

Theory of Heat-Pulse Propagation in a Phonon Gas

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(Received 31 May 1968)

A theory of the propagation of heat pulses in a phonon system has been constructed. It is based on the solution of the Boltzmann equation for the phonon distribution function in the presence of a heat source and in the relaxation-time approximation. The result obtained is valid at all times, whether longer than, shorter than, or comparable to the mean phonon relaxation time, and provides a basis for quantitative interpretation of the experimental data.

1. INTRODUCTION

RECENTLY there has been much activity in the experimental study of the propagation of heat pulses in solids. These experiments are intended to yield various interesting properties of the thermal carriers responsible for the heat transmission, for example, their velocities and their mean free paths due to scattering. In metals at very low temperature the thermal carriers are essentially the electrons, whereas in dielectrics or in metals at higher temperature¹ they are mainly the phonons. For a general review of the situation of heat-pulse experiments the readers are referred to von Gutfeld.² One notices immediately from his article that the theoretical understandings of the experimental results are rather qualitative. The difficulty lies in the fact that one is usually working in situations in which the transmission time of the heat pulse along the sample is comparable to the mean relaxation time of the carriers. In such cases the transport of heat cannot be described by the ordinary heat conduction equation. Hence it is of theoretical interest to construct a theory capable of describing the physics of heat-pulse propagation valid at all times. Then it would be possible to interpret the

experimental data more quantitatively, and also to correlate various observations in a coherent manner. In this paper we shall be concerned exclusively with the transmission of heat pulses by phonons. The theoretical analysis is based on the Boltzmann equation of the phonon distribution function. The main task consists of solving the Boltzmann equation in the relaxation-time approximation and in the presence of a heating source (prescribing a phonon-production rate). The solution obtained represents a new result which has not been considered before.³ It leads to a description of the transport of heat which is valid at all times. Only after time long compared to the characteristic times in the sample does one recover the usual heat conduction result. Before we go into the details of our theoretical analysis let us review briefly the usual experimental setup in a heat-pulse experiment. It consists mainly of three elements: the heater, the sample, and the detector. There are various kinds of heater and detectors. For our purpose we shall take them to be thin metallic films evaporated onto the ends of the sample (Fig. 1). The whole assembly is immersed in a heat bath (usually liquid helium) at temperature T_0 . The experiment is started (at $t=0$) by passing a current pulse through the heater. The generated heat pulse now travels down the sample and is detected. The detector is usually a superconducting film near its transition temperature so that its resistivity is very sensitive to heating or temperature change and is therefore a convenient quantity to measure.

In Sec. 2 the generation of heat in the heating metallic film is discussed. It is shown that under some simplifying assumptions one can treat the heater as a phonon source for the sample. In Sec. 3 the Boltzmann equation of the phonon distribution function in the sample in the presence of phonon generation at the interface is studied. Using the relaxation time approximation, a solution to the Boltzmann equation is found which describes the space- and time-dependent deviation of the phonon

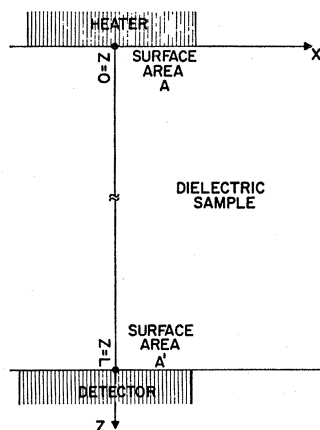


FIG. 1. Heat-pulse experiment schematics.

¹In a metal, the thermal energy is mainly contained in the lattice vibrations (or phonons) except at very low temperature ($T \lesssim 1^\circ\text{K}$). See Eq. (2) and the discussion following it.

²R. J. von Gutfeld, in *Physical Acoustics*, edited by Warren P. Mason (Academic Press Inc., New York, to be published), Vol. V.

³Our result can describe the transient behavior of the phonon distribution after it has been disturbed far from thermal equilibrium and before the distribution relaxes to one which can be characterized by local thermodynamic variables like temperature and phonon drift velocity and their derivatives. The behavior of the phonon system in this latter limit has been studied by many authors, and particularly comprehensive discussions can be found in R. A. Guyer and J. A. Krumhansl, *Phys. Rev.* **148**, 766 (1966); **148**, 778 (1966).

distribution function from total thermal equilibrium. In Sec. 4, the result is discussed in various limiting cases, in particular, the heat conduction (diffusion) or long-time limit. In Sec. 5, the detection of the pulse is briefly considered. It is followed by concluding remarks on the applicability of the present theory to explain various experiments.

2. PHONON PRODUCTION BY HEATER

We begin by studying how the heater plays the role of an external time-dependent phonon source for the sample and thus provides the necessary boundary condition for the phonon Boltzmann equation in the sample to be studied in Sec. 3. As mentioned before, the heat-pulse experiment begins when one passes a current pulse through the heating metallic film at $t=0$. Thus the current $I(t)$ is a known function of time. It is beyond the scope of this paper to investigate in detail the time-dependent behavior of the coupled electron-phonon system in the metal to study how heat is generated. One makes the simple assumption that the film remains always in thermal equilibrium. In other words, it is characterized by a temperature $T_H(t)$ which is time-dependent. For simplicity we take T_H to be uniform in the film. The assumption of thermal equilibrium is valid if the thermal relaxation time (time for thermalization) of electron-phonon system is very short compared to the width of the current pulse. This condition is usually satisfied in real experimental situations.² Then denoting the total energy of the system by $E_H(t)$, whose dependence on time is entirely through $T_H(t)$, we have the energy balance equation

$$\frac{\partial}{\partial t} E_H(t) = I^2(t) R_H - \left(\frac{\partial}{\partial t} E_H(t) \right)_{\text{loss}}, \quad (1)$$

where R_H is the resistance of the film. The first term on the right-hand side represents the input power which is just Joule heating in our case. It will, of course, take on different forms if the film is heated by other means, e.g., by a laser beam. The second term represents the heat loss to the environment. The film loses its heat both to the heat bath and to the sample. In actual systems the latter loss is by far the dominant one. This unfortunately creates a slight difficulty, namely, that one cannot, in principle, decouple the energy-conservation equation (1) from the heat-propagation equation in the sample to be discussed later. The reason is that part of the heat contained in the heat pulse transmitted into the sample will at a later time diffuse or reflect back to the film. However, in order not to complicate our analysis, we shall make the reasonable assumption that the total amount of heat which will eventually reach the film at later times is small compared to the initial energy input. This condition is satisfied if the sample is large.

