Propagation of nondecaying TA phonons

N. M. Guseinov and I. B. Levinson

L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR
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A study has been made of the propagation of superthermal phonons (ω > T₀) in crystals in which the spontaneous decay of those modes for which it is allowed by the conservation laws occurs more rapidly than elastic scattering by defects. Under such conditions the excitation energy is concentrated in the nondecaying modes. A kinetic equation has been derived to describe the propagation of nondecaying phonons. Besides elastic scattering, it involves effective decay which accompanies elastic scattering with conversion of the nondecaying mode into a decaying mode. In the long wavelength region of the spectrum this equation leads to a linear dependence of the propagation time τ of the phonon disequilibrium on the distance r, the velocity v being lower than that of sound. Recent experiments on the propagation of phonons in GaAs are analyzed critically and it is shown that the experimental results can be explained naturally by the theory developed in the present work.

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INTRODUCTION

It is now certain that the propagation of nonequilibrium phonons created on cooling and recombination of an electron-hole plasma in a semiconductor, and also on injection from "hot" metallic sources, is accompanied by the decay of phonons.4 Such phonon propagation was first discussed in Refs. 4 to 6, where an isotropic model was used with a single, naturally decaying, phonon branch. In real crystals there are, also, nondecaying modes, usually belonging to the lowest TA (transverse-acoustic) branch of the spectrum. This is not of special significance in those crystals for which fast conversion of nondecaying to decaying modes occurs on elastic scattering by defects, and the occupation numbers of these modes are evaned out. However, in very pure crystals a situation is possible when decay takes place more frequently than elastic scattering; in this case phonons will accumulate in nondecaying modes and the model4-6 is inapplicable. It has been stimulated by experiments in which the propagation and lifetime of high-frequency nondecaying TA modes in GaAs and other similar crystals have been studied.5-8

It has been found4-6 that the propagation time of the phonon disequilibrium from source to detector, t, depends linearly on the distance r between them, i.e., r/t is about one third of the velocity of the slowest transverse sound. The following interpretation was given for this. On stopping the photoexcitation of the plasma the carriers emit longwave LO phonons which decay into shortwave LA and TA phonons. Only the nondecaying TA phonons which occupy the frequency region near 1.7 THz (which corresponds to a wavelength of about 8 Å) survive. The group velocity of such shortwave TA phonons is 0.8×10⁹ cm·s⁻¹, which determines the signal peak arrival time. It is assumed that 1.7-THz phonons are not scattered over a length r<2 µm.

However, as will be shown in Sec. 5, the 1.7-THz TA phonon free path due only to scattering by isotopes is not more than 3 µm, so that an interpretation based on ballistic propagation of these phonons over a distance of the order of millimeters must be considered doubtful.

In the present work the propagation of nondecaying TA phonons is studied with account taken of their elastic scattering.1-3 A TA phonon can convert into a LA phonon on elastic scattering, and the latter on decaying leads to the appearance of lower frequency TA phonons. The result is effective decay of TA phonons that accompanies their propagation. It will be shown that such a propagation regime enables the linear relation r/t and other experimental results to be explained.

1. THE KINETIC EQUATION IN CLOSED FORM FOR NONDECAYING MODES

The propagation of nonequilibrium phonons is described by the kinetic equation

\[ \frac{\partial}{\partial t} + v(q) \nabla \cdot \begin{pmatrix} n(q) \sigma(q) \nabla S(q) + J(q) \end{pmatrix} \]

for the distribution function n(q,t). Here q indicates the phonon mode, i.e., the wave vector q and the branch number σ; v is the group velocity, S and J are collision terms responsible for anharmonic three-phonon processes (decay and fusion) and for elastic scattering by point defects. It is assumed below that the occupation numbers of the nonequilibrium phonons of interest are small, n<1, and their frequency is above the temperature describing the equilibrium phonons, ω > T₀. Under these conditions the scattering of nonequilibrium phonons can be neglected, and only spontaneous decay need be considered for nonequilibrium phonons, ignoring fusion of phonons and induced decays.1-3 In what follows, the occupation numbers of decaying modes are denoted by n₁(q) and of nondecaying modes by n(q); the choice of the indices l and s is related to the fact that in the isotropic model the decaying modes belong to the longitudinal branch and the nondecaying to the transverse branch.
The collision term $S$ for $\sigma < 1$ is linearized in $\sigma$; for a decaying mode it takes the form

$$S(q) = -n(q)\tau(q) + B(q),$$

where $\tau$ is the spontaneous decay time and the incoming term is of the form

$$B(q) = \int dq'n(q')P(q' - q).$$

$P$ is here the probability of a phononous decay of a phonon $q'$; it differs from zero only for $\omega_q > \omega_{q'}$. The integral covers all decaying modes, i.e.

