isotope effects: The experimental effect on the peak energy is constant and approximately equal to 2×10^{-2} eV; on the other hand, the effect on the half-width at temperatures higher than 100°K probably disappears only in appearance. If we subtract the isotopic contribution, i.e., the last term in the right member of Eqs. (8), from the experimental values of the half-widths W_H^2 and W_D^2, we obtain the data of Fig. 5; it can be seen that both the points due to the U_H and U_D centers can be made to fall on the same curve, within the limits of the experimental error.

Finally, we have studied the temperature dependence of the peak energy $\bar{\epsilon}(T)$ and the half-width $W(T)$. We know the symmetry properties of the continuum modes which give rise to the temperature dependence, but *a priori*, it is difficult to estimate the values of displacement or frequency effects. We have estimated the overall effect by means of two effective frequencies ω_1 and ω_2, which, together with the infrared-active local mode, describe the whole electron-phonon interaction [see Eqs. (8) and (9)]. The values of the two effective frequencies and of the coefficients E, B, and A of Eqs. (8) and (9), determined from the experimental data of Figs. 2 and 3, are reported in Table II. From the frequency distribution reported by Karo[14] for the perfect crystals, one finds that the two frequencies ω_1 and ω_2 fall around the maximum in the transverse optic branch and in the longitudinal acoustic branch, respectively. The considerable difference between ω_1 and ω_2 suggests that a configurational coordinate model with one frequency is not sufficient to describe the optical absorption of the U center or, perhaps, of other centers.

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Role of Longitudinal and Transverse Phonons in Lattice Thermal Conductivity of GaAs and InSb

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The lattice thermal conductivity of GaAs and InSb has been calculated in the temperature range 2–300°K taking into consideration the details of phonon dispersion relations resulting in the separate contributions of the longitudinal and transverse phonons. This analysis gives a much better fit to the experimental data throughout the whole range of temperature than is given by Callaway's formulation which is based on Debye's phonon spectrum, a choice of relaxation time for phonon-phonon scattering relaxation time which is valid for longitudinal phonons, and the use of the average phonon velocity.

1. INTRODUCTION

THE problem of lattice thermal conductivity of substances in which heat is primarily carried by phonons, has been thoroughly investigated in recent years.[1–10] An exact treatment of the problem, however, is hampered by the lack of knowledge of the crystal vibration spectra and the anharmonic forces, and by the difficulty of obtaining exact solutions of the Boltzmann equation. A much-simplified model was proposed by Callaway assuming a Debye phonon spectrum consisting of one average (acoustic) branch and making several assumptions as to the form of the three-phonon-scattering relaxation times. This model has been successfully applied to a number of substances.[11–14]

Holland[15] has studied lattice thermal conductivity of GaAs and InSb in the temperature range 1.7° to 300°K and compared the experimental results with the analysis based on Callaway's model which could not explain the entire temperature dependence of thermal conductivity. There is a change in the slope in the experimental data at about 80°K in InSb and about

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TABLE I. Values of ω_1, ω_2, ω_3 and velocities of longitudinal and transverse phonons.

Substance	ω_1	ω_2 (10^{13} cps)	ω_3	V_{T1}	V_{T2} (10^5 cm/sec)	V_L
GaAs	1.34	1.48	4.27	2.48	0.90	5.24
InSb	0.73	0.81	2.22	2.28	0.82	3.77

100°K in GaAs. This change in slope can be explained by two-mode conduction. Such a change in slope was also observed in Si and Ge[16] where Callaway's phenomenological model could not explain the temperature dependence of lattice thermal conductivity at temperatures from beyond 800°K to the melting point. Any attempt to fit the theoretical curve with the experimental data at high temperatures leads to a poor fit with the experimental results at the other temperatures. Transverse phonons play an important role in thermal conduction and if one calculates the separate contribution of transverse and longitudinal phonons and then the total thermal conductivity due to phonons, the entire temperature dependence of lattice thermal conductivity in Si and Ge is explained very well. Parrott,[17] after attempting to fit the experimental data on Si-Ge alloys, also concluded that thermal conduction at high temperatures is primarily due to transverse phonons, which conclusion is supported by our analysis[18] too. The excellent agreement obtained between theory and experiment for InSb and GaAs also supports the present approach of separation into longitudinal and transverse phonons.

