Determination by the "warm" pulse method of the kinetic characteristics of phonons in crystals with impurities

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The inelastic relaxation times of nonequilibrium phonons have been measured by the "warm" pulse method in $Y_{3-x}Lu_xAl_5O_{12}$ and $BeAl_{2-x}Cr_xO_4$ solid solutions. The relation between the rates of elastic and inelastic phonon relaxation is shown for the case of YAG:Lu solid solutions. The influence of heat-transfer processes on the time characteristics of nonequilibrium phonon signals is discussed for the case in which the signals propagate in crystal diffusively.

The propagation of nonequilibrium phonons (NP) in a crystal can be accompanied by their interaction with various forms of excitations and defects of the crystal lattice. If phonons in a crystal containing defects do not interact with one another, then their propagation can be described by the usual diffusion equation. But if the phonon energy or the particulars of the structure of the specimens studied are such that phonon-phonon interactions become possible in the process of diffusion, then the propagation process becomes more complicated, leading to a characteristic change in the time behavior of the recorded signal.[1,2]

We have described[3] a method for obtaining the temperature dependence of the elastic scattering relaxation time τ_0 in crystals under conditions in which NP propagate diffusively. For a "warm" generator ($\Delta T \ll T_0$) (T_0 is the thermostat temperature, ΔT is the excess of the injector temperature above the thermostat temperature), when the occupation numbers of phonons injected from the film heater into the specimen are little different from the equilibrium numbers, the arrival time of the maximum of the nonequilibrium phonon signal satisfies $t_M(T) \propto \tau_0^{-1}(T) \propto T^4$.

When one analyzes the diffusive motion of NP the dependence of the arrival time of the maximum of the NP signal or of the point of the maximum steepness (before the maximum) on thermostat temperature, power dissipated in the film injector, etc., are studied. The trailing edge of the nonequilibrium phonon signal usually remains outside the investigation; in the case of a plane source with $\Delta T \ll T_0$ this has a time dependence $S(t) \propto t^{-1/2}$. One would expect that after a certain lapse of time for the NP forming the trailing edge of the signal, energy exchange processes would become probable, which would promote the establishment of equilibrium in the NP-thermostat system, which should make the nonequilibrium phonon signal disappear and sharply increase the steepness of the $S(t) \propto t^{-1/2}$ preceding it. Such an increase in the steepness of the bolometer signal could be interpreted as the result of the switching-on of phonon-phonon interactions in the NP-thermostat system and the actual time corresponding to this moment could be interpreted as the relaxation time of inelastic phonon-phonon interaction.

An experimental finding of the increase in steepness of the trailing edge of the signal $S(t)$ in the diffusive NP propagation regime would also enable the temperature and impurity or crystal lattice defect concentration dependence of the phonon-phonon scattering time to be studied together with the dependence of phonon-impurity scattering, and in this way determine the whole set of times characterizing the kinetics of phonon processes in materials with intense phonon scattering, which is the aim of the present work.

INVESTIGATION OF INELASTIC PHONON INTERACTION IN $Y_{3-x}Lu_xAl_5O_{12}$ AND $BeAl_{2-x}Cr_xO_4$

In the present work the main emphasis was directed to a study of nonequilibrium phonon signals for long propagation times, which required some updating of the method which we had used earlier. The main difference was the broadening of the frequency range of the measuring circuit into the low-frequency region which enabled errors in measuring signals at times up to 0.01 s to be avoided. Apart from this, steps were taken to improve the signal-to-noise ratio.

The nonequilibrium phonon signals recorded by the bolometer in a specimen of $Y_{2.65}Lu_{0.35}Al_5O_{12}$ are shown in Fig. 1 for a number of thermostat temperatures. The curves of Fig. 1a for $t \gg t_M$ are shown in Fig. 1b on a log-log plot with the curves normalized to some fixed amplitude and with the time intervals of measuring the signals expanded. For observation times $\gtrsim 10^3$ μs the sharp increase in the steepness of the curves is clearly seen.

