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ROLE OF THE DEBYE TEMPERATURE IN THE LATTICE THERMAL CONDUCTIVITY OF SILICON

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This paper is a theoretical study of the effect of the variation in the Debye temperature Θ_D with temperature on the lattice thermal conductivity of Si in the temperature range 2-300 K. Expressions for the three-phonon scattering relaxation rates previously proposed by Sherma *et al.* are used here. The percentage changes in the lattice thermal conductivity due to the Debye temperature for the transverse and the longitudinal phonons are studied separately.

It has now been established that phonon-phonon scattering plays a very important role in the analysis of the lattice thermal conductivity of a sample at high as well as low temperatures. The three-phonon scattering processes have been studied by a number of researchers [1-9]. The phonon-phonon scattering processes are divided into two groups: normal processes (N-processes) and umklapp processes (U-processes), where the momentum is conserved in the former and not conserved in the latter. Several expressions have been proposed for the three-phonon scattering relaxation rate ('C_{3ph}^{-1}) for N and U-processes, which are reported in Table 1. These rates indicate that the Debye temperature is an important factor in the estimation of the phonon-phonon scattering relaxation rate. Using these scattering relaxation rates, several workers [10-14] have calculated the lattice thermal conductivity (K) for different samples. It has become well known that phonon-phonon scattering plays a very important role in the analysis of the lattice thermal conductivity of a sample.

It has been found that the three-phonon scattering processes predominate over other scattering processes at high temperatures. At the same time, these processes at low temperatures are not negligibly small, an

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play and important role even in the vicinity of the conductivity maxima. Some studies [15, 16] have been made to calculate the phonon conductivities of different samples having different values of Θ_D. However, in these studies the effect of the variation in Θ_D with temperature on lattice thermal conductivity was not taken into account. Dubey and Verma [17] found that, when the relaxation is principally due to boundary scattering, the lattice thermal conductivity is proportional to Θ_D^{-2}. Klemens [2] showed that the lattice thermal conductivity is proportional to $\Theta_D^{-2}T^3$. Recently, Awad [18] studied the effect of the variation in Θ_D with temperature on the phonon conductivity of Ge in the frame of the Dubey model [9].

Table 1 The combined scattering relaxation rates. In these expression, w_1 and w_2 are the transverse phonon frequencies at $\frac{1}{2}k_{...}$ and $k_{....}$ respectively, w_3 and w_4 are the same for the longitudinal phonons, w_D is the Debye frequency, α is a constant, k_{max} is the zone boundary of the first Brillouin zone and m is the temperature exponent

	Combined scattering relaxation rates	Frequency range
Callaway [3]	$``C_c^{-1} = `C_B^{-1} + `C_{pt}^{-1} + (B_1 + B_2)\,w^2T^3$	0-w_D
Holland [4]	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_{TN}\,wT^4$	0-w_1
	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_{TU}\,w^2/\sinh(\bar{n}w/K_BT)$	w_1-w_2
	$`C_{c,L}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_L\,w^2T^3$	0-w_4
Joshi and Verma [5]	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_T\,wT^m$	0-w_2
	$`C_{c,L}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_L\,w^2T^m$	0-w_4
SDV model [6]	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_{T,I}\,wT^{m..}(T)e^{-\Theta/\alpha T}$	0-w_2
	$`C_{c,L}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + B_{L,I}\,w^2T^{m..}(T)e^{-\Theta/\alpha T} + B_{L,II}\,w^2T^{m...}(T)e^{-\Theta/\alpha T}$	0-w_4
Dubey and Misho [8]	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + (B_{TN} + B_{TU}e^{-\Theta/\alpha T})\,wT^m$	0-w_2
	$`C_{c,L}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + (B_{LN} + B_{LU}e^{-\Theta/\alpha T})\,w^2T^m$	0-w_4
Dubey [9]	$`C_{c,T}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + (B_{TN,I} + B_{TU,I}e^{-\Theta/\alpha T})\,wT^{m..}(T)$	0-w_2
	$`C_{c,L}^{-1} = `C_B^{-1} + `C_{pt}^{-1} + (B_{LN,I} + B_{LU,I}e^{-\Theta/\alpha T})\,w^2T^{m..}(T) + (B_{LN,II} + B_{LU,II}e^{-\Theta/\alpha T})\,w^2T^{m...}(T)$	0-w_4

The aim of the present work is to study the effect of the Debye temperature variation with temperature on the lattice thermal conductivity of Si. The percentage changes in the lattice thermal conductivity have also been studied separately for the transverse and longitudinal phonons. The results of the calculations are obtained in the frame of the Sharma-Dubey-Verma (SDV) model within the temperature range 2-300 K.

