Lattice Thermal Conductivity of Si in the Temperature Range (2–1400) °K

K. S. Dubey and G. S. Verma
Physics Department, Banaras Hindu University, Varanasi-5, India
(Received 17 April 1972)

In the present paper we have determined the temperature dependence of the three-phonon relaxation rate, which is valid for the entire temperature range (2–1400) °K for Si and is expressed as $\tau_{3ph}^{-1} \propto T^{m(T)} e^{-\theta/\alpha T}$ The exponent m, which depends upon temperature, is determined with the help of Guthrie's expression as given in the Sharma–Dubey–Verma model. Using the SDV model we have calculated the lattice thermal conductivity of Si in the temperature range (2–1400) °K and found excellent agreement between experimental and calculated results.

Recently Sharma, Dubey, and Verma (SDV)[1,2] proposed a model for the phonon conductivity of insulators. In the SDV model the three-phonon relaxation rate is of the form $\tau_{3ph}^{-1} \propto g(\omega) T^{m(T)} e^{-\theta/\alpha T}$. The exponent m is a continuous function of temperature. This idea is borrowed from Guthrie's[3,4] work. Guthrie has also shown that both normal and umklapp processes are bounded by the same temperature dependence. In the SDV model there is no explicit distinction between normal and umklapp processes. However, in the present model, a distinction is made between longitudinal phonons and transverse phonons and also between class-I and class-II processes. In class-I three-phonon scattering events, the carrier phonon is annihilated by combination, and in class-II the annihilation takes place by splitting. Following Klemens,[5] $g(\omega) = \omega^2$ for longitudinal phonons and $g(\omega) = \omega$ for transverse phonons. For the sake of simplicity of calculation, the same frequency dependence is taken for both class-I and class-II scattering processes. As a matter of fact, in the high-temperature region phonon conductivity is not very sensitive to the frequency dependence of τ_{3ph}^{-1}. However, it is very sensitive to the temperature dependence of τ_{3ph}^{-1}. Class-II events occur only for longitudinal phonons and are more frequent in the high-temperature region. For high-frequency phonons, the phonon-density space available for splitting events is larger than for events which consist of annihilation by combination. Thus in the high-temperature region, $[\tau_{3ph}^{-1}]_{L,II} \gg [\tau_{3ph}^{-1}]_{L,I}$ in the combined expression $[\tau_{3ph}^{-1}]_L = [\tau_{3ph}^{-1}]_{L,I} + [\tau_{3ph}^{-1}]_{L,II}$.

The lattice thermal conductivity of Si has been described recently by Joshi, Tewari, and Verma[6] and by Joshi and Verma.[7] Detailed references of earlier work are given in these articles. Holland[8] distinguished between longitudinal and transverse phonons and used the following form of three-phonon relaxation rate:

$[\tau_{3ph}^{-1}]_L = B_L \omega^2 T^3$ for the entire temperature range up to 1300 °K,

$[\tau_{3ph}^{-1}]_{Tu} = 0$ for $\omega < \omega_1$,

$[\tau_{3ph}^{-1}]_{Tu} = B_{Tu} [\omega^2/\sinh(\hbar\omega/k_B T)]$ for $\omega_1 < \omega < \omega_2$,

$[\tau_{3ph}^{-1}]_{TN} = B_{TN} \omega T^4$ for $0 < \omega < \omega_1$.

Guthrie drew attention to the fact that in the high-temperature range, the three-phonon relaxation rate should be proportional to T both for longitudinal and transverse phonons, i.e., $\tau_{3ph}^{-1} \propto T$. Fulkerson[9] also pointed out that his experimental results in the temperature range (100–1300) °K cannot be explained by the Holland formulation. For the phonon-phonon scattering relaxation rate, Joshi and Verma[7] used the following expressions:

$$\tau_{pp}^{-1} = \tau_{4ph}^{-1} + \tau_{3ph}^{-1} .$$

The frequency and temperature dependences of τ_{3ph}^{-1} are given in Table I. The frequency and temperature dependence of four-phonon processes is given by

$$\tau_{4ph}^{-1} = B_H \omega^2 T^2 .$$

In the present model we have used the following expressions for the phonon relaxation rates (Dubey[1,2] *et al.*):

$$[\tau_{3ph}^{-1}]_T = B_{T,I} \omega\, T^{m_{T,I}(T)} e^{-\theta/\alpha T} ,$$

$$[\tau_{3ph}^{-1}]_L = B_{L,I} \omega^2 T^{m_{L,I}(T)} e^{-\theta/\alpha T} + B_{L,II} \omega^2 T^{m_{L,II}(T)} e^{-\theta/\alpha T} .$$