The arrival time of the maximum of the nonequilibrium phonon signal t_M (Fig. 1a) carries information about elastic scattering of phonons by the mass defect of the ions $Y^{3+} \rightleftarrows Lu^{3+}$; we have analyzed its temperature dependence previously.[3] The results in Fig. 1b, which reflect the sharp increase in the steepness of the signal at the tails of the curves, are analyzed first.

In the warm injector regime ($\Delta T \ll T_0$) an increase in the curvature of the trailing edge of the signal arises later for a lower thermostat temperature and correspondingly a lower injected phonon temperature, since $T_0 \approx T_H$ (T_H is the temperature of the film heater). Analysis of the results of the kind shown in Fig. 1b, with a correction for the real temperature of the film heater[3] taken into account, gives for the time for the start of the increase in steepness τ^* a temperature dependence on the form[1)] $\tau^* \propto T^{-n}$, where $n \approx 5$, which enabled us to assume that the observed change in the time dependence of the trailing edge of the signal is the result of inelastic processes in the NP-thermostat system, and τ^* is the relaxation time for inelastic scattering of phonons.

Theoretical estimates of the phonon-phonon interaction relaxation time in $Y_3Al_5O_{12}$ on the basis of the known first- and second-order elastic constants[4] give the value $\tau^* \approx 10^{-2}$–10^{-3} s which is in good order-of-magnitude agreement with the results obtained.

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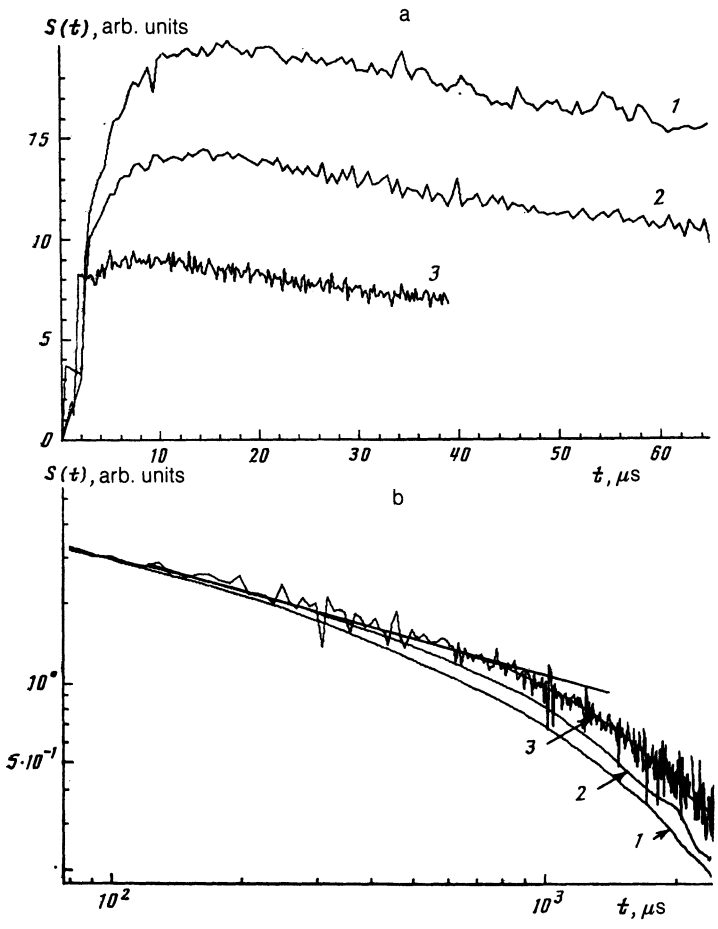

FIG. 1. a—Signals recorded by the bolometer for the propagation of NP in the diffusive regime in a specimen of $Y_{2.65}Lu_{0.35}Al_5O_{12}$; specimen length $L = 5$ mm for different values of T_0: *1*) $T_0 = 3.78$ K; *2*) 2.85 K; *3*) 2.25 K; b—trailing edge of the signals shown in Fig. 1a (curves *1–3*); straight line *4*–the dependence $S \propto t^{-1/2}$.