Scattering relaxation rates and phon conductivity integral

The lattice thermal conductivity can be expressed as

$$K = K_T + K_L \tag{1}$$

where K_T and K_L are the phonon conductivities arising from the transverse and longitudinal phonons, respectively, and can be given by

$$K_T = \frac{c_0}{v_{T_1}} \int_0^{\Theta_1/T} {}^{\backprime}C_{c,T} \,(1 + R_1X^2T^2)^2 (1 + 3R_1X^2T^2)^{-1} X^4 e^X (e^X - 1)^{-2} \mathrm{d}X$$
$$+ \frac{c_0}{v_{T_2}} \int_0^{\Theta_2/T} {}^{\backprime}C_{c,T} \,(1 + R_2X^2T^2)^2 (1 + 3R_2X^2T^2)^{-1} X^4 e^X (e^X - 1)^{-2} \mathrm{d}X \tag{2}$$

$$K_L = \frac{c_0}{2v_{L_1}} \int_0^{\Theta_3/T} {}^{\backprime}C_{c,L} \,(1 + R_3X^2T^2)^2 (1 + 3R_3X^2T^2)^{-1} X^4 e^X (e^X - 1)^{-2} \mathrm{d}X$$
$$+ \frac{c_0}{2v_{L_2}} \int_0^{\Theta_4/T} {}^{\backprime}C_{c,LT} \,(1 + R_4X^2T^2)^2 (1 + 3R_4X^2T^2)^{-1} X^4 e^X (e^X - 1)^{-2} \mathrm{d}X \tag{3}$$

where $c_0 = (K_B / 3\pi^2)(K_BT / \bar{n})^3$, $R_i = r_i (K_B / \bar{n})^2$, K_B is the Boltzmann constant, h is the Planck constant divided by 2π, the v's are the velocities of the corresponding modes, the Θ's are the temperatures corresponding to the Brillouin zone boundary (for more details, see [10-12]), and $C_{c,T}$ and $C_{c,L}$ are the combined scattering relaxation times due to the transverse and longitudinal phonons, respectively; these terms can be expressed as

$${}^{\backprime}C_{c,T}^{-1} = {}^{\backprime}C_B^{-1} + {}^{\backprime}C_{pt}^{-1} + {}^{\backprime}C_{3ph,T}^{-1} \tag{4}$$

$${}^{\backprime}C_{c,L}^{-1} = {}^{\backprime}C_B^{-1} + {}^{\backprime}C_{pt}^{-1} + {}^{\backprime}C_{3ph,L}^{-1} \tag{5}$$

where C_B^{-1} and C_{pt}^{-1} are the scattering relaxation rates of the boundary and the point defect, respectively [2, 19].

The three-phonon scattering relaxation rates are given by the SDV model [6, 7] as

$$'C_{3ph,T}^{-1} = B_{T,I}\, wT^{m_{T,I}(T)} e^{-\Theta_D/\alpha T} \tag{6}$$

$$'C_{3ph,L}^{-1} = B_{L,I}\, w^2 T^{m_{L,I}(T)} e^{-\Theta_D/\alpha T} + B_{L,II}\, w^2 T^{m_{L,II}(T)} e^{-\Theta_D/\alpha T} \tag{7}$$

where Θ_D is the Debye temperature of the sample and α is a constant. The values of the temperature exponents $m(T)$ can be calculated from the following relations:

$$m_I(T) = X_{\max.}(e^{X_{\max.}} - 1)^{-1} + 0.5X_{\max.} + [\ln(1 + \Theta/T)/\ln(T)] \tag{8}$$

$$m_{II}(T) = 0.5X_{\max.} e^{0.5X_{\max.}} (e^{X_{\max.}} - 1)^{-1} + 0.5 + [\ln(1 + \Theta/T)/\ln(T)] \tag{9}$$

where $X_{\max.} = hw_{\max.}/K_BT$, the B's are the corresponding scattering strength, and suffixes, T, L, I and II represent the transverse phonons, the longitudinal phonons, a class I event and a class II event, respectively.

