Anomalies in the propagation of heat pulses in solid solutions of yttrium-rare-earth-aluminum garnets

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The propagation of nonequilibrium phonons is predominantly a diffusive propagation in garnets in which the atomic concentration of yttrium and the rare-earth metals Lu, Yb, and Dy exceeds 5% (the atoms of yttrium and those of these rare-earth metals can substitute for each other). At a rare-earth-metal concentration $\simeq$25% there is an anomalous increase in the mean free path of nonequilibrium phonons, whose propagation may become ballistic.

In this letter we report the use of the heat-pulse method[1] to study the relaxation of nonequilibrium phonons in isomorphic garnet solid solutions $(Y_{1-C}R_C)_3Al_5O_{12}$, where R = Lu, Yb, or Dy.

The nonequilibrium phonons are excited by a brief current pulse $t_p = 70$ ms, which is applied to a gold heater deposited on the end of the sample, which is $L = 0.4$–1.0 cm long. The nonequilibrium phonons are detected by an indium bolometer at the opposite end of the sample. The temperature of the superconducting junction of the bolometer is varied over the interval $T = 2.0$–3.4 K by applying an external magnetic field. The heater temperature T_h is calculated by the method of Ref. 2 under the assumption of strong elastic scattering in the solid solutions. The power dissipated in the heater during the experiments was $P_h = (0.2\text{–}2.0)\times 10^3$ W/cm^2 ($T_h \simeq 18$–30 K). We found no significant differences in the results for the various rare-earth metals.

We first consider the case of relatively low power levels, $P_h \simeq 0.2\times 10^3$ W/cm^2. In the garnet with $C = 0$, and with L up to 0.5 cm, there was a ballistic propagation of nonequilibrium phonons, and it was possible to clearly distinguish longitudinal and shear components of the signal (curve 1 in Fig. 1). In the matrix with $C = 1$ we also observed a ballistic response, but there was also a significant diffusive component, apparently due to isotopic scattering by the R atoms (curve 5 in Fig. 1).

As the substitutional impurity is added to the garnet matrix, i.e., with increasing C (or $1 - C$), the elastic Rayleigh scattering by the isomorphic substitutional atoms intensifies, and the mean free path of the nonequilibrium phonons decreases. The ballistic component of the bolometer response decreases and becomes substantially distorted by a diffusive background that develops. At C, $1 - C \gtrsim 0.05$–0.07, the propagation of the nonequilibrium phonons is predominantly diffusive, and the phonon-nonequilibrium signal received by the bolometer has a corresponding bell shape.

Unexpectedly, we found an anomalous change in the nature of the propagation of the nonequilibrium phonons for the garnet of the composition $C_c \simeq 0.25$. In these samples we again observed a ballistic propagation of the nonequilibrium phonons. At $C = 0.2$ and 0.34, i.e., near the critical composition of the solid solution, the propaga-

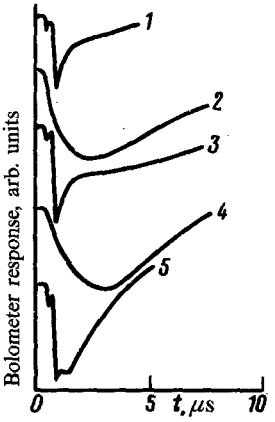

FIG. 1. Time evolution of the bolometer response for various compositions of the solid solution $(Y_{1-C}R_C)_3Al_5O_{12}$ for samples $L = 0.4$ cm long. The bath temperature is 2.11 K; $P_h = 0.2\times10^3$ W/cm^2. 1—$C = 0$; 2—$C = 0.2$; 3—$C = 0.25$; —$C = 0.34$; 5—$C = 1$ (R = Lu).

tion of the nonequilibrium phonons is diffusive (curves 2, 3, and 4 in Fig. 1).

In experiments with large values $P_h = 2\times10^3$ W/cm^2, in which the temperature of the injected nonequilibrium phonons rises, and the scattering by atoms of the impurity and isotopes intensifies, the phonon-nonequilibrium signal at the bolometer is diffusive for all samples except that with $C = 0$. Figure 2 shows the arrival time of the phonon-nonequilibrium peak versus the composition of the solid solution. Again in this case we see an anomaly: At $C = C_c$ the diffusion coefficient of the nonequilibrium phonons is at a maximum, corresponding to an increase in the mean free path of the nonequilibrium phonons. The apparent explanation for this sharp change in the nature of the propagation of the nonequilibrium phonons at both large and small values of P_h is a pronounced weakening of the elastic scattering of nonequilibrium phonons by substitutional atoms near $C = C_c$.

In general, the Y and R atoms, which replace each other, are distributed at

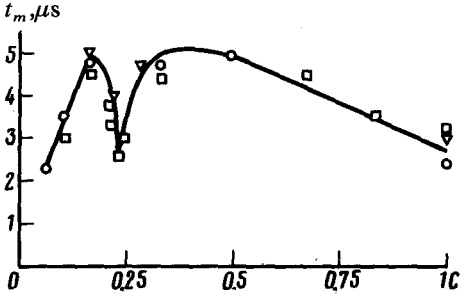

FIG. 2. Time at which the maximum of the phonon nonequilibrium appears versus the composition of the solid solution $(Y_{1-C}R_C)_3Al_5O_{12}$ ($P_h = 2\times10^3$ W/cm^2, bath temperature of 2.11 K, samples $L = 0.5$ cm long). □—Lu; ∇—Yb; ○—Dy.

random among the corresponding lattice sites. This random distribution causes a local perturbation of the phonon state density near the vibration frequencies of the impurity atoms and a scattering of nonequilibrium phonons by these atoms at low impurity concentrations, C, $1 - C \ll 1$. With increasing concentration of the randomly distributed substitutional atoms, i.e., at a certain composition of the solid solution, an *ordered* arrangement of Y$\rightleftarrows$R atoms may form because of, for example, a predominant occupation of certain nonequivalent Y positions by R atoms in the unit cell of the garnet. The rate of elastic scattering of nonequilibrium phonons decreases in this ordered solid solution, and the nonequilibrium phonons propagate ballistically or with a larger diffusion coefficient.

The asymmetry of the curve in Fig. 2 with respect to $C = 0.5$ can be explained on the basis of a transformation of the phonon spectrum in the substitutional solid solutions and the existence of critical concentrations, at which the atoms of one species (the impurity) form clusters, whose dimensions tend toward infinity and fill the entire crystal at[3] $C = C_c$. In our experiments, the value $C_c \simeq 0.25$ correlates well with the percolation threshold for cubic crystals.[4]

This interpretation can be tested by measuring the thermal conductivity and x-ray spectra of solid solutions of this sort. For the thermal conductivity of this solid solution, for example, we would expect an asymmetry of the curve with respect to $C = 0.5$ because of the effective scattering by low-frequency resonant modes near isolated heavy rare-earth metal atoms[3] and singularities near $C = C_c$.

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