$$\int dq_1...dq_n\sum_{\sigma} \frac{d^3q}{(2\pi)^3}...,$$

where $V_{\omega_q}$ is that part of the Brillouin zone where phonons of the branch $\sigma$ are decaying. If the mode $q$ is nondecaying, the outgoing term $-n/\tau$ is missing in Eq. (2); the incoming term $B$ is of the same form as Eq. (3).

For elastic scattering by defects

$$I(q) = -n(q)\tau^* + A(q),$$

where $\tau^*$ is the elastic scattering relaxation time and the incoming term is

$$A(q) = \int dq'n(q')W(q' - q).$$

The integration here is over all modes, both decaying and nondecaying; the probability $W$ contains the factor $\delta(\omega_q - \omega_{q'})$.

We now assume that decay is the most rapid process for decaying modes, that is,

$$\tau < \tau^*, \tau/r,$$

where $\tau$ and $r$ are the characteristic scales of temporal and spacial nonuniformity. Under these conditions phonons accumulate in nondecaying modes and, as will be seen,

$n_\sigma \gg n_\omega$.

We will also assume that there are no fewer nondecaying than decaying modes, i.e., that in the important frequency region

$$\rho_\sigma(\omega) > \rho_\omega(\omega),$$

where $\rho_\sigma$ and $\rho_\omega$ are the densities of states of nondecaying and decaying modes:

$$\rho_\omega(\omega) = \int dq_1\delta(\omega_q - \omega), \quad \rho_\sigma(\omega) = \int dq_1\delta(\omega_q - \omega).$$

The integral determining $\rho_\sigma$ has a meaning similar to Eq. (4) with the only difference that integration is over the region $V_{\omega_q}$, that part of the Brillouin zone where the modes of branch $\sigma$ are nondecaying.

We now go on to simplify the kinetic equation. On the basis of relation (7) the left-hand part of Eq. (1) and the outgoing term $n/\tau^*$ can be neglected by comparison with the outgoing term $n/\tau$. Using Eqs. (8) and (9) the integration in $A$ can be limited to the nondecaying region. We then obtain the local equation

$$n/\tau - B(n) = A(n),$$

which can be regarded as a linear integral equation in $n$. Its formal solution can be written in the form

$$n(q) = \int dq'A(q')\tau(q' - q).$$

The Green function $\tau$ of the operator in the left-hand part of Eq. (11) which enters in here has the following physical interpretation. Suppose a decaying phonon $q'$ had been created; decaying and passing through various intermediate states, it finally enters the range of nondecaying modes. The time which the phonon spends in the process of decaying in the elementary volume of phase space of decaying modes $dq$ near $q$ is $\tau(q' - q)dq$. Evidently $\tau(q' - q)$ differs from zero only for $\omega_q < \omega_{q'}$.

We turn to the equation for a nondecaying mode. Substituting the distribution of Eq. (12) into $B$, we find

$$B(q) = \int dq'n(q')P(q' - q) = B(q'),$$

where

$$P(q' - q) = \int dqW(q' - q)\eta(q' - q),$$

$$\eta(q' - q) = \int dq_1\tau(q_1)P(q_1 - q).$$

It is easy to see that $P(q' - q)$ differs from zero only for $\omega_q < \omega_{q'}$. The incoming term written in the form of Eq. (13) describes the appearance of nondecaying phonons of lower frequencies due to the effective decay mentioned in the Introduction. The path of this decay is seen in the structure of the kernel $P$: on elastic scattering, a nondecaying phonon $q'$ transforms into a decaying phonon $q$, which on decay finally leads to the appearance of a nondecaying phonon $q$ of lower frequency. The factor $\eta$ is the relative probability that upon decay of $q'$, just the phonon $q$ should appear.

