

transition probabilities. In comparing such results to theoretical band-structure calculations this, however, is rather a virtue because it would allow a comparison not only with the total density of states of the conduction band but also with the relative fractions having different symmetries. The above illustrates both the complimentary nature of electron-transition measurements (between core states and the conduction band) and induced-electron-emission measurements and the

benefits which can result from a combination of these experiments.

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## Phonon Conductivity of InAs

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The low-temperature thermal conductivity  $K$  of undoped  $n$ -type InAs has been measured between 5 and 300 °K, and analyzed on the basis of three different approaches, each including boundary-effect, mass-difference-scattering, and phonon-phonon processes. Various required parameters, mainly the phonon frequencies at zone boundary, have been deduced for InAs from known values of these parameters in the III-V compounds Si and Ge. Callaway's expression of  $K$  is found to give satisfactory agreement from low temperature up to temperatures slightly beyond the maximum. At low temperatures boundary and mass-difference scattering predominate. The Holland two-mode formulation explains the temperature dependence of  $K$  in the entire temperature range by emphasizing the predominant contribution of transverse phonons at low and high temperatures. The more elaborate approach, two-mode heat conduction using three-phonon relaxation times deduced from Guthrie's analysis and a quadratic relation ( $q, \omega$ ), suggests that at high temperatures the contribution of longitudinal phonons to  $K$  becomes comparable to that of transverse phonons. Moreover, four-phonon processes have been tentatively included in order to fit the experimental data in the higher-temperature range. It appears that no conclusion as to the relative contributions of transverse and longitudinal phonons may at present be satisfactorily established for InAs at high temperature.

### INTRODUCTION

In the absence of electron-phonon scattering, three main phonon scatterings are generally taken into account in the low-temperature thermal conductivity  $K$  of pure insulators and semiconductors: boundary-effect, mass-difference-scattering, and phonon-phonon interactions. While the first two mechanisms are in satisfactory agreement with experimental results, three-phonon processes are still not well defined. Different relaxation times have been considered for the latter, depending on

the type of the processes (N or U), the polarization mode, and the temperature interval.

The Callaway expression<sup>1</sup> for phonon conductivity, which includes the above three mechanisms, considers an average acoustic branch in the Debye approximation. It has been shown that this expression can account for the data from low temperature up to temperatures slightly higher than the maximum in  $K$ . A refined approach to the problem has been carried out by Holland,<sup>2</sup> who distinguished between conduction by transverse and longitudinal phonons. This formulation has been successfully

applied to several semiconductor materials, allowing one tentatively to establish the respective contributions of transverse and longitudinal phonons to  $K$ , and to account for the change in the slope of the curve  $K(T)$  in the high-temperature region above the maximum in  $K$ . More recently, Guthrie<sup>3</sup> has analyzed the temperature dependence of three-phonon processes, assuming a relaxation time  $\tau$  of the form  $\tau^{-1} \sim T^m$ , where  $m$  is a function of temperature. The Guthrie results have been used by Joshi and Verma<sup>4</sup> to analyze the lattice thermal conductivity of silicon.

In the present work, the thermal conductivity  $K$  of undoped  $n$ -type InAs has been measured between 5 and 300 °K, and in Sec. I of this paper, the data have been first analyzed on the basis of the Callaway and Holland formulations. In Sec. III, the Guthrie analysis has been applied to InAs. The deduced three-phonon relaxation times and the corresponding temperature intervals have been taken into account in the analysis of the phonon conductivity of the material. This has been done in order to compare the conclusion obtained from Holland's formulation to that deduced on the basis of a probably more appropriate approach. It is to be pointed out that various parameters, mainly phonon frequency limits, which are involved in the latter formulations have been deduced for InAs from comparison of known values of these parameters in the III-V compounds Ge and Si.

### I. THEORETICAL FORMULATIONS

#### Callaway's Formulation

The simple Callaway expression<sup>1</sup> for phonon conductivity  $K$  is

$$K = \frac{k}{2\pi^2 V} \int_0^{\omega_D} \tau_c \frac{\hbar^2 \omega^2}{k^2 T^2} \frac{e^{\hbar \omega / kT}}{(e^{\hbar \omega / kT} - 1)^2} \omega^2 d\omega,$$

where  $k$  is the Boltzmann constant,  $V$  the average phonon velocity, and  $\omega_D = k\Theta_D/\hbar$ ,  $\Theta_D$  being the Debye temperature. The combined relaxation time  $\tau_c^{-1} = \sum_i \tau_i^{-1}$  is

$$\tau_c^{-1} = V/L + A \omega^4 + B \omega^2 T^3.$$

$\tau_B^{-1} = V/L$  is due to boundary effect,<sup>5</sup> and the Casimir characteristic length  $L$  is given by  $L = 2\pi^{-1/2} \times s^{1/2}$ , where  $s$  is the cross-sectional area of the sample.  $\tau_p^{-1} = A \omega^4$  is related to mass-difference scattering,<sup>6</sup> where the parameter  $A$  is given by the Klemens relation

$$A = \frac{V_0}{4\pi V^3} \sum_i f_i (1 - m_i/\bar{m})^2.$$

$V_0$  is the atomic volume,  $f_i$  the atomic fraction of the  $i$ th isotope of mass  $m_i$ , and  $\bar{m}$  the average atomic mass.

