

## Analysis of Phonon Conductivity: Application to Si

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Callaway's model of lattice thermal conductivity has been modified by introducing an arbitrary relation between the phonon frequency and the phonon wave vector, to take account of (i) the phonon dispersion, and (ii) the number of phonon states in the wave-vector space, allowing for separate contributions of the transverse and the longitudinal phonons. Three-phonon relaxation times that have different temperature dependence for different temperature ranges have been used. Application to silicon has been made successfully. It is observed that the four-phonon processes play an important role at high temperatures, and that the major contribution to the lattice thermal conductivity comes from the transverse phonons.

### INTRODUCTION

A SATISFACTORY formulation of the lattice thermal conductivity of solids at low temperatures has been given by Callaway.<sup>1</sup> On account of its simple form, many authors<sup>2-7</sup> have studied and applied his model to a number of thermal conductivity data. The success of his model lies in the fact that the Debye approximation for the phonon frequency  $\omega$  as a function of the phonon wave vector  $q$ , is a valid assumption at low temperatures, where only low-frequency phonons are excited. At high temperatures, however, high-frequency phonons whose dispersive nature cannot be ignored, become more important in thermal transport. Moreover, a distinction between the longitudinal and the transverse polarization branches is also required to be made in view of the possibly different relaxation times for the corresponding phonons. Taking these two points into consideration, Holland<sup>8</sup> modified the Callaway model, making it applicable at all temperatures, and applied it to silicon and germanium quite successfully. His formulation has drawn the attention of many authors who have applied it to a number of materials.<sup>9-11</sup>

Holland<sup>8</sup> has taken account of the phonon dispersion by considering the longitudinal and the transverse polarization branches separately, and by using suitably averaged values of the phonon velocity for the low- and the high-frequency phonons (the two transverse polarization branches are assumed to be degenerate).

On account of the strong temperature dependence of the phonon distribution function, one does not expect such a weighted averaged value of the phonon velocity to be a constant at all temperatures. According to Holland,<sup>8</sup> the low-frequency phonons have an average velocity that is more than two times greater than that of the high-frequency phonons in silicon. We shall see later that such a large difference between the two is not justified. In fact, a proper distinction between the phonon group and phase velocities is required to be made for at least high-frequency phonons, and in this regard the phonon velocity bears an ambiguous meaning.

Still more important is the fact that even at present, we lack exact analytical expressions for the different phonon relaxation times. On account of the complex structure of the Brillouin zone and the strong temperature dependence of the phonon distribution function, the relaxation time expressions have a complicated dependence on the phonon frequency and the temperature  $T$ . These expressions cannot be of much help in analyzing the thermal conductivity of solids even if their exact functional forms are known. For practical purposes, it is desirable to express the phonon frequency and temperature dependence of the relaxation times by simple exponents. This is not a very serious problem at low temperatures, where the phonon scattering due to crystal boundaries, and to isotopes and point defects, etc., is more important, since the corresponding relaxation times are sufficiently accurately known. At high temperatures, the three-phonon processes dominate over the others, and one has to be careful about the accuracy of the corresponding relaxation time expressions. Guthrie<sup>12</sup> has suggested that the temperature dependence of the three-phonon relaxation times can be effectively expressed by  $T^{-m}$ , where  $m$  is an exponent that is a function of  $T$ . It is difficult to express  $m$  as a function of temperature and to use it in the analysis of thermal conductivity. One, therefore, has to seek an

<sup>1</sup> J. Callaway, Phys. Rev. **113**, 1046 (1959).

<sup>2</sup> J. Callaway and M. C. V. Baever, Phys. Rev. **120**, 1149 (1960).

<sup>3</sup> A. M. Toxen, Phys. Rev. **122**, 450 (1961).

<sup>4</sup> B. K. Agrawal and G. S. Verma, Phys. Rev. **126**, 24 (1962).

<sup>5</sup> B. K. Agrawal and G. S. Verma, Phys. Rev. **128**, 603 (1962).

<sup>6</sup> B. K. Agrawal and G. S. Verma, Physica **28**, 599 (1962).