We shall now study Eq. (1). The total energy of the

film E_H consists of two parts, an electronic part which is proportional to $(T_H)^2$ and a phonon part proportional to $(T_H)^4$, i.e.,

$$E_H(t) = \left\{ \frac{1}{2} [\gamma T_H(t)]^2 + \frac{1}{4} b [T_H(t)]^4 \right\} \times V. \quad (2)$$

The quantity V is the volume of the film and the constants γ and b are characteristic constants of the particular metal in question. The two contributions are comparable at very low temperature, $T_H \sim 1^\circ\text{K}$. Thus, in usual situations, the phonon part dominates. The energy-loss term arising from the heat transfer across the metal-dielectric interface is quite complicated. However, the situation is much simplified if one ignores the role of the electrons and assumes that the dominant mechanism is phonon transmission. A thorough discussion of this topic of heat transfer across solid boundaries may be found in the articles by Little⁴ and by von Gutfeld *et al.*⁵

We shall now outline the derivation of the energy-loss term in Eq. (1). Heating of the film increases the phonon population. Since we have assumed thermal equilibrium at all times, the change in the phonon distribution function is simply given by

$$\delta n_{\lambda\mathbf{p}}(t) = n_{\lambda\mathbf{p}}^0(T_H(t)) - n_{\lambda\mathbf{p}}^0(T_0), \quad (3)$$

$$n_{\lambda\mathbf{p}}^0(T) = [\exp(\hbar\omega_{\lambda\mathbf{p}}^{(H)}/k_B T) - 1]^{-1}, \quad (4)$$

where λ and \mathbf{p} denote the polarization and wave vector of the phonon with frequency $\omega_{\lambda\mathbf{p}}^{(H)}$. Next we calculate the number of these extra phonons striking a unit area of heater-sample surface per unit time. This can be readily found to be

$$\delta n_{\lambda\mathbf{p}}(t) \times (\hat{p}_z/\hat{p}) v_{\lambda}^{(H)} \{ \tau_{\lambda\mathbf{p}}^{(H)} [1 - \exp(-1/\tau_{\lambda\mathbf{p}}^{(H)})] \} \quad (\hat{p}_z > 0), \quad (5)$$

where $v_{\lambda}^{(H)}$ is the sound velocity for branch λ , and $\tau_{\lambda\mathbf{p}}^{(H)}$ is some effective phonon scattering time. The appearance of the phonon relaxation is obvious by considering the two limiting cases. In the case of no scattering ($\tau^{(H)} \rightarrow \infty$) all the phonons within a distance $v_{\lambda}^{(H)} \times (\hat{p}_z/\hat{p})$ will hit the surface within 1 sec. Now scattering will reduce this number and, in fact, when $\tau^{(H)} \ll 1$ sec, only those phonons within a distance of the order of a mean free path $v^{(H)}\tau^{(H)}$ from the surface can reach it. Finally we consider the generation of phonons in the insulator sample by the phonons [Eq. (5)] striking the surface. Such a process is in general very complicated and explicit results have been obtained only for some simple cases.⁶ For our purpose, we shall simply describe this process by a transmission coefficient $\alpha(\lambda\mathbf{p}, \lambda'\mathbf{p}')$ which represents the probability of creating a phonon ($\lambda\mathbf{p}$) in the sample by a phonon ($\lambda'\mathbf{p}'$) in the metal incident upon the surface. One property of that is obvious is that

⁴ W. A. Little, *Can. J. Phys.* **37**, 334 (1959).

⁵ R. J. von Gutfeld, A. H. Nethercot, Jr., and J. A. Armstrong, *Phys. Rev.* **142**, 436 (1966).

⁶ See Ref. 4 and H. Kolsky, *Stress Wave in Solids* (Oxford University Press, London, 1953).

it must be zero when $p_z < 0$. Thus the total number of phonons of the kind $(\lambda \mathbf{p})$ produced in the sample per unit time and per unit area of the surface is

$$\begin{aligned} \dot{\eta}_{\lambda \mathbf{p}}(t) &= 0 \quad (p_z < 0) \\ &= \sum_{\lambda' \mathbf{p}'} \alpha(\lambda \mathbf{p}, \lambda' \mathbf{p}') \delta n_{\lambda' \mathbf{p}'}(t) v_{\lambda'}^{(H)} \{ \tau_{\lambda' \mathbf{p}'}^{(H)} \\ &\quad \times [1 - \exp(-1/\tau_{\lambda' \mathbf{p}'}^{(H)})] \} \quad (p_z > 0). \end{aligned} \quad (6)$$

It will be discussed in more detail in Sec. 3 that these generated phonons are to be treated as being localized in the region near the heater (Fig. 1). Now from Eq. (6) one can immediately calculate the rate of energy loss as

$$+ \left(\frac{\partial E_H(t)}{\partial t} \right)_{\text{loss}} = \left(\sum_{\lambda \mathbf{p}} \hbar \omega_{\lambda \mathbf{p}} \dot{\eta}_{\lambda \mathbf{p}}(t) \right) \times A, \quad (7)$$

where A is the contact area of the heater with the sample and $\omega_{\lambda \mathbf{p}}$ is the phonon frequency in the dielectrics. Combining Eq. (7) with Eqs. (1)–(3), the unknown time-dependent temperature of the film $T_H(t)$ may be determined. This allows us to determine $\dot{\eta}_{\lambda \mathbf{p}}(t)$ explicitly as a function of time. We shall conclude this section by discussing briefly the qualitative feature of the spectrum of the emitted phonons. For this purpose the transmission coefficient $\alpha(\lambda \mathbf{p}, \lambda' \mathbf{p}')$ is simply taken as $\alpha \delta_{\lambda \lambda'} \delta_{\mathbf{p} \mathbf{p}'}$, where α is a constant. Then Eq. (6) says that the spectral distribution of the phonons generated in the sample is approximately proportional to the change in the thermal spectrum in the heater film.

