

# Propagation of nonequilibrium acoustic phonons in solid solutions of aluminum-based garnets

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It is shown that nonequilibrium phonons injected into a garnet lattice by a thermal pulse propagate predominantly quasidiffusively when the power dissipated at the heater is in the range  $10^2$ – $10^4$  W/cm<sup>2</sup>. Anomalous propagation of the nonequilibrium phonons is discovered at 25% concentrations of rare earth metals substituting yttrium atoms. An explanation of the observed anomaly is suggested.

## 1. FORMULATION OF THE PROBLEM AND THE EXPERIMENTAL METHOD

Yttrium-aluminum garnets (YAG) activated by the ions of rare earth metals (REM) are widely used as the active media in lasers<sup>1</sup> and, because of their small damping coefficient for acoustic waves, in making acoustoelectrics.<sup>2,3</sup> In order to usefully modify the acoustic parameters of solid solutions of aluminum based yttrium-rare-earth garnets, it is necessary to understand the relaxation processes leading to the thermal equilibrium of the phonon excitations.

An effective way of studying phonon kinetics is to study the propagation of nonequilibrium phonons through the crystal (the method of "heat pulses"). Here we shall apply this method to the solid solutions of aluminum based garnets.

YAG crystals and the solid solutions in which yttrium is substituted by a rare-earth metal  $R$  ( $R = \text{Dy}, \text{Yb}, \text{Lu}$ ) were grown in a molybdenum container by the method of horizontal crystallization. The REM concentration was controlled by x-ray methods. The investigated samples had the form of parallelepipeds of cross section  $1.0 \times 1.0$  cm<sup>2</sup> and of length 0.4–1.2 cm along  $\langle 100 \rangle$  or  $\langle 110 \rangle$  crystallographic axes. We deposited a gold heater of dimensions  $0.1 \times 0.1$  cm<sup>2</sup> on one of the optically polished faces, while on the opposite face we placed an indium bolometer in the form of a meander. The measurements were conducted in the temperature interval  $T = 2.0$ – $3.4$  K. The superconducting transition temperature of the bolometer was changed by means of an external magnetic field.

In order to generate a flow of nonequilibrium phonons, current pulses of length 70 ns and repetition rate 50–100 Hz were applied to the heater. The power dissipated at the heater was in the  $10^2$ – $10^4$  W/cm<sup>2</sup> range. Assuming strong elastic scattering and neglecting any heat transfer to the bath we calculated the temperature of the heater according to Ref. 4; estimates of temperatures of the measured nonequilibrium phonons were also given in Ref. 4.

Bolometer signals, corresponding to the arrival of the nonequilibrium phonons, were collected by the BCI-280 data acquisition system and were automatically recorded.

The influence on the observed relationships of using different REM was not investigated.

## 2. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

1. A regime of ballistic propagation of nonequilibrium phonons was observed over the whole investigated temperature interval in pure garnet crystals or in garnets with small amounts of impurities,  $(\text{Y}_{1-c}\text{R}_c)_3\text{Al}_5\text{O}_{12}$ ,  $c \lesssim 0.03$ , where the sample length  $L$  was up to 0.5 cm. Clearly distinguishable were the longitudinal and the transverse components of the signals; different transverse modes were not resolved because their velocities were nearly equal.

2. An estimate of the nonequilibrium phonons temperature  $T_{\text{ph}}$  measured by the bolometer was based on experiments on long ( $L > 0.5$  cm) crystals with small impurity concentrations ( $c \approx 0.03$ ). By increasing the power  $P_h$  dissipated at the heater it was possible in such samples to follow transition from the ballistic nonequilibrium phonon propagation regime to the diffusive regime in which the form of the nonequilibrium phonon signals registered by the bolometer displays the characteristic inverted bell form (see Fig. 1).