<sup>7</sup> C. T. Walker and R. O. Pohl, Phys. Rev. **131**, 1433 (1963).

<sup>8</sup> M. G. Holland, Phys. Rev. **132**, 2461 (1963).

<sup>9</sup> C. M. Bhandari and G. S. Verma, Phys. Rev. **138**, A288 (1965).

<sup>10</sup> C. M. Bhandari and G. S. Verma, Phys. Rev. **140**, A2101 (1965).

<sup>11</sup> Y. P. Joshi, M. D. Tiwari, and G. S. Verma, Phys. Rev. (to be published).

<sup>12</sup> G. L. Guthrie, Phys. Rev. **152**, 801 (1966).

alternative. It should be noticed that Holland<sup>8</sup> has not made use of this fact.

In the present work, we have assumed a quadratic expression for the phonon wave vector as a function of the phonon frequency so as to be able to express the phonon group and phase velocities as functions of the phonon frequency. We have assumed that the Brillouin zone is spherically symmetric, and that the two transverse polarization branches are degenerate. We also define different temperature ranges such that for each range, one constant value of  $m$  is used in the three-phonon relaxation time expressions. An application of the formulation has been made to silicon, getting good agreement with the experimental results. It has been found necessary to include the four-phonon relaxation times in the thermal conductivity analysis of silicon at temperatures above 400°K.

### FORMULATION

Callaway<sup>1</sup> expressed the lattice thermal conductivity as a sum of two terms. One of the terms, which we shall refer to as the first one, consists of a single integral and has the structure that one would have obtained if one had ignored the fact that the normal processes by themselves do not cause any thermal resistivity, and had added the various inverse relaxation times as usual. The second term, usually called the correction term, has a much more complicated form and is obtained because the normal and umklapp processes behave differently. However, Callaway has shown that the correction term is usually small compared to the first term at low temperatures, where the phonon scattering due to the crystal boundaries and isotopes, etc., is important. We believe that the three-phonon umklapp processes dominate over the normal ones at high temperatures, so that the correction term is again small. We shall, therefore, neglect the correction term at all temperatures, although it definitely forms a finite fraction of the total. The contribution of the optical phonons, if any, is also assumed to be negligible. We further assume that the Brillouin zone is spherically symmetric, and that the three polarization branches, one longitudinal and two transverse, contribute separately to the thermal conductivity. The contribution of each branch can be given by an expression of the form

$$K_i = \frac{1}{6\pi^2} \int \tau_c v_g^2 \frac{\hbar^2 \omega^2}{kT^2} \frac{e^{\hbar\omega/kT}}{(e^{\hbar\omega/kT} - 1)^2} q^2 dq, \quad (1)$$

where the integration is performed over the first Brillouin zone. Here  $\tau_c$  is the total relaxation time, which will be defined later, and  $v_g$  is the phonon group velocity corresponding to the polarization branch under consideration.

In real crystals, the two transverse polarization branches are degenerate in only certain symmetry directions. Nevertheless, the phonon frequencies of the two

branches are usually very near to each other and much different from the corresponding longitudinal phonon frequency for most of the directions of the wave vector in the reciprocal lattice space; it is often a good assumption to take the transverse polarization branches to be degenerate. The contribution of either of the transverse polarization branches is then given by the same expression.

It is desirable to express Eq. (1) as an integral in terms of the phonon frequency  $\omega$ . A convenient approximation that is often used is to assume a linear relation between  $\omega$  and  $q$ , which is, in fact, far from reality.  $\omega$  is generally a function of the magnitude as well as the direction of the wave vector  $\mathbf{q}$ . Under the present assumption that the Brillouin zone is spherically symmetric,  $\omega$  is a function of  $q$  only. Looking at Eq. (1), one finds it convenient to express  $q$  as a function of  $\omega$ . This function has a form that is a characteristic of the crystal, and is determined by its structure and constituting atoms. Thus, for a monoatomic one-dimensional crystal lattice with nearest-neighbor force constant<sup>13</sup> only,  $\omega$  is a sine function of  $q$ . Experimental studies<sup>14-17</sup> of the dispersion curves show that the phonon frequency  $\omega$  is generally an increasing function of  $q$ , with the  $\omega$ - $q$  curve gradually bending towards the  $q$  axis. There are, however, exceptions to this rule.<sup>18</sup> It appears that it is possible to express  $q$  as a polynomial in  $\omega$ . In order to avoid the determination of the parameters that would be involved, and the complication that would arise in evaluating Eq. (1), we shall write  $q$  as a very simple function of  $\omega$ . Since we are interested in showing how such an approximation leads to better results, we assume