3. PHONON BOLTZMANN EQUATION IN SAMPLE

The phonon distribution in the sample is described by the function $N_{\lambda \mathbf{p}}(\mathbf{r}, t)$. To avoid further complication we assume that the dimension of the sample in the x and y directions is large compared to the length L along z . This allows us to neglect complicated surface effects. The dynamical development of the phonon distribution function is determined by the Boltzmann equation⁷ (for $0 < z < L$)

$$\begin{aligned} \frac{\partial}{\partial t} \delta N_{\lambda \mathbf{p}}(\mathbf{r}, t) + \mathbf{v}_{\lambda \mathbf{p}} \cdot \nabla \delta N_{\lambda \mathbf{p}}(\mathbf{r}, t) &= -(\partial N_{\lambda \mathbf{p}} / \partial t)_{\text{collision}} \\ &\quad + (\partial N_{\lambda \mathbf{p}} / \partial t)_{\text{source}}, \end{aligned} \quad (8)$$

where $\mathbf{v}_{\lambda \mathbf{p}}$ is the group velocity of the phonon defined by

$$\mathbf{v}_{\lambda \mathbf{p}} = \nabla_{\mathbf{p}} \omega_{\lambda \mathbf{p}}. \quad (9)$$

The function $\delta N_{\lambda \mathbf{p}}(\mathbf{r}, t)$ is used to represent the deviation of the phonon distribution from the complete thermal equilibrium distribution at the ambient temperature T_0 , i.e.,

$$\begin{aligned} \delta N_{\lambda \mathbf{p}}(\mathbf{r}, t) &= N_{\lambda \mathbf{p}}(\mathbf{r}, t) - N_{\lambda \mathbf{p}}^0(T_0), \\ N_{\lambda \mathbf{p}}^0(T_0) &= [\exp(\hbar \omega_{\lambda \mathbf{p}} / k_B T_0) - 1]^{-1}. \end{aligned} \quad (10)$$

⁷ R. E. Peierls, *Quantum Theory of Solids* (Pergamon Press, Inc., London, 1955).

The first term on the right-hand side of Eq. (8) is the collision integral describing the rate of change of the phonon distribution function due to scattering of phonons. More will be said of this term later. The last term in Eq. (8) describes the rate of phonon production due to an external source. We will now determine its form using the result obtained in Sec. 2. Let us first recall that the function $N_{\lambda \mathbf{p}}(\mathbf{r}, t)$ represents the number of phonons of the type $(\lambda \mathbf{p})$ per unit volume at position \mathbf{r} (and time t). Such a description necessarily implies that one must treat the phonons as wave packets which are simultaneously quite well localized in momentum and coordinate space. Hence for phonons with wavelengths comparable to the size of the crystal, the concept of a local distribution is no longer valid. In Sec. 2 we found $\dot{\eta}_{\lambda \mathbf{p}}(t)$ [Eq. (6)], the rate of phonon production per unit area in the sample by the heating film. One immediately has the relation

$$\int_{\text{sample volume}} d^3 \mathbf{r} \left(\frac{\partial N_{\lambda \mathbf{p}}(\mathbf{r}, t)}{\partial t} \right)_{\text{source}} = A \times \dot{\eta}_{\lambda \mathbf{p}}(t). \quad (11)$$

To determine $(\partial N / \partial t)_{\text{source}}$ explicitly, we first make use of the concept of the localization of the phonons which implies that the phonons produced are localized within quantum distances from the boundary ($z=0$). Furthermore, we assume that the dimensions of surface area A are small compared to those of the sample. With these conditions we may treat the heater as a point source and obtain

$$(\partial N_{\lambda \mathbf{p}} / \partial t)_{\text{source}} = A \delta^3(\mathbf{r}) \dot{\eta}_{\lambda \mathbf{p}}(t), \quad (12)$$

which obviously satisfies Eq. (11). The Boltzmann equation (8) will offer a complete description if the collision term is specified. In the following discussion two types of phonon scattering processes will be considered. The first case is the simpler one in which the dominant phonon scattering process is the absorption of phonons by other quantum-mechanical systems. The reemissions of phonons may be ignored because they occur at a much later time (long compared to the passage of the heat pulse through the sample). In this case the solution to the Boltzmann equation is very simple, describing what will be called a ballistic flow. The energy in the pulse is carried by the individual phonons created at the boundary. They travel down the crystal with their respective group velocities until being absorbed and thus losing their share of energy. The second case is the more complicated one, in which the phonon scattering processes considered conserve the total amount of energy in the phonon system. This leads to the existence of another type of flow besides the ballistic one and is referred to as the thermal flow. This is the result of thermalization or thermal mixing of the phonon distribution. It will be seen that the initial flow is ballistic in nature; then gradually the thermal part of the phonon distribution is built up at the expense of the ballistic

part. Finally, after a time extremely long compared to the average relaxation time, the thermal part approaches a local equilibrium form. In this limit the heat flow may be adequately described by the ordinary heat-conduction equation.

