Nonlocal phonon heat conductivity

I. B. Levinson

L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR
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It is well known that in dielectrics where the scattering of phonons by static defects dominates there the heat conductivity coefficient is infinite due to the "runaway" of low-frequency phonons. We show that in actual fact the heat conductivity in such a situation has a nonlocal character, i.e., the energy flux at a given point is determined by the temperature distribution in the whole of space. Physically, the nonlocal effect is connected with the fact that the energy is transported by low-frequency subthermal phonons which have a diffusion length of the order of macroscopic dimensions.

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INTRODUCTION

In most dielectrics and semiconductors the main source for the scattering of phonons are static defects (isotopes, impurities). Therefore, if we exclude the case of very-low-frequency phonons for which the propagation proceeds ballistically, the phonon propagation mechanism is almost always diffusion. However, it is important to realize that phonon-phonon interactions can modify considerably the diffusion even if they occur more rarely than collisions with defects. The fact is that the diffusion coefficient depends strongly on the frequency: $D(\omega)\sim\omega^{-4}$, while the phonon-phonon processes can change the spectral composition of the phonon distribution; they can thus affect also the effective diffusion coefficient. Of course, for such an affect to occur the duration of the process must be longer than the characteristic time of the phonon-phonon interactions.

The propagation mechanism caused by the simultaneous action of scattering by defects and three-phonon anharmonicity was considered in Refs. 1 to 3, where it was assumed that the occupation numbers of nonequilibrium phonons were small: $n(\omega)\ll 1$, and hence that, of all the three-phonon processes, only decay processes were important. Another situation is also possible when $n(\omega)\approx 1$; in that case, besides the decay processes, the fusion processes are important. The simultaneous action of decays and fusion leads to the phonon distribution in a small region becoming Planckian, i.e., a local temperature $T(\mathbf{r}, t)$ is established. The aim of the present paper is to obtain an equation describing the propagation of the temperature. Such a problem is non-trivial and arises because the usual heat conduction equation for T does not exist in the situation considered (it is well known[4] that the thermal conductivity coefficient becomes infinite when the dominating scattering is by static defects).

1. STATEMENT OF THE PROBLEM

To elucidate the physical reasons why it is impossible to write down a heat conduction equation when scattering by defects dominates it is instructive to try to derive this equation.

The diffusion equation for the occupation numbers, taking anharmonic processes into account, has the form

$$[\partial/\partial t - D(\omega)\nabla^2]n(\omega, \mathbf{r}, t) = S\{n(\omega, \mathbf{r}, t)\}, \tag{1}$$

where on the right-hand side we have the collisional term for anharmonic processes. We assume that the occupation numbers $n(\omega)$ of all phonon branches are the same for a given frequency ω, since not only the directions of the phonon motion but also their polarizations are mixed in the scattering by defects. Correspondingly, $D(\omega)$ is some average over directions and polarizations.

It is natural to substitute into (1) the Planck distribution

$$n(\omega, \mathbf{r}, t) = n(T(\mathbf{r}, t)\,|\,\omega),\quad n(T|\omega) \equiv (e^{\omega/T}-1)^{-1}, \tag{2}$$

and to set up the energy balance, assuming $\int d\omega\rho(\omega)\omega\ldots$, where $\rho(\omega)\propto\omega^2$ is the density of states (the total one, for three branches). The right-hand side then vanishes (since the total energy is conserved in phonon-phonon processes) and we get the equation

$$\partial\varepsilon/\partial t + \operatorname{div}\mathbf{q} = 0, \tag{3}$$

where the energy density is

$$\varepsilon(T) = \int_0^\infty d\omega\,\rho(\omega)\omega n(T|\omega) = \frac{\pi^4}{15}\rho(T)T^3 \sim T^4 \tag{4}$$

and the energy flux

$$\mathbf{q} = \nabla\mu,\quad \mu(T) = \int_0^\infty d\omega\,\rho(\omega)\omega D(\omega) n(T|\omega). \tag{5}$$

Formally we have, indeed, obtained an equation for T. However, one sees easily that if $D(\omega)\propto\omega^{-4}$ the integrand in the integral for μ behaves as ω^{-2}, i.e., the integral diverges. This means that, on the one hand, although almost all the phonon energy is concentrated in the region of frequencies $\omega\approx T$, the energy flux is transported by low-frequency phonons $\omega\ll T$; on the other hand, in the region of the low frequencies which transport the energy the distribution is non-Planckian.

We must therefore evaluate the flux $\mathbf{q}$ differently. Let $\bar{\omega}$ be that limiting frequency above which (when $\omega\gg\bar{\omega}$) the distribution is quasi-equilibrium, i.e., Planckian. We assume that this distribution, i.e., in fact $T(\mathbf{r}, t)$, is known and use it to find the distribution

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$n(\omega, \mathbf{r}, t)$ in the range $\omega \lesssim \bar{\omega}$. Afterwards we use the distribution thus found to evaluate the flux:

$$\mathbf{q}(\mathbf{r}, t) = \int_0^\infty d\omega\, \rho(\omega)\omega[-D(\omega)\nabla n(\omega, \mathbf{r}, t)]. \tag{6}$$

As a result we find the flux as a functional of the temperature: substituting $\mathbf{q}\{T\}$ into (3) we find an equation for T. However, we shall see that the relation between $\mathbf{q}$ and T is nonlocal in character, i.e., the flux at a given point depends on the temperature in the whole region where $T \neq 0$. Therefore, in contrast to the usual heat conduction equation, which is a differential equation, the equation we get for T will be an integro-differential equation.

