Transition between different nonequilibrium phonon propagation regimes in solid solutions of garnets

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It is shown that nonequilibrium phonons injected by a heat pulse into a garnet lattice with a rare earth metal impurity propagate by diffusion, by quasidiffusion or in the nonlocalized phonon heat conductivity regime, depending on the power dissipated in the heater. It is shown that the relationships observed agree reasonably well with theoretical ideas.

1. STATEMENT OF THE PROBLEM; EXPERIMENTAL METHOD

Nonequilibrium phonons injected into a specimen by means of, for example, a metal heater can propagate in it ballistically or by diffusion. The nature of the diffusive motion is, in general, made rather complicated by intense phonon scattering and, at high energies, by the participation of anharmonic decay processes. A number of theoretical and experimental papers devoted to these questions have appeared recently.[1–5]

Crystals of yttrium aluminum garnet (YAG) are a convenient material for studying the diffusive motion of injected phonons. The yttrium atoms in YAG are replaced isomorphously by atoms of rare earth metals (REM, R) providing a controlled concentration of centers for elastic scattering of phonons. The large dimensions of the elementary cell of YAG ($a_0 = 1.2\times 10^{-7}$ cm) are responsible for a low Debye temperature for acoustic phonons, $T_D^{ac} = 170$ K (Ref. 6). The low value of T_D^{ac} increases appreciably the effectiveness of both elastic and anharmonic decay processes. Since the heat capacity of the acoustic part of the spectrum satisfies $C_v^{ac} \sim N \sim a_0^{-3}$, where N is the number of elementary cells per cm^3, the power level limit on the heater P_H necessary to bring about the regime of nonlocal phonon thermal conductivity (NPC) is considerably lowered.[2]

Apart from the independent study of different modes of propagation of nonequilibrium phonons in YAG, the investigations are also of interest in that YAG:R is widely used in acousto-electronic apparatus because of the small attenuation of acoustic waves.[7,8] The latter is produced by the scattering of phonons by the REM atoms. The study of the processes by means of which a phonon system relaxes to thermal equilibrium, by the heat pulse method used in the present work, will aid in understanding how the REM in YAG affects the attentuation of acoustic waves.

In $(Y_{1-c}R_c)_3Al_5O_{12}$ solid solutions, phonon propagation for $c \gtrsim 0.1$ and a heater temperature T_H close to the bath temperature $T_b = 3.4$ K is already diffusional due to the high rate of elastic relaxation.[4] Changes in the purely diffusional phonon motion would be expected as the power P_H dissipated in the heater increases. It will be essentially quasidiffusional because of the participation of anharmonic (decay) processes.[1)1] In addition, in very short specimens of length $L \approx l_H \approx 0.1$ cm (l_H is a characteristic dimension of the metallic phonon injector) for large P_H, a stationary temperature can be established in a region l_h^3 with propagation of the injected phonons in the NPC regime.[2,9]

The present work is devoted to a detailed study of the "transformation" of the diffusional motion of nonequilibrium phonons in YAG:R as their temperature increases.

Crystals of substitutional YAG:R solid solutions based on dysprosium were used in this work, grown by the method of horizontal crystallization in a molybdenum container. The concentration of dysprosium atoms was monitored by x-ray methods. The specimens studied were in the form of a parallelepiped of cross-section 1.0×1.0 cm^2 and length 0.2–1.0 cm along the ⟨100⟩ or ⟨110⟩ crystal axes. A gold heater of dimensions $S_H = 0.4\times 0.4$ cm^2 was deposited onto one of the optically polished ends of the specimen and an indium bolometer of dimensions 0.3×0.25 cm^2 onto the other in a meandering shape.

The specimen lengths and the dimensions of the heater and bolometer used in the experiment corresponded to an essentially "plane" case for the injected phonon flux. Measurements were made in the temperature range $T = 2.0$–3.4 K. The superconducting transition temperature was varied by switching on an external magnetic field. Current pulses of duration 50–100 ns and repetition frequency 100–200 GHz were fed to the heater to generate the nonequilibrium phonon flux. The specific power dissipated in the heater varied within the limits 10^{-2}–10^2 W·mm^{-2}. The heater temperature was calculated by taking account of the strong elastic scattering in solid solutions[10] and the heat transfer to the helium bath as found by Danil'chenko *et al.*[11,12] An example of such a calculation is shown in Fig. 1. The extention of the region of weak dependence of T_H on P_H up to $P_H \approx 0.1$ W·mm^{-2} is due to the heat transfer from the heater to the helium.[11,12] A sharp jump in T_H is observed in the region of $P_H \approx 0.5$–1.0 W·mm^{-2} due to boiling of the helium and a reduction in heat transfer. A further jump takes place in the curve $T_H \propto P_H^{1/4}$ corresponding to the complete separation of the heater from the substrate due to the strong elastic scattering of phonons. The bolometer signal corresponding to the onset of phonon nonequilibrium was received on a VS1–280 signal storage system and was registered on a recorder.