$$q = \omega v^{-1} (1 + \alpha \omega). \quad (2)$$

An equally good choice is to take a cubic term instead of the quadratic one in Eq. (2). This would modify the results to some extent, but the main features would remain of the same nature. Moreover, whether a quadratic or higher degree term should be taken in Eq. (2) will depend upon what choice proves to be the best representative of the observed phonon dispersion.

It is evident that in Eq. (2), the constant  $v$  is the low-frequency phonon velocity corresponding to the polarization branch under consideration. The constant  $\alpha$  is evaluated by using the fact that the total number of phonon states per polarization branch in a given crystal

<sup>13</sup> J. de Launey, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2, p. 237.

<sup>14</sup> B. N. Brockhouse, in *Aarhus Summer School Lectures, 1963* edited by Thor A. Bak (W. A. Benjamin, Inc., New York, 1964), p. 221.

<sup>15</sup> P. S. Yen and Y. P. Varshni, *Phys. Rev.* **164**, 895 (1967).

<sup>16</sup> J. L. Waren, J. L. Yarnell, G. Dolling, and R. A. Cowley, *Phys. Rev.* **158**, 805 (1967).

<sup>17</sup> W. J. L. Buyers, *Phys. Rev.* **153**, 923 (1967).

<sup>18</sup> A. A. Maradudin, in *Aarhus Summer School Lectures, 1963* edited by Thor A. Bak (W. A. Benjamin, Inc., New York, 1964), pp. 466, 499.

is equal to the number of the lattice points available. If  $n$  is the number of lattice points per unit volume of the crystal, then one can show that

$$\alpha = \omega_m^{-1} [(6n\pi^2)^{1/3} v / \omega_m - 1], \quad (3)$$

where the subscript  $m$  denotes the maximum value of  $\omega$ . Subscripts  $L$  and  $T$  should be attached to  $\alpha$ ,  $v$ , and  $\omega_m$  in Eq. (3) to specify the polarization branches.

The phonon phase velocity  $v_p$  and the phonon group velocity  $v_g$  are given by  $\omega/q$  and  $\partial\omega/\partial q$ , respectively. Using Eq. (2), one can express  $v_p$  and  $v_g$  in terms of  $\omega$ . We rewrite Eq. (1) as

$$K_i = \frac{k^4 T^3}{6\pi^2 \hbar^3} \int_0^{\Theta/T} \frac{v_g}{v_p^2} \frac{x^4 e^x}{(e^x - 1)^2} dx, \quad (4)$$

where  $x = \omega \hbar / kT$  and  $\Theta = \omega_m \hbar / k$ . Equation (4) differs from that given by Holland<sup>8</sup> in that here we have  $v_g/v_p^2$  instead of  $1/v$ . It is evident that  $v_p^2/v_g$  has the dimensions of velocity and is a constant for a given phonon. But it is not proper to call  $v_p^2/v_g$  the phonon velocity, since it is not a velocity in the ordinary sense. According to Holland,  $v$ , the phonon velocity, which occurs in place of  $v_p^2/v_g$ , is a sufficiently rapidly decreasing function of  $\omega$  such that its averaged value for low-frequency phonons is more than two times larger than that for the high-frequency phonons. Since  $v_g$  decreases more rapidly than  $v_p$  as  $\omega$  increases, one cannot expect the high-frequency value of  $v_p^2/v_g$  to be very small compared to the low-frequency value. This point is clear at least for those phonons for which  $v_g$  tends to zero. Using Eq. (2), one can express  $v_p^2/v_g$  in terms of  $\omega$  and find that this does not vary rapidly with  $\omega$ . We have found that  $v_p^2/v_g$  is almost a constant for the longitudinal phonons, and that its value at  $\omega = \omega_m$  is about one and a half times the low-frequency value for the transverse phonons in silicon.