The percentage change in the lattice thermal conductivity and three-phonon scattering relaxation rate can be expressed as

$$\% K_s = \frac{K_s(c) - K_s(T)}{K_s(c)} \cdot 100 \tag{10}$$

where $K_x(c)$ and $K_x(T)$ are the lattice thermal conductivity when Θ_D is taken as a constant ($\Theta_D(c)$) and as a function of temperature ($\Theta_D(T)$), respectively, and s represents either T or L. The percentage change in the Debye temperature can be given by

$$\% \Delta\Theta_D = \frac{\Theta_D(c) - \Theta_D(T)}{\Theta_D(c)} \cdot 100 \tag{11}$$

Through use of the SDV model, the lattice thermal conductivity has been studied for different values of the Debye temperature. The results obtained are shown in Fig. 1. The percentage change in the lattice thermal conductivity has been calculated for Si, and the relevant results are plotted in Fig. 2 for both the transverse and longitudinal phonons. The variation in the lattice thermal conductivity at a constant temperature (30 K) has also been calculated by assuming different values of $\%\Delta\Theta_D$ such as $\%\Delta\Theta_D = 5, 10, 15, \ldots, 30$. This variation is illustrated in Fig. 3.

Results and discussion

The constants and the parameters used in the present calculations are taken from the earlier report of Dubey and Verma [17]. The temperature-dependent Debye temperature $\Theta_D(T)$ of Si is taken from the work of Flubacher *et al.* [20]. The results obtained are shown in Figs 1-3. With the help of these Figures, one can conclude the following points.

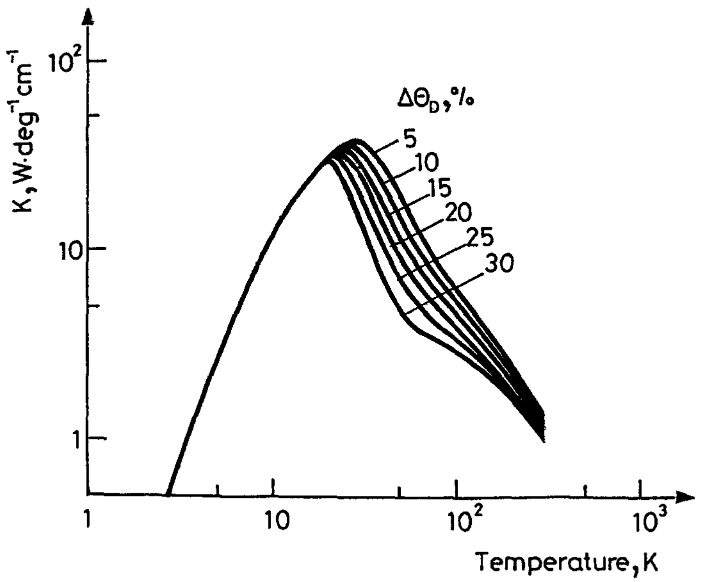

Fig. 1 The lattice thermal conductivity of Si in the temperature range 2-300 K for different values of the Debye temperature

1. Figure 1 reveals that the position of the conductivity maximum in the K *vs.* T curve moves towards higher temperatures on any increase in the Debye temperature. Thus, one can conclude that the Debye temperature is one of the factors responsible for assigning the maximum value of conductivity. It is also clear from this Figure that at very low temperatures (say below 20 K) the lattice thermal conductivity is independent of the percentage change in the Debye temperature, which is in agreement with the fact that at temperatures below and near the maxima 'C_B^{-1} and 'C_{pt}^{-1} predominate over 'C_{3ph}^{-1} for both the transverse and longitudinal phonons; moreover, at low values of temperature, the factor $e^{-\Theta. / \alpha T}$ is negligibly small. At a little higher

temperature, just beyond the conductivity maximum, the lattice thermal conductivity decreases very rapidly with increasing percentage change in the Debye temperature at each temperature. The basic reason for this kind of variation resides in the dominant contribution of the phonon-phonon scattering to the thermal resistance. At higher temperatures, both $e^{-\Theta.\,/\alpha T}$ and the temperature exponent $m(T)$ tend to unity resulting in the T dependence of the phonon-phonon scattering relaxation rate for both polarization branches. As a result, at each temperature the lattice thermal conductivity varies very slowly with $\%\Delta\Theta_D$. It can also be confirmed that the nature of the K *vs*. T curve is almost the same for all values of $\%\delta\Theta_D$.

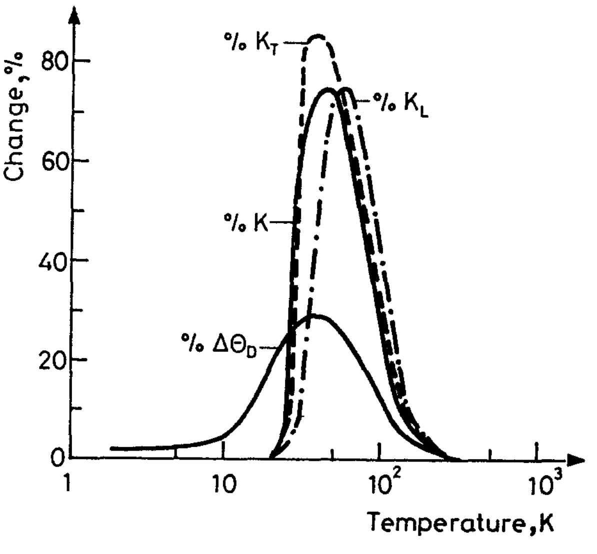

Fig. 2 The percentage change of the lattice thermal conductivity (K) of Si due to the variation of the Debye temoerature (Θ_D) in the temperature range 2-300 K. % K_T and % K_L refers to the percentage change in the lattice thermal conductivity for transverse and longitudinal phonons respectively