Just as for a decaying mode, the integration in the term $A$ for nondecaying mode $q$ can also be limited to the nondecaying region and we take

$$A(q) = \int dq'n(q')W(q' - q) = A(q).$$

It is convenient to divide the outgoing term in two, putting

$$1/\tau(q) = 1/\tau(q) + 1/\tau(q'),$$

Here

$$\frac{1}{\tau(q)} = \int dqW(q' - q')$$

is the time for elastic scattering of a nondecaying phonon $q$ relative to its conversion into a decaying phonon. This time can obviously be considered as the lifetime for effective scattering. Furthermore

$$\frac{1}{\tau(q') = \int dq'W(q'' - q')}$$

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is the elastic-scattering time of a nondecaying phonon \( q \) relative to its transformation into a nondecaying phonon.

The kinetic equation for nondecaying modes can now be written as:

\[
\frac{\partial}{\partial t} \rho_q + \nabla \cdot \left( \tau_q \rho_q \right) = -\rho_q + \mathcal{I}(q),
\]

where

\[
\tau_q = -\frac{\partial \rho_q}{\partial t} + \mathcal{I}(q),
\]

\[
\mathcal{I}(q) = -\frac{\partial q}{\partial t} + \mathcal{J}(q).
\]

This equation is closed for the distribution function \( \rho_q \) of the nondecaying modes (the index \( \mathcal{I} \) is omitted for simplicity). \( S \) describes an effective decay and \( \mathcal{J} \) the elastic scattering between nondecaying modes.

2. LONGWAVE PHONONS; SELF-SIMILAR GREEN FUNCTION

The propagation of nondecaying phonons far from the source of their excitation is determined by the Green function of Eq. (20), i.e., by the solution of this equation for a point and instantaneous source. We shall look for the Green function, assuming the source to be monochromatic with frequency \( \omega \) and to use instead of \( q \) of the set of variables \( (a, e, w) \), where

\[
\frac{\partial}{\partial t} G(a, e, w, t) + \nabla \cdot \left( \tau(a, e, w) \nabla G(a, e, w, t) \right) = \delta(a, e, w) \delta(t),
\]

where

\[
\tau(a, e, w) = \frac{4\pi}{3} \left( \frac{a^3}{m} \right) \nu_i(e),
\]

\[
\nu_i(e) = \frac{1}{k_v} \int d\omega \left( \frac{\partial}{\partial \omega} \rho_{\omega}(e, w) \right).
\]

The existence of a self-similar solution for \( G \) is related to the fact that for longwave phonons all quantities have a power-law dependence on the phonon frequency \( \omega \) and the nature of this dependence is not connected with anisotropy. It is therefore convenient to indicate the frequency explicitly and to use instead of \( q \) of the set of variables \( (a, e, w) \), where

\[
\frac{\partial}{\partial t} G(a, e, w, t) + \nabla \cdot \left( \tau(a, e, w) \nabla G(a, e, w, t) \right) = \delta(a, e, w) \delta(t),
\]

where

\[
\tau(a, e, w) = \frac{4\pi}{3} \left( \frac{a^3}{m} \right) \nu_i(e),
\]

\[
\nu_i(e) = \frac{1}{k_v} \int d\omega \left( \frac{\partial}{\partial \omega} \rho_{\omega}(e, w) \right).
\]

If the elastic scattering obeys the Rayleigh law, then

\[
\tau(a, e, w) = \frac{1}{k_v} \nu_i(e),
\]

\[
\nu_i(e) = \frac{1}{k_v} \int d\omega \left( \frac{\partial}{\partial \omega} \rho_{\omega}(e, w) \right).
\]

The regions \( V_a \) and \( V_w \) are conical with origin at \( q = 0 \); the solid angles \( \Delta V_a \) and \( \Delta V_w \) correspond to them. The integration of Eq. (4) over decaying modes can thus be written in the new variables in the following way:

\[
\int d\omega = \sum \int d\omega \Delta V_a \rho_{\omega}(e, w),
\]

where \( \Delta V_a \) is an element of solid angle in the direction \( e \) and

\[
\mathcal{J}(q) = -\frac{\partial q}{\partial t} + \mathcal{J}(q).
\]
and find a solution in the form
\[ G_\lambda(e, u|\eta, \xi) = A_\lambda f_\lambda(e, u|\eta, \xi). \]  
(42)  
The factor \( A \) is written so that the function \( f \) can be chosen to be dimensionless.