Case 1: Phonon Absorption

In case 1 we consider the situation in which the dominant phonon scattering process is absorption by impurity states. A familiar and interesting example is the resonant phonon absorption by paramagnetic impurities in the presence of a variable magnetic field. A review of the theory of spin-lattice interaction may be found in the article by Tucker.⁸ Another example is the absorption of phonons by shallow neutral impurity states.⁹ However, here one cannot vary the resonant absorption frequency and it occurs at rather high frequencies ($\omega \gtrsim 10^{11}$ sec⁻¹, $\hbar\omega/k_B T \gtrsim 3^\circ\text{K}$). In the relaxation-time approximation the collision integral for such process is extremely simple, namely,

$$-\left(\frac{\partial N_{\lambda p}}{\partial t}\right)_{\text{collision}} = -\frac{\delta N_{\lambda p}(\mathbf{r}t)}{\tau_{\lambda p}^{(1)}}, \quad (13)$$

where $\tau_{\lambda p}^{(1)}$ denotes the branch- and wave-number-dependent phonon relaxation time or the inverse of the rate of phonon absorption.¹⁰ Equation (13) says that the extra phonon disappears according to the absorption rate and the phonon distribution function is reduced to complete thermal equilibrium. Substituting Eqs. (12) and (13) into Eq. (8), we obtain

$$(\partial/\partial t + \mathbf{v}_{\lambda p} \cdot \nabla + 1/\tau_{\lambda p}^{(1)}) \delta N_{\lambda p}^{(1)}(\mathbf{r}t) = A \delta^3(\mathbf{r}) \dot{\eta}_{\lambda p}(t). \quad (14)$$

A superscript (1) will be used to denote that we are dealing with case 1. This equation can be readily solved and the solution is

$$\delta N_{\lambda p}^{(1)}(\mathbf{r}t) = \int d^3 r' \int dt' g_{\lambda p}^{(1)}(\mathbf{r}-\mathbf{r}', t-t') \times (A \delta^3(\mathbf{r}') \dot{\eta}_{\lambda p}(t')), \quad (15)$$

where $g^{(1)}$ is the Green's function defined as the response to a unit pulse

$$(\partial/\partial t + \mathbf{v}_{\lambda p} \cdot \nabla + 1/\tau_{\lambda p}^{(1)}) g_{\lambda p}^{(1)}(\mathbf{r}-\mathbf{r}', t-t') = \delta^3(\mathbf{r}-\mathbf{r}') \delta(t-t'). \quad (16)$$

The solution which satisfies the boundary condition that $g=0$ at infinite past ($t-t' \rightarrow -\infty$) is

$$g_{\lambda p}^{(1)}(\mathbf{r}-\mathbf{r}', t-t') = 0 \quad (t < t') \\ = \exp\left(-\frac{t-t'}{\tau_{\lambda p}^{(1)}}\right) \delta^3(\mathbf{r}-\mathbf{r}' - \mathbf{v}_{\lambda p}(t-t')) \quad (t > t'). \quad (17)$$

⁸ E. B. Tucker, in *Physical Acoustics*, edited by Warren P. Mason (Academic Press Inc., New York, 1966), Vol. IV, Part A.

⁹ P. C. Kwok, *Phys. Rev.* **149**, 666 (1966).

¹⁰ Formal study of the collision integral may be found in the references quoted in Ref. 3.

In Sec. 4 we have occasion to use the more formal expression for $g_{\lambda p}^{(1)}$, namely,

$$g_{\lambda p}^{(1)}(\mathbf{r}-\mathbf{r}', t-t') = (\partial/\partial t + \mathbf{v}_{\lambda p} \cdot \nabla + 1/\tau_{\lambda p}^{(1)})^{-1} \times \delta^3(\mathbf{r}-\mathbf{r}') \delta(t-t'). \quad (18)$$

Substituting expression (17) for the Green's function in (15), we obtain the explicit solution to the Boltzmann equation

$$\delta N_{\lambda p}^{(1)}(\mathbf{r}t) = A \delta(x - \gamma_x z) \delta(y - \gamma_y z) \\ \times \exp\left(-\frac{z}{v_{\lambda p}^z \tau_{\lambda p}^{(1)}}\right) \frac{1}{v_{\lambda p}^z} \dot{\eta}_{\lambda p}\left(t - \frac{z}{v_{\lambda p}^z}\right), \quad (19) \\ \gamma_x = v_{\lambda p}^x / v_{\lambda p}^z, \quad \gamma_y = v_{\lambda p}^y / v_{\lambda p}^z,$$

where we have used the property that $\dot{\eta}_{\lambda p} = 0$ for $p_z < 0$ and have assumed that the z component of the phonon phase velocity $v_{\lambda p}^z$ is positive for positive p_z . The nature of the solution (19) is obvious. It describes a collimated ballistic flow in which the initial pulse shape of each phonon component remains unchanged. The velocity of the propagation is simply the group velocity $\mathbf{v}_{\lambda p}$ (or $v_{\lambda p}^z$ along the z direction). Its amplitude is decreasing due to phonon absorption. The attenuation suffered by any component of the flow at a position \mathbf{r} depends on the number of mean free paths it has traversed.

Case 2: Energy-Conserving Phonon Scattering

The next case is more complicated; we assume that the dominant phonon scattering processes conserve the total energy of the phonons. Examples of such are phonon-phonon interaction including both normal and Umklapp processes. If the normal phonon processes dominate the collisions, then not only the phonon energies but also the phonon momenta are conserved. This makes the situation slightly more complicated. However, no new concepts other than those discussed below need be introduced. We shall come back to this point at the end of the section. Other examples of such processes are elastic scatterings of phonons by impurities, defects and dislocations, etc. In this case the collision integral in the Boltzmann equation $(\partial N/\partial t)_{\text{collision}}$ has (a) the property of conserving phonon energies, namely,

$$\sum_{\lambda p} \hbar \omega_{\lambda p} \left(\frac{\partial N_{\lambda p}}{\partial t}\right)_{\text{collision}} = 0. \quad (20)$$