The situation described here is still more critical than in the nonlocal phonon hydrodynamics where both the energy and its flux are determined by phonons with $\omega \approx T$ and only the momentum flux, i.e., the viscosity, is determined by phonons with $\omega \ll T$.[5]

2. COLLISIONAL TERM IN THE "SUB-THERMAL" REGION OF THE SPECTRUM

To obtain the equation for the distribution function in the low-frequency region we consider first the collisional term which, for normal three-phonon processes has the following form:

$$S\{n(\omega)\} = {}^1/_2 \int\int d\omega'\, d\omega''\, \rho'\rho''\delta(\omega-\omega'-\omega'')A(\omega; \omega', \omega'')[-n(n'+n''+1) + n'n''] + \int d\omega' \int d\omega''\, \rho'\rho''\delta(\omega+\omega'-\omega'')A(\omega''; \omega, \omega')[n''(n+n'+1)-nn']. \tag{7}$$

We have written here for simplicity $n = n(\omega), n' = n(\omega')$, and so on. The factor A arises after averaging the square of the matrix element (together with the delta-functions expressing the momentum conservation law) over angles and polarizations. It is clear that

$$A(\omega; \omega', \omega'') = A(\omega; \omega'', \omega'). \tag{8}$$

For frequencies below the Debye frequency ω_D we have

$$A(\lambda\omega; \lambda\omega', \lambda\omega'') = A(\omega; \omega', \omega''), \tag{9}$$

which together with the conservation law $\omega = \omega' + \omega''$ means that A depends only on the ratio ω'/ω''.

In the situation of interest to us $n(\omega)$ is the same as $n(T|\omega)$ for $\omega \gg \bar{\omega}$ (while $\bar{\omega} \ll T$) and is small in the region of phonons with $\omega \lesssim \bar{\omega}$. In that case Eq. (7) simplifies considerably in the "sub-thermal" region $\omega \ll T$. In the second term in (7) in the important region of integration the phonons ω' and ω'' are "thermal," i.e., $\omega' \approx \omega'' \approx T$; at the same time, in the first term the phonons ω' and ω'' are "subthermal," i.e., $\omega' \approx \omega'' \approx \omega \ll T$. The first term is thus small as far as phase volume is concerned and we can drop it. In the second term we replace n' and n'' by the Planck occupation numbers and expand in $\omega'' - \omega' = \omega$. As a result we get

$$S\{n(\omega)\} = -n(\omega)/\hat{\tau}(T, \omega) + w(T), \tag{10}$$

where

$$w(T) = \frac{4\pi^4}{15} A_0 \rho(T)^2 T = \frac{1}{\tau(T)} \sim T^5, \tag{11}$$

$$\frac{1}{\hat{\tau}(T, \omega)} = \frac{4\pi^4}{15} A_0 \rho(T)^2 \omega = w(T)\frac{\omega}{T} \sim T^4\omega, \tag{12}$$

and A_0 is the value of $A(\omega; \omega', \omega'')$ when $\omega'/\omega'' = 0$. For comparison we write down again the time for the spontaneous decay of a phonon of frequency ω:

$$1/\tau(\omega) = {}^1/_{60} A_1 \rho(\omega)^2 \omega \sim \omega^5. \tag{13}$$

Here A_1 is some average of $A(\omega; \omega', \omega'')$ in the region $\omega'/\omega'' \approx 1$. Since usually A_0 and A_1 are of the same order of magnitude, $\tau(T)$ is of the order of the time for spontaneous decay for $\omega \approx T$. We can thus assume $\tau(T)$ to be the time required to establish the Planck equilibrium at the level T. It is clear from (10) and (11), (12) that the collisional term vanishes when we substitute the Planck distribution $n(\omega) = T/\omega$, and thus $\hat{\tau}(T, \omega)$ is the time required to establish quasi-equilibrium at the level ω if at the level T quasi-equilibrium is already established. The lower the level ω the slower quasi-equilibrium is established there.

It is expedient to elucidate for what follows how fast the energy is transferred from the level T to the lower levels. The rate of energy transfer to a level below ω is

$$\dot{\varepsilon}(T \to \omega) = \int_0^\omega d\omega'\, \rho(\omega')\omega' w(T) = {}^1/_4 w(T)\rho(\omega)\omega^2. \tag{14}$$

In fact, the whole of the energy enters in a section of the spectrum $\omega' \approx \omega$, since frequencies $\omega' \ll \omega$ contribute little to the integral. The reciprocal time for transferring all the energy from the level T to the level ω is

$$\frac{1}{\bar{\tau}(T, \omega)} = \frac{\dot{\varepsilon}(T \to \omega)}{\varepsilon(T)} = \frac{4\pi^4}{15} w(T)\left(\frac{\omega}{T}\right)^4 \sim T\omega^4. \tag{15}$$

It is important to note that this time is longer than the time needed to establish quasi-equilibrium at the level ω.

3. DISTRIBUTION IN THE NON EQUILIBRIUM REGION OF THE SPECTRUM AND EVALUATION OF THE FLUX

We now write Eq. (1) in the low-frequency region $\omega \ll T$ as follows:

$$\left[\frac{\partial}{\partial t} - D(\omega)\nabla^2 + \frac{1}{\hat{\tau}(T, \omega)}\right] n(\omega, \mathbf{r}, t) = w(T). \tag{16}$$

Here T is a function of $\mathbf{r}$ and t with large characteristic variation scales $\bar{r}$ and $\bar{t}$

$$\bar{t} \gg \tau(T), \tag{17a}$$

$$\bar{r} \gg l(T) = [D(T)\tau(T)]^{1/2}; \tag{17b}$$

on the right-hand sides of the inequalities we have the time and length for establishing quasi-equilibrium at the level T. Considering T to be a given function of $\mathbf{r}$ and t, we must find $n(\omega, \mathbf{r}, t)$. In the present paper we restrict ourselves to a study of the initial temperature distribution in an infinite medium. In such a situation

we have in the low-frequency region

$$n(\omega, \mathbf{r}, t)=n(T_0|\omega) \text{ when } \mathbf{r}\to\infty,\ t=0, \tag{18}$$

where T_0 is the temperature of the heat bath.

