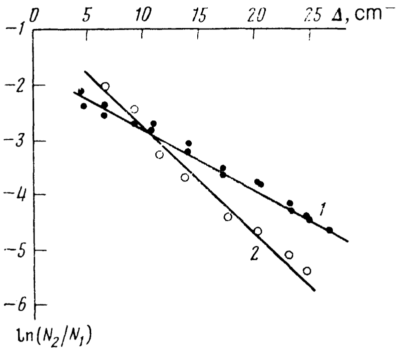

FIG. 1. The transformation of the phonon spectrum at the transition from the ballistic (1) to the diffusive (2) propagation regime. The distance to the heater was 0.5 mm. Values of N_2/N_1 measured at the maximum of the pulse (1) and after 1.5 μsec (2).

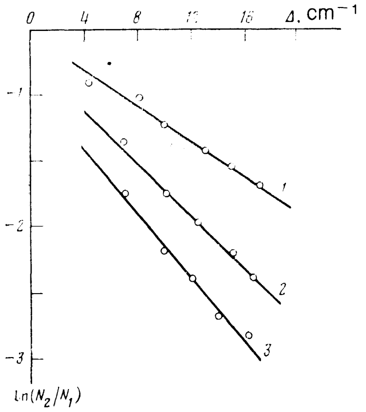

FIG. 3. Spectra of nonequilibrium phonons at different distances from the heater: 1) 1 mm; 2) 2.2 mm; 3) 4 mm. The energy of the heat pulse was 25 μJ.

phonon spectrum is described by an exponential function [the Planck function for $n(\nu)\ll 1$]. The slope of the lines (the color temperature of the phonon distribution) and the value of the intercept on the ordinate axis (the extent of the nonequilibrium) depend appreciably on the experimental conditions—an increase in the energy of the heat pulse leads to an increase in the color temperature and simultaneously to a reduction in the extent of the nonequilibrium (Fig. 2). A similar effect is also observed as a result of reducing the distance between the heater and the phonon detector (Fig. 3). Such a behavior of the phonon spectrum does not correspond to the existing models of quasidiffusive (diffusion with decay) phonon motion,[6-8] and further theoretical analysis is required for its explanation.

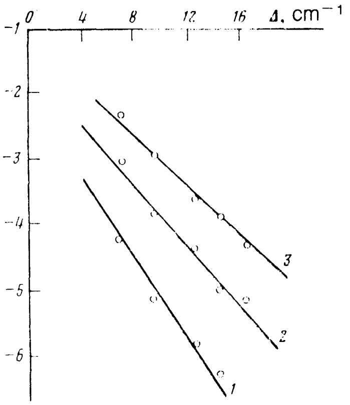

FIG. 2. Spectra of the diffusive component for values of the energy of the heat pulse: 1) 20 μJ; 2) 6 μJ; 3) 2.5 μJ. The distance to the heater was 1.5 mm.

A clear feature was observed in the nature of the deformation of the phonon spectrum: while at large distances from the heater ($r \gtrsim 1$ mm) the changes noted above were entirely monotonic, a sharp growth in the degree of equilibrium was recorded in the subsurface layer ($r \lesssim 0.4$ mm) on using heat pulses with energy 10 μJ and higher. It can be seen from Fig. 4 that under these conditions the frequency dependence of the occupation numbers (curve 2) corresponded to a situation of equilibrium heating (i.e., for $\nu \to 0$ the ratio $n(\nu)/[n(\nu)+1] \to 1$). The formation in the crystal of a local uniformly heated region corresponds in principle to the existing ideas[3] about the formation of a hot spot. At the same time, numerical estimates of the magnitudes of the thermal energy and of the depth of heating necessary to establish local equilibrium in fluorite, give values noticeably different from those which were realized in the experiment. According to Kazakovtsev and Levinson,[3] for the establishment of thermal equilibrium in a local region of a crystal containing excess phonons it is sufficient that the diffusion displacement (l_d) of phonons of the characteristic frequency ν_T over a time equal to the lifetime with respect to anharmonic decay, should not exceed the minimum dimension r of the heated region, i.e.,

$$r > l_d = [D(\nu_T)\tau_a(\nu_T)]^{1/2}, \qquad (1)$$

where $D(\nu_T) = a\nu_T^{-4}$ is the diffusion coefficient,[5] due to elastic scattering by point defects (it is just this process which is most probable at low temperatures), and $\tau_a(\nu_T) = b\nu_T^{-5}$ (Ref. 9) is the inverse probability of anharmonic decay. The value $\nu_T = 3kT/h$ corresponds to the maximum in the phonon energy distribution at the steady local temperature T. In the low temperature region when the vibrational properties of crystals are well described by the Debye model, the value of T can be determined from the relation[10]

$$Q/V = {}^1/_{10}\pi^2(kT)^4(\hbar\bar{v})^{-3}. \qquad (2)$$

Here Q is the energy, V the volume of the heated region, and $\bar{v}$ the value of the velocity of sound averaged over the modes.