The value of  $T_{\text{ph}}$  was calculated in the following way. From the data of Fig. 1 it is possible to determine the mean

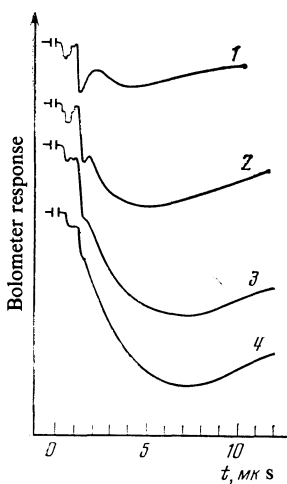


FIG. 1. Time dependence of the bolometer response for different powers dissipated at the heater for a  $(\text{Y}_{0.97}\text{Yb}_{0.03})_3\text{Al}_5\text{O}_{12}$  crystal of length  $L = 0.73$  cm and for bolometer temperature  $T_b = 3.4$  K: (1)  $P_h = 0.2 \times 10^3$  W/cm<sup>2</sup>; (2)  $P_h = 0.88 \times 10^3$  W/cm<sup>2</sup>; (3)  $P_h = 1.8 \times 10^3$  W/cm<sup>2</sup>; (4)  $P_h = 5 \times 10^3$  W/cm<sup>2</sup>.

free path  $l$  of nonequilibrium phonons in the following two cases:

1) When the signal corresponding to the nonequilibrium ballistic propagation of phonons is still developing while there is already influx of diffusive phonons (curve 1 in Fig. 1), in which case  $l \approx L$  to a good approximation;

2) When the signal registered by the bolometer has a clearly diffusive character (curves 3 and 4 in Fig. 1), and when the arrival time  $t_m$  of its maximum is much larger than its flight time  $t_f \approx L/\bar{v}$ , where  $\bar{v}$  is the mean phonon speed equal in our experiments,  $5.6 \times 10^5$  cm/s. In this case one can estimate  $l$  by comparing the experimental curve with the solution of the nonstationary heat equation for a point source<sup>5,6</sup>:

$$\Delta T \sim t^{-3/2} e^{-L^2/4Dt}, \quad (1)$$

where  $\Delta T$  is the temperature increase at the bolometer relative to the equilibrium temperature,  $t$  is the observation time and  $D = \bar{v}l/3$  is the thermal conductivity. Below we shall evaluate the adequacy of Eq. (1) for the analysis of the experimental data.

The values of  $l$  obtained with such estimates were compared with the values obtained from the rate of the Rayleigh scattering:

$$l_0^{-1} = \delta \omega_D / \bar{v} (T_{ph}/T_D)^4, \quad (2)$$

where  $\omega_D$  and  $T_D$  are the Debye frequency and temperature of the crystal (for YAG  $T_D = 750$  K),<sup>7</sup> and  $\delta$  is a measure of crystal imperfection (in the notation of Ref. 4). The use of Eq. (2) to estimate  $T_{ph}$  assumes that the rate of elastic scattering on a given quantity of substitutional impurities ( $c \approx 0.03$ ,  $\delta \approx 0.05$ ) is maximized in comparison with possible contributions from the scattering on structural defects, various structural strains, isotopic scattering, etc. That is, the defect measure  $\delta$  is solely determined by the controllable concentration of the impurities introduced.

The temperature  $T_{ph}$  of the phonons detected by the bolometer calculated according to the suggested procedure is shown by the filled circles in Fig. 2.

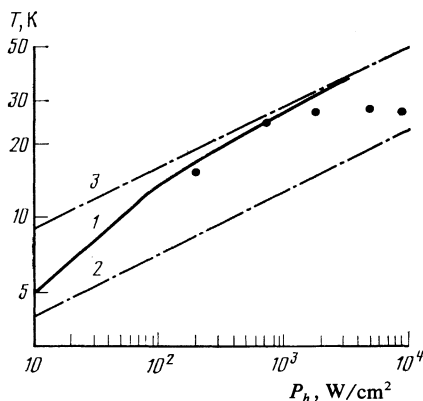


FIG. 2. Temperature of the nonequilibrium phonons registered at the bolometer vs power dissipated at the heater: filled circles correspond to the  $(Y_{0.97}Yb_{0.03})_3Al_5O_{12}$  crystals of lengths  $L = 0.73$  cm and  $0.39$  cm; curves 1 and 3 are calculated according to Ref. 4, while curve 2 is calculated according to the acoustic matching theory. The bolometer temperature is  $T_b = 3.32$  K.