Unfortunately it is impossible to obtain from Eq. (16) a general expression for n, because $\hat{\tau}$ depends (through T) on $\mathbf{r}$. Therefore we limit ourselves to begin with to the linear case of a weak initial temperature perturbation

$$|T(\mathbf{r}, 0)-T_0|\ll T_0 \tag{19}$$

and we put

$$T=T_0+\delta T,\quad n(\omega)=n(T_0|\omega)+\delta n(\omega). \tag{20}$$

Linearizing (16) with respect to δT and δn and limiting ourselves for the sake of simplicity to the one-dimensional case we get

$$\left[\frac{\partial}{\partial t}-D(\omega)\frac{\partial^2}{\partial z^2}+\frac{1}{\hat{\tau}_0(\omega)}\right]\delta n(\omega, z, t)=\theta w_0. \tag{21}$$

Here $\theta=\delta T/T_0$ and the subscript 0 indicates that a quantity is evaluated for $T=T_0$. Changing the dimensionless variables

$$\sigma=\omega/T_0,\quad \zeta=z/l_0,\quad s=tw_0, \tag{22}$$

we find the distribution

$$\delta n(\sigma, \zeta, s)=\int_0^s ds'\int_{-\infty}^{+\infty} d\zeta'\,\theta(\zeta', s')$$
$$\times(4\pi)^{-1/2}\sigma^2(s-s')^{-1/2}\exp\left\{-(s-s')\sigma-\frac{(\zeta-\zeta')^2}{4(s-s')}\sigma^4\right\} \tag{23}$$

and we evaluate the flux

$$q=q_0\int_0^s ds'\int_{-\infty}^{+\infty} d\zeta'\,\theta(\zeta', s')Q(\zeta-\zeta', s-s'). \tag{24}$$

Here

$$q_0=4\varepsilon_0 D_0/l_0, \tag{25}$$

$$Q(\zeta, s)=\frac{15}{16\pi^{5/4}}s^{-3/4}\zeta\int_0^\infty\frac{d\sigma}{\sigma}\sigma^6\exp\left\{-s\sigma-\frac{\zeta^2}{4s}\sigma^4\right\}. \tag{26}$$

The kernel Q is odd in ζ and has the following asymptotic behavior:

$$\begin{aligned} &Q(\zeta, s)\approx\zeta s^{-15/4},\quad \sigma\approx s^{-1}\quad (s^5\gg\zeta^2),\\ &Q(\zeta, s)\approx\zeta^{-2}\operatorname{sign}\zeta,\quad \sigma\approx s^{1/4}|\zeta|^{-1/2}\quad (s^5\ll\zeta^2). \end{aligned} \tag{27}$$

We indicate also those σ which are important in the integral of (26) at the given s and ζ.

4. EQUATION FOR THE TEMPERATURE

Noting that $\delta\varepsilon=4\varepsilon_0\theta$ and substituting (24) into (3) we find an equation for the temperature

$$\frac{\partial}{\partial s}\theta(\zeta, s)+\frac{\partial}{\partial\zeta}\int_0^s ds'\int_{-\infty}^{+\infty} d\zeta'\,\theta(\zeta', s')Q(\zeta-\zeta', s-s')=0. \tag{28}$$

This equation can be further simplified. This is most easily accomplished through a Laplace transformation $s\to p$ and a Fourier transformation $\zeta\to k$. The transform of the kernel is then $Q\to -iM$ where

$$M(k, p)=2\int_0^\infty d\zeta\sin k\zeta\int_0^\infty ds\,e^{-ps}Q(\zeta, s). \tag{29}$$

After the transformation, Eq. (28) can be solved. The transform of the solution is

$$\theta(k, p)=\theta(k)G(k, p), \tag{30}$$

where the transform of the Green function of Eq. (28) is

$$G(k, p)=[p+kM(k, p)]^{-1}, \tag{31}$$

and $\theta(k)$ is the Fourier transform of the initial distribution $\theta(\zeta, s=0)$. The characteristic values of k are small, since they are of the order of $\bar{\zeta}^{-1}$, where $\bar{\zeta}$ is, for instance, the width L of the initial distribution of $\delta T(z, t=0)$, measured in units l_0.

We now consider the singularities of G in p for fixed small k. We note first that

$$M(k, p)=k^{3/5}H(pk^{-4/5}), \tag{32}$$

where

$$H(w)=\frac{15}{4\pi^4}\int_0\frac{dx}{x}\frac{1}{w+\varphi(x)},\quad \varphi(x)=x+x^{-4}. \tag{33}$$

The function $H(w)$ is analytic in the w-plane with a cut $(-a, -\infty)$, where $a=5\times 2^{-8/5}$ (see the Appendix). Moreover

$$H(0)=(3/4\pi^3)\operatorname{cosec}\pi/5=c=0.04115\ldots, \tag{34}$$

$$H(w)=(75/16\pi^4)\ln w/w,\quad |w|\to\infty. \tag{35}$$

Using these properties of H and the fact that k is small, we can verify that the denominator of G vanishes only in the point $p=-ck^{8/5}$, where G has a first-order pole with residue 1. Furthermore, G has singularities at the cut $(-ak^{2/5}, -\infty)$, but one can show (see the Appendix) that the contribution from the cut is small for small k. We can thus assume that

$$G(k, p)=[p+ck^{8/5}]^{-1}. \tag{36}$$

This is equivalent to replacing $M(k,p)$ by $M(k,0)$, i.e., the kernel $Q(\zeta, s)$ by $K(\zeta)\delta(s)$, where

$$K(\zeta)=\int_0^\infty ds\,Q(\zeta, s)=c_K|\zeta|^{-6/5}\operatorname{sign}\zeta,\quad c_K=\frac{9}{20\pi^4}\Gamma\left(\frac{3}{5}\right). \tag{37}$$