The dependence of τ_0 and τ^* on the composition of the Y–Lu garnet solid solution are shown in Fig. 2. Both curves have a similar nonmonotonic character, from which we can state that there is a connection between the rates of elastic and inelastic NP scattering in the solid solution studied.

In the diffusive regime the signal recorded by the bolometer is formed by the whole phonon distribution injected into the object studied from the film heater. In addition, different groups of NP contribute variously in different parts of the signal. Notably the trailing edge of the signal is formed mainly by higher frequency phonons than the maximum, i.e., for a given heater temperature the time τ^* characterizes higher-frequency phonons rather than the phonons forming the maximum of the recorded signal, for which τ_0 is characteristic of the elastic-scattering relaxation time.

It is for this reason that one must apply a certain degree of caution to the estimates given below of the diffusion length of NP relative to inelastic scattering

$$l = (D\tau^*)^{1/2} = \left(\frac{\overline{V}^2 \tau_0 \tau^*}{3}\right)^{1/2},$$

where $\overline{V}$ is the acoustic wave velocity averaged over all polarizations. If we use the values of τ_0 and τ^* from Fig. 2, then it turns out that l is greater than or comparable with the specimen length L. This confirms the fact that the group of NP in the specimens of Y–Lu garnets studied which form the maximum of the nonequilibrium phonon signal propagate diffusively without interacting with one another. Taking into account the fact that τ^* characterizes higher-frequency phonons than those which determine the maximum of the recorded signal (i.e., that in estimating the diffusion length we used a low value of τ^*), the condition for the existence of a NP diffusive regime is satisfied better under our experimental conditions.

The absolute values of τ_0 and τ^* obtained as functions of

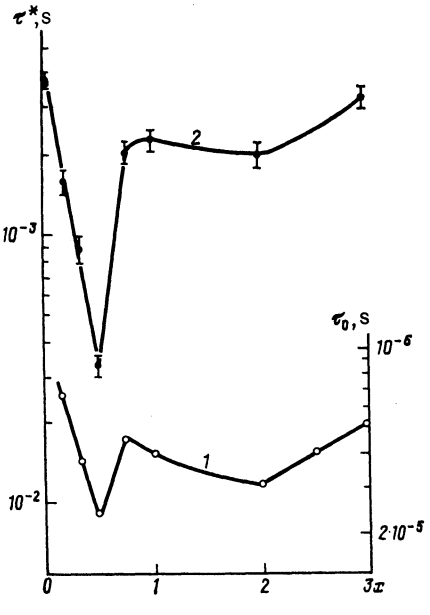

FIG. 2. Concentration dependence of the (*1*) elastic, τ_0, and (*2*) inelastic, τ^*, relaxation times for the interaction of NP in $Y_{3-x}Lu_xAl_5O_{12}$ solid solutions; $T_0 = 2.56$ K.

the composition of the Y–Lu solid solution and of the temperature (Figs. 1 and 2) allow us to estimate how likely a NP quasidiffusive regime is to occur,[1,4] i.e., the possibility of even one decay of a nonequilibrium phonon in the time required for them to diffuse through the specimen. This should be observed for $l < L$, which is really satisfied in the specimens studied at $T_H \geqslant 4$–5 K. Then τ^* becomes comparable with t_M and the influence of decay processes on the temporal position of the main part of the nonequilibrium phonon signal, its maximum.[2]

The results of a similar investigation in $BeAl_{2-x}Cr_xO_4$ solid solutions are shown in Fig. 3. It follows from Fig. 3 that the time τ^*, determined from analysis of the time dependence of the trailing edge of the nonequilibrium phonon pulse signal, depends nonmonotonically on the chromium impurity concentration.