Substituting Eq. (42) into the left hand part of Eq. (20), we have
\[ \frac{\partial}{\partial t} + \nabla \cdot \mathbf{u}\nabla \cdot \mathbf{u} = \frac{\partial}{\partial t} + \nabla \cdot \mathbf{u} A_\lambda \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi). \]  
(43)  
Substituting Eq. (42) into \( J \), we obtain
\[ A_\lambda (e, u|\eta, \xi) = \frac{\partial}{\partial t} \left( e \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi) \right) \]  
(44)  
Transformation of the incoming term in \( \mathbf{S} \) is somewhat more complicated. Taking Eq. (24) into account, we have
\[ J_\lambda(e, u|\eta, \xi) = \frac{\partial}{\partial t} \left( e \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi) \right) \]  
(45)  
Substituting here Eqs. (38), (40) and (42) we find
\[ J_\lambda(e, u|\eta, \xi) = \frac{\partial}{\partial t} \left( e \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi) \right) \]  
(46)  
where
\[ x = \alpha(e, u), \quad \eta = \eta(e, u), \quad \xi = \xi(e, u). \]  
Noticing that
\[ \mathbf{S} = \sum \frac{\partial}{\partial \xi} \left( e \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi) \right) \]  
and integrating over \( e \) instead of \( \eta \), we obtain
\[ \mathbf{S}_\lambda(e, u|\eta, \xi) = \frac{\partial}{\partial t} \left( e \frac{\partial}{\partial \xi} f_\lambda(e, u|\eta, \xi) \right) \]  
(47)  
where \( \mathbf{S} = \alpha(e, u) \). Collecting together the separate terms of the kinetic equation (20) and noting that \( \xi \) and \( \xi \) depend in the same way on \( \alpha(e, u) \), it can be seen that the frequency drops out everywhere except for the integration limit \( \xi \). However, \( \alpha(e, u) \) is finite, so that this condition is self-similar solution actually exists.

The parameter \( \alpha(e, u) \) is found from the condition of conservation of the total energy of nondecaying phonons
\[ E(t) = \int d\alpha \epsilon(e, \mathbf{u}|\eta, \xi), \]  
(49)  
where the energy density of the nondecaying phonons is
\[ e(e, \mathbf{u}|\eta, \xi) = \int d\alpha \epsilon(e, \mathbf{u}|\eta, \xi), \]  
(50)  
while the function \( f \) (without indices) is the mean:
\[ \mathbf{S}_\lambda(e, u|\eta, \xi) = \frac{\partial}{\partial t} \left( e \frac{\partial}{\partial \xi} f(e, u|\eta, \xi) \right) \]  
(51)  
On integrating over \( \xi \) and \( \xi \) as was done by Kazakovtsev and Levinson, \( ^{1} \) it can be confirmed that \( E(t) \) is independent of \( t \) only if \( \alpha = \alpha(e) \), and the upper limit in Eq. (50) can be replaced by infinity; this leads to the requirement \( \mathbf{J} = 0 \). The parameter \( \alpha(e) \) can also be found in a different way \( ^{3} \) by considering the total energy flux through a sphere of radius \( r \):  
\[ \mathbf{Q}(r) = \int d\mathbf{S} \int d\alpha \epsilon(e, \mathbf{u}|\eta, \xi), \]  
(52)  
where
\[ \mathbf{S} = \alpha(e, u|\eta, \xi), \]  
(53)  
On integrating first over \( \xi \) and then over \( \xi \), it can be confirmed that \( \mathbf{Q}(r) \) is independent of \( \xi \) only if \( r \) and \( \alpha(e) \) are finite. The factor \( A \) is determined by the energy of the source \( \mathbf{E} \). If the function \( \epsilon(e, \mathbf{u}|\eta, \xi) \) is normalized according to the condition
\[ \int d\mathbf{S} (\mathbf{S} - \mathbf{S}) = 0, \]  
(54)  
we obtain
\[ \mathbf{E} = k_\lambda A_\lambda \frac{\partial}{\partial \xi} \left( \mathbf{S} - \mathbf{S} \right). \]  
(55)  
The quantities in square brackets are in fact independent of \( \alpha(e, \mathbf{u}|\eta, \xi) \). In the region
\[ u < \alpha, \quad \xi = \xi(u|\eta, \xi), \quad r > r_\lambda \tau \]  
(57)  
there is thus a self-similar solution for the Green function
\[ G_\lambda(e, u|\eta, \xi) = A_\lambda f_\lambda(e, u|\eta, \xi), \]  
(58)  
with scaling variables of Eq. (41) and normalization according to Eqs. (55) and (56). This solution does not depend on the source frequency \( \omega \), but only on its total energy. If the source is spread over the spectrum in some frequency interval \( \mathcal{A} \), the solution of Eq. (58) and its region of applicability Eq. (57) do not change. In other words, after some effective decays the system forgets everything about the source except its total energy.