The flux can thus be written as follows:

$$q(\zeta, s)=q_0\int_{-\infty}^{+\infty} d\zeta'\,\theta(\zeta', s)K(\zeta-\zeta'). \tag{38}$$

The nonlocal nature of the connection between the flux and the temperature is now clear. The kernel K decreases so slowly that its second moment diverges and values of $|\zeta-\zeta'|$ of the order of $\bar{\zeta}$ are important in the integral (38). This means that the flux in each point depends on the temperature distribution in the whole diffusion region. At the same time the fact that there is no integration over time in (38) means that the flux

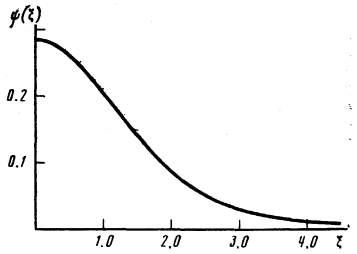

FIG. 1.

is established fast, i.e., the process is quasi-stationary.

Turning to the coordinate-time representation we get from (36) the temperature distribution for an initial point perturbation

$$G(\zeta, s)=\frac{1}{\pi}\int_0^\infty dk\cos k\zeta\, e^{-csk^{8/5}}=s^{-3/5}\Psi(\zeta s^{-8/5}), \tag{39}$$

where

$$\Psi(\xi)=\frac{5}{8\pi}\int_0^\infty \frac{dy}{y}\, y^{3/5}\cos(\xi y^{3/5})\, e^{-cy}. \tag{40}$$

For comparison we write down the function G for normal diffusion

$$G(\zeta, s)=s^{-1/2}\Psi_0(\zeta s^{-1/2}),\quad \Psi_0(\xi)=(4\pi)^{-1/2}e^{-1/4\xi^2}. \tag{41}$$

The function $\Psi(\xi)$ is positive, decreases monotonically with increasing argument, and has the following properties:

$$\Psi(0)=c_1=\frac{1}{\pi}\Gamma\left(\frac{13}{8}\right)c^{-3/5}, \tag{42}$$

as $\xi\to\infty$, we have

$$\Psi(\xi)=c_2\xi^{-13/3},\quad c_2=\frac{1}{\pi}\cos\frac{3\pi}{10}\Gamma\left(\frac{13}{5}\right)c, \tag{43}$$

$$\int_0^\infty d\xi\,\Psi(\xi)=1/2. \tag{44}$$

The function $\Psi(\xi)$ is inconvenient for numerical tabulation, as it contains the small numerical parameter c. Splitting it off we write

$$\Psi(\xi)=c'\psi(c'\xi),\quad c'=c^{-5/8}=7.346\ldots$$

$$\psi(\xi)=\frac{1}{\pi}\int_0^\infty dy\cos\xi y e^{-y^{8/5}}.$$

We give in Fig. 1 the function $\psi(\xi)$.

5. DISCUSSION OF THE RESULTS

A formal comparison of the nonlocal and the local diffusion, i.e., a comparison of (30) and (41), shows that when there is nonlocal diffusion the scale of the spatial distribution varies like $t^{5/8}$ instead of like $t^{1/2}$ for normal diffusion. This difference is not very large; more important is that when the diffusion is nonlocal ahead of the front the temperature drops as a power, as $z^{-13/5}$, instead of exponentially as in local diffusion.

The temporal and spatial scales of nonlocal diffusion are connected by the relation $s\approx\zeta^{8/5}$. This means, for instance, that if the temperature distribution initially occupies a region of width ζ, the time after which that region is appreciably broadened is $s\approx\zeta^{8/5}$.

To visualize a picture of the processes which take place in nonlocal diffusion we must ascertain the frequencies at which the phonons transfer the energy. When evaluating the integral (37) the important value is $s\approx\zeta^{2/5}$, as is readily checked by using the asymptotic expression (27). Hence it follows that $\sigma\approx\zeta^{-2/5}$ is important in the integral (26). This means that the energy flux q is transferred by phonons of frequency

$$\bar\omega\approx T_0(L/l_0)^{-3/5}. \tag{45}$$

One can check that this is just the frequency below which the distribution (23) ceases to be in quasi-equilibrium, i.e., $\delta n\neq\delta T/\omega$. Equation (45) can be rewritten as

$$D(\bar\omega)\,\hat\tau_0(\bar\omega)\approx L^2, \tag{46}$$

i.e., the energy is transported by those phonons for which the diffusion length is of the order of macroscopic dimensions after a time needed for their absorption by thermal phonons. In other words, below $\bar\omega$ quasi-equilibrium is violated because the phonons diffuse away from the excited region. We note that a different situation is also *a priori* possible, namely is $\hat\tau_0(\bar\omega)\approx t$, which would mean that quasi-equilibrium cannot be established during the time of the process.

One sees easily that the characteristic time for the process

$$t\approx\tau_0(L/l_0)^{3/5} \tag{47}$$

is of the order of the time needed to transfer energy from the level T to the level $\bar\omega$:

$$t\approx\bar\tau_0(\bar\omega). \tag{48}$$

This time is longer than the time $\hat\tau_0(\bar\omega)$ needed to establish equilibrium at the level $\bar\omega$. This is just the reason why the process is quasi-stationary. The range of the kernel $Q(\zeta, s)$ in s is of the order $\zeta^{-2/5}$, which in dimensional units is just $\hat\tau_0(\bar\omega)$.