Finally, from Eqs. (2) and (4) one obtains

$$K_i = \frac{k^4 T^3}{6\pi^2 v \hbar^3} \int_0^{\Theta/T} \frac{x^4 e^x}{(e^x - 1)^2} \frac{(1 + \beta T x)^2}{(1 + 2\beta T x)} dx, \quad (5)$$

where  $\beta = \alpha k / \hbar$ .

### RELAXATION TIMES

There can be many phonon-scattering processes that lead to thermal resistivity in semiconductors. It has been shown that the thermal conductivity at the lowest temperatures can be explained very well on the basis of the boundary scattering.<sup>19-22</sup> According to Casimir,<sup>19</sup> the boundary scattering relaxation time  $\tau_b$

is given by  $L/v$ , where  $v$  is the phonon velocity and  $L$  is a characteristic length associated with the specimen under study. Since boundary scattering is important for low-frequency phonons, it is immaterial whether  $v$  is the group or phase velocity. It is sufficient to use a low-frequency value for  $v$ . Holland has used the same weighted averaged value of  $v$  for all the polarization branches. There seems, however, no reason why  $\tau_b$  should be the same for all phonons. Studying Casimir's derivation, we believe that it is a better approximation to use the respective low-frequency velocity for the different polarization branches. That this is a better choice also appears from the recent work of Hamilton and Parrott.<sup>23</sup> The characteristic length  $L$  is determined by crystal dimensions and is assumed to be the same for all phonons. The value of  $L$  given by theory usually does not give a good agreement with experiment. It is better to treat  $L$  as an unknown parameter which is assigned the value that gives the best fit to the experimental results. This value differs from that given by theory by a factor approximately equal to unity.

The scattering due to isotopes, point-defects, etc., is the most important relaxation process at temperatures about the conductivity maximum. At such temperatures, the high-frequency phonons are not excited to a large extent, and it is reasonable to use  $A\omega^4$  for the corresponding inverse relaxation time  $\tau_{pt}^{-1}$ . This expression was initially obtained by Klemens,<sup>24</sup> and is particularly valid for low-frequency phonons.<sup>25,26</sup> A careful analysis of Klemens's approach shows that  $A$  should increase with frequency, and that it should be considerably different for that longitudinal phonon whose frequency lies above the maximum of the transverse polarization branches. As the longitudinal phonons are found to contribute little<sup>23</sup> to the thermal conductivity, one need not take this possibility into consideration.

The three-phonon processes dominate over the others at high temperatures. These processes are not negligibly small at low temperatures, and play an important part even in the vicinity of the conductivity maximum. It is difficult to give a simple expression for the three-phonon relaxation time  $\tau_{3ph}$ . In general,  $\tau_{3ph}$  is different for the longitudinal and the transverse phonons. Many authors<sup>8,27-30</sup> have calculated approximate expressions for  $\tau_{3ph}$ . Their results suggest that it is a reasonably good approximation to assume the frequency dependence of  $\tau_{3ph}^{-1}$  to be  $\omega$  and  $\omega^2$  in the

<sup>23</sup> R. A. H. Hamilton and J. E. Parrott, *Phys. Rev.* **178**, 1284 (1969).

<sup>24</sup> P. G. Klemens, *Proc. Phys. Soc. (London)* **68**, 1113 (1955).

<sup>25</sup> B. K. Agrawal, *J. Phys.* **C2**, 252 (1969).

<sup>26</sup> P. C. K. Kwok, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic Press Inc., New York, 1966), Vol. 20, p. 214.

<sup>27</sup> P. G. Klemens, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1958), Vol. 7.

<sup>28</sup> P. G. Klemens, *Phys. Rev.* **119**, 507 (1960).

<sup>29</sup> C. Herring, *Phys. Rev.* **95**, 954 (1954).

<sup>30</sup> P. G. Klemens, *Proc. Roy. Soc. (London)* **A208**, 108 (1957).