We now show that the self-similar distribution (58) corresponds to a broadening, linear in time, of the phonon disequilibrium. In the range of Eq. (57) the upper limit in Eq. (50) can be put as \( \mathcal{A} = \infty \); on changing to integration over \( \eta \), we find
\[ e(e, \mathbf{u}|\eta, \xi) = \epsilon(e, \mathbf{u}|\eta, \xi), \]  
(59)  
where
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It can be seen from this that with the passage of time the distribution of the energy density is transformed in similar way. The energy flux density behaves in just the same way. The relaxation times for elastic scattering and for effective decay ~and the ensuing linear nature of the broadening of the way. 

The question of whether the system of equations for the self-similar functions $f_\alpha$ contains small parameters is very important. It can be seen that all the quantities in this system are due only to the angular and polarization dependences of the spectrum and of scattering probabilities. If we assume that small parameters do not arise from such quantities, the functions $f_\alpha$ and $\Phi$ also do not contain small parameters. Under these conditions it can be confirmed that for a linear broadening of the phonon disequilibrium the corresponding velocity $v_l$ is of the order of the velocity of sound $v$. Actually, however, in many cases the number of nondecaying modes in the low frequency region appreciably exceeds the number of decaying modes: $p_\alpha \gg P_\alpha$ [see Eq. (3)]. In this case $t \ll c_0$, i.e., elastic scattering of a nondecaying phonon with conversion into a decaying phonon takes place considerably more rarely than without conversion. In other words, effective decay takes place more rarely than elastic scattering. In this case, one can pass from Eq. (20) to the diffusion equation for the function

$$n(u, t) = n_0(e, t).$$

It is important to emphasize, however, that the transition to the diffusion equation does not destroy the linear transformation of the spatial scale, and only a reduction in the speed of the rate $r_l$ of broadening of the phonon disequilibrium results. We shall consider this situation in the isotropic model in order not to burden the discussion with unimportant complications.

### 3. DIFFUSION OF NONDECAYING PHONONS IN THE ISOTROPIC MODEL

In the isotropic model all longitudinal $4\alpha$ modes are decaying and all transverse $T4$ modes are nondecaying. We also assume the probability of scattering not to depend on the direction of the phonon momenta, but only on the polarizations. Under these conditions

$$\tau_L = \frac{1}{\ln \eta} = \tau_{4\alpha,L}(u),$$

$$\tau_T = \tau_{T4}(u),$$

while it follows from the principle of detailed balancing that

$$\tau_{4\alpha,L}(u)/\tau_{T4}(u) = \pi_{4\alpha}(u)/\pi_{T4}(u).$$

For all frequencies $\omega$ for which the $T4$ branch exists, the ratio (64) is small; in the long wavelength region it is $\tau_{4\alpha,L}/\tau_{T4} \approx 0.1$, and it is even smaller in the short wavelength region.