In $BeAl_2O_4$ specimens subjected to γ-radiation up to doses of 10^7–10^8 rd defects are formed, produced by the displacement of the light Be atoms.[6] We have investigated such crystals and found that in them the time τ^* decreases after irradiation, which indicates the connection between intense inelastic phonon scattering processes and the number of defects in the crystal lattice.

The correlation of intense inelastic and elastic phonon scattering in crystals in the form of substitutional Y–Lu garnet solid solutions was first observed and to all appearances has a universal character. This effect was evidently observed in studies[7] of the kinetics of nonequilibrium phonons of a narrow spectral range in CaF_2:Zr by the method of optical generation and detection. A nonmonotonic dependence of the inelastic scattering relaxation time on the doping concentration was obtained.

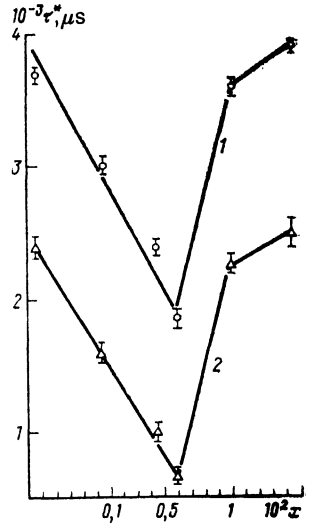

FIG. 3. Concentration dependence of the relaxation times for inelastic scattering in $BeAl_{2-x}Cr_xO_4$ solid solutions; *1*) $T_0 = 3$ K; *2*) 3.8 K.

DISCUSSION OF THE RESULTS; INVESTIGATION OF THE INFLUENCE OF HEAT TRANSFER PROCESSES ON THE TIME CHARACTERISTICS OF THE RECORDED SIGNAL

When discussing the results obtained in our experiments V. I. Kozub (A. F. Ioffe Physicotechnical Institute) suggested that the features of the behavior of the recorded signal (the arrival time of the nonequilibrium phono maximum and the time dependence of the trailing edge of the signal) can be determined to a certain extent by processes of heat transfer from the surface of the specimen studied.

We carefully analyzed a number of results of experiments described below to ascertain the influence of heat transfer processes on the formation of the heater temperature and on the form of the NP signals recorded by the bolometer. Our conclusions reached enabled us to exclude completely in the measurements the influence of heat transfer on the results given above, in particular by limiting the values of the power dissipated in the heater and also due to the corresponding choice of the temperature range of the thermostat.

At the basis of our method of NP generation[2] lies the well known means of heating a metal film heater by a current pulse. The film heater is prepared by thermal deposition of gold onto one end of the specimen studied; the recording of the nonequilibrium phonon signal is achieved at the opposite end of the specimen by a superconducting bolometer made of tin or indium film. Under the conditions of our experiment a noticeable change in heat transfer at the surface of the heater is possible became helium starts to boil as the thermal energy of the electric current dissipated in the metal film increases or when measurements are carried out in the region of the λ-point. We will analyze both these situations.

The total energy of the NP signal recorded by the bolometer (area under the curve) is shown in Fig. 4a as a function of the power P_H dissipated in the heater for a specimen in contact with the helium bath. A nearly linear dependence between the energy of the signal recorded by the bolometer and P_H is observed up to a power $P_H \leqslant 0.3$ W/mm^2,