2. Form Fig. 2, it can be seen that the percentage change in the lattice thermal conductivity is directly proportional to the percentage change in the Debye temperature, and the nature of the variation is similar in both polarization branches. At very low temperatures, where the boundary and point defect scattering are the only relevant modes of scattering of phonons, the contributions of the longitudinal and transverse phonons to the total lattice thermal conductivity of Si are approximately in the ratio 1:5 [17], which

results in $\%K$ being independent of $\%\delta\Theta_D$. One can also see, that, at any certain temperature beyond the percentage change maximum, $\%K_L$ is greater than $\%K_T$, which can be attributed to the presence of the umklapp processes in both class *I* and class *II* events. It is of interest that the results obtained here are similar to the earlier finding of Awad [18].

3. The theoretical calculations shown in Fig. 3 indicate that, at constant temperature, $\%\ K$ increases as $\%\Delta\Theta_D$ increases. At each value of $\%\Delta\Theta_D$, the value of the percentage change in the lattice thermal conductivity for the transverse phonons is larger than that obtained for the longitudinal phonons. This stems from the fact that the percentage contribution of the transverse phonons exceeds the percentage contribution of the longitudinal phonons. However, in the temperature range 15-100 K, the transverse and longitudinal phonons make comparable contributions [17]. The same behaviour was also observed by Awad [18] in the lattice thermal conductivity of Ge based on the Dubey model [9].

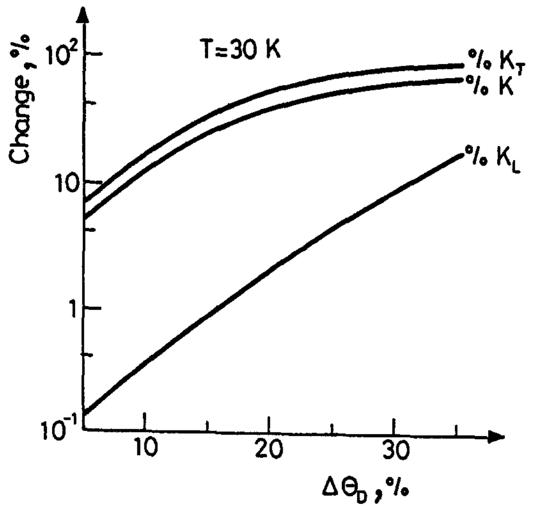

Fig. 3 The percentage change of the lattice thermal conductivity (K) of Si due to the percentage change of the Debye temperature (Θ_D) in the constant temperature T=30 K. $\%\ K_T$ and $\%\ K_L$ represent the percentage change in the lattice thermal conductivity for the transverse and longitudinal phonons respectively

4. In fact, within the temperature range 40-100 K, the Debye temperature appears to be the reason for the discrepancy between the theoretical and experimental lattice thermal conductivities of Si [17]. It is instructive to sug-

gest that these discrepancies can be modified by using the temperature-dependent Debye temperature $\Theta_D(T)$ for the whole range of temperatures, instead of taking it as a constant, which is relevant to the work of Awad [18].

5. It should be noted that the umklapp processes are characterized by the exponential temperature dependence $e^{-\Theta_{\cdot} / \alpha T}$, and this is the reason why the SDV model is quite successful in this study.

6. In conclusion, the present calculations provide significant account of the effect of the Debye temperature variation with temperature on the lattice thermal conductivity of Si.

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Zusammenfassung — Dieses Manuskript ist eine theoretische Untersuchung des Einflusses der Veränderung der Debey-Temperatur ΘD mit der Temperatur auf die Gitterwärmeleitfähigkeit von Si im Temperaturbereich 2-300 K. Unlängst von Sherma *et al.* vorgeschlagene Ausdrücke für die drei Phononen-Streuungsrelaxations-Geschwindigkeiten fanden dabei Anwendung. Die prozentuelle Änderung der Gitterwärmeleitfähigkeit in Abhängigkeit von der Debey-Temperatur wurde für transversale als auch für longitudinale Phononen separat untersucht.