The three-phonon processes, U as well as N types, are assumed to have the same relaxation

time at all temperatures:  $\tau_{3p}^{-1} = B \omega^2 T^3$ . Such a relaxation rate is open to criticism, for it appears to be valid only for low-frequency longitudinal phonons in N processes.<sup>2</sup>

#### Holland's Formulation

Holland has calculated<sup>2</sup> separately the contributions towards thermal conductivity of both polarization modes:

$$K = K_T + K_L.$$

$K_T$  is the contribution of transverse phonons:

$$K_T = K_{T1} + K_{T2},$$

$$K_{T1} = \frac{2}{3} \frac{k}{2\pi^2 V_{T1}} \int_0^{\omega_1} (\tau_B^{-1} + \tau_p^{-1} + \tau_{TU}^{-1} + \tau_{TN}^{-1})^{-1} \times \frac{\hbar^2 \omega^2}{k^2 T^2} \frac{e^{\hbar \omega / kT}}{(e^{\hbar \omega / kT} - 1)^2} \omega^2 d\omega,$$

and

$$K_{T2} = \frac{2}{3} \frac{k}{2\pi^2 V_{T2}} \int_{\omega_1}^{\omega_2} (\tau_B^{-1} + \tau_p^{-1} + \tau_{TU}^{-1} + \tau_{TN}^{-1})^{-1} \times \frac{\hbar^2 \omega^2}{k^2 T^2} \frac{e^{\hbar \omega / kT}}{(e^{\hbar \omega / kT} - 1)^2} \omega^2 d\omega.$$

$K_L$  is the contribution of longitudinal phonons:

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

In the above expressions,  $\omega_2$  and  $\omega_3$  are, respectively, the frequencies of the transverse and longitudinal acoustic branches at the zone boundary.  $\omega_1$  is the frequency of the transverse branch corresponding to  $\frac{1}{2}q_{\max}$ , where  $q_{\max}$  is the wave vector at the zone boundary.  $V_{T1}$  and  $V_{T2}$  are the velocities of transverse phonons in the intervals  $(0, \omega_1)$  and  $(\omega_1, \omega_2)$ , respectively.  $V_L$  is the velocity of longitudinal phonons.

The relaxation times  $\tau_B$  and  $\tau_p$  are relative to boundary-effect and mass-difference scattering. The average phonon velocity involved in the expressions of  $\tau_B$  and  $\tau_p$  is given by

$$V^{-1} = \frac{1}{3} V_L^{-1} + \frac{2}{3} V_{T1}^{-1}.$$

The relaxation rates due to three-phonon processes for transverse phonons are given by

$$(\tau_{TN})_{\omega < \omega_1}^{-1} = B_{TN} \omega T^4,$$

$$(\tau_{TN})_{\omega_1 < \omega < \omega_2}^{-1} = B'_{TN} \omega T,$$

$$(\tau_{TU})_{\omega < \omega_1}^{-1} = 0,$$

$$(\tau_{TU})_{\omega_1 < \omega < \omega_2}^{-1} = B_{TU} \omega^2 / \sinh(\hbar \omega / kT),$$

and for longitudinal phonons,

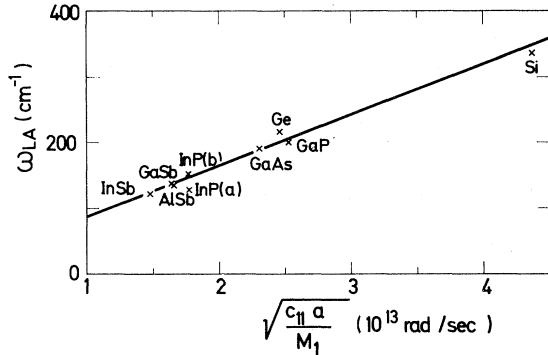


FIG. 1. Values of the longitudinal acoustic frequency  $\omega_{LA}$  at the zone boundary vs  $(c_{11}a/M_1)^{1/2}$  for the different III-V compounds Ge and Si (see for Ge: Refs. 9 and 10; Si, AlSb, GaAs, GaSb, InSb: Refs. 9 and 11; GaP: Ref. 11, 12, and 14; and InSb: Refs. 11-13).