<sup>19</sup> H. B. G. Casimir, *Physica* **5**, 595 (1938).

<sup>20</sup> R. Berman, F. E. Simon, and J. M. Ziman, *Proc. Phys. Soc. (London)* **A220**, 171 (1953).

<sup>21</sup> R. Berman, E. L. Foster, and J. M. Ziman, *Proc. Phys. Soc. (London)* **A231**, 130 (1955).

<sup>22</sup> P. Carruthers, *Rev. Mod. Phys.* **33**, 92 (1961).

case of transverse and longitudinal phonons, respectively. We have neglected the exponential factor in the expression for  $\tau_{3\text{ph}}$  given by Klemens,<sup>30</sup> and assume that the normal and umklapp processes have the same frequency dependence. Truly speaking, the thermal conductivity is not very sensitive to the frequency dependence of  $\tau_{3\text{ph}}$  particularly at high temperatures. This is evident if one neglects all the other relaxation times at high temperatures and evaluates Eq. (5). On the other hand, the thermal conductivity is very much sensitive to the temperature dependence of  $\tau_{3\text{ph}}^{-1}$ . Guthrie<sup>12</sup> has pointed out that a fixed temperature dependence of the type  $T^3$  or  $T^4$  is valid for low temperatures only, and that it changes as the temperature does. He has shown that  $\tau_{3\text{ph}}^{-1}$  can be effectively taken proportional to  $T^m$ , where  $m$  is an exponent that depends on the temperature. The low-temperature values of  $m$  are 3 and 4 for the longitudinal and the transverse phonons, respectively, and  $m$  tends to unity at high temperatures,<sup>31</sup> irrespective of the phonon polarization state and the nature of the process, normal or umklapp. These results are in contradiction to Holland's<sup>8</sup> assumption. It is very difficult to determine an exact variation of  $m$  with temperature. A possible approximation, crude yet instructive, is to define temperature ranges for each of which a constant value of  $m$  may be taken as valid. This apparently leads to a large number of parameters. These parameters are not arbitrary, however, since the relaxation time expressions should be smoothly joined at the temperatures that define the temperature ranges. Thus, a single parameter is associated with  $\tau_{3\text{ph}}$ .

It has been observed<sup>31,32</sup> that at temperatures of the order of 1000°K, the thermal conductivity falls with temperature more rapidly than expected if only three-phonon processes are considered. Such behavior of the thermal conductivity can be explained in terms of an additional phonon-scattering process that is more strongly temperature-dependent than the three-phonon processes. We believe that the four-phonon processes play an important role at high temperatures.<sup>32,33</sup> This point was first suggested by Pomeranchuk<sup>34-36</sup> who calculated  $\tau_{4\text{ph}}$ , the four-phonon relaxation time, given by

$$\tau_{4\text{ph}}^{-1} = B_H \omega^2 T^2. \quad (6)$$

The total relaxation time  $\tau_c$  is now given by

$$\tau_c^{-1} = \tau_B^{-1} + \tau_{\text{pt}}^{-1} + \tau_{3\text{ph}}^{-1} + \tau_{4\text{ph}}^{-1}, \quad (7)$$

where the  $\tau$ 's have been defined above. In order to specify the polarization branches, one has to attach subscripts  $L$  and  $T$  to the terms occurring in Eq. (7).

In view of the assumed degeneracy of the transverse

branches, the total lattice thermal conductivity  $K$  is given by

$$K = K_L + 2K_T. \quad (8)$$

Using Eqs. (5) and (7), explicit expressions for  $K_L$  and  $K_T$  can be written. We notice that  $K$  involves only two integrals corresponding to  $K_L$  and  $K_T$ .