The theoretical model developed[3] was based on the assumption of the existence at the initial moment of time of a sharply limited uniform heating of a region of the crystal, while in the experiments described here the penetration depth of phonons changed continuously, starting from zero values, and the dimensions of the heated region could not, therefore, be strictly determined. It is, however, not difficult to show that in the situation when the phonon penetration depth in the crystal does not exceed the dimensions of the heater, the indeterminacy in the value of r does not, practically, prevent the applicability of the criteria given above. For a plane propagation geometry the volume of the heated region is determined by the product $S \times r$ (S is the area of the heater), which by using Eq. (1) and (2) enables the criteria for the establishment of equilibrium to be formulated

$$l_d/r = D_0(Q/S)^{-9/8} r^{1/8} < 1, \qquad (3)$$

where

$$D_0 = (ab)^{1/8}(3/h)^{9/2}[10\pi^{-2}(\hbar\bar{v})^3]^{-9/8}.$$

It follows directly from here that in such a geometry the possibility of the establishment of equilibrium depends very weakly (as the eighth root) on the depth of phonon penetration into the crystal. Given a reasonable value for r (for example, conforming to the limiting distance from the heater at which the establishment of equilibrium could still be observed), this enables the amount of energy Q required for equilibrium heating to be estimated and to compare this value with the values realized in the experiment. Calculations carried out by using Eq. (3) and values known from experiments: $\bar{v} = 4.1 \times 10^5$ cm/s, $a = 1.4 \times 10^{46}$ cm/s (Ref. 5) and $b = 6.9 \times 10^{60}$ cm/s (Ref. 9), showed that for the dimensions $S = 1$ mm^2 (the true area of the heater) and $r = 0.4$ mm, the required amount of energy $Q = 60\ \mu$J is about 10 times greater than the experimental value. The disagreement between the predictions of the model and the experimental values is not only of a quantitative, but also of a qualitative nature. As follows from Eq. (3), even an insignificant excess of energy density above the threshold value should lead to the establishment of equilibrium at distances appreciably exceeding the threshold, while in the experiment equilibrium is observed only in the subsurface region of the crystal.

The nature of the disagreement between the model calculations and the experimental results can be explained by estimating the temperature established in the experiment and the lifetime of phonons of the characteristic frequency ν_T. According to Eq. (2), a heat pulse of energy 13.5 μJ can heat a local region of fluorite with volume 0.4 mm^3 up to a temperature of 17 K, i.e., close to the value determined from the slope of line 1 in Fig. 4. Making use of the experimental results given by Baumgartner *et al.*[9] on the probabilities of anharmonic processes in fluorite crystals, it can be concluded that the lifetime of phonons with frequency $\nu_T = 1.1$ THz, corresponding to such a temperature, is $\tau_a(\nu_T) = 1 \times 10^{-5}$ sec, which is noticeably greater than the duration of the pulse recorded. Under the experimental conditions, therefore, anharmonic phonon-phonon interactions could not lead to the establishment of equilibrium and to explain the results obtained it is necesary to draw on an additional mechanism for the transformation of phonon frequencies.

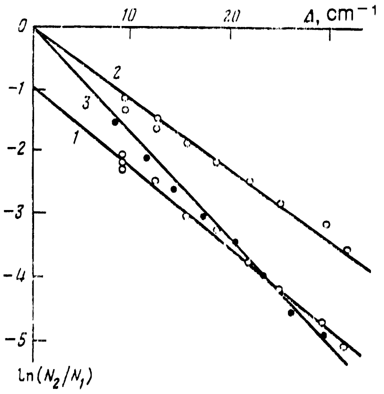

FIG. 4. The establishment of thermal equilibrium in the subsurface layer of the crystal ($r = 0.4$ mm). The energies of the pulses were: 1) 3–7 μJ; 2) 13.5 μJ. 3) The heater covered with paraffin. Temperatures determined from the slopes of the straight lines: 1) 11 K; 2) 13.5 K; 3) 8.3 K.

From the fact that equilibrium is only established in the subsurface region of the crystal, it can be assumed that this other mechanism is the return of phonons moving diffusively to the surface and the transformation of their frequencies due to interaction either with surface defects or with conduction electrons of the metallic film heater. The appreciable role of the return of phonons is confirmed by the results of additional experiments in which the heater was covered outside by a relatively thin (of the order of 0.1 mm) layer of solidified paraffin. The paraffin film could not influence the phonon properties of the crystal, but changes the boundary conditions for propagation of phonons in the heater. The results of the experiments are shown in Fig. 4. As can be seen from the figure, the deposition of the paraffin film (curve 3) leads to the establishment of equilibrium at appreciably smaller values of the energy introduced into the crystal than in experiments with a free heater. The results obtained indicate convincingly the appreciable part played by the return of phonons from the crystal in the process of establishing equilibrium. The paraffin film, having a disordered structure is then an additional transformer of phonon frequencies. (The possibility of increasing the effective anharmonicity in disordered media was shown by Levinson[11] and was found experimentally.[12])

The process for a hot spot to arise on the generation of heat pulses at the surface of a crystal is thus characterized by different rules from the case of bulk generation of nonequilibrium phonons. Interaction with surface defects and the metal film promotes the formation of an additional channel for the transformation of phonon frequencies, thanks to which local equilibrium can be established at appreciably lower values of the thermal energy, but only in the subsurface region of the crystal. The temporal evolution of a hot spot is accompanied by a lowering of its temperature without a change in the characteristic dimensions. The phonon spectrum outside the limits of the hot spot, which remains nonequilibrium under all conditions, is described by an exponential Planck function with a temperature which changes in the process of propagation. The mechanism for the formation of this spectrum requires further study.

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