An immediate consequence of this is a conservation equation for the phonon energy density. Multiplying Eq. (8) by $\hbar \omega_{\lambda p}$ and summing over (λp) , one gets

$$\frac{\partial}{\partial t} E(\mathbf{r}t) + \nabla \cdot \mathbf{J}_E(\mathbf{r}t) = P_H(\mathbf{r}t), \quad (21)$$

where E is the phonon energy density, \mathbf{J}_E is the energy current, and P_H is the power input by the heater. They

are given by

$$E(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \delta N_{\lambda p}(\mathbf{r}t), \quad (22)$$

$$\mathbf{J}_E(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \mathbf{v}_{\lambda p} \delta N_{\lambda p}(\mathbf{r}t), \quad (23)$$

$$\begin{aligned} P_H(\mathbf{r}t) &= \sum_{\lambda p} \hbar\omega_{\lambda p} \left(\frac{\partial N_{\lambda p}}{\partial t} \right)_{\text{source}} \\ &= A \delta^3(\mathbf{r}) \sum_{\lambda p} \hbar\omega_{\lambda p} \dot{\eta}_{\lambda p}(t). \end{aligned} \quad (24)$$

Another property of the collision term is (b) that it vanishes when the distribution function $N_{\lambda p}(\mathbf{r}t)$ corresponds to a local equilibrium distribution $\tilde{N}_{\lambda p}(\mathbf{r}t)$ which is a Planck distribution characterized by a space- and time-dependent temperature function $T(\mathbf{r}t)$, i.e.,

$$\left(\frac{\partial N_{\lambda p}}{\partial t} \right)_{\text{collision}} = 0 \text{ for } N_{\lambda p}(\mathbf{r}t) = \tilde{N}_{\lambda p}(\mathbf{r}t), \quad (25)$$

$$\begin{aligned} \tilde{N}_{\lambda p}(\mathbf{r}t) &= N_{\lambda p}^0(T(\mathbf{r}t)) \\ &= \{ \exp[\hbar\omega_{\lambda p}/k_B T(\mathbf{r}t)] - 1 \}^{-1}. \end{aligned} \quad (26)$$

At present the function $T(\mathbf{r}t)$ must be considered as arbitrary and not be interpreted as a physically measurable local temperature. In the relaxation-time approximation the collision term takes the form

$$\left(\frac{\partial N_{\lambda p}}{\partial t} \right)_{\text{collision}} = (\delta N_{\lambda p}(\mathbf{r}t) - \delta \tilde{N}_{\lambda p}(\mathbf{r}t)) / \tau_{\lambda p}^{(2)}, \quad (27)$$

which obviously satisfies Eq. (25). The quantity $\tau_{\lambda p}^{(2)}$ is the phonon collision time, and $\delta \tilde{N}$ is the deviation of the local equilibrium distribution from the complete thermal equilibrium function:

$$\begin{aligned} \delta \tilde{N}_{\lambda p}(\mathbf{r}t) &= \tilde{N}_{\lambda p}(\mathbf{r}t) - N_{\lambda p}^0(T_0) \\ &= N_{\lambda p}^0(T(\mathbf{r}t)) - N_{\lambda p}^0(T_0). \end{aligned} \quad (28)$$

Using Eq. (27), one finds that the Boltzmann equation in this case is

$$\begin{aligned} (\partial/\partial t + \mathbf{v}_{\lambda p} \cdot \nabla + 1/\tau_{\lambda p}^{(2)}) \delta N_{\lambda p}^{(2)}(\mathbf{r}t) &= A \delta^3(\mathbf{r}) \dot{\eta}_{\lambda p}(t) \\ &+ \delta \tilde{N}_{\lambda p}(\mathbf{r}t) / \tau_{\lambda p}^{(2)}. \end{aligned} \quad (29)$$

This equation differs from Eq. (14) for case 1 by having an extra term proportional to a yet undetermined local equilibrium distribution function. The solution to (29) can be immediately written as

$$\delta N_{\lambda p}^{(2)}(\mathbf{r}t) = \delta N_{\lambda p}^{(B)}(\mathbf{r}t) + \delta N_{\lambda p}^{(th)}(\mathbf{r}t). \quad (30)$$

The first term, which shall be referred to as the ballistic part, is given by

$$\begin{aligned} \delta N_{\lambda p}^{(B)}(\mathbf{r}t) &= \int d^3r' \int dt' g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t') \\ &\times A \delta^3(\mathbf{r}') \dot{\eta}_{\lambda p}(t'). \end{aligned} \quad (31)$$

This expression is identical to the result obtained for case 1 [Eq. (15)] except that $\tau_{\lambda p}^{(1)}$ has been replaced by $\tau_{\lambda p}^{(2)}$ (hence the change of superscript on the Green's function). One point to keep in mind is that $\delta N^{(B)}$ is completely determined once the source term is known. The other term in Eq. (30), which shall be called the thermal part, is

$$\begin{aligned} \delta N_{\lambda p}^{(th)}(\mathbf{r}t) &= \int d^3r' \int dt' g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t') \\ &\times \frac{1}{\tau_{\lambda p}^{(2)}} \delta \tilde{N}_{\lambda p}(\mathbf{r}'t') \\ &= \int d^3r' \int dt' g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t') \frac{1}{\tau_{\lambda p}^{(2)}} \\ &\times [N_{\lambda p}^0(T(\mathbf{r}t)) - N_{\lambda p}^0(T_0)]. \end{aligned} \quad (32)$$

In contrast to the ballistic term this expression is not completely determined because it still contains an unknown function $T(\mathbf{r}t)$. To solve for $T(\mathbf{r}t)$ one must now make use of the energy-conservation equation (21). The result is an integro-differential equation determining $T(\mathbf{r}t)$:

$$\frac{\partial}{\partial t} E^{(th)}(\mathbf{r}t) + \nabla \cdot \mathbf{J}_E^{(th)}(\mathbf{r}t) = P_{\text{eff}}(\mathbf{r}t), \quad (33)$$

where

$$E^{(th)}(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \delta N_{\lambda p}^{(th)}(\mathbf{r}t), \quad (34)$$

$$\mathbf{J}_E^{(th)}(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \mathbf{v}_{\lambda p} \delta N_{\lambda p}^{(th)}(\mathbf{r}t). \quad (35)$$