On the whole the spatial energy transport in nonlocal diffusion must be the following. Let some phonons be excited in some region of space. In the spectral region $\omega\gg\bar\omega$ a quasi-equilibrium distribution is established, but phonons with $\omega\lesssim\bar\omega$ leave the spatial region of the excitation rapidly. In the excitation region there occurs thus a Planck distribution which is depleted at $\omega\lesssim\bar\omega$. Phonon-phonon processes tend to restore the complete Planck distribution, and as a result there occurs in the excitation region an energy flux downwards along the spectrum: from the level $\omega\approx T$ to the level $\omega\approx\bar\omega$. Phonons with $\omega\lesssim\bar\omega$ transfer energy into the originally unexcited region of space where the spectral region $\omega\lesssim\bar\omega$ turns out to be, in contrast, enriched. Thanks to phonon-phonon processes there arises here an energy flux upwards along the spectrum: from the level

$\omega \approx \bar{\omega}$ to the level $\omega \approx T$.

All this means that the temperature propagation process from a point z_1 to a point z_2 proceeds so to speak in three stages: 1) an energy transport downwards along the spectrum in the point z_1 from the level T to the level $\bar{\omega}$; 2) spatial energy transport from the point z_1 to the point z_2 through diffusion at the level $\bar{\omega}$; 3) energy transport upwards along the spectrum in the point z_2 from the level $\bar{\omega}$ to the level T. The diffusion time is $L^2/D(\bar{\omega}) = \hat{\tau}_0(\bar{\omega})$. It is shorter than the time for the spectral transfer $\bar{\tau}_0(\bar{\omega})$. The bottleneck for the process is thus the spectral transfer.

6. NONLINEAR CASE

It is important to emphasize that all qualitative conclusions and order-of-magnitude estimates are valid for any geometry also for the nonlinear case, for instance, when in some region of the crystal there is an initial temperature T much higher than the bath temperature T_0, which we can take to be $T_0 = 0$ in such a situation.

To verify this we estimate first the phonon occupation numbers. The condition for quasi-equilibrium is clearly

$$\hat{\tau}(T, \omega) \ll \min\{t, L^2/D(\omega)\}. \tag{49}$$

The solution of this inequality gives $\omega \ll \bar{\omega}$ where $\bar{\omega}$ is the level at which the quasi-equilibrium is established. We introduce the dimensionless variables

$$s = t/\tau(T), \quad \zeta = L/l(T), \quad \sigma = \omega/T, \quad \bar{\sigma} = \bar{\omega}/T. \tag{50}$$

The resultant expressions for $\bar{\omega}$ can be different, depending on the relation between s^5 and ζ^2. In the quasi-uniform case ($s^5 \ll \zeta^2$) we have

$$\hat{\tau}(T, \bar{\omega}) \approx t, \quad \text{or} \quad \bar{\sigma} \approx s^{-1}, \tag{51}$$

i.e., it is not possible to establish equilibrium below $\bar{\omega}$ during the time of the process. In the quasi-stationary case ($s^5 \gg \zeta^2$) we have

$$\hat{\tau}(T, \bar{\omega}) \approx L^2/D(\bar{\omega}), \quad \text{or} \quad \bar{\sigma} \approx \zeta^{-2/5}, \tag{52}$$

i.e., equilibrium below $\bar{\omega}$ is prevented from being established by the diffusive departure from the excited region.

Thus, for $\omega \gg \bar{\omega}$ we have $n = n(T|\omega)$. For $\omega \lesssim \bar{\omega}$, as to order of magnitude,

$$n(\omega) \approx w(T) \min\{t, L^2/D(\omega)\}. \tag{53}$$

Hence we get

$$n \approx \zeta^2 \sigma^4, \quad \sigma^4 \ll s\zeta^{-2}; \tag{54}$$

$$n \approx \zeta^2 s^{-1}, \quad \sigma^4 \gg s\zeta^{-2}. \tag{55}$$

One sees easily that these occupation numbers are much *smaller* than the quasi-equilibrium ones $n = \sigma^{-1}$. As $\omega \to 0$ we have $n(\omega) \propto \omega^4$ so that the flux (6) is finite.

We now estimate the flux, writing $q = q_1 + q_2$, where q_1 and q_2 are the fluxes transported by phonons with $\omega < \bar{\omega}$ and $\omega > \bar{\omega}$, respectively. To estimate q_2 we can assume the occupation numbers to be quasi-equilibrium ones, so that

$$q_2 \approx L^{-1} \bar{\omega}^3 \rho(\bar{\omega}) D(\bar{\omega}) (T/\bar{\omega}). \tag{56}$$

For q_1 the following estimate holds

$$q_1 \approx L^{-1} \int_0^{\bar{\omega}} d\omega\, \rho(\omega) D(\omega) n(\omega), \tag{57}$$

where the occupation numbers $n(\omega)$ must be taken from (54) and (55). Let the situation be quasi-stationary. In that case $s\zeta^{-2} \gg \bar{\sigma}$ and $n(\omega)$ is given by Eq. (54), and $\bar{\omega}$ by Eq. (52). Estimating the integral (57) and expression (56) we find

$$q_1 \approx q_2 \approx q \approx q(T) \zeta^{-3/5}, \tag{58}$$

where $q(T)$ is obtained from q_0 by the substitution $T_0 \to T$. Substituting the flux (58) in the energy conservation law (3) written in the form

$$\varepsilon(T)/t \approx q/L, \tag{59}$$

we find $s \approx \zeta^{8/5}$ which agrees with our quasi-stationarity assumption. If we assume that the situation is quasi-uniform, we have $s\zeta^{-2} \ll \bar{\sigma}$ and in the $n(\omega)$ distribution there are below $\bar{\omega}$ two regions corresponding to (54) and (55). The contribution to the integral (57) comes from the high-frequency range and

$$q_1 \approx q(T)\, s \ln(s^{-1}\zeta^{3/5}). \tag{60}$$

Substituting $\bar{\omega}$ from (51) into (56) we have

$$q_2 \approx q(T) s \zeta^{-1} \ll q_1. \tag{61}$$

We now get from (59) $s \approx \zeta (\ln \zeta)^{-1/2}$ which, however, does not agree with our assumption about quasi-uniformity.