Calculation of the separate contributions of longitudinal phonons and transverse phonons requires a detailed knowledge of vibration spectra of the solids under consideration. If the phonon spectrum is similar to that of Si and Ge where the transverse branch becomes almost flat from $q_{\max/2}$ to $q_{\max}$ at the zone boundary, one needs, for the calculation of thermal-conductivity values of the zone boundary, frequencies for the longitudinal (ω_3) and transverse (ω_2) acoustic branches and the frequency ω_1 corresponding to $q_{\max/2}$ for transverse branch where the umklapp processes are supposed to start. Fortunately, for GaAs the complete phonon spectrum is available from the neutron-scattering studies by Waugh and Dolling.[19] For InSb the values of the zone-boundary frequencies are obtained from infrared-absorption data.[20]

2. FORMULATION

The lattice thermal conductivity K is separated into two parts, (i)K_T, which is the contribution of transverse phonons and (ii)K_L, which is the contribution of longitudinal phonons. The thermal conductivity due to transverse phonons K_T is given by

$$\begin{aligned}K_T&=K_{T1}+K_{T2}\\&=(K_T)_0{}^{\omega_1}+(K_T)_{\omega_1}{}^{\omega_2}\\&=\frac{2}{3}\frac{k}{2\pi^2}\Big[(V_T{}^{-1})_{\omega<\omega_1}\int_0^{\omega_1}[\tau_B{}^{-1}+\tau_{pt}{}^{-1}+\tau_{TN}{}^{-1}]^{-1}\\&\qquad\times\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}}{(e^{\hbar\omega/kT}-1)^2}\omega^2\,d\omega\\&\quad+(V_T{}^{-1})_{\omega_1<\omega<\omega_2}\int_{\omega_1}^{\omega_2}[\tau_B{}^{-1}+\tau_{pt}{}^{-1}+\tau_{TU}{}^{-1}+\tau_{TN}{}^{-1}]^{-1}\\&\qquad\times\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}}{(e^{\hbar\omega/kT}-1)^2}\omega^2\,d\omega\Big],\end{aligned}\tag{1}$$

where V_T is the velocity of transverse phonons which is assumed to be constant from 0 to ω_1, where it changes abruptly and then remains constant from ω_1 to ω_2. Here ω_2 is the zone-boundary frequency for the transverse phonons and ω_1 is the frequency corresponding to $q_{\max/2}$ where umklapp processes are supposed to start. Below ω_1, umklapp processes are absent. In the above equation the phonon dispersion relation is assumed to be isotropic.

The relaxation time for normal processes for transverse phonons is given by

$$\begin{aligned}(\tau_{TN}{}^{-1})_{\omega<\omega_1}&=B_{TN}\omega T^4,\\(\tau_{TN}{}^{-1})_{\omega_1<\omega<\omega_2}&=B_{TN}{}'\omega T.\end{aligned}\tag{2}$$

The relaxation time for umklapp processes for transverse phonons is given by

$$\begin{aligned}(\tau_{TU}{}^{-1})&=B_{TU}\omega^2/\sinh(\hbar\omega/kT)\quad\text{for}\quad\omega_1<\omega<\omega_2,\\&=0\qquad\qquad\qquad\qquad\text{for}\quad\omega<\omega_1.\end{aligned}\tag{3}$$

In the calculation of point-defect and boundary-scattering relaxation times, average phonon velocity V_S is used, which is given by

$$V_S{}^{-1}=\tfrac{1}{3}(2V_T{}^{-1}+V_L{}^{-1})\tag{4}$$

where V_L is the velocity of longitudinal phonons. The relaxation times for the boundary and point-defect scattering are given by

$$\tau_B{}^{-1}=V_S/LF,\tag{5}$$

$$\begin{aligned}\tau_{pt}{}^{-1}(\text{mass-diff.})&=A\omega^4\\&=(V_0/4\pi V_S{}^3)\textstyle\sum_i f_i(1-m_i/\bar{m})^2,\end{aligned}\tag{6}$$

where the characteristic length L is given by $2R$ for a specimen of circular cross section of radius R and is equal to $1.12S^{1/2}$ for a rectangular cross section of area of cross section equal to S, F is the geometrical factor, V_0 is the atomic volume, m_i is the mass of the ith species of the atom and $\bar{m}$ is the average atomic mass. Similarly,

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TABLE II. Parameters obtained from the analysis of thermal-conductivity data.

Substance	τ_B 10^{-6} sec	A (10^{-44} sec^3)	B_L(10^{-23} sec deg^{-3})	B_{TN}(10^{-23} deg^{-4})	B_{TU}(10^{-18} sec)
GaAs	1.68	0.46	1.75	41.62	6.01
InSb	3.33	2.77	5.84	34.73	4.03

the contribution due to longitudinal phonons is given by

$$K_L=\frac{1}{3}\frac{k}{2\pi^2V_L}\int_0^{\omega_3}[\tau_B^{-1}+\tau_{pt}^{-1}+\tau_{LN}^{-1}+\tau_{LU}^{-1}]^{-1}\times\frac{\hbar^2\omega^2}{k^2T^2}\frac{e^{\hbar\omega/kT}}{(e^{\hbar\omega/kT}-1)^2}\omega^2d\omega. \quad (7)$$

It is assumed that a relaxation time of the form $\tau^{-1}=B_L\omega^2T^3$ takes care of both normal and umklapp processes for longitudinal phonons.