In the same figure we give the theoretical dependence of the heater temperature  $T_h$  on the dissipated power<sup>4</sup> (curve 1) calculated assuming that the elastic scattering rate is only controlled by the introduced impurities.

For small powers  $P_h < 10^2$  W/cm<sup>2</sup> curve 1 approaches curve 2, which corresponds to the acoustic matching between the heater and the crystal and to its ballistic cooling (bolometer temperature  $T_b = 3.4$  K); for  $P_h > 10^2$  W/cm<sup>2</sup> curve 1 approaches curve 3, which corresponds to a significant tearing of the heater away from the crystal caused by the strong return of the emitted phonons as a result of elastic scattering.<sup>4</sup>

Comparison between the calculated values  $T_{ph}$  and  $T_h$  shows that  $T_{ph} < T_h$ . Therefore, the relation  $T_{ph} \sim 3T_h$  usually used to estimate the temperature of the nonequilibrium phonons measured by the bolometer is not fulfilled.<sup>8</sup> This discrepancy increases with the power dissipated at the heater. The reduction of  $T_{ph}$  received by the bolometer is clearly caused by inelastic relaxation processes e.g., decay. Estimates of the length  $l_d$  of the free path associated with the decay processes shows that for crystals with  $L \approx 1$  cm the decay processes can be significant even for  $P_h > 5 \cdot 10^2$  W/cm<sup>2</sup>, i.e.,  $l_d < L$  for  $T_{ph} \approx 3T_h > 60$  K. As  $P_h$  increases the importance of decay processes increases which is consistent with the observed relationship between  $T_h$  and  $T_{ph}$  (Fig. 2).

It is necessary to notice that the calculated limiting values of  $T_h$  (curve 3, Fig. 2) can be reduced even at  $T > 10$  K [Ref. 9] because of the departures of the specific heat of gold from the Debye law which is assumed in Ref. 4. This is confirmed by the results of Ref. 10, where a significant tearing-away of the heater from its base resulted from roughening of the surface on which the heater is deposited. It was found that  $T \sim 100$  K for  $P \gtrsim 10^2$  W/cm<sup>2</sup>.

3) The diffusive regime for propagation of phonon instabilities in solid crystal solutions was observed for values of  $c$ , satisfying  $(1 - c) \geq 0.05 - 0.07$ . For such concentrations the ballistic signal was very weak and was measured definitively only for short samples ( $L < 0.3 - 0.4$  cm) at  $T = 2.1$  K. This is a reasonable result. Indeed, with an increase in concentration of substitutional impurities the rate of elastic scattering and the temperature of injected nonequilibrium phonons increase, while the length of their mean free path drops and they propagate predominantly by diffusion.

The nonequilibrium phonon signal received by the bolometer has an analogous form (Fig. 3). The following question arises: to what extent can the observed curves be described as solutions to the diffusion equation (1)? In order to clarify this question we conducted experiments to investigate the arrival time  $t_m$  for the maximum deviation from equilibrium of the phonons as a function of the crystal length. It was found that  $t_m \sim L^2$  only for long crystals of  $(Y, Lu, Dy)_3Al_5O_{12}$ , that is, in garnet matrices based on aluminum. The diffusive character of nonequilibrium phonon propagation in such crystals is caused by the scattering on trace impurities, on inhomogeneities, and by the contribution from the isotope scattering.