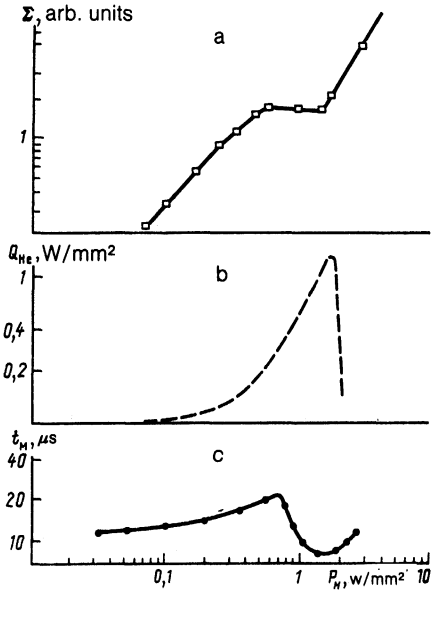

FIG. 4. a—Dependence of the total energy of the signal recorded by the bolometer (area under the curve) Σ on the power P_H dissipated in the film heater in a specimen of $Y_{2.65}Lu_{0.35}Al_5O_{12}$ for $L = 4.6$ mm, $T_0 = 3.75$ K; b—dependence of the thermal energy liberated from the film heater into the helium Q_{He} on the electrical power dissipated in it (according to Danil'chenko *et al.*[8]), c—dependence of the time of arrival of the signal maximum of nonequilibrium phonons, t_M, on the power dissipated in the heater P_H in a specimen of $Y_{2.65}Lu_{0.35}Al_5O_{12}$ for $L = 4.6$ mm, $T_0 = 3.75$ K.

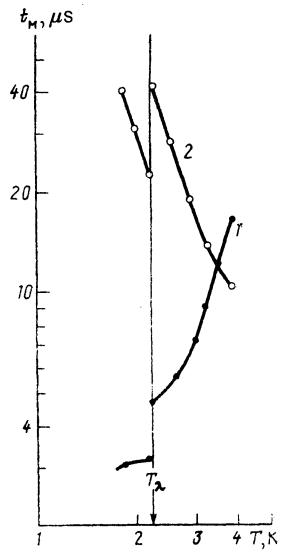

FIG. 5. Temperature dependence of the time of arrival of the maximum of the NP signal t_M in the specimens: *1*) $Y_{2.65}Lu_{0.35}Al_5O_{12}$ $L = 4.6$ mm; *2*) LiF; $L = 3$ mm (dislocation density 10^7 cm^{-2}), $P_H \leqslant 10^{-2}$ W/mm^2.

i.e., the heat removed from the film heater to the helium changes insignificantly as P_H increases. For $P_H > 0.5$ W/mm^2 the curve sharply flattens and is almost horizontal up to a value of $P_H \approx 1$–2 W/mm^2. In our opinion this is produced by intense heat removal from the metal film heater to the helium. A similar conclusion was drawn by Danil'chenko *et al.*[8] who showed that in the interval $P_H \approx 0.3$–2 W/mm^2 the energy flux to the helium grows, associated with its bubble boiling at the heater surface. This result[8] for experimental conditions close to ours is shown schematically in Fig. 4b.

The dependence of the arrival time of the NP signal maximum in a $Y_{2.65}Li_{0.35}Al_5O_{12}$ specimen is shown in Fig. 4c for the same thermostat temperature T_0 as for the dependence in Fig. 4a. It follows from a comparison of the dependence in Figs. 4a and 4c that the reduction in the arrival time of the NP signal maximum is related to the intense release of phonons from the film heater into liquid helium. In the power interval corresponding to the section of nonlinear $\Sigma(P_H)$ dependence (Fig. 4a) not only does the arrival time of the signal maximum then change (Fig. 4b), but also its shape (half width, trailing edge[2]), which can be associated with an appreciable change in the output coefficient of phonons or of separate spectral groups of phonons in the process of development of bubble boiling and the transition to film boiling of helium at the heater surface as its temperature rises (i.e., the power dissipated in the heater).

We have thus shown experimentally that the change in the conditions of heat transfer from the film heater to the helium as the power dissipated in it increases to $P_H > 0.1$ W/mm^2 for a given thermostat temperature T_0 above the λ-point leads to a change in the time characteristics and the shape of signal recorded for NP propagating through the specimen in the diffusive regime, which must be taken into account in analyzing the results of experiments on diffusive propagation of NP.