$$\tau_{LN}^{-1} + \tau_{LU}^{-1} = B_L \omega^2 T^3.$$

## II. RESULTS AND ANALYSIS

### Experimental

The thermal conductivity  $K$  was measured between 5 and 300 °K, using a steady-state longitudinal-heat-flow technique, described elsewhere.<sup>7</sup> The undoped single crystals of InAs were  $n$  type, with a room-temperature electrical resistivity of  $13 \times 10^{-3} \Omega \text{ cm}$  and a carrier concentration of  $2.5 \times 10^{16} \text{ cm}^{-3}$ . The  $K$  samples were rectangular parallelepipeds with a cross-sectional area of  $6 \text{ mm}^2$ , and the long dimension was approximately in the  $\langle 111 \rangle$  direction. The experimental results are shown in Figs. 3 and 4.

### Determination of Required Velocities and Frequencies

In the above formulations, the values of various parameters are required to carry out the analysis of the experimental data. The velocities  $V_{T1}$  and  $V_L$  in InAs were calculated from the elastic constants measured by Gerlich.<sup>8</sup> The velocity  $V_{T2}$  of high-frequency phonons ( $\omega_1 < \omega < \omega_2$ ) was determined assuming for the ratio  $V_{T2}/V_{T1}$  in InAs the same value (0.363) as in GaAs and InSb.<sup>9,10</sup>

The phonon frequencies  $\omega_2$  and  $\omega_3$  at the zone boundary in InAs were determined, using the known values of these parameters in Ge, Si, and other III-V compounds. This was performed by plotting (Fig. 1) the values of longitudinal acoustic frequency  $\omega_{LA}$  (i. e.,  $\omega_3$ ) at the zone boundary as a function of  $(c_{11}a/M_1)^{1/2}$  for various compounds for which the frequencies  $\omega_{LA}$  are known. In a cubic crystal with two atoms per unit cell,  $M_1$  is the mass of the heavier atom,  $c_{11}$  one of the second-order elastic constants, and  $a$  the interatomic distance.

A similar plot was carried out for longitudinal

optical frequency  $\omega_{LO}$  at the zone boundary as a function of  $(c_{11}a/M_2)^{1/2}$ , where  $M_2$  is the mass of the lighter atom (Fig. 2). Both curves in Figs. 1 and 2 are approximately linear, and the deduced values for  $(c_{11}a/M_1)^{1/2} = 1.62 \times 10^{13}$  and  $(c_{11}a/M_2)^{1/2} = 2.01 \times 10^{13} \text{ rad sec}^{-1}$  in InAs are, respectively,  $\omega_{LA} = \omega_3 = 133 \text{ cm}^{-1}$  and  $\omega_{LO} = 200 \text{ cm}^{-1}$ . The value thus obtained for  $\omega_{LA}$  is close to that given by Spitzer.<sup>15</sup> Moreover, from the plot of  $\omega_{TA}$  as a function of  $\omega_{LO}$  given by Mitra<sup>11</sup> for different diamond-structure materials, the value  $\omega_{TA} = 55 \text{ cm}^{-1}$  was deduced for InAs. On the other hand, the acoustic transverse frequency  $\omega_1$  was calculated, assuming the same ratio  $\omega_2/\omega_1$  in InAs as in GaAs and InSb ( $\omega_2/\omega_1 = 1.105$ ), because these materials exhibit the same structure.<sup>9,10</sup>

### Comparison of Theory and Experiment

The mass-difference-scattering parameter  $A$  was calculated on the basis of the Klemens relation applied to a compound:  $A = 0.24 \times 10^{-44} \text{ sec}^3$ . The Casimir length  $L$  was deduced from the cross section of the sample:  $L = 0.277 \text{ cm}$ . The Debye temperature was taken equal to 250 °K.

The fit of the experimental data on the basis of the Callaway expression is shown in Fig. 3. The agreement was obtained from low temperatures up to  $\sim 90 \text{ °K}$  for the parameters given in Table I.

The adopted value  $A_{\text{expt}}$  is about three times the calculated one  $A_{\text{theor}}$ . The origin of this discrepancy is presently not understood and may be due to an additional impurity or lattice defect scattering. The small difference between the experimental value  $L_{\text{expt}}$  and the calculated one  $L_{\text{theor}}$  can be because of specular reflection, or finite length of the sample. However, an additional electron-phonon scattering cannot be ruled out, and might perhaps account for the experimental data near the maximum, using the calculated values of  $A$  and  $L$  instead of the adjusted ones.