### APPLICATION TO SILICON

The lattice thermal conductivity of silicon has been analyzed by numerical integration using the relaxation times listed in Table I. An order-of-magnitude estimation of the parameters is obviously not difficult. An exact estimation, however, requires simultaneous consideration of all the relaxation times, and one has to perform complicated integrations numerically. We have done this to obtain a final set of parameters that lead to the best fit to the experimental results.<sup>33,37-39</sup> Table II lists all the parameters used. Holland has already given values of  $\Theta_L$  and  $\Theta_T$ , making explicit use of the phonon dispersion in silicon.<sup>40</sup> He has also calculated the low-frequency phonon velocities,  $v_L$  and  $v_T$ , using elastic constants of silicon. We have made use of the atomic weight  $a$  and density  $\rho$  of silicon, and of Avogadro's number, to calculate the number of lattice points per unit volume of silicon crystal, keeping in mind that there are two atoms per lattice point. Finally,  $\alpha_L$  and  $\alpha_T$  are calculated making use of Eq. (3).

The low-temperature data of Holland and Neuringer<sup>38</sup> require a value of  $L$  that is slightly different than that given by theory (Table II). Glassbrenner and Slack<sup>33</sup> have reported a theoretical value of  $L$  that is an order of magnitude larger than what our analysis requires.<sup>41</sup>

TABLE I. Inverse relaxation time expressions used in the analysis of the phonon conductivity of silicon.

Type of scattering	Symbol	Transverse	Longitudinal	Temperature range (°K)
Boundary <sup>a</sup>	$\tau_b^{-1}$	$v_T/L$	$v_L/L$	all temperatures
Isotope, etc. <sup>b</sup>	$\tau_{\text{pt}}^{-1}$	$A\omega^4$	$A\omega^4$	all temperatures
Three-phonon <sup>c</sup>	$\tau_{3\text{ph}}^{-1}$	$B_T\omega T^4$	$B_L\omega^2 T^3$	$T \leq 43$
		$B_{T1}\omega T^3$	$B_{L1}\omega^2 T^3$	$43 \leq T \leq 190$
		$B_{T2}\omega T^2$	$B_{L1}\omega^2 T^2$	$190 \leq T \leq 280$
		$B_{T3}\omega T$	$B_{L2}\omega^2 T$	$280 \leq T$
Four-phonon <sup>d</sup>	$\tau_{4\text{ph}}^{-1}$	$B_H\omega^2 T^2$		High temperatures

<sup>a</sup> See Ref. 19.

<sup>b</sup> See Ref. 24.

<sup>c</sup> See Refs. 8, 27-30; the temperature dependence of the three-phonon relaxation times is based on the results of Ref. 12.

<sup>d</sup> See Refs. 34, 35.

<sup>37</sup> H. R. Shanks, P. D. Maycock, P. H. Sidles, and G. C. Danielson, Phys. Rev. **130**, 1743 (1963).

<sup>38</sup> M. G. Holland and L. J. Neuringer, in *Proceedings of the International Conference on the Physics of Semiconductors, Exeter, 1962* (The Institute of Physics and The Physical Society, London, 1962), p. 475.

<sup>39</sup> W. Fulkerson, J. P. Moore, R. K. Williams, R. S. Graves, and D. L. McElroy, Phys. Rev. **167**, 765 (1968).

<sup>40</sup> B. N. Brockhouse, Phys. Rev. Letters **2**, 256 (1959).

<sup>41</sup> The reported value of  $L$  appears to suffer from a misprint. This value is so large that its use in the analysis leads to a particularly large phonon conductivity at low temperature, with the result that one is compelled to seek some extra scattering mechanism, which we do not believe to exist.

<sup>31</sup> E. F. Steigmeier and I. Kudman, Phys. Rev. **141**, 767 (1966).

<sup>32</sup> E. F. Steigmeier and I. Kudman, Phys. Rev. **132**, 508 (1963).

<sup>33</sup> C. J. Glassbrenner and G. A. Slack, Phys. Rev. **134**, A1058 (1964).

<sup>34</sup> I. Pomeranchuk, Phys. Rev. **60**, 820 (1941).

<sup>35</sup> I. Pomeranchuk, J. Phys. USSR **4**, 259 (1941).

<sup>36</sup> I. Pomeranchuk, J. Phys. USSR **7**, 197 (1942).

TABLE II. Parameters used in the analysis of the phonon conductivity of silicon.