The function $P_{\text{eff}}(\mathbf{r}t)$ denotes the known effective power (density) driving the thermal part of the energy density and is given by

$$P_{\text{eff}}(\mathbf{r}t) = P_H(\mathbf{r}t) - \frac{\partial}{\partial t} E^{(B)}(\mathbf{r}t) - \nabla \cdot \mathbf{J}_E^{(B)}(\mathbf{r}t), \quad (36)$$

$$E^{(B)}(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \delta N_{\lambda p}^{(B)}(\mathbf{r}t), \quad (37)$$

$$\mathbf{J}_E^{(B)}(\mathbf{r}t) = \sum_{\lambda p} \hbar\omega_{\lambda p} \mathbf{v}_{\lambda p} \delta N_{\lambda p}^{(B)}(\mathbf{r}t). \quad (38)$$

For reference purposes, Eqs. (33) and (36) are written more explicitly as¹¹

$$\begin{aligned} \int d^3r' \int dt' \sum_{\lambda p} \left\{ \left[\hbar\omega_{\lambda p} \frac{\partial}{\partial t} g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t') + \hbar\omega_{\lambda p} \right. \right. \\ \left. \left. \times \mathbf{v}_{\lambda p} \cdot \nabla g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t') \right] \times \left[N_{\lambda p}^0(T(\mathbf{r}t)) \right. \right. \\ \left. \left. - N_{\lambda p}^0(T_0) \right] \right\} / \tau_{\lambda p}^{(2)} = P_{\text{eff}}(\mathbf{r}t), \end{aligned} \quad (39)$$

¹¹ We shall assume here that the solution for $T(\mathbf{r}t)$ is unique.

$$P_{\text{eff}}(\mathbf{r}t) = A \delta^3(\mathbf{r}) \sum_{\lambda p} \hbar \omega_{\lambda p} \dot{\eta}_{\lambda p}(t) + A \sum_{\lambda p} \hbar \omega_{\lambda p} \delta(x - \gamma_p z) \\ \times \delta(y - \gamma_p z) \exp\left(-\frac{z}{v_{\lambda p} \tau_{\lambda p}^{(2)}}\right) \left(\frac{1}{v_{\lambda p} \tau_{\lambda p}^{(2)}}\right) \\ \times \dot{\eta}_{\lambda p}\left(t - \frac{z}{v_{\lambda p}}\right). \quad (40)$$

Therefore, in the present case the phonon distribution consists first of all of a decaying ballistic part, characterized by an angular distribution which is entirely in the positive z direction as $\dot{\eta}_{\lambda p} = 0$ for $p_z < 0$. The phonon distribution also contains a thermal part which is gradually building up as a consequence of phonon energy conservation. Its space and time behavior is, in general, very complicated except in certain limiting cases considered in Sec. 4. One property of the thermal part that we may notice from Eq. (32) is that the angular distribution is quite isotropic, implying a backflow of the phonons which are generated originally only in the positive z direction.

We will now make a few remarks on the generalization of our present treatment to more complicated cases. First of all, let us suppose that the dominant phonon scattering mechanism is a normal process. Then besides energy conservation, Eq. (2), one also has phonon momentum conservation, i.e.,

$$\sum_{\lambda p} \hbar \mathbf{p} \left(\frac{\partial N_{\lambda p}}{\partial t} \right)_{\text{normal collision}} = 0. \quad (41)$$

This leads to a momentum density conservation law similar to Eq. (21) for the energy. Furthermore, the collision integral has the property of vanishing when the phonon distribution is equal to a "drifted" local equilibrium distribution:

$$\tilde{N}_{\lambda p}^{(d)}(\mathbf{r}t) = \{ \exp[\hbar \omega_{\lambda p} - \hbar \mathbf{p} \cdot \mathbf{V}(\mathbf{r}t) / k_B T(\mathbf{r}t)] - 1 \}^{-1}. \quad (42)$$

The vector function $\mathbf{V}(\mathbf{r}t)$ may be interpreted as a local phonon drift velocity if it is much smaller than the sound velocities. In the relaxation-time approximation, the collision integral is simply

$$-(\delta N_{\lambda p} - \delta \tilde{N}_{\lambda p}^{(d)}) / \tau_{\lambda p}^{(N)}. \quad (43)$$

Now the Boltzmann equation may be solved as before. The solution is similar to that of case 2, i.e., Eqs. (30)–(32), except that $\tau^{(2)}$ is replaced by $\tau^{(N)}$ and that the local equilibrium distribution function which appears in the thermal part is replaced by the drifted distribution $\tilde{N}^{(d)}$. Now there are four unknown functions, $T(\mathbf{r}t)$ and $\mathbf{V}(\mathbf{r}t)$, instead of one and we need the additional momentum-conservation equations (three components) to determine them.

Finally, we remark on the most general case in which all kinds of phonon scattering processes are present and are equally important. These different processes relax

the phonon distribution in different ways. However, in the relaxation-time treatment, no fundamental complications arise because the collision integral can be systematically decomposed into appropriate terms.¹² Then one can solve the Boltzmann equation as before and make use of the conservation equations (which may now contain damping terms) to determine $T(\mathbf{r}t)$ and $\mathbf{V}(\mathbf{r}t)$ if they have been introduced.

4. APPROXIMATION SOLUTIONS OF LOCAL TEMPERATURE FUNCTION

The exact solution to Eq. (40) for the local temperature function $T(\mathbf{r}t)$ is obviously impossible to obtain. We shall thus restrict ourselves to consider limiting cases for which approximate solutions can be calculated. First of all, let us substitute the explicit form of the Green's function $g_{\lambda p}^{(2)}(\mathbf{r}-\mathbf{r}', t-t')$ [Eq. (17) with $\tau_{\lambda p}^{(1)}$ replaced by $\tau_{\lambda p}^{(2)}$] into Eq. (39). We get

$$\sum_{\lambda p} \frac{\hbar \omega_{\lambda p}}{\tau_{\lambda p}^{(2)}} \delta \tilde{N}_{\lambda p}[T(\mathbf{r}t)] - \int_0^t dt' \sum_{\lambda p} \frac{\hbar \omega_{\lambda p}}{(\tau_{\lambda p}^{(2)})^2} \\ \times \exp\left(-\frac{t-t'}{\tau_{\lambda p}^{(2)}}\right) \times \delta \tilde{N}[T(\mathbf{r}-\mathbf{v}_{\lambda p}(t-t'), t')] \\ = P_{\text{eff}}(\mathbf{r}t), \quad (44)$$