2. EXPERIMENTAL RESULTS AND DISCUSSION

As has already been mentioned, the diffusional regime of propagation of nonequilibrium phonons is attained in a

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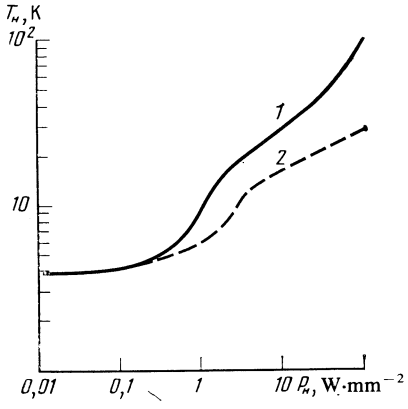

FIG. 1. The calculated dependence of the temperature of the heater T_H on the power dissipated in it P_H, taking account of heat transfer to the helium;[12] $T_b = 3.4$ K, $c = 0.1$. Curve 1—according to Kazakovtsev and Levinson[10] taking account of the temperature dependence of the heat capacity of the heater material (gold);[14] curve 2—the case of acoustic matching.

crystal with $c \gtrsim 0.1$. A weak ballistic signal for such REM concentrations was only observed in short ($L < 0.2$–0.3 cm) specimens at $T_b = 2$ K. The form of the nonequilibrium phonon signal received by the bolometer was of the characteristic diffusional bell-shape (Fig. 2). In addition, the shape of the tailing edge (see the inset to Fig. 2) and the arrival time of the nonequilibrium phonon maximum t_m (Fig. 3) change with the power dissipated in the heater P_H. The form of the dependence of the time t_m on the crystal length L also changes (Fig. 4).

The curves of Figs. 2–4 can be fairly simply interpreted within the framework of the work of Kazakovtsev and Levinson[1] and of Levinson.[2] The motion of the phonons is purely diffusive for small $P_H < 0.1$ W·mm^{-2} and the solution of the time-dependent equation of thermal conductivity can be used to analyze the excess temperature of the bolometer over the thermostat temperature in the case of a plane phonon source:

$$\Delta T \infty t^{-3/2} \exp(-L^2/4Dt), \qquad (1)$$

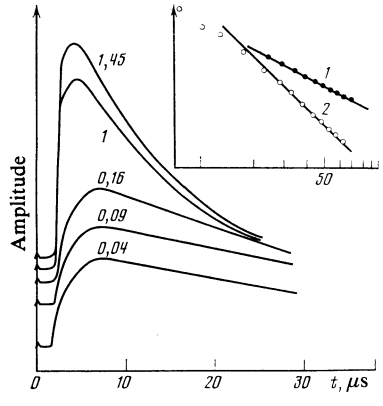

FIG. 2. The nonequilibrium phonon signal received by the bolometer. $T_b = 3.4$ K, $c = 0.1$, the value of P_H in W·mm^{-2} is shown against each curve. The magnitude of the signal as a function of the observation time is shown in the inset on a double logarithmic scale ($L = 3.1$ mm, $S_H = 9$ mm^2): 1) $P_H \lesssim 0.1$ W·mm^{-2}; 2) $P_H \gtrsim 1$ W·mm^{-2}.

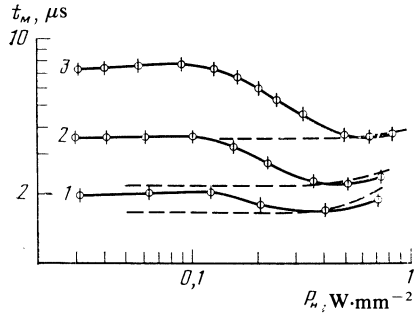

FIG. 3. The dependence of the arrival time of the maximum of the nonequilibrium phonons t_m on the power dissipated in the heater ($T_b = 3.4$ K, $c = 0.1$): 1) $L = 0.2$ cm, 2) $L = 0.27$ cm, 3) $L = 0.4$ cm. The dashed lines show how the relation transforms when there is no heat transfer to the helium.

where ΔT is the excess bolometer temperature compared with equilibrium; t is the time of observation; $D = vl/3$ is the thermal diffusivity; v is the mean phonon velocity, in our experiments equal to 5.6×10^5 cm·s^{-1}; and l is the mean free path of the nonequilibrium phonons.