$a$ (at. wt)	28.1
$\rho$ (density)	$2.33 \text{ g cm}^{-3}$ <sup>a</sup>
$\Theta_T$	$210^\circ\text{K}$ <sup>b</sup>
$\Theta_L$	$570^\circ\text{K}$ <sup>b</sup>
$v_T$	$5.86 \times 10^5 \text{ cm sec}^{-1}$ <sup>b</sup>
$v_L$	$8.48 \times 10^5 \text{ cm sec}^{-1}$ <sup>b</sup>
$\alpha_T$	$51.9 \times 10^{-15} \text{ sec}$
$\alpha_L$	$3.94 \times 10^{-15} \text{ sec}$
$L^\circ$	$2.600(0.295) \text{ cm}^d$
$L^\circ$	$0.716(0.660) \text{ cm}^d$
$A$	$0.248 \times 10^{-44} \text{ sec}^3$
$B_T$	$4.17 \times 10^{-13} \text{ deg}^{-4}$
$B_L$	$1.54 \times 10^{-23} \text{ sec deg}^{-3}$
$B_H$	$1.70 \times 10^{-20} \text{ sec deg}^{-2}$

<sup>a</sup> See Ref. 39.

<sup>b</sup> See Ref. 8.

<sup>c</sup> See Ref. 33.

<sup>d</sup> See Ref. 41; the bracketed value is experimental and the other is theoretical.

<sup>e</sup> See Ref. 38.

The evaluation of  $A$  is often carried out by binomially expanding the expression for  $\tau_c$  at low temperatures, treating  $\tau_{pt}^{-1}$  as a small quantity compared to  $\tau_b^{-1}$ . It is a question of common reasoning that such expansion is not valid even at low temperatures, since the inequality  $\tau_{pt}^{-1} < \tau_b^{-1}$  does not hold for all values of  $x$  lying within the integration limits. A binomial expansion

is not valid particularly when the upper integration limit is replaced by infinity. What one can do is to evaluate the integrals numerically without any approximation of this type. Nevertheless, a binomial expansion of  $\tau_c$  is valid at high temperatures, where  $\tau_{pt}^{-1} < \tau_{3ph}^{-1}$  is satisfied for values of  $x$  within the integration limits. The value of  $A$  given in Table II leads to a good agreement with the data of Glassbrenner and Slack.<sup>33</sup> The dashed curve in Fig. 1 is the fit with the data of Holland and Neuringer.<sup>38</sup> The agreement is poor. We have observed that a 20% lower value of  $A$  proves satisfactory for these data. The difference between the two values of  $A$  that the two sets of data require may be due to the different crystalline structures and impurity contents of the corresponding specimens.

It has been observed that good agreement with experimental results is obtained only up to about  $400^\circ\text{K}$ , if one does not consider the four-phonon processes. The dotted curve in Fig. 1 shows the possible behavior of  $K$  in the absence of four-phonon processes. By taking  $\tau_{4ph}$  into consideration, one can obtain agreement with experiment up to temperatures as high as  $1400^\circ\text{K}$ .

We have found that the agreement of theory with experiment requires a small contribution of the longitudinal phonons. At low temperatures,  $K_L$  is about