With the present expressions for three-phonon relaxation rates, which are based upon Guthrie's ideas, it is possible to explain the phonon-conduc-

TABLE I. Temperature dependence of τ_{3ph}^{-1} in the different temperature regions.

τ_{3ph}^{-1}	Transverse	Longitudinal	Temperature
	$B_T \omega T^4$	$B_L \omega^2 T^3$	$T \leq 43$
	$B_{T_1} \omega T^3$	$B_L \omega^2 T^3$	$43 \leq T \leq 190$
	$B_{T_2} \omega T^2$	$B_{L_1} \omega^2 T^2$	$190 \leq T \leq 280$
	$B_{T_3} \omega T$	$B_{L_2} \omega^2 T$	$280 \leq T$

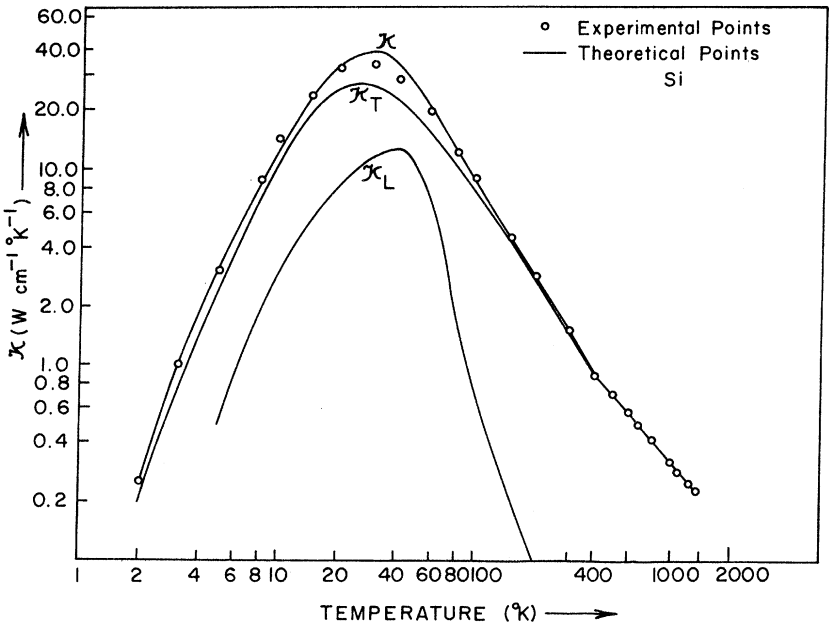

FIG. 1. Comparison of the theoretical values of the phonon conductivity of Si with the experimental values in the temperature range 2–1400 °K. κ_T is the contribution of transverse phonons, and κ_L is the contribution of longitudinal phonons.

tivity results without invoking four-phonon processes.

Joshi and Verma used $\vec{q} = (\omega/\vec{V})(1 + r\omega)$ for replacing V_g/V_p^2 in the conductivity integrals. Here V_g is the group velocity and V_p is the phase velocity. In the present model we have used a cubic term[10] instead of a quadratic one, i.e., $\vec{q} = (\omega/\vec{V})(1 + r\omega^2)$. The latter expression gives a better representation of a dispersion curve.[11] Real dispersions may be expected to approximate to some degree the q proportional to arcsinω relations found in simple linear lattices, and the Taylor's expansion of arcsinω consists of a sum of odd powers with positive coefficients for even terms. It may be argued that if dispersion is included in terms of modified phase and group velocities, some allowance should also be made for the effect of dispersion in the frequency dependence of re-