According to Eq. (1), the trailing edge of the bolometer response satisfies $\Delta T \propto t^{-1/2}$ for $t \to \infty$, which agrees well with the results shown by curve 1 of the inset to Fig. 2, while $t_m \propto L^2$, which also agrees with the results shown by curve 1 of Fig. 4.

If in Eq. (1) the mean free path is determined by the rate of elastic Rayleigh scattering:

$$l_0^{-1} \sim \eta(\omega_D/v)(T_{ph}/T_D)^4, \qquad (2)$$

where ω_D and T_D are the Debye frequency and temperature of the crystal (for the acoustic part of the YAG spectrum; $T_D \approx 170$ K (Ref. 6)); T_{ph} is the phonon temperature recorded by the bolometer; and η is a measure of the crystal "defectiveness,"[10] then according to the simplest model of acoustic matching of the heater and the crystal, $t_m \propto P_H$, while in the experiment t_m is practically independent of P_H over the power range studied, 0.01–0.1 W·mm^{-2} (see Fig. 3). This means that for $P_H \lesssim 0.1$ W·mm^{-2} the temperature of the injected phonons depends weakly on P_H, which agrees with

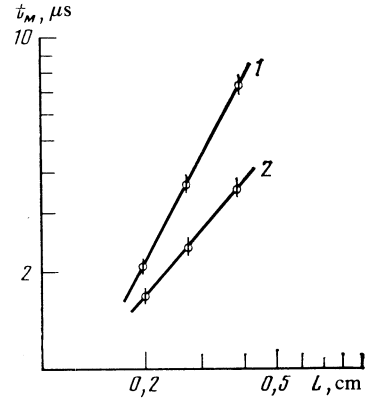

FIG. 4. The dependence of the arrival time of the maximum of the nonequilibrium phonons t_m on the specimen length L ($T_b = 3.4$ K, $c = 0.1$): 1) $P_H \lesssim 0.1$ W·mm^{-2}; 2) $P_H \gtrsim 1$ W·mm^{-2}.

the corresponding part of the curve in Fig. 1 and is brought about by the strong heat transfer from the film injector to the helium bath.

The power region $P_H \gtrsim 0.1$ W·mm^{-2} is a characteristic interval in the dependence of t_m on P_H (Fig. 3). A "stepwise" reduction in t_m occurs in this region (the step is naturally spread out). We note that for such values of $P_H \gtrsim 0.1$ W·mm^{-2} an appreciable increase in T_H is observed and, consequently, in the temperature of the injected phonons (see Fig. 1), which according to Eq. (2) must lead to an increase in t_m. We interpret the reduction of t_m as the starting up of inelastic phonon-phonon interaction (decay processes). We make some estimates of the phonon mean free path under conditions of elastic scattering and decay:

$$l=(\bar{v}/\omega_D)(3\gamma\eta)^{-1/2}(T_{ph}/T_D)^{-9/2}; \quad (3)$$

where γ is the phonon-phonon interaction parameter which for YAG is $\approx 4\times 10^{-4}$; $\eta \approx 0.1$. The value of l becomes less than 1 cm, i.e., the maximum specimen lengths used in the experiment, at $T_{ph} \approx 10$–12 K. By using the familiar relation $T_{ph} \approx 2.8\ T_H$, we come to the conclusion that decay processes become important in our experiments even for $T \gtrsim 4$ K, which does not exceed T_b by much. We repeat again that the high efficiency of phonon scattering processes in YAG:R is due to the low Debye temperature of acoustic phonons.

The propagation regime of nonequilibrium phonons becomes quasidiffusional after decay processes start up: in accordance with Kazakovtsev and Levinson[1] the trailing edge of the diffusional bell becomes steeper: $\Delta T \to t^{-1}$ as $t \to \infty$ (curve 2 of the inset to Fig. 2).

When P_H is increased further the injected phonons can propagate in the NPC regime, in which case near the phonon injector their distribution can be described by the Planck distribution. The possibilities of realizing this regime experimentally have been analyzed in detail by Ivanov, *et al.*[9] Here we only note that the relaxation $t_m \propto P_H^{1/2}$ is characteristic for the NPC regime,[2] which is indeed observed experimentally for short specimens.

The $t_m(L,P_H)$ relations for specimens of various lengths for $P_H = 1$–10 W·mm^{-2} are shown in Fig. 5. For a specimen with $L = 0.2$ cm we have $t_m \propto P_m^{1/2}$, which corresponds to NPC. As the specimen length increases, the curves flatten since the NPC makes an ever-decreasing contribution to the bolometer response and the region of quasidiffusional phonon motion broadens, for which t_m is independent of P_H (Ref. 1).