where $\delta \tilde{N}$ is the function defined by Eqs. (26) and (28). If we use the formal solution (18) for the Green's function, Eq. (39) may be written alternatively as

$$\sum_{\lambda p} \left[1 + \tau_{\lambda p}^{(2)} \left(\frac{\partial}{\partial t} + \mathbf{v}_{\lambda p} \cdot \nabla \right) \right]^{-1} \left(\hbar \omega_{\lambda p} \frac{\partial}{\partial t} + \hbar \omega_{\lambda p} \mathbf{v}_{\lambda p} \cdot \nabla \right) \\ \times \delta \tilde{N}_{\lambda p}[T(\mathbf{r}t)] = P_{\text{eff}}(\mathbf{r}t). \quad (45)$$

These two expressions are, of course, equivalent but they are respectively more appropriate forms to be used for each of the two limiting cases considered below.

Case 1: Short-Time Limit

We first consider the behavior of $T(\mathbf{r}t)$ at times short compared to the relaxation time $\tau^{(2)}$ (some average of $\tau_{\lambda p}^{(2)}$). In this short-time limit Eq. (44) is the more useful expression to deal with. The reason is that the second term on the left-hand side may be neglected compared to the first since it is smaller by a factor of the order $t/\tau^{(2)}$. Therefore, we have approximately

$$\sum_{\lambda p} \frac{\hbar \omega_{\lambda p}}{\tau_{\lambda p}^{(2)}} \delta \tilde{N}_{\lambda p}[T(\mathbf{r}t)] \cong P_{\text{eff}}(\mathbf{r}t). \quad (46)$$

To solve for $T(\mathbf{r}t)$ we assume that the difference between $T(\mathbf{r}t)$ and the ambient temperature T_0 is small, i.e.,

$$\sigma(\mathbf{r}t) = (T(\mathbf{r}t) - T_0) / T_0 \ll 1. \quad (47)$$

¹² J. Callaway, Phys. Rev. 113, 1046 (1959).

Thus according to Eq. (28), the deviation $\delta\tilde{N}$ to linear order in σ is

$$\delta\tilde{N}_{\lambda\mathbf{p}}[T(\mathbf{r}t)] = S_{\lambda\mathbf{p}}^0\sigma(\mathbf{r}t), \quad (48)$$

$$S_{\lambda\mathbf{p}}^0 = N_{\lambda\mathbf{p}}^0(T_0)(1 + N_{\lambda\mathbf{p}}^0(T_0))(\hbar\omega_{\lambda\mathbf{p}}/k_B T_0).$$

Thus Eq. (46) becomes

$$\sigma(\mathbf{r}t) \cong \left(\sum_{\lambda\mathbf{p}} \frac{S_{\lambda\mathbf{p}}^0 \hbar\omega_{\lambda\mathbf{p}}}{\tau_{\lambda\mathbf{p}}^{(2)}} \right)^{-1} P_{\text{eff}}(\mathbf{r}t). \quad (49)$$

This expression says that the deviation of the local temperature function from T_0 is proportional to the effective power input into the thermal part of the distribution. A note of caution is appropriate here. One must not interpret $T(\mathbf{r}t)$ as the actual local temperature in the sense that it can be measured by an external thermometer. The phonon distribution still contains a ballistic part and the thermal part is still a long way from attaining a local equilibrium form. Thus one may regard $T(\mathbf{r}t)$ as a temporary theoretical construct by which one expresses the solution of the Boltzmann equation.

Case 2: Long-Time Limit

Next we turn our attention to obtaining an approximate solution to $T(\mathbf{r}t)$ at times long compared to $\tau^{(2)}$. By this time the ballistic part of $\delta\tilde{N}$ would have almost been completely attenuated. The solution in this limit is meaningful only if we are working with samples that are much longer than the mean free path $l^{(2)} = v\tau^{(2)}$. In this case, Eq. (45) is more appropriate to use. One assumes that $\delta\tilde{N}^{(th)}$ is relaxing towards the local equilibrium distribution $\delta\tilde{N}$ and, in the same time, the variation of $T(\mathbf{r}t)$ in space and time is becoming less rapid. When such a limit is approached, one may expand the inverse derivative operator in Eq. (45). Retaining only the leading terms in the expansion and using the linear approximation for $\delta\tilde{N}$, we obtain the familiar heat-conduction equation describing the diffusion of heat:

$$\frac{\partial}{\partial t} \sigma(\mathbf{r}t) - \frac{\kappa}{b} \nabla^2 \sigma(\mathbf{r}t) = -P_{\text{eff}}(\mathbf{r}t), \quad (50)$$

where

$$b = \sum_{\lambda\mathbf{p}} \hbar\omega_{\lambda\mathbf{p}} S_{\lambda\mathbf{p}}^0, \quad (51)$$

$$\kappa = \frac{1}{3} \sum_{\lambda\mathbf{p}} \hbar\omega_{\lambda\mathbf{p}} (v_{\lambda\mathbf{p}})^2 S_{\lambda\mathbf{p}}^0 \tau_{\lambda\mathbf{p}}^{(2)}. \quad (52)$$

The quantity κ is the thermal conductivity. In deriving Eq. (52) we have also assumed cubic symmetry. The nature of the solution to Eq. (50) needs no further discussion. In the present limit the function $T(\mathbf{r}t)$ does indeed represent the physically measurable local temperature. Let us remark that if the phonon collisions are completely normal, the equations determining the local temperature $T(\mathbf{r}t)$ and the local phonon drift velocity $\mathbf{V}(\mathbf{r}t)$ in the present limit will assume a damped

waveform rather than the diffusive form (50).¹³ This corresponds to the well-known second sound mode of heat or temperature propagation.