Since scattering within the $T4$ branch takes place more frequently than scattering with $T4\rightarrow L4$ conversion, i.e., more frequently than effective decay, we can study the quasi-diffusion equation (64) instead of Eq. (20) for the isotropic part of the distribution $\pi_{4\alpha}(u) = \pi(u)$. It has the following form:

$$\left(\frac{d}{dt} - D(u) \nabla^2\right) n(u, r, t) = -\delta(n(u) - \delta(n(u))),$$

where the diffusion coefficient is

$$D(u) = \frac{1}{\tau_{4\alpha,L}(u)} \tau_{T4}(u),$$

and the term responsible for decay is

$$\delta(n(u)) = \frac{n(u)}{\tau_{4\alpha,L}(u)} + \int dw \pi_{4\alpha}(u) n(u') P(u' \rightarrow u).$$

Here $\pi_{4\alpha}(u)$ is the group velocity of a $T4$ phonon and the kernel $P$ is obtained from the kernel $P$ of Eq. (13) by averaging over $\alpha$ and $\epsilon$.

In the long wavelength region Eq. (65) belongs to the type of equations studied elsewhere. For a point source it has the self-similar solution

$$G(u, r, t) = A(u)(\frac{t}{r})^\frac{\beta}{2},$$

where

$$\beta = \frac{C}{c_0},$$

and $C$ is a number of the order unity, dependent on the form of the function $h(u)$ which is obtained from the function $\delta(u)$ occurring in Eq. (40) by averaging over $\alpha$ and $\epsilon$. The factor $A$ is expressed through the source energy $E$, the normalization (55) yields

$$E = -4\pi A^2 \pi_{4\alpha}(u) \pi_{T4}(u) \tau_{4\alpha,L}(u) \tau_{T4}(u).$$

The quantities in square brackets do not, in fact, depend on frequency.

The energy density propagating from a point source is

$$\varepsilon(r, t) = \frac{E}{4 \pi r} \left(\frac{t}{2r}\right)^{\frac{\beta}{2}},$$

where

$$E = \frac{16 \pi^3}{15} \left(\frac{E}{m}\right)^\frac{\beta}{2}. \left(\frac{t}{2r}\right)^{\frac{\beta}{2}}.$$
It follows from normalizing \( f \) according to Eq. (55) that \( \frac{1}{4} \int dx^2 g(x) = 1 \). (75)

The asymptotic functions \( g \) can be found by using Eqs. (70) and (71); they are:

\[
g(0) = c_1, \quad g'(0) = 0, \quad g(z) = c_2 z^2 (z \rightarrow 1),
\]

where \( c_1 \) and \( c_2 \) are numbers of the order unity.

It follows from Eq. (73) that the transition to the diffusion equation does not alter the nature of the relation between the temporal and spatial scales; it remains linear. The reason is that diffusion is accompanied by decay with the mean phonon frequency decreasing and the diffusion coefficient increasing. Since the functions \( f \) and \( g \) do not contain small parameters, the propagation velocity in the diffusion regime is of the order of \( s \), which is appreciably less than the velocity of sound. The time evolution of \( \tau \) at a fixed position in space is shown in Fig. 1; the distribution of \( E \) at a fixed instant is shown in Fig. 2.

The results obtained allow a clear physical interpretation in terms of phonon generations4,8 produced by effective decay. The generation with frequency \( \omega \) lives for a time \( \tau = \frac{\omega}{2} \alpha y \) and during this time propagates over a diffusion length \( l(\omega) \) by diffusion. Since \( l(\omega) \) increases rapidly with decreasing \( \omega \), the total distance \( r \) traversed by \( TA \) phonons from the source by the given instant \( t \) is of the order of the diffusion length \( l(\omega) \) for the last generation, i.e., the one that exists at the moment \( t \). This means that phonons with frequency determined by the relation

\[
\frac{\omega}{\alpha y} = \frac{r}{1/\tau_0(\omega)} \geq l(\omega)
\]

reach a detector at a distance \( r \). On the other hand, since the lifetime \( \tau = \frac{\omega}{2} \alpha y \) also increases rapidly with decreasing \( \omega \), the frequency of the generation existing at the moment \( t \) is determined by the condition

\[
\tau = \tau_0(\omega) \geq l(\omega)
\]

