Lattice thermal conductivity of Mg_2Ge in the temperature range 4–800°K

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The Sharma-Dubey-Verma model of two-mode conduction, similar to the Holland model, has been applied to explain the phonon conductivity data of Mg_2Ge in the temperature range 4–800°K. The temperature exponents $m(T)$ for the three-phonon scattering relaxation rates have been calculated for both class-I and class-II events following Guthrie. Excellent agreement has been found between theoretical and experimental values of the phonon conductivity of Mg_2Ge over a wide range of temperature. Separate percentage contributions of transverse and longitudinal phonons have also been calculated in the temperature range of investigation.

I. INTRODUCTION

Recently Martin[1] has measured the phonon conductivity of Mg_2Ge, Mg_2Si, and Mg_2Sn in the temperature ranges 4–700°K, 3–300°K, and 4–700°K, respectively, and has interpreted his results using Holland's model[2] of two-mode conduction. He used $\tau_{3ph,T}^{-1} \propto \omega T^4$ and $\tau_{3ph,L}^{-1} \propto \omega^2 T^3$ in the entire temperature range, where $\tau_{3ph,T}^{-1}$ and $\tau_{3ph,L}^{-1}$ are the three-phonon scattering relaxation rates for transverse and longitudinal phonons, respectively. These relations are actually valid only in the low-temperature regions.

Recently[3–8] we have proposed a modification of Holland's model following the work of Guthrie.[9,10] This is known as the Sharma-Dubey-Verma (SDV) model. In the SDV model, phonon-phonon scattering events are classified into two groups: class-I events in which the carrier phonon is annihilated by combination, and class-II events in which the annihilation takes place by splitting. The three-phonon scattering relaxation rate used in the SDV model is similar to the Klemens[11–13] expression. Using this approach, the phonon conductivity of several semiconductors[3–8] was calculated previously, and good agreement was found between calculated and experimental values of phonon conductivity.

Martin tried to explain his results using the Holland model and found good agreement between theory and experiment at high temperature but noted discrepancies at low temperatures. In interpreting the same experimental data, we have been able to obtain excellent agreement in the high-temperature region as well as in the low-temperature region. However, some discrepancies remain in the temperature region near the conductivity maximum. In the present work, the phonon conductivity of

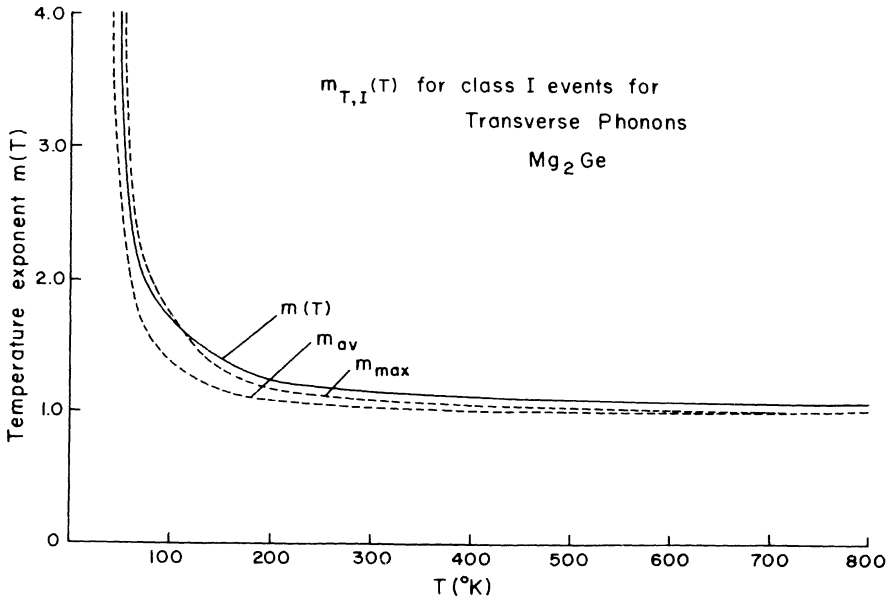

FIG. 1. $m(T)$ for the transverse phonon for class-I events in Mg_2Ge. m_{max} is the upper bound of Gutherie. m_{av} is the average of the upper and lower bounds of Guthrie.

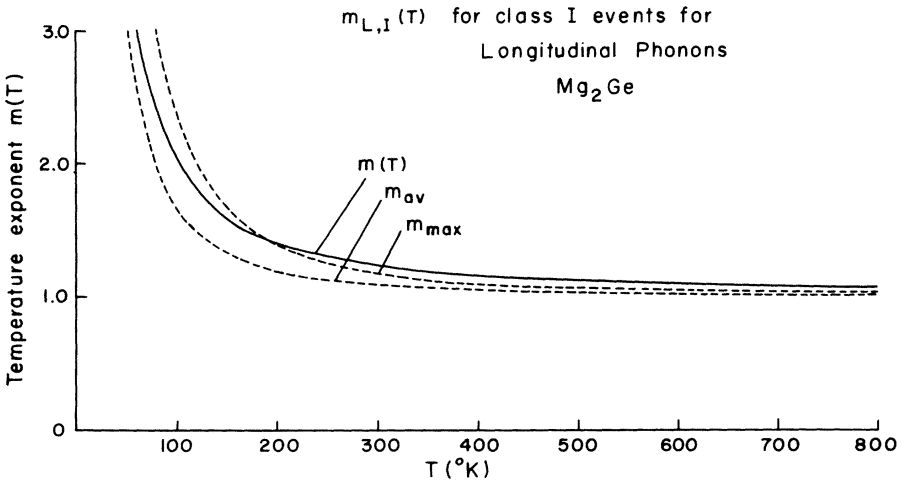

FIG. 2. $m(T)$ for the longitudinal phonon for class-I events in Mg_2Ge. m_{max} is the upper bound of Guthrie. m_{av} is the average of upper and lower bounds of Guthrie.

Mg_2Ge has been calculated under two distinct sets of assumptions using the SDV model. The first assumption is that at room temperature each phonon contributes equally, i.e., at 300° K the transverse and longitudinal phonon contribute 66.7% and 33.3%, respectively, to the total phonon conductivity. The second assumption is that at high temperatures, all the heat is carried by the transverse phonon alone.

The temperature exponents $m(T)$ for class-I events for transverse phonons and for class-I and class-II events for longitudinal phonons have been calculated, and these values of $m(T)$ are incorporated in the present investigation. Separate percentage contributions of the longitudinal and transverse phonon have also been calculated in both approaches in the temperature range of investigation.

II. CONDUCTIVITY INTEGRAL AND TEMPERATURE EXPONENT $m(T)$

According to the Sharma-Dubey-Verma model (details are given in Refs. 3–8), the three-phonon scattering relaxation rates are given by

$$\tau_{3ph,T}^{-1} = B_{T,I}\omega T^{m_{T,I}(T)} e^{-\Theta/\alpha T}, \quad (1)$$

since only class-I events are possible for the transverse phonon. For the longitudinal phonon one obtains