5. DETECTION OF HEAT PULSE

We shall now consider the detection of the heat pulse. The amount of heat flowing across the sample-detector interface can be calculated from the rate of phonon generation in the detector. Denoting the phonon distribution function of the detector by $n'_{\lambda\mathbf{p}}$ and the phonon transmission coefficient from the sample to the detector film by $\alpha'(\lambda\mathbf{p}, \lambda'\mathbf{p}')$, one finds the rate per unit area is

$$\left(\frac{\partial n'_{\lambda\mathbf{p}}}{\partial t} \right)_{\text{in}} = 0 \quad (p_z < 0)$$

$$= \sum_{\lambda'\mathbf{p}'} \alpha'(\lambda\mathbf{p}, \lambda'\mathbf{p}') |v_{\lambda'\mathbf{p}'\text{,eff}}| (p_z'/p') [\delta N_{\lambda'\mathbf{p}'}(\mathbf{r}t)] \quad (p_z > 0, \quad z=L), \quad (53)$$

where

$$|v_{\lambda'\mathbf{p}'\text{,eff}}| = |v_{\lambda'\mathbf{p}'}| \{ \tau_{\lambda'\mathbf{p}'\text{,eff}}^{(1,2)} [1 - \exp(-1/\tau_{\lambda'\mathbf{p}'\text{,eff}}^{(1,2)})] \}. \quad (54)$$

Equation (53) is, of course, similar to Eq. (6). The total input power is then given by

$$P_{\text{in}}(t) = \int_{A'} \int dxdy \sum_{\lambda\mathbf{p}} \hbar\omega'_{\lambda\mathbf{p}} \left(\frac{\partial n'_{\lambda\mathbf{p}}}{\partial t} \right)_{\text{in}}. \quad (55)$$

The integration is over the contact area A' between the sample and the detector (Fig. 1). This is the quantity that one hopes to measure directly. However, in the usual experimental situations, it is only possible to make indirect measurement of P_{in} , for example, by measuring the temperature change in the detector. Assuming that the rate of thermalization is fast, one can determine the time-dependent temperature $T_D(t)$ if one knows the total energy $E_D(t)$ as a function of T_D and the rate of energy loss. It turns out that, in most cases, the loss of heat is through the contact with the sample. In other words, the heat loss is due to reradiation of phonons back into the sample. From previous considerations, one can immediately write the expression as

$$\left(\frac{\partial}{\partial t} -E_D(t) \right)_{\text{loss}} = A' \times \sum_{\lambda} \alpha''(\lambda\mathbf{p}, \lambda'\mathbf{p}') \hbar\omega_{\lambda\mathbf{p}} [n_{\lambda\mathbf{p}}^{(0)'} \times (T_D(t) - n_{\lambda'\mathbf{p}'}^{(0)'}(T_0))] |v_{\lambda'\mathbf{p}'\text{,eff}}| (p_z'/p'), \quad (56)$$

where α'' is the transmission coefficient from the detector to the sample. We have also made use of the assumption of thermal equilibrium. Now Eqs. (55) and (56), together with the energy balance equation

$$\frac{\partial}{\partial t} -E_D(t) = \frac{\partial}{\partial t} E_D[T_D(t)] = P_{\text{in}}(t) - \left(\frac{\partial}{\partial t} -E_D(t) \right)_{\text{loss}}, \quad (57)$$

¹³ See, for example, P. C. Kwok, *Physics* 3, 221 (1967).

will enable us to determine $T_D(t)$, or $T_D(t) - T_0$, caused by the arrival of the heat pulse.

We have succeeded above in constructing a kinetic theory of the propagation of heat pulse in dielectric materials. Our result is equally applicable to the experimental situation in which the transit time of the heat pulse is short as well as long compared to the mean phonon collision time. However, it is obvious that for the intermediate time regime the result is so complicated that it seems very difficult to extract useful information from it. The simplest situation is when the heat flow is mainly ballistic in nature. In this case the result is straightforward and various properties of the phonons can be studied directly. For example, one can study the dispersion and damping of high-frequency phonons which cannot be achieved by ordinary ultrasonic means. It is possible in heat-pulse experiments because the phonons injected into the sample are predominantly at the thermal frequency of the heater (see Sec. 2). Thus by varying the power input into the heater, i.e., changing the heater temperature $T_H(t)$, one can excite various high-frequency phonons in the sample.

Our result in the long-time limit looks simple also. However, it must be borne in mind that we have considered cases in which only one scattering mechanism dominates the phonon collisions. When several are present, the result becomes more complicated and depends rather intricately on the relative importance of different relaxation times. Furthermore, one may now have to consider the arrival of those portions of the heat pulse that have suffered reflections off the boundary surfaces in the x and y directions.

We will conclude by commenting on the feasibility of measuring second sound in solids by the heat-pulse method. In principle, this can be done if the following conditions are satisfied. First of all, one should work at a temperature low enough that the umklapp processes are negligible compared to the normal processes. Secondly, the sample should be as pure and free of dislocations and other types of defects as possible (the ideal case is a pure single crystal). These two conditions are intended to reduce the momentum nonconserving phonon collision processes which damp the second-sound mode. Finally, the length of the sample must be very long compared to the average mean free path of the normal processes to ensure adequate thermalization of the phonon distribution necessary for the appearance of the collective second-sound mode. These conditions are extremely difficult (nearly impossible) to satisfy in practice for most solids (with the obvious exception of solid helium¹⁴). Thus the measurement of second sound in solids by heat-pulse methods remains an experimental challenge.

ACKNOWLEDGMENTS

The author wishes to thank Dr. N. Shiren, Dr. M. Pomerantz, Dr. R. von Gutfeld, Dr. P. Marcus, and Dr. T. Schultz for many valuable discussions at the various stages of the work and for their reading of the manuscript.

¹⁴ C. C. Ackerman, B. Bertman, H. A. Fairbank, and R. A. Guyer, *Phys. Rev. Letters* **16**, 789 (1966).