Eliminating from Eqs. (77) and (78) the frequency of phonons arriving at the detector, we find the arrival time \( t \rightarrow r/\tau \). The actual frequency of detected phonons is much less than the source frequency:

\[
\omega / \alpha y = \left[ \frac{r}{1/\tau(\omega)} \right]^{-1/2}.
\]

4. SCATTERING TIMES IN GaAs

Scattering by isotopes

An exact formula for the probability of scattering by isotopes in a crystal with one atom per unit cell is given in Ref. 12. It is easy to generalize this expression for the case of scattering in a crystal with several atoms per cell and to obtain the following expression for the scattering rate:

\[
\frac{1}{\tau_0(\omega)} = \frac{\pi}{2} \alpha y \beta y (\omega) \sum \frac{M_s^2}{M_T^2} \langle \epsilon \rangle \langle \epsilon \rangle \langle \epsilon \rangle
\]

Here \( q \) and \( q' \) are the initial and final phonon momenta, \( \alpha \) and \( \alpha' \) are the initial and final branches, \( \alpha \) is the number of atom in the cell, and the polarization vectors are averaged according to

\[
\sum M_s \langle \epsilon \rangle = \frac{\pi}{2} \alpha y \beta y \omega \sum \frac{M_s^2}{M_T^2} \langle \epsilon \rangle
\]

Equation (80) with Eq. (82) inserted is equivalent to an earlier form13 if the difference in the way of normalizing the polarization vectors is taken into account.

For the acoustic branch in the longwave region \( |q|^2 \approx 1 \), so that an isotropic scattering time is obtained for a cubic crystal; it is convenient to express the time in the following way:

\[
\frac{1}{\tau_0(\omega)} = \eta_{\alpha y} (\omega) \delta / \omega \alpha y (\omega) ^2,
\]

where \( \delta \) is the Debye frequency and \( \eta_{\alpha y} (\omega) = 1/12n_s (\omega) \beta y (\omega) \delta / \omega \alpha y (\omega) \). (83)

Decay

In the isotropic model the \( TA \) phonons are nondecaying, while \( LA \) phonons decay via two channels:

\[
LA \rightarrow LA + TA, \quad TA + TA.
\]

(85)

This statement is valid both in the long wavelength and short wavelength regions of the spectrum. In the isotropic model the decay of \( TA \) phonons is forbidden by the dispersion (if there is no dispersion, collinear decays are possible). Anisot-
The magnitude of the anisotropy does not depend strongly on frequency, so that the prohibition is relaxed more easily in the long wavelength region where the dispersion is small. This has been illustrated by numerical calculations of the two-phonon density of states and the fact that the anisotropy of the velocity of sound in GaAs is close to that which follows from the model indicated above attest to this.

The probability of decay of long wavelength phonons can be conveniently written in the following form:

\[ \frac{1}{\tau_{\text{e}}(\mathbf{q}, \omega)} = \gamma \left( e_{\text{Ga}}(\alpha/\alpha_{\text{Ga}})^{1/2} \right) \tag{86} \]

where \( \gamma \) is an adiabatic parameter of the order of \( \alpha_{\text{Ga}}/\alpha_{\text{Ga}}^{2} \) and is independent of the frequency. Accurate values of \( \gamma \) can be calculated by using measured third order elastic moduli. As far as we know, such a calculation has not been made for GaAs, but it has been made for Ge, admittedly in the isotropic model. Since the phonon spectra of GaAs and of Ge are very close, we used for GaAs the values of \( \gamma \) obtained for Ge:

\[ \gamma_{\text{Ga}} \approx 2 \times 10^{-3} \quad \gamma_{\text{GaAs}} \approx 4 \times 10^{-4} \]

5. DISCUSSION OF EXPERIMENTS

It was estimated \(^{2-7,9}\) that short wavelength T4 phonons of frequency about 1.7 THz and wavelength 8 Å propagate in GaAs over a distance of the order of a few mm without being scattered by isotopes. Since the usual estimates of the mean free path according to a formula like (83) give an appreciably smaller value, it has been suggested \(^{4}\) that these phonons, which lie close to the zone edge, are weakly scattered by isotopes or that the oscillation amplitude of the single iso- 
apes in the 0.9 THz region is slight, justifying the use of the long wavelength description. We now find the scattering time in the important region of 0.9 THz. In this frequency region Eq. (83) can be used for isotope scattering, where

\[ \eta_{\text{GaAs}} = 3.5 \times 10^{-3} \]