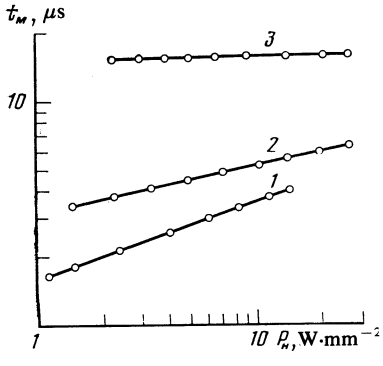

FIG. 5. Arrival time of the maximum of the nonequilibrium phonons t_m vs the power P_H dissipated in the heater, for specimens of different length ($T_b = 3.4$ K, $c = 0.1$, $S_H = 1$ mm^2): 1) $L = 0.2$ cm, 2) $L = 0.4$ cm, 3) $L = 0.9$ cm.

We shall make some quantitative estimates. The threshold poewrs are given by Kazakovtsev and Levinson:

$$P_e = 0.92\eta^{-1}\frac{\hbar v_s}{(a_0)^3}t_p^{-1}, \quad (4)$$

at which the ballistic regime for cooling the injector goes over to diffusional,[10] and

$$P_a = 0.64(\eta\gamma)^{-4/9}\frac{\hbar v_s}{a_0^3}\left[\frac{d}{a_0}\frac{v_s}{\bar{v}_s}\right]^{1/9}t_p^{-1}, \quad (5)$$

at which the diffusional cooling goes over to thermal conductivity.[13] In Eqs. (4) and (5) t_p is the duration of the heating pulse, v_s and $\bar{v}_s$ are the mean velocities of sound in the substrate and heater and d is the heater thickness. It is easy to show that for the parameters of the materials we used and $t_p \approx 10^{-7}$ s, we have $P_a/P_e \approx 20$ and $P_e \approx 0.03$ W·mm^{-2}.

The estimates obtained are in reasonable agreement with the experimental results given above. In fact the phonon propagation regime is already diffusional at 0.1 W·mm^{-2}, which corresponds to the estimate of $P_e \approx 0.03$ W·mm^{-2}. Pure diffusional motion changes to quasidiffusional (for long specimens) in the region of $P_H \approx 1$ W·mm^{-2} and to NPC (short specimens). This value of $P_H \approx$ W·mm^{-2} agrees with the estimate of $P_a \approx 0.6$ W·mm^{-2}. It is not surprising that there are differences between the observed and calculated magnitudes of the threshold values of $P_{e,a}$. Expressions (4) and (5) are derived, in particular, without taking account of heat transfer from the heater to the liquid helium. The influence of the fundamental role of the conditions of heat transfer from the heater to the helium in producing the different regimes of phonon kinetics is illustrated by a calculation shown in Fig. 1. In addition we have studied the influence of the heat transfer conditions experimentally.

A thin thermally insulating convering was deposited on the metal heater film to reduce the heat transfer from the surface of the heater into the helium. This gave rise to the region on the t_m-P_H dependence (see Fig. 3) where a reduction in the arrival time of the nonequilibrium phonon maximum was previously observed, shifting in the direction of smaller values of P_H and for efficient thermal insulation of the film, the region in which the dependence of t_m on P_H changed sharply was not observed at all.

The results obtained for efficient thermal insulation are shown in Fig. 3 by the dashed lines. The independence of t_m on the magnitude of the heater power P_H and the nearly linear dependence $t_m \propto L^n$, $n \approx 1$ (curve 2 of Fig. 4) were characteristic of them, in complete agreement with Eq. (1). The removal of heat transfer to the helium thus increases the temperature of the injected phonons, brings in decay processes earlier and broadens in the direction of smaller values the range of values of P_H for which the phonon motion is quasidiffusional. We were unable to observe the regions of

pure diffusional motion under thermal insulation conditions.

The various propagation regimes for phonons in yttrium aluminum garnet alloyed with dysprosium, traced in this work, are on the whole in satisfactory agreement with existing theoretical ideas about the form of the dependence temperature of the nonequilibrium phonons, the specimen length, the conditions of heat transfer from the heater, etc. The conditions for the specimen length, power dissipated in the heater, etc., have been obtained when each propagation regime is realized: diffusion, quasidiffusion or the nonlocal phonon thermal conductivity regime. The investigations carried out on the propagation of phonons under such specific conditions of bath temperature, of P_H and a number of other parameters enables the correctness of expressions (2) and (3) which we have used for the rates of elastic and inelastic relaxation to be analyzed for crystals with large impurity concentration and in solid solutions.

In conclusion the authors thank Yu. V. Galyaev for his interest in the work and I. B. Levinson for valuable discussions and for allowing us to learn about the results of his calculations before their publication.

[1)]The first observations of quasidiffussional motion were made by Bron *et al.*[3] and by Ivanov and Khazanov.[4]

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