The results of the analysis of the experimental

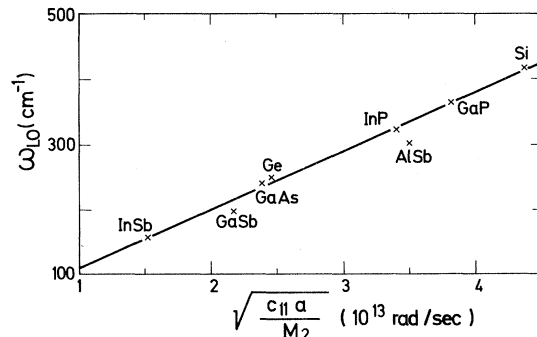


FIG. 2. Values of the longitudinal optical frequency  $\omega_{LO}$  at the zone boundary vs  $(c_{11}a/M_2)^{1/2}$  for the different III-V compounds Ge and Si (see references in caption of Fig. 1).

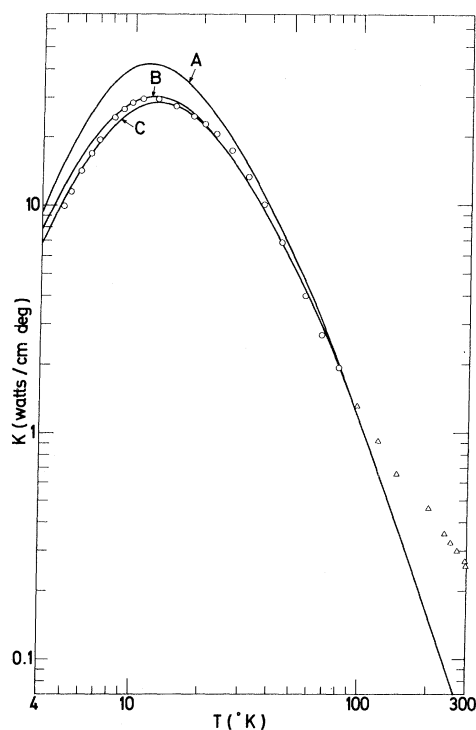


FIG. 3. Variation of the thermal conductivity  $K$  of InAs as a function of temperature. Solid lines are calculated on the basis of Callaway's expression (curve A:  $L=0.277$  cm,  $A=0.24 \times 10^{-44}$  sec<sup>3</sup>,  $B=6.6 \times 10^{-23}$  sec deg<sup>-3</sup>; curve B:  $L=0.277$  cm,  $A=0.8 \times 10^{-44}$  sec<sup>3</sup>,  $B=6.6 \times 10^{-23}$  sec deg<sup>-3</sup>; curve C:  $L=0.23$  cm,  $A=0.8 \times 10^{-44}$  sec<sup>3</sup>,  $B=6.6 \times 10^{-23}$  sec deg<sup>-3</sup>).

data on the basis of Holland's formulation are shown on Fig. 4. A satisfactory fit between 5 and 300 °K was obtained, using the experimental values  $L_{\text{expt}}$  and  $A_{\text{expt}}$ , the above deduced velocities and frequencies, and the adjusted parameters  $B_{\text{TN}}$ ,  $B_{\text{TU}}$ , and  $B_{\text{L}}$ . All these values are given in Table II.

It can be seen, that a dominant contribution of transverse phonons to thermal conductivity is exhibited at low and high temperatures, whereas longitudinal phonons make a comparable contribution at intermediate temperatures. Similar conclusions have already been obtained in other semiconductors on the basis of the same formulation (see Refs. 2 and 16-18). A dominant contribution

TABLE I. Experimental and calculated values of various parameters involved in the analysis, using Callaway's expression.

$V$	$L_{\text{expt}}$	$L_{\text{theor}}^a$	$A_{\text{expt}}$	$A_{\text{theor}}^a$	$B$
$2.54 \times 10^5$ cm/sec	0.23 cm	0.277 cm	$0.8 \times 10^{-44}$ sec <sup>3</sup>	$0.24 \times 10^{-44}$ sec <sup>3</sup>	$6.6 \times 10^{-23}$ sec deg <sup>3</sup>

<sup>a</sup>Calculated values.

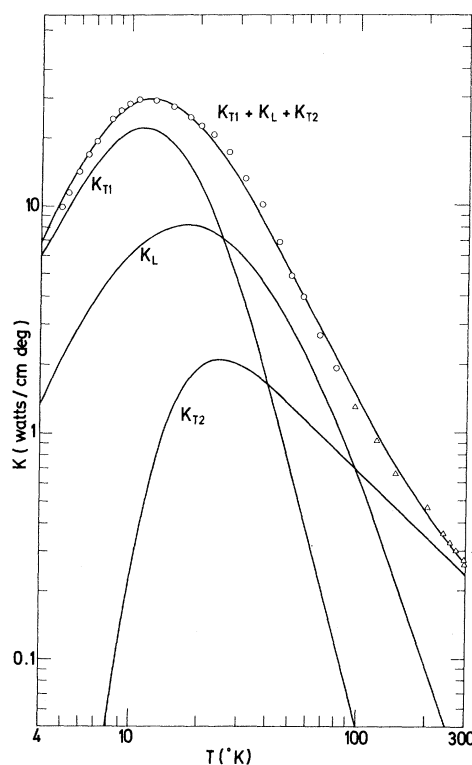


FIG. 4. Analysis of the thermal conductivity  $K$  of InAs, using Holland's formulation. Solid lines are calculated curves,  $K_{\text{T1}}$  and  $K_{\text{T2}}$  are the contributions of transverse phonons, and  $K_{\text{L}}$  the contribution of longitudinal phonons.