Even in solid garnet solutions such that  $1 - c \sim 0.07 - 0.1$  the exponent  $n$  in the dependence of  $t_m$  on  $L$  does not

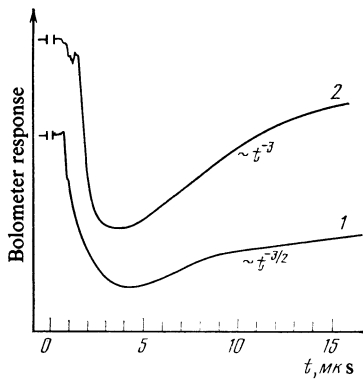


FIG. 3. Time dependence of the bolometer response for various temperatures: (1)  $T_b = 3.32$  K; (2)  $T_b = 2.12$ . The crystal is  $(Y_{0.9}Dy_{0.1})_3Al_5O_{12}$ , its length is  $L = 0.4$  cm, and the power is  $P_h = 0.54 \times 10^3$  W/cm $^2$ .

exceed 1.5 and over a broad interval of concentrations it is close to one (Fig. 4).<sup>2)</sup>

Without analyzing the fine structures in Fig. 4, we notice one more important difference between the bolometer response in our experiments and in diffusive propagation regime: the arrival time  $t_m$  of the maximum phonon deviation from equilibrium is weakly dependent on the power dissipated by the heater. Within the interval of investigated powers  $t_m \sim P_h^{1/6-1/4}$ , whereas in the diffusive regime (1) with only elastic scattering on impurities  $t_m \sim T_{ph}^4 \sim P_h$ .

All these facts allow us to conclude that in our experiments on solid garnet solutions a quasidiffusive regime of nonequilibrium phonon propagation arises.<sup>11</sup> This is a diffusive propagation with a diffusion constant which varies as a result of the energetic phonon relaxation mechanism. In the case where inelastic processes are dominated by decay one finds for the quasidiffusive regime

$$t_m \sim L^n P_h^k, \quad (3)$$

where  $n > 10/9$  and  $k \approx 0$ , which is close to the experimentally observed relationship. The deviation of  $k$  from zero could be caused in our experiments either by an alternation of the nonequilibrium phonon propagation regime between diffusive and quasidiffusive, or by the appearance of the effects of nonlocal thermal conductivity<sup>12</sup> for  $P_h \gtrsim 10^3$  W/cm $^2$ . For the nonlocal thermal conductivity regime (linear case)

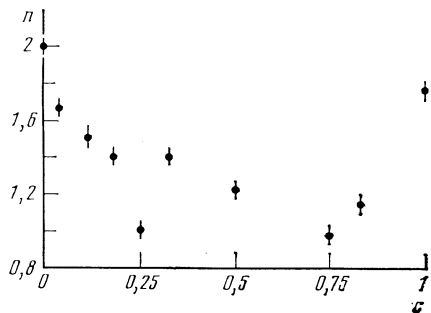


FIG. 4. Dependence of the exponent in  $t_m \sim L^n$  on the concentration of the solid solution  $(Y_{1-c}R_c)_3Al_5O_{12}$  for  $P_h = 2 \times 10^3$  W/cm $^2$  and  $T_b = 3.34$  K.

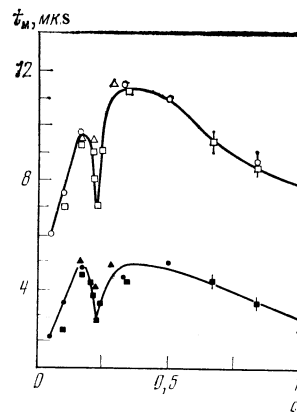


FIG. 5. Arrival time of the maximum of the phonon deviation from equilibrium vs concentration of the solid solutions  $(Y_{1-c}R_c)_3Al_5O_{12}$ : (O—Dy,  $\Delta$ —Yb,  $\square$ —Lu), filled points correspond to  $T_b = 2.15$  K while the open points correspond to  $T_b = 3.34$  K. for all samples  $L = 0.5$  cm and  $P_h = 2 \times 10^3$  W/cm $^2$ .

$$t_m \sim L^{1/2} P_h^{1/4}. \quad (4)$$

A nonzero  $k$  can also occur because of some other inelastic energy relaxation process more effective than decay processes (for example, resonant scattering by the excitations in the yttrium garnet phonon spectrum which are caused by the introduction of the REM).<sup>3)</sup>

In the regime of quasidiffusive propagation of nonequilibrium phonons ( $c, 1-c > 0$ ;  $P_h \sim 2 \times 10^3$  W/cm $^2$ ) we traced the dependence of  $t_m$  for a sequence of solid garnet solutions.<sup>3)</sup> This is shown in Fig. 5 for the cases  $Y_3Al_5O_{12} \rightarrow (Lu, Yb, Dy)_3Al_5O_{12}$  in samples of equal length. With an increase in  $c$  or  $1-c$ , that is, as the aluminum based garnet is doped, one observes an increase in  $t_m$ . This is a result of an increase in the rate of elastic scattering at the isomorphous substitutional atoms.