The sharp change in the conditions of heat transfer from the film heater into helium at the λ-point of helium ($T = 2.17$ K) also shows up in the characteristics of NP signals. The temperature dependence of the arrival time of the NP signal maximum $t_M(T)$ in specimens of $Y_{2.65}Lu_{0.35}Al_5O_{12}$ and LiF for $P_H = 10^{-2}$ W/mm^2 is shown in Fig. 5. The form of the $t_M(T)$ temperature dependence in these materials is different, but in both cases an increase in heat transfer at the λ-point reduces the arrival time of the NP signal maximum.

The shapes of the signals at temperatures above and below the λ-point are also appreciably different. Such results are shown in Fig. 6 for a LiF specimen ($P_H \leqslant 10^{-2}$ W/mm^2). For the signal obtained at $T_0 < T_\lambda$ the extended trailing edge characteristic for the diffusive regime of NP propagation in a defect crystal is practically absent. Such a signal shape below the λ temperature could also reflect not only the nature of the heat transfer from the film heater but also a change in the conditions for the emergence of phonons into the helium through the opposite end of the specimen where the bolometer is positioned. The sharp increase in the number of phonons passing through the solid-helium II boundary for phonons of frequency > 100 GHz was observed by Heim *et al.*[9] In our case of diffusive motion of phonons they may interact many times with the specimen surface in the vicinity of the bolometer, which can also contribute to their increased emergence into the helium.

These experimental results of the study of the influence

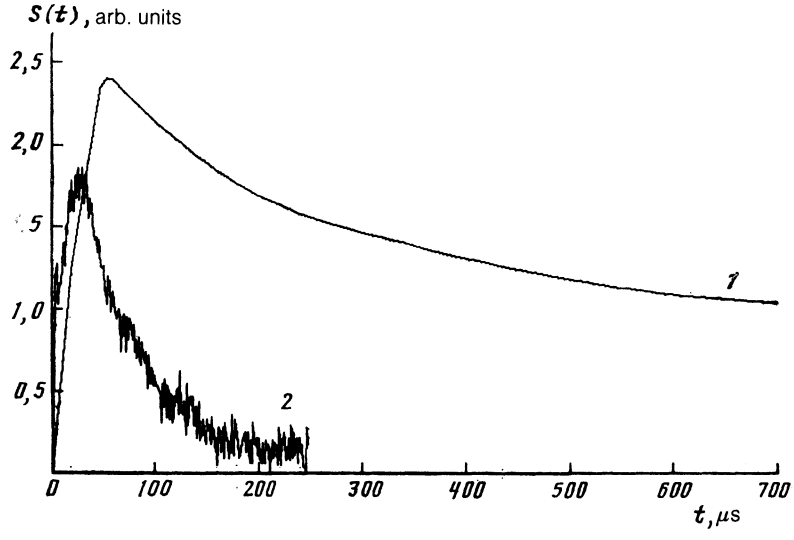

FIG. 6. Typical form of NP signals recorded by the bolometer in the region of the λ-point: *1*) $T_0 > T_\lambda$ ($T_0 = 2.2$ K); *2*) $T_0 < T_\lambda$ ($T_0 = 2.06$ K).

of heat transfer processes on the time characteristics of the NP signal recorded by the bolometer in pulse experiments show that obtaining reliable quantitative results, within the framework of the method used, is possible when heat transfer in the process of the experiments carried out does not change or can be taken into account in a proper manner. This requirement can be satisfied by limiting the temperature range of the thermostat temperatures studied to $T_0 > T_\lambda$ and the dynamic range of powers to $P_H \leqslant 10^{-2}$–10^{-1} W/mm^2, which we have done in the present work in carrying out experiments to study the kinetic characteristics of phonon scattering.

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[1)] We note that the determination of an accurate value of n for the $\tau^*(T)$ dependence on the basis of results of the form of Fig. 1b is complicated in view of the relative smoothness of the process by which the steepness increases and the comparatively narrow temperature range of the measurements, 2.2–3.8 K.

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