This gives

\[ \tau_{\text{GaAs}} = 0.3 \mu s, \quad \tau_{\text{Ga}} = 3 \mu s. \]

According to Eq. (86) the decay time of 0.9 THz LA phonons \( \tau_{\text{LA}} \approx 20 \) ns. It is seen that decaying phonons decay much more rapidly than they are scattered and that assumption (7) is satisfied in the important frequency region.
A weak dependence of signal arrival velocity on propagation direction is observed in the experiments; for example, the velocity in the three principal directions [100], [110] and [111] varies for GaAs from $1 \times 10^3$ to $0.8 \times 10^3$ cm s$^{-1}$. It seems that this scatter lies within the limits of the experimental accuracy, but in principle a dependence on direction can exist. We point to the following fact in this connection. In the quasidiffusion approximation the energy flow from a point source in a cubic crystal possesses spherical symmetry, independently of whether the isotropic model is used or not; the reason is that in a cubic crystal the diffusion tensor reduces to a scalar. Therefore, in the quasidiffusion approximation the signal velocity in GaAs should be the same in all directions. However, the accuracy of this approximation is in fact not very high; it is determined by the parameter $a$.

Violation of the quasidiffusion approximation can lead to some anisotropy. The so-called "velocity bunching" was also studied experimentally, when phonons were excited by scanning a laser spot with velocity $u$ over a length $L$, with the detector located on the continuation of the line at a distance $d$ from its end. In this case the phonon energy density at the place where the detector is positioned, i.e., its signal, is according to Eq. (79)

$$s(t) = \frac{1}{\hbar} \frac{d}{dt} \int_{E_d}^{E_f} \frac{1}{x(x-\varepsilon)^{1/2}} \frac{L}{x(t-f)} \frac{d\varepsilon}{x(t-f)},$$

(89)

Here $W$ is the laser power, $\beta$ the fraction of the quantum energy $hv$ transformed into LO phonons on relaxation of electron-hole pairs, $E_d$ is the width of the forbidden band, $R$ the reflection coefficient, $L=L'+d$ is the distance from the initial scanning point to the detector, and $t=0$ corresponds to the start of the scanning.

For numerical calculations of $e(t)$ the function $g$ was approximated by the expression

$$g(z)=A(1+az)^{-4},$$

(90)

satisfying the asymptote (76) and the normalization (55). The parameter $a$ can be found from the measured$^{15}$ maximum spatial distribution of the signal $e(t)$ from a point source. Using the signal in GaAs for $t=2.05 \mu s$, we find $a=0.98$. The results of a numerical calculation of $e(t)$ are shown in Fig. 3. It can be seen that as the scanning speed $u$ decreases, the steep leading and mildly sloping rear edges change over to mild leading and steep rear edges, and for $u=z$ the signal is symmetrical. It is just this behavior that was observed experimentally.$^7$

The numerically calculated dependence of the peak signal amplitude, $\max e(t)$, on the scanning velocity $u$ is shown in Fig. 4 and agrees with measured value quite well. The experimental points for $u=z=0$ fall on a straight line with slope $-1$, corresponding to a relation $\max e(t) \sim u^{-1}$. This relation follows from Eq. (89) at $u=0$.

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A short account of the present work has been published.$^1$


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FIG. 3. Calculated detector signal $(L=2.59 \text{ mm}, d=0.42 \text{ mm})$ for different scanning speeds: a) $u=3.6$; b) $u=2.47$; c) $u=0.29$; d) $u=0.1$ (in units of $10^3$ cm s$^{-1}$). One division on the abscissa axis is $5 \mu s$; the arrows indicate the measured signal peak arrival time. The signals are normalized to the same amplitude at the maximum.

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FIG. 4. Dependence of the peak signal amplitude on the scanning speed $(L=2.59 \text{ mm}, d=0.42 \text{ mm})$. The points are experimental, the curve is calculated; for normalization the theoretical curve is made to coincide with the experimental points at $u=0$. The authors are grateful to L. Shchur for the numerical calculations.

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