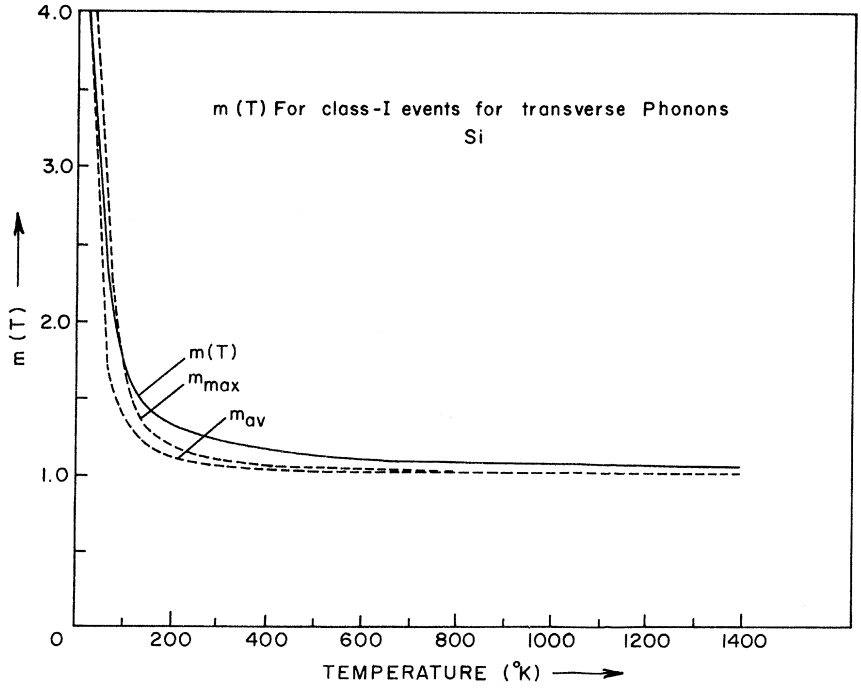

FIG. 2. Temperature dependence of the exponent $m(T)$ for transverse phonons for three-phonon class-I events. m_{max} is the maximum limit obtained by Guthrie, and m_{av} is the average value of upper and lower bounds of Guthrie. $m_{T,I}(T)$ is the value of exponent $m(T)$ used in the present calculation.

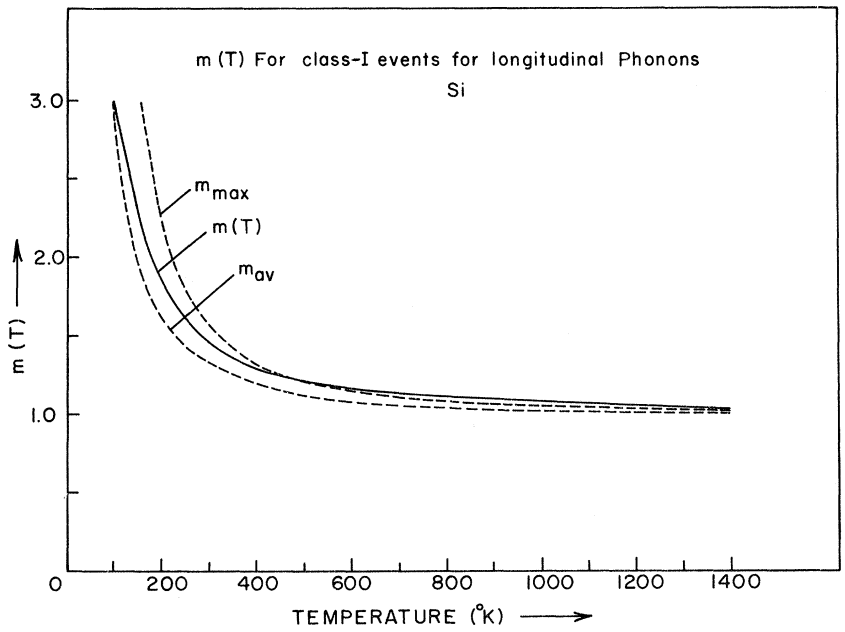

FIG. 3. Temperature dependence of the exponent $m(T)$ for longitudinal phonons for three-photon class-I events. m_{max} is maximum limit obtained by Guthrie, and m_{max} is the average of upper and lower bounds of Guthrie. $m_{L,I}(T)$ is the value of exponent m (T) used in the present calculation.

laxation times. However, this effect has not been considered in the present model.