In the nonlinear case, as in the linear one, the nonlocal temperature transfer is thus quasi-stationary. The energy flux

$$q \approx q(T)[L/l(T)]^{-3/5} \tag{62}$$

is transported by phonons of frequency $\bar{\omega} = T[L/l(T)]^{-2/5}$, for which the diffusion length during the time to establish quasi-equilibrium $[D(\bar{\omega})\hat{\tau}(T, \bar{\omega})]^{1/2}$ is of the order of the characteristic dimensions L. The duration of the process

$$t \approx \tau(T)[L/l(T)]^{3/5} \tag{63}$$

is of the order of the time for energy transfer from the level of frequencies $\omega \approx T$ to the level $\omega \approx \bar{\omega}$, i.e., of the order of $\bar{\tau}(T, \bar{\omega})$.

The considerations given here allow us to answer semi-quantitatively a number of questions which arise in processes of strongly nonlinear phonon energy transport in crystals at low bath temperatures. We consider, for instance, a conducting film on the surface of a dielectric (or a doped surface layer of a high-resistance semiconductor), heated by a current pulse to a temperature T much higher than the bath temperature. What is the time t needed for the film to cool off? We write down the energy balance

$$E(t)\approx qt, \tag{64}$$

where $E(T)$ is the energy of the film per cm² (due to phonons and electrons). Substituting here q from (62) and using (63) to eliminate L we find

$$t\approx\tau(T)\,[E(T)/\varepsilon(T)l(T)]^{1/4}. \tag{65}$$

The meaning of L in this problem becomes clear if we multiply (62) and (63) and substitute into (64). We then get

$$E(T)\approx L\varepsilon(T), \tag{66}$$

i.e., L is the thickness of the substrate, which is heated until the time the film cools off.

7. CRITERIA

In conclusion we indicate exactly in which region the nonlocal heat conduction mechanism which we have discussed operates. We recall first that by assumption the crystal is governed by defects rather than by anharmonicity, i.e.,

$$\delta=\tau(T)/\tau_I(T)\gg 1, \tag{67}$$

where $\tau_I(\omega)$ is the time for scattering by defects, in terms of which we can express the diffusion coefficient

$$D(\omega)=\tfrac{1}{3}v^2\tau_I(\omega); \tag{68}$$

v is some average sound velocity. Moreover, since the energy flux transported by subthermal phonons with frequency $\bar\omega$ was evaluated using the diffusion Eq. (6), we must require that the diffusion equation (1) be valid in the region of frequencies $\bar\omega$, i.e., that we have

$$v\tau_I(\bar\omega)\ll L. \tag{69}$$

Using (46) we see easily that (69) is equivalent to the inequality

$$\tau_I(\bar\omega)\ll\hat\tau(\bar\omega), \tag{70}$$

i.e., a phonon $\bar\omega$ is scattered many times by defects before it is absorbed by quasi-equilibrium phonons. Substituting into (69) the frequency $\bar\omega$ from (45) we find an upper bound for L; a lower bound follows from (17b). We can write these limitations as follows:

$$\delta^{-1/2}\ll L/v\tau(T)\ll\delta^{1/3}. \tag{71}$$

It is very instructive to consider the location of the region (71) on a plot where the abscissa is the defect content δ and the ordinate the length L measured in mean free path lengths $v\tau(T)$: see Fig. 2. Above the thick line the duration t of the processes is longer than $\tau(T)$ and here it is possible to establish a local temperature $T(\mathbf{r},t)$ so that one can speak of "heat" transfer. This includes the second sound (SS) region, the local heat conduction (LHC1) region determined by defects, the (shaded) region of nonlocal heat conduction, discussed by us, and the region (LHC2) to be discussed below. Under the thick line the anharmonic processes are unimportant, there is no local temperature, and phonons of different frequencies propagate independently. This includes the regions of ballistic propagation (B) and of diffusive propagation (D).

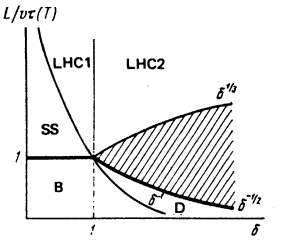

FIG. 2. Regions in which different mechanisms for phonon non-equilibrium transfer are realized.

Inequality (70) is violated in region LHC2, i.e., the phonons which transport the energy do not manage to be scattered by defects and therefore the diffusion approximation is not valid for those phonons. In the region LHC2 the phonons transporting the energy propagate ballistically. A detailed analysis, which will be published separately, shows that there occurs then a local thermal conductivity with a heat conduction coefficient which is determined not only by scattering by defects, but also by anharmonic processes.