3. RESULTS

The lattice thermal conductivity of GaAs and InSb has been measured in the temperature range 1.7 to 300°K.[15] An analysis of the data on the basis of Callaways's formulation shows its failure to explain the temperature dependence of the thermal conductivity in the entire temperature range especially beyond about 100°K. In the present analysis, which is based on the calculation of separate contributions of longitudinal and transverse phonons, good agreement between theory and experiment has been obtained for the entire temperature range. The dip in the thermal-conductivity curve, however, requires some sort of resonant scattering of phonons. The value of zone boundary frequencies for transverse (ω_2) and longitudinal (ω_3) phonons as well as the frequency ω_1 which corresponds to the lower limit of umklapp processes for transverse phonons are given in Table I.

The last three columns of Table I give the velocities of transverse and longitudinal phonons. Waugh and Dolling have obtained the dispersion relation for the normal mode of vibration of GaAs from neutron scattering studies. Values of the parameters for GaAs given in Table I are from Waugh and Dolling. For the present calculations the dispersion relations have been assumed to be isotropic. For InSb the phonon frequencies ω_2 and ω_3 at the zone boundary are taken from infrared absorption results. For ω_1 it has been assumed that the ratio ω_1/ω_2 for InSb is same as in GaAs as both of them possess the same structure. The phonon velocities for InSb are taken from Potter.[21] V_{T2} is obtained by assuming that V_{T2}/V_{T1} remains the same in InSb as in GaAs.

The values of the parameters A, B_L, B_T, B_{TU} and the relaxation time for the boundary scattering as obtained from the analysis of the thermal-conductivity data of GaAs and InSb are given in Table II. For GaAs the characteristic length of the sample is 0.729 cm, the geometrical factor F is 0.75 and the average phonon velocity V_S is 3.3×10^5 cm/sec. The theoretical estimate of the boundary-scattering relaxation time from these constants is $\tau_B=1.66\times10^{-6}$ sec. The ratio $(\tau_B)_{\text{expt}}/(\tau_B)_{\text{theo}}$ is of the order unity. The theoretical estimate of A for GaAs is 0.465×10^{-44}, whereas the experimental values of A from Table II is also the same. Thus, for GaAs, excellent agreement is obtained between the theoretical estimate and the experimental values of the boundary and point-defect scattering relaxation times. For InSb the characteristic length of the sample is 0.554 cm, the geometrical factor F is 0.83 and the average phonon velocity V_S is 2.3×10^5 cm/sec. This gives $\tau_B=2\times10^{-6}$ sec and the ratio of the experimental value of τ_B to this theoretical estimate of τ_B is 1.67. Also, A_{theo} for InSb is 0.923×10^{-44} sec^3 which gives $(A_{\text{expt}}/A_{\text{theo}})\sim3$.

The results of calculations for GaAs are shown in Fig. 1 which shows good agreement between theory and experiment for the entire temperature range from 1.7 to 300°K except near the dip in the conductivity curve beyond the maximum at about 30°K. The contributions towards thermal conductivity of low-frequency transverse phonons K_{T1}, high-frequency transverse phonons K_{T2}, longitudinal phonons K_L, and the resultant conductivity K are shown separately in Fig. 1. Figure 2 shows the comparison of the curve calculated on the

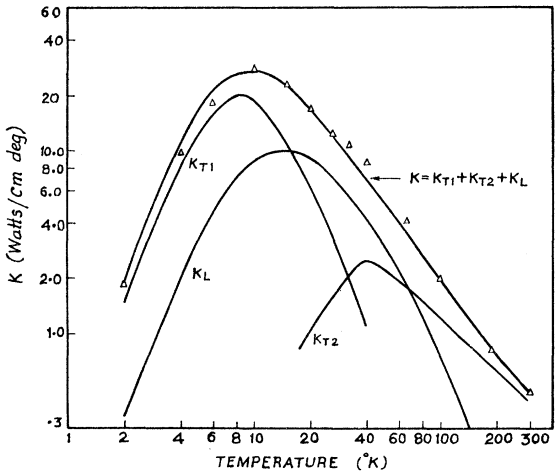

FIG. 1. Thermal conductivity of GaAs. K_{T1} is the contribution of transverse phonons of frequencies lying between 0 and ω_1, K_{T2} is the contribution of transverse phonons of frequencies lying between ω_1 and ω_2, K_L is the contribution of longitudinal phonons, and K is the total thermal conductivity.