The values of the parameters which have been used to calculate the phonon conductivity of Si in the temperature range 2–1400 °K are given in Table II. It may be seen in Fig. 1 that except near the conductivity maximum, i.e., in the temperature range 40–100 °K where there is some slight discrepancy, the agreement between theory and experiment is excellent. At high temperatures the contribution of longitudinal phonons is 1% to 5% of the total conductivity, and the rest is due to transverse phonons. This result is in agreement with the high-temperature results of Hamilton and Parrott[12] for Ge at 400 °K. At low temperatures the contribution of longitudinal phonons is as high as 25% of the total conductivity, which is correct in the sense that there are two transverse modes of each longitudinal mode and that the velocity of transverse phonons is nearly half of that of longitudinal phonons.

In cases where the relaxation is principally due to boundary scattering, the thermal conductivity should be proportional to the product of the heat capacity and the group velocity, or κ is proportional to $V_g \times (T/\Theta_D)^3$. For small wave numbers, V_g is proportional to Θ_D^1, so that κ is proportional to Θ_D^{-2} or to V^{-2}. Klemens has also shown in his review article that κ is proportional to $\Theta_D^{-2} T^3$. Thus in the temperature range where boundary scattering of phonons dominates over other phonon-scattering processes, the contribution of longitudinal and transverse phonons towards phonon conductivity is approximately in the ratio 1:5. In the temperature range 15–100 °K, the longitudinal phonons and transverse phonons make comparable contributions. The values of $m(T)$ at different temperatures for transverse phonons and longitudinal phonons, as well as for class-I and class-II events, are shown in Figs. 2–4. These values of m in general lie in the neighborhood of the upper bound of m obtained on the basis of Guthrie's relations.

The success of the SDV model in explaining the phonon-conductivity results of silicon is quite significant in the sense that four-phonon processes

TABLE II. Parameters which are used in the calculation of phonon conductivity of Si in the temperature range 2–1400 °K.

(V_T) $(0 < \omega < \omega_1) = 5.86 \times 10^5$ cm/sec
(V_T) $(\omega_1 < \omega < \omega_2) = 2.0 \times 10^5$ cm/sec
(V_L) $(0 < \omega < \omega_4) = 8.48 \times 10^5$ cm/sec
(V_L) $(\omega_4 < \omega < \omega_3) = 4.24 \times 10^5$ cm/sec
$\theta_1 = 180$ °K $\theta_4 = 350$ °K
$\theta_2 = 210$ °K $\theta_3 = 570$ °K
$L = 2.600$ mm (theor.)
$L = 0.295$ cm (expt.)
$A = 0.247 \times 10^{-44}$ sec^3
$B_{T,I} = 1.43 \times 10^{-6}$ deg^{-m}
$B_{L,II} = 3.90 \times 10^{-17}$ sec deg^{-m}
For [100] direction
$r_1 = 6.3897 \times 10^{-28}$ sec^2
$r_2 = 1.934 \times 10^{-27}$ sec^2
$r_4 = 0$
$r_3 = 5.588 \times 10^{-29}$ sec^2
$\theta = 658$ °K, $\alpha = 1.3$
$a = 5.4307 \times 10^{-8}$ cm^{-1}

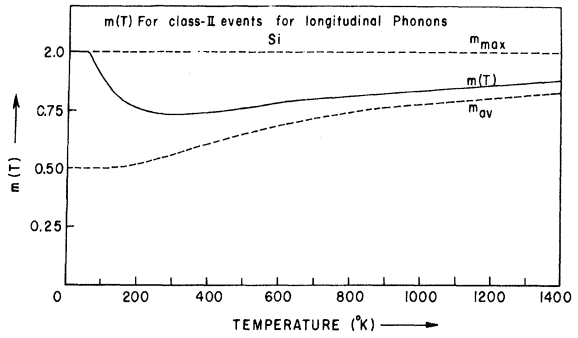

FIG. 4. Temperature dependence of the exponent $m(T)$ for longitudinal phonons for three-phonon class-II events. m_{max} is the maximum limit obtained by Guthrie, and m_{av} is the average of upper and lower bounds of Guthrie. $m_{L,II}(T)$ is the value of exponent $m(T)$ used in the present calculation.