Nonlocal heat conduction can be realized in InSb for $T=10$ K in specimens of a few mm size. We shall assume first that the only defects are isotopes. In that case

$$1/\tau_I(\omega)=1.9\cdot10^{10}\ \text{sec}^{-1}\cdot x^4,\quad x=\omega/\omega_D,$$

where the Debye frequency $\omega_D=2.63\times10^{13}$ sec^{-1} $=200$ K. To estimate the anharmonic times we use the relaxation times found from the heat conduction[6] substituting in them $\hbar\omega/2.8$ for kT. We then find

$$1/\tau(\omega)=7.4\cdot10^9\ \text{sec}^{-1}\cdot x^3\ (TA\text{-phonons}),$$
$$1/\tau(\omega)=2.2\cdot10^9\ \text{sec}^{-1}\cdot x^3\ (LA\text{-phonons}).$$

One can show that the averaging over the polarization is with a weight v^{-3}, where v is the sound speed of the appropriate branch. This gives for the average over the branches ($v_T=1.8\times10^5$ cm/sec; $v_L=3.1\times10^5$ cm/sec)

$$1/\tau(\omega)=7\cdot10^9\ \text{sec}^{-1}\cdot x^3.$$

For thermal phonons with $\omega=28$ K we have $\tau_I(T)=1.4\times10^{-7}$ sec, $\tau(T)=2.7\times10^{-6}$ sec at $T=10$ K, i.e., $\delta=19$. Using the average velocity $v=2\times10^5$ cm/sec we find the limits of L which for a given δ bound the

regions of the nonlocal heat conduction: $L_{max}=1.4$ cm and $L_{min}=0.12$ cm. It is clear that if we take other defects into account the region where nonlocal heat conduction occurs widens. The scattering of phonons by free electrons is negligible under the conditions considered for pure samples with $n \lesssim 10^{15}$ cm^{-3}.

Finally, the divergence of the integral μ in (5) could be removed by taking umklapp processes into account. In umklapp processes, apart from the low-frequency phonon ω, two phonons ω' and ω'' which lie near the Brillouin-zone boundary take part: $\omega+\omega' \to \omega''$. The relaxation time $\tau_U(\omega)$, when umklapp processes are taken into account, depends on a number of factors, on whether the temperature of the phonons ω' and ω'' is T or T_0, whether the condition $\tau(\omega) \gg \tau_I(\omega)$ is satisfied for these phonons, or whether the energy of ω' small or large compared to T_0. One can show, however, that in all those cases the time $\tau_U(\omega)$ increases not faster than ω^{-1} as $\omega \to 0$. Therefore, at sufficiently low frequencies just that time will determine the diffusion coefficient, and the integral μ will be finite. Neglecting umklapp processes we assume that $\tau_U(\bar{\omega}) \gg \tau_I(\bar{\omega})$ which, of course, imposes some upper bound on L. However, this limitation is not very stringent because as $\tau_U(\omega)$ contains at low temperatures an exponentially large factor $\exp(\omega_D'/T)$, where ω_D' is of the order of the Debye frequency.

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APPENDIX

1. The asymptotic behavior of $H(w)$ at large w can be found by splitting the integral with respect to x into two integrals: from 0 to 1 and from 1 to ∞. In the first integral the value $x \ll 1$ is important and we can put $\varphi(x)=x^{-4}$, in the second $x \gg 1$ is important and we can put $\varphi(x)=x$. Singularities of $H(w)$ occur at those w for which the equation $w+\varphi(x)=0$ has a solution $x>0$. It is clear that such solutions exist only if $w<0$ and $|w|>\min\varphi(x) \equiv a$. Therefore $H(w)$ is defined in the w-plane with the cut $(-a, -\infty)$.

2. To evaluate the contribution to $G(k, s)$ from the cut we change from integration with respect to p to integration with respect to $w=pk^{-2/5}$. This contribution is then

$$G'(k, s)=ck^{6/5}\frac{1}{2\pi i}\int_{-\infty}^{-a}\frac{du}{u^2}e^{usk^{2/5}}[F_+(u)-F_-(u)], \tag{72}$$

where the values on the edges of the cut are

$$F_\pm(u)=F(u\pm i\varepsilon), \quad \varepsilon \to +0. \tag{73}$$

One sees easily that

$$F_+(u)-F_-(u)=\int_0^\infty \frac{dx}{x}\frac{-2i\varepsilon}{[\varphi(x)+u]^2+\varepsilon^2}$$
$$=-2\pi i\int_0^\infty \frac{dx}{x}\delta[\varphi(x)+u]=-2\pi i\chi(u), \tag{74}$$

where

$$\chi(u)=|x_1(u)-4x_1(u)^{-4}|^{-1}+|x_2(u)-4x_2(u)^{-4}|^{-1}, \tag{75}$$

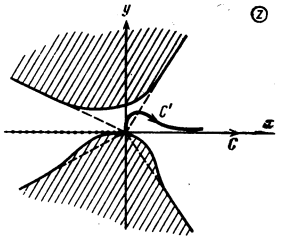

FIG. 3. Complex z-plane. We have shaded the regions where $\mathrm{Re}\,\varphi(z)>0$. The dots indicate the cut. The angles of the asymptotes (the dashed lines) are $\pm\pi/2\alpha$ and $\pm 3\pi/2\alpha$.

and $x_{1,2}(u)$ are the roots of the equation $\varphi(x)+u=0$. We now get

$$G'(k, s)=-ck^{2/5}\Phi(sk^{2/5}), \tag{76}$$

where

$$\Phi(\eta)=\int_{-\infty}^{-a}\frac{du}{u^2}e^{u\eta}\chi(u). \tag{77}$$

Since $\chi(u)=\frac{1}{4}|u|^{-1}$ as $u \to -\infty$, the function $\Phi(\eta)$ is bounded by a constant $\Phi(0)$ of order unity and the contribution from the cut is small like $k^{6/5}$.