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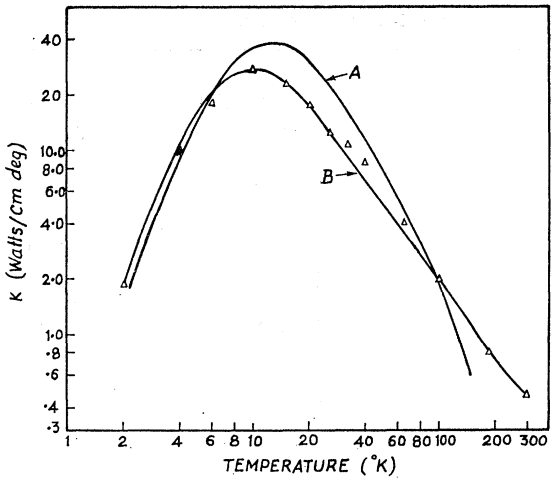

FIG. 2. Thermal conductivity of GaAs. Curve A: Callaway model. Curve B: Present analysis. Parameters for curve A: $\tau_B=1.66\times10^{-6}$ sec, $(B_1+B_2)=3.6\times10^{-23}$ sec deg^{-3} and $A=0.466\times10^{-44}$ sec.^{3}. Parameters for curve B are same as in Fig. 1.

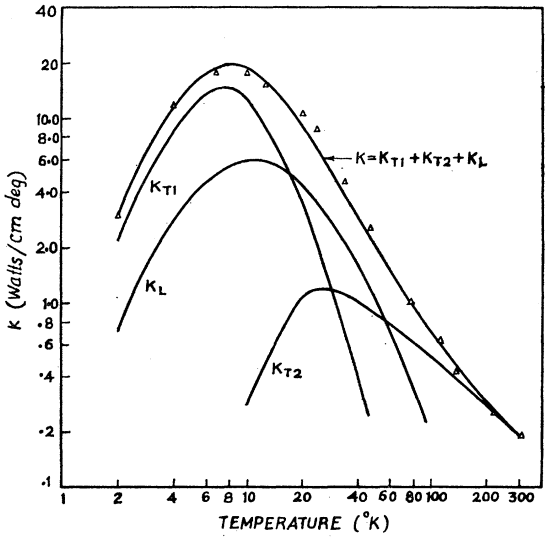

FIG. 3. Thermal conductivity of InSb. K_{T1} is the contribution of transverse phonons of frequencies lying between 0 and ω_1, K_{T2} is the contribution of transverse phonons of frequencies lying between ω_1 and ω_2, K_L is the contribution of longitudinal phonons and K is the total thermal conductivity.

basis of the formulation presented in this paper with that obtained on the basis of Callaway's model and it is obvious from this figure that the approach presented in this paper gives much better agreement with the experimental results. It may be noted in this connection that Callaway's formulation makes no distinction between longitudinal and transverse phonons and the sum over phonon polarization is set equal to three and an average phonon velocity V is used everywhere including the calculation of boundary and point-defect scattering relaxation times. The relaxation time for normal processes is taken to be proportional to $\omega^{-2}T^3$ which is true only for low-frequency longitudinal phonons. The temperature and frequency dependence of relaxation time for umklapp processes is assumed to be same as that of normal processes, which is not valid either at low temperatures or at high temperatures. Similarly, for InSb, the results of calculation are shown in Fig. 3 which also shows the different contributions K_{T1}, K_{T2}, K_L and the resultant conductivity separately. It is again obvious from this figure that the agreement between theory and experiment is good in the entire temperature range.

4. CONCLUSIONS

(1) The present analysis of the lattice thermal conductivity of GaAs and InSb in the temperature range 1.7 to 300°K reveals the important role which the transverse phonons play. Good agreement between theory and experiment over the entire temperature range is obtained only if one considers the separate contributions of transverse phonons and longitudinal phonons.

(2) Callaway's formulation which is based on Debye's approximation of the phonon spectrum and the use of relaxation time for phonon-phonon scattering which is valid for longitudinal phonons, does not explain the entire temperature dependence of the thermal conductivity. Any attempt to fit the theory with high-temperature data leads to misfit at rest of the temperatures.

(3) For GaAs there is excellent agreement between the theoretical estimates and the experimental values of the relaxation times for boundary scattering and impurity scattering. For InSb $(\tau_B)_{\text{expt}}/(\tau_B)_{\text{theo}}=1.67$ and $(\tau_{pt})_{\text{theo}}/(\tau_{pt})_{\text{expt}}$ is of the order three.

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