of transverse phonons at low temperature is expected because of their lower velocity. However, the predominance of the contribution of transverse phonons at high temperatures is still a matter of discussion.<sup>19</sup> One may ask whether such a conclusion obtained on the basis of Holland's model is not due to the theoretical formulation itself, and more precisely, to the choice of the three-phonon relaxation times adopted for longitudinal and transverse phonons. It is known that at high tempera-

TABLE II. Parameters used in the analysis on the basis of Holland's formulation.

$V_{\text{T1}} (\omega < \omega_1)$	$2.1 \times 10^5$ cm/sec
$V_{\text{T2}} (\omega_1 < \omega < \omega_2)$	$0.76 \times 10^5$ cm/sec
$V_{\text{L}}$	$4.34 \times 10^5$ cm/sec
$V$	$2.54 \times 10^5$ cm/sec
$L_{\text{expt}}$	0.23 cm
$\omega_1$	$0.94 \times 10^{13}$ rad sec <sup>-1</sup>
$\omega_2$	$1.04 \times 10^{13}$ rad sec <sup>-1</sup>
$\omega_3$	$2.45 \times 10^{13}$ rad sec <sup>-1</sup>
$A_{\text{expt}}$	$0.8 \times 10^{-44}$ sec <sup>3</sup>
$B_{\text{TN}}$	$0.19 \times 10^{-10}$ deg <sup>-4</sup>
$B_{\text{TU}}$	$6.5 \times 10^{-18}$ sec
$B_{\text{L}}$	$1.7 \times 10^{-23}$ sec deg <sup>-3</sup>

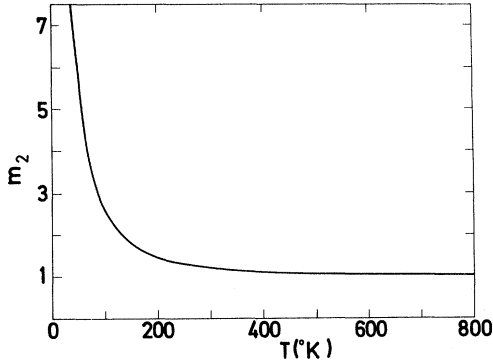


FIG. 5. Variation with temperature of the upper limit  $m_2$  of the exponent  $m$  in the relaxation rate  $\tau^{-1} \sim T^m$  for three-phonon processes in InAs.

tures the three-phonon relaxation times vary as  $T^{-1}$ , thus giving rise to a temperature dependence of the form  $K \sim T^{-1}$ . If this is the case for transverse phonons in the Holland formulation, the relaxation time in the latter for longitudinal phonons is assumed to vary as  $T^{-3}$  in all temperature ranges. Thus, one expects  $K_L$  to decrease rapidly with increasing temperature in the high-temperature range, leading to a predominant contribution of transverse phonons to heat conduction. Consequently, it was felt interesting to carry out the analysis of the phonon conductivity of InAs on the basis of a more appropriate approach for relaxation times. This is performed in Sec. III using the analysis of Guthrie for the temperature dependence of three-phonon processes.

### III. RELAXATION TIMES AND CORRESPONDING TEMPERATURE INTERVALS

In this section, the phonon conductivity of InAs is analyzed on the basis of the Guthrie results for three-phonon processes.

Guthrie has shown that for a three-phonon relaxation rate of the form  $\tau_{3p}^{-1} \sim g(\omega) T^m$ , the temperature-dependent exponent  $m$  exhibits an upper limit  $m_2$  which is given by

$$m_2 = x_0 [2(e^{x_0} - 1)^{-1} + 1] - 1,$$

with  $x_0 = \hbar\omega_0/kT$ , where  $\omega_0$  is the frequency of optical phonons at the zone boundary.