3. To evaluate the asymptotic behavior of $\Psi(\xi)$ as $\xi \to \infty$ we make the substitution $y=(\xi/c)^{5/3}x^{8/5}$. After this we find the integral

$$J_\alpha(\lambda)=\int_0^\infty dx\cos\lambda x e^{-\lambda x}$$
$$\alpha=8/5, \quad \lambda=c^{-3/5}\xi^{3/5}\to\infty. \tag{78}$$

This integral is the real part of the integral

$$\int_C dz\, e^{\lambda\varphi(z)}, \quad \varphi(z)=iz-z^\alpha, \tag{79}$$

where the contour C is the real semi-axis $x>0$, while the function $\varphi(z)$ is defined in the z-plane with a cut $(0, -\infty)$. One can easily check that in the stationary points z_0 of the function $\varphi(z)$ we have $\mathrm{Re}\,\varphi(z_0)<0$ (when $3/2<\alpha<2$). The main contribution to the integral therefore comes from the point $z=0$, where $\mathrm{Re}\,\varphi(0)=0$. The steepest descent in the point $z=0$ goes along the imaginary y-axis in the direction of $y>0$. Deforming the contour to C' (see Fig. 3) and expanding

$$e^{\lambda\varphi(iy)}=e^{-\lambda y}[1-\lambda y^\alpha e^{i\pi\alpha/2}-\dots], \tag{80}$$

we find ($\lambda \to \infty$)

$$J_\alpha(\lambda)=\lambda^{-\alpha}\Gamma(\alpha+1)\cos\frac{\pi}{2}(\alpha-1) \quad (3/2<\alpha<2). \tag{81}$$

Using these results we get the asymptotic behavior of $\Psi(\xi)$ given in (43).

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Parametric amplified echo

Yu. M. Bun'kov and T. V. Maksimchuk

Institute of Physics Problems, USSR Academy of Sciences
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Amplification of a nuclear induction signal was obtained by exciting a spin system with a parametric pumping pulse. The experiments were performed on Mn^{55} nuclei in $CsMnF_3$ under conditions of coupled nuclear-electron precession. A theory is constructed for the formation of the parametric amplified echo excited by a high-power parametric pumping pulse.

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The spin-echo method makes it possible in a number of cases to obtain an echo signal of higher intensity than one of the exciting radiofrequency (RF) pulses. One can then speak of an RF amplifier of sorts, which produces also a time delay. This effect has been named "amplified" echo and was first observed in a ferromagnet for a system of long-wave spin waves.[1] It was observed for the same system[2] that replacement of the second resonant rf pulse by an RF pulse at double the frequency greatly enhances the amplification effect. The dynamics of the oscillations of the long-wave spin waves and their interaction with RF fields have a number of nonlinear features that hinder both the use of the effect and the development of a quantitative theory. The amplified-echo effect can be obtained in principle in systems with dynamic frequency shift, in which the spin echo is formed with the aid of a frequency-modulation (FM) mechanism (concerning the FM echo mechanism see, e.g., the review[3]). So far, however, this effect has not been obtained experimentally via the FM signal-formation mechanism.

Another echo-formation mechanism, in which echo amplification is possible, is parametric echo. We report here experimental observation of the amplified-echo effect via the parametric-echo mechanism. The parametric echo is produced in systems of oscillators on which it is possible to act directly both in resonant and in parametric fashions. Among the spin systems, these include electron spin systems in many magnetically ordered substances, as well as a system of nuclear spins under conditions of coupled nuclear-electron precession.

Parametric echo was first observed on Mn^{55} nuclei in a number of antiferromagnets.[4] To produce this echo, an RF pulse was applied to the spin system and was followed, after a time delay t_{12}, by an RF pulse having double the frequency and a magnetic field polarized along the constant magnetic field. This pulse excited the spin system parametrically. The specific nature of the direct parametric excitation of spin system makes the signal echo produced at the instant $2t_{12}$ an effect of first order in the RF pulse amplitude (and not of second order as in the Hahn or in the FM echo).

One of us and Gladkov[4,5] investigated theoretically and experimentally the mechanism of parametric echo formation at low amplitudes of the exciting pulses. With increasing amplitude of the amplitude of the RF parametric pumping pulse, we succeeded in observing the amplified echo effect. Experiments aimed at observing parametrically amplified echo were carried out on the system of Mn^{55} nuclei in the antiferrmagnet $CsMnF_3$ at 1.5 K and an NMR frequency 500 MHz. The experiments were performed with the parametric-echo spectrometer described in Ref. 5. To increase the parametric pumping power, a pulsed voltage up to 2 kV was applied to the oscillator and amplifier tubes of the double-frequency oscillator (G4-37A), so that the pulse RF power could be raised to 10 W.

The direct effect of the amplified echo is that the perpendicular magnetization of the sample, which forms the spin-echo signal, turns out to be larger than the perpendicular magnetization induced by the first pulse. In the experiment this reduces to a higher intensity of the echo signal than the intensity of the induction signal after the first pulse. The possibility of obtaining an echo signal exceeding in intensity the RF field of the resonant pulse is determined by the coupling between the RF field and the spin system. In the case of nuclear spin systems with low susceptibility, this is an extremely difficult task and is not considered in the present article.

Unfortunately, direct observation of the induction signal intensity following the resonant RF pulse is strongly hindered by the "dead" time of the receiving system, which amounts to 5 μsec. Therefore the intensity of the induction signal was estimated by the three-pulse procedure described in Chapter 5 of Ref. 5. At a maximum parametric buildup pulse, we succeeded in obtaining a parametric-echo signal of intensity four times higher than that of the induction signal.

For a quantitative investigation of the effect of the amplified parametric echo, we used as the amplified signal a spin-echo FM signal produced by two resonant

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