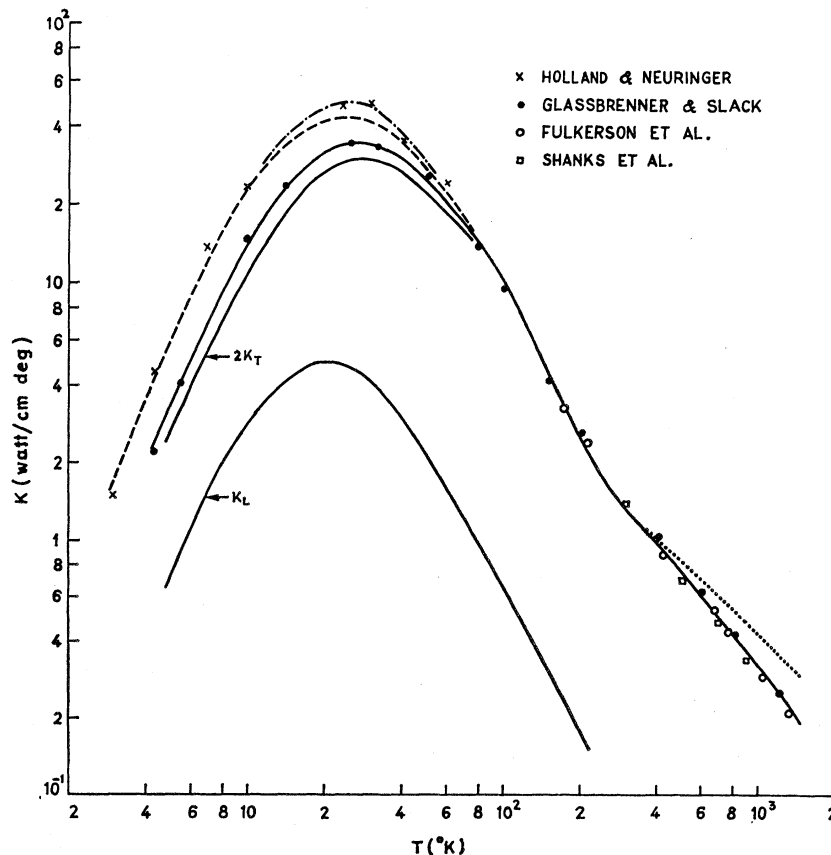


FIG. 1. Lattice conductivity of silicon. The solid curves are the analysis of the data of Glassbrenner and Slack. Curves marked with  $K_L$  and  $2K_T$  represent the contributions of the longitudinal and the transverse phonons, respectively. The third solid curve represents the total lattice conductivity and is equal to  $K_L + 2K_T$ . The dashed and dot-dashed curves are the analysis of the data of Holland and Neuringer. The dashed curve corresponds to a value of  $A$  equal to that given in Table II; and the dot-dashed curve, to a 20% lower value. The dotted curve shows the trend of the theoretical results in the absence of four-phonon processes.

one-fourth of  $2K_T$ , the transverse contribution. Such a result is expected because of the fact that there are two transverse branches and that the phonon velocity  $v_T$  is smaller than  $v_L$ . In other words, the transverse phonon states are more densely packed in energy space than the longitudinal phonon states. At high temperatures,  $K_L$  is below 10% of the corresponding transverse contribution. These results are in agreement with the recent results of Hamilton and Parrott<sup>23</sup> in the case of germanium. That the transverse phonons give a major contribution to lattice thermal conductivity was also pointed out earlier by Parrott.<sup>42</sup> It should be noticed that in Holland's analysis, the longitudinal contribution is small compared to the transverse one at low and high temperatures only. At about 100°K, the two are comparable. In Fig. 1, we have plotted  $K_L$  and  $2K_T$  for comparison. It will be noticed that at high temperatures, the two have almost the same slope, and it is not possible to decide from the high-temperature thermal conductivity data how much one or the other of the two contributes.

Finally, one point that deserves attention is that it is difficult to simplify the integral in Eq. (5) to express  $K$  as a simple function of temperature. This is because of the fact that each of the relaxation times is dominant over the others at one temperature or the other. At high temperatures, one can approximate  $x^2 e^x / (e^x - 1)^2$  to unity for  $x < 1$ . The boundary scattering may be

<sup>42</sup> J. E. Parrott, Proc. Phys. Soc. (London) **81**, 726 (1961).

neglected. The integrals so obtained can easily be evaluated analytically.

### CONCLUSION

The most significant point of the present analysis is the introduction of an arbitrary relation between the phonon frequency and the phonon wave vector. We do so because we believe that it is possible to find some empirical relation between  $\omega$  and  $q$  that represents the phonon dispersion in a crystal most effectively so far as thermal conductivity is concerned. We have indicated that the most important relaxation process is the three-phonon one, and that one has to be particularly careful about its temperature dependence. In the present case we have written the lattice conductivity as the sum of two integral expressions rather than three, as done by Holland,<sup>8</sup> to represent the separate contributions of the longitudinal and transverse phonons. It is recalled that the transverse phonons give a major contribution to the lattice conductivity of silicon at all temperatures.

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