At low temperatures, Herring<sup>20</sup> has derived the three-phonon relaxation times for N processes:

$$\tau_{3pL}^{-1} \sim \omega^2 T^3 \text{ for longitudinal phonons,}$$

$$\tau_{3pT}^{-1} \sim \omega T^4 \text{ for transverse phonons.}$$

The above expressions are valid in the nondispersive region of the spectrum. Guthrie has shown that  $\tau_{3pT}^{-1} \sim \omega T^4$  is thus valid for  $T < \hbar\omega_{Tsd}/4.9k$ , where  $\omega_{Tsd}$  is the frequency limit of nondispersion on the transverse acoustic branch. In the same

manner,  $\tau_{3pL}^{-1} \sim \omega^2 T^3$  is found to be valid for  $T < \hbar\omega_{Lsd}/3.85k$ , where  $\omega_{Lsd}$  is the frequency limit of nondispersion on the longitudinal branch. In the absence of known values of these limits, it was approximated that  $\omega_{Tsd} = \omega_{Lsd} = \omega_2$  (the frequency limit of transverse phonons at zone boundary). The corresponding temperature limits in InAs for  $\tau_{3pT}^{-1} \sim \omega T^4$  and  $\tau_{3pL}^{-1} \sim \omega^2 T^3$  are, respectively,  $T < 16$  and  $< 21$  °K. Because of the small gap between the two values, we adopted for simplification the limit  $T < 16$  °K in both cases. On the other hand, according to the results obtained by Herring at high temperature for N processes ( $\tau_{3pL}^{-1} \sim \omega^2 T$  for longitudinal phonons, and  $\tau_{3pT}^{-1} \sim \omega T$  for transverse phonons), it was assumed that the frequency dependence at all temperatures for  $\tau_{3p}^{-1}$  was  $\omega^2$  for longitudinal phonons, and  $\omega$  for transverse phonons. Moreover the temperature intervals were chosen using the approximation that each interval corresponds to a constant integer value of  $m$ , in agreement with the Guthrie condition  $m < m_2$ . Figure 5 gives the variation of  $m_2$  as a function of temperature for InAs. The relaxation rates and the corresponding temperature intervals are given in Table III. Guthrie's analysis was supposed to be applicable for N and U processes. In the absence of information about U processes, it was assumed that the deduced relaxation times and corresponding temperature intervals are also valid for U processes.

On the other hand, relaxation rates for boundary effect were taken equal to

$$\tau_{BT}^{-1} = V_T/L \text{ for transverse phonons,}$$

$$\tau_{BL}^{-1} = V_L/L \text{ for longitudinal phonons,}$$

where  $V_T$  and  $V_L$  are, respectively, the velocities of low-frequency transverse and longitudinal phonons, calculated above. Thus, the combined relaxation times  $\tau_c$  used in the following analysis are

$$\tau_{cT}^{-1} = V_T/L + A\omega^4 + \tau_{3pT}^{-1}$$

and

$$\tau_{cL}^{-1} = V_L/L + A\omega^4 + \tau_{3pL}^{-1},$$

where  $A\omega^4$  is relative to mass-difference scattering, and  $\tau_{3pT}^{-1}$  and  $\tau_{3pL}^{-1}$  are shown in Table III. It is worth

TABLE III. Relaxation rates  $\tau_{3p}^{-1}$  for three-phonon processes and the corresponding temperature intervals.

Transverse phonons	Longitudinal phonons	Temperature intervals (°K)
$B_{T4}\omega T^4$	$B_{L3}\omega^2 T^3$	$T \leq 16$
$B_{T3}\omega T^3$	$B_{L2}\omega^2 T^2$	16–88
$B_{T2}\omega T^2$	$B_{L2}\omega^2 T^2$	88–130
$B_{T1}\omega T$	$B_{L1}\omega^2 T$	$T \geq 130$

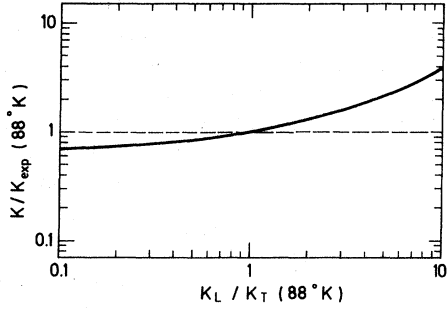


FIG. 6. Variation of  $K/K_{\text{exp}}$  vs  $K_L/K_T$  at 88°K for InAs.  $K_{\text{exp}}$  is the experimental thermal conductivity.  $K_T$  and  $K_L$  are the contributions of transverse and longitudinal phonons, respectively, and  $K = K_T + K_L$ .

noticing already that no fit could be obtained with the above relaxation times  $\tau_{cT}$  and  $\tau_{cL}$  for  $T > \sim 130$ °K. It was found that the introduction in  $\tau_c$  (for  $T > 130$ °K) of the four-phonon relaxation time given by Pomeranchuk<sup>21</sup> ( $\tau_H^{-1} = B_H \omega^2 T^2$ ) enables us to fit the experimental data above 130°K, as explained below.

#### IV. THEORETICAL FORMULATION AND ANALYSIS

The contribution of each polarization branch to the phonon conductivity is given by the expression

$$K = \frac{1}{6\pi^2} \int \tau_c V^2 \frac{\hbar^2 \omega^2}{k^2 T^2} \frac{e^{\hbar\omega/kT}}{(e^{\hbar\omega/kT} - 1)^2} q^2 dq,$$

the integration interval being the Brillouin zone.

In Holland's formulation, the integration is performed, assuming a linear relation between  $\omega$  and  $q$ . In the present approach, we used a more realistic approximation adopted by Joshi and Verma<sup>4</sup> in a similar analysis on Si:

$$q = \omega V_i^{-1} (1 + \alpha_i \omega),$$

where  $\alpha_i$  ( $i$  representing L or T) is a characteristic parameter of the branch considered and  $V_i$  is the group velocity of low-frequency phonons of polarization  $i$ . For the determination of  $\alpha_L$  and  $\alpha_T$ , the above relation was applied at the zone boundary, using the values of  $\omega_2$  and  $\omega_3$ , and  $q_{\text{max}} = (6n\pi^2)^{1/3}$ , where  $n$  is the number of lattice points per unit volume of the crystal.

Assuming, as in the case of Holland's model, that the transverse branches are degenerate, and using the above quadratic relation ( $q, \omega$ ) the phonon conductivity can be written<sup>4</sup>

$$K = K_L + K_T,$$

where

$$K_L = \frac{k^4 T^3}{6\pi^2 V_L \hbar^3} \int_0^{\theta_L/T} \tau_{cL} \frac{x^4 e^x}{(e^x - 1)^2} \frac{(1 + \beta_L T x)^2}{1 + 2\beta_L T x} dx,$$

and

$$K_T = \frac{k^4 T^3}{3\pi^2 V_T \hbar^3} \int_0^{\theta_T/T} \tau_{cT} \frac{x^4 e^x}{(e^x - 1)^2} \frac{(1 + \beta_T T x)^2}{1 + 2\beta_T T x} dx,$$

with

$$x = \hbar\omega/kT, \quad \theta_L = \hbar\omega_3/k,$$

$$\theta_T = \hbar\omega_2/k, \quad \beta_L = \alpha_L k/\hbar, \quad \beta_T = \alpha_T k/\hbar.$$

The analysis was carried out using the following procedure. The fit was established for the successive temperature intervals, from low to high temperature, using in each interval the appropriate relaxation times. Because of the continuity condition of  $K$  at the interval limits, there are only two adjustable parameters (one  $B_L$  and one  $B_T$ ) for the three-phonon processes. In the low-temperature range ( $T < 16$ °K) the fit is independent of  $B_L$  and  $B_T$ . The values  $B_{L3}$ ,  $B_{T4}$  are determined from the values of  $B_{L2}$  and  $B_{T3}$  deduced from the plot at 88°K (Fig. 6) of the curve  $(K/K_{\text{exp}})_{88^\circ\text{K}}$  as a function of  $(K_L/K_T)_{88^\circ\text{K}}$ , where  $K_{\text{exp}}$  is the experimental thermal conductivity and  $K = K_L + K_T$ . It can be seen that the agreement ( $K/K_{\text{exp}} = 1$ ) at 88°K is obtained for  $K_L \approx K_T$ . With the deduced parameters the fit can be made up to  $\sim 130$ °K. All the parameters used are listed in Table IV, and the fit of the experimental data is shown in Fig. 7.

In the temperature range  $T > \sim 130$ °K, the experimental thermal conductivity decreases more rapidly than  $T^{-1}$ , and as the three-phonon relaxation times  $\tau_{3pT}$  and  $\tau_{3pL}$  in this interval vary as  $T^{-1}$ , no fit is possible on the basis of three-phonon processes alone. At high temperatures, it has been suggested that four-phonon processes be taken into account in order to explain a temperature dependence of phonon conductivity greater than  $T^{-1}$ . We looked for such possibility, and the four-phonon relaxation time of Pomeranchuk ( $\tau_H^{-1} = B_H \omega^2 T^2$ ) was included, at  $T > 130$ °K, in the combined relaxation times  $\tau_{cL}$  and  $\tau_{cT}$ . However, the introduction of two different parameters  $B_{HL}$  and  $B_{HT}$ , respectively,

TABLE IV. Parameters used in the analysis of the phonon conductivity on the basis of Guthrie's three-phonon relaxation times, and a quadratic ( $q, \omega$ ) relation.

$V_L$	$4.34 \times 10^5$ cm/sec
$V_T$	$2.1 \times 10^5$ cm/sec
$L_{\text{exp}}$	0.18 cm
$A_{\text{exp}}$	$0.8 \times 10^{-44}$ sec <sup>3</sup>
$\theta_L$	187°K
$\theta_T$	80°K
$\alpha_L$	$5.36 \times 10^{-14}$ sec
$\alpha_T$	$1.54 \times 10^{-13}$ sec
$B_{L3}$	$1.3 \times 10^{-22}$ sec deg <sup>-3</sup>
$B_{T4}$	$1.85 \times 10^{-11}$ deg <sup>-4</sup>
$B_H$	$6.9 \times 10^{-22}$ sec deg <sup>-2</sup>

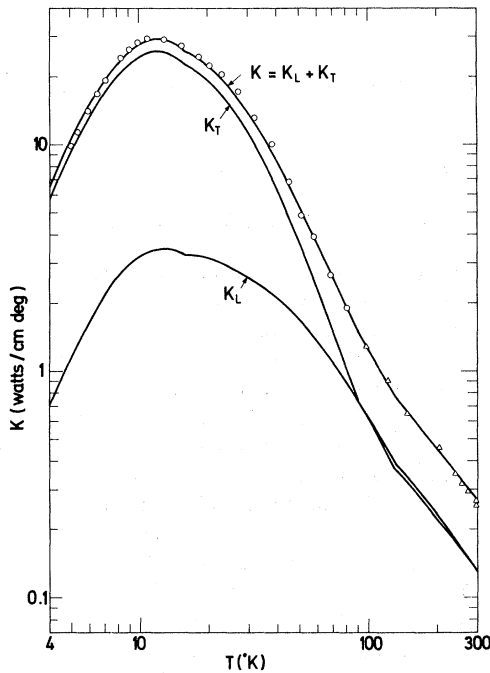


FIG. 7. Analysis of the thermal conductivity of InAs, using, for three-phonon processes, relaxation times deduced from Guthrie's work. Above 130 °K, four-phonon processes are included.

in  $\tau_{3\phi L}^{-1}$  and  $\tau_{3\phi T}^{-1}$  would lead to undetermined values of these parameters. Thus the fit was carried out, assuming  $B_{HL} = B_{HT} = B_H$ . This explains the comparable contributions of  $K_L$  and  $K_T$  to  $K$  in the temperature range 130–300 °K (Fig. 7). The appropriate value of  $B_H$  was found to be  $6.9 \times 10^{-22}$  sec deg<sup>-2</sup>. As four-phonon processes are generally considered to be important at temperatures higher than the Debye temperature, an alternative approach might be to introduce  $\tau_H$  only in the combined relaxation time of transverse phonons, which corresponds to a lower value for  $\theta_T$ . This would lead to a contribution of  $K_L$  higher than that of  $K_T$  at about 300 °K.

The striking feature arising from this analysis concerns the comparable contributions of longitudinal and transverse phonons to the heat conduction at high temperature, such behavior being in disagreement with the conclusion derived above from Holland's formulation.

However, it is to be recalled that a similar analysis by Joshi and Verma<sup>4</sup> of the phonon conductivity of silicon has led to a different conclusion concerning the role of longitudinal phonons. It was found that the latter give a small contribution to the lattice conductivity of the material at all temperatures. Thus, the major contribution of transverse phonons, particularly at high temperature, was consistent with the results deduced from Holland's model.

One might therefore ask whether this difference between the roles of longitudinal phonons in the high-temperature heat conduction in silicon and InAs is due to the theoretical formulation used, or is somewhat characteristic of the thermal behavior of the materials. This requires further theoretical investigation and similar studies on other materials.

#### CONCLUSION

The above analyses of the phonon conductivity of InAs have been carried out on the basis of various approaches, looking tentatively for an investigation of the relative contributions of transverse and longitudinal phonons towards heat conduction.

As previously observed on other materials, Callaway's expression was found to give a satisfactory fit of the experimental data in the region of predominant boundary and mass-difference scatterings.

Holland's formulation was found to explain the temperature dependence of the phonon conductivity in the entire temperature range (5–300 °K). This approach suggests the dominant contribution of transverse phonons at low and high temperatures. If the low-temperature behavior is in agreement with what one may expect according to the lower velocity of transverse phonons, the observed major contribution of the latter at high temperatures is still questionable. The choice of three-phonon relaxation times adopted in this formulation enhances the contribution of transverse phonons at high temperature.

Such a conclusion is consistent with the results obtained from the analysis performed on the basis of more appropriate three-phonon relaxation rates given by Guthrie and using a more realistic quadratic ( $q, \omega$ ) relation. If the contribution of transverse phonons is dominant at low temperature, it is found that, with increasing temperature, the relative contribution of longitudinal phonons increases and becomes, at high temperature, comparable to that of transverse phonons. Moreover, it was shown tentatively that the introduction of four-phonon processes in this approach may account for the thermal conductivity at higher temperatures ( $T > 130$  °K).

However, the present work shows that the determination of the relative contributions of transverse and longitudinal phonons to the heat conductivity of InAs at high temperature depends on the choice of the scattering rates for phonon-phonon processes, and this cannot presently be conclusively established.

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