Unexpectedly, one observes in this series of experiments an anomalous change in the character of the nonequilibrium phonon propagation for garnets with composition  $c_k \sim 0.25$ . Namely, one observes a sharp drop in  $t_m$ . This means that the diffusion constant and, consequently, the mean free path peak at  $c = c_k$ . This increase in  $l$  at  $c = c_k$  allowed us to observe for small values of  $P_h < 0.2 \times 10^3$  W/cm $^2$  even the ballistic nonequilibrium phonon propagation regime in crystals of the same composition.

Therefore, an anomaly at  $c = c_k$  is superimposed on a general quasidiffusive character of nonequilibrium phonon propagation in systems of solid garnet solutions. This anomaly can be qualitatively interpreted in terms of a model of randomly distributed Y and R substitution atoms over corresponding vertices of the crystallographic lattice.

At small concentrations of impurities,  $c(1-c) \ll 1$ , this random distribution causes excitations of the phonon density of states near the vibration frequencies of the impurity atoms, which in turn scatter both thermal and nonequilibrium phonons. This excitation of the phonon spectrum of YAG partially determines the change in the absorption of acoustic waves in yttrium-rare earth aluminum garnets.<sup>3)</sup>

As the concentration of randomly distributed substitu-

tional atoms increases is possible for some composition of the solid solution to form an ordered distribution of  $Y \leftrightarrow R$  atoms. For example, R atoms could preferentially occupy some inequivalent Y positions in the unit cell of the garnet. The elastic scattering rate of the phonons is reduced in such ordered solid solutions and the phonons propagate either ballistically or with a large diffusion constant.

Asymmetry of the curves in Fig. 5 relative to  $c = 0.5$  can be explained within the context of the changes of the phonon spectrum in solid substitutional solutions and by the existence of critical concentrations at which atoms of one kind (impurities) form clusters whose size tends to infinity permeating the whole crystal at  $c = c_k$ .<sup>14</sup> We note that in our experiments the value  $c_k \approx 0.25$  correlates well with the value for the percolation threshold in cubic crystals.<sup>15</sup>

Measurements of thermal conductivity and x-ray spectra of such solid solutions could confirm the validity of this interpretation of the anomaly at  $c_k \approx 0.25$ . Thus, for the thermal conductivity of the investigated solid solutions one should expect asymmetry in the curve about  $c = 0.5$  at low temperatures ( $T = 2-4$  K) because of the effective scattering on the low frequency resonant modes near isolated heavy REM atoms.<sup>3,14</sup> One also expects anomalies near  $c = c_k$ .

In conclusion, the authors are grateful to Yu. V. Gulyaev for his interest in the work, to S. F. Akhmetov for preparing the samples of the garnets, and to I. B. Levinson for numerous useful discussions.

<sup>11</sup>Note that presence of the surface layer which was damaged during the process of optical polishing always causes separation of the heater from its base and an increase of its temperature relative to the acoustic match-

ing model. We conducted qualitative experiments regarding the influence of the surface treatment of  $T_{ph}$  registered by the bolometer. Etching the surfaces of the investigated samples in boiling  $H_2SO_4$  for six hours removed the surface layer and caused the length of the phonon mean free path to increase as a result of the decrease in  $T_h$ .

<sup>2</sup>In the above analysis of the heater performance under a very elastically scattering load ( $c \approx 0.03$ ), we used the diffusion equation to estimate the free path mean length. We note that at such concentrations  $n = 1.8$  and because of this inconsistency it is necessary to consider estimates of the nonequilibrium phonon temperatures given in Fig. 2.

<sup>3</sup>A short exposition of the present section can be found in Ref. 13.

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