are not invoked in the high-temperature region. This is notable since the latest calculation of Joshi *et al.*[6,7] for Si indicated that four-phonon processes must be included to explain the high-temperature behavior of silicon. Thus the present calculations establish that if one takes the temperature dependence of three-phonon scattering events properly into account, one can interpret the results quite successfully. Such a temperature dependence is bound to lead to a T dependence for longitudinal and transverse phonons in the high-temperature regions. In the low-temperature region, one has a T^4 dependence for transverse phonons and a T^3 dependence for longitudinal phonons, in keeping with Herrings's relations. The transition from T dependence to T^4 or T^3 dependence is continuous with the same three-phonon scattering strength adjusted for the entire temperature range, from 20 to 1400 °K. The use of a dispersion relation $\vec{q} = (\omega/\vec{V})(1+r\omega^2)$ in calculating the group velocities for the different regions of the dispersion curves and replacing V_g/V_p^2 in the conductivity integrals makes the whole calculation more realistic.

Although there is no explicit distinction between normal and umklapp processes in the SDV model, this is unlikely to create difficulties when umklapp processes dominate over normal process, which is generally the case except for isotopically pure materials at low temperatures.[13–15] For example, in Ge and Si it has been shown that the correction term due to normal processes in the Callaway theory, i.e., the β term, is negligible. However, normal processes are also included in the expression for the combined relaxation time τ_C^{-1}. At temperatures beyond the conductivity maximum, $\tau_u^{-1} > \tau_N^{-1}$, while at temperatures near the maximum and below the maximum, $\tau_B^{-1} + \tau_{pt}^{-1} > \tau_N^{-1}$. Thus in substances such as Ge and Si, τ_u^{-1} makes a dominant contribution towards thermal resistance. The umklapp processes are characterized by the exponential temperature dependence $e^{-\theta/\alpha T}$, and this is the reason that the present expressions for three-phonon relaxation rate in the SDV model are so successful in fitting the experimental results.

ACKNOWLEDGMENTS

The authors wish to express their thanks to Professor K. S. Singwi for his interest in the present work. One of us (K.S.D.) is also grateful to C.S.I.R., India for providing financial assistance in the form of a fellowship.

[1]P. C. Sharma, K. S. Dubey and G. S. Verma, Phys. Rev. B **4**, 1306 (1971).
[2]K. S. Dubey and G. S. Verma, Phys. Rev. B **4**, 4491 (1971).
[3]G. L. Guthrie, Phys. Rev. **152**, 801 (1966).
[4]G. L. Guthrie, Phys. Rev. B **3**, 3573 (1971).
[5]P. G. Klemens, *Solid State Physics* (Academic, New York, 1958), Vol. 7, p. 1.
[6]Y. P. Joshi, M. D. Tewari, and G. S. Verma, Phys. Rev. B **1**, 642 (1970).
[7]Y. P. Joshi and G. S. Verma, Phys. Rev. B **1**, 750 (1970).
[8]M. G. Holland, Phys. Rev. **132**, 2401 (1964).
[9]W. F. Fulkerson, R. K. Moore, R. S. Graves, and D. L. McIroy, Phys. Rev. **167**, 765 (1968).
[10]P. C. Sharma, K. S. Dubey and G. S. Verma, Phys. Rev. B **3**, 1985 (1971).
[11]B. N. Brockhouse, Phys. Rev. Lett. **2**, 256 (1959).
[12]R. A. H. Hamilton and J. E. Parrott, Phys. Rev. **178**, 1284 (1969).
[13]B. K. Agarwal and G. S. Verma, Phys. Rev. **128**, 603 (1962).
[14]J. Callaway and M. C. V. Baeyer, Phys. Rev. **120**, 1149 (1960).
[15]H. Jackson and C. T. Walker, Phys. Rev. B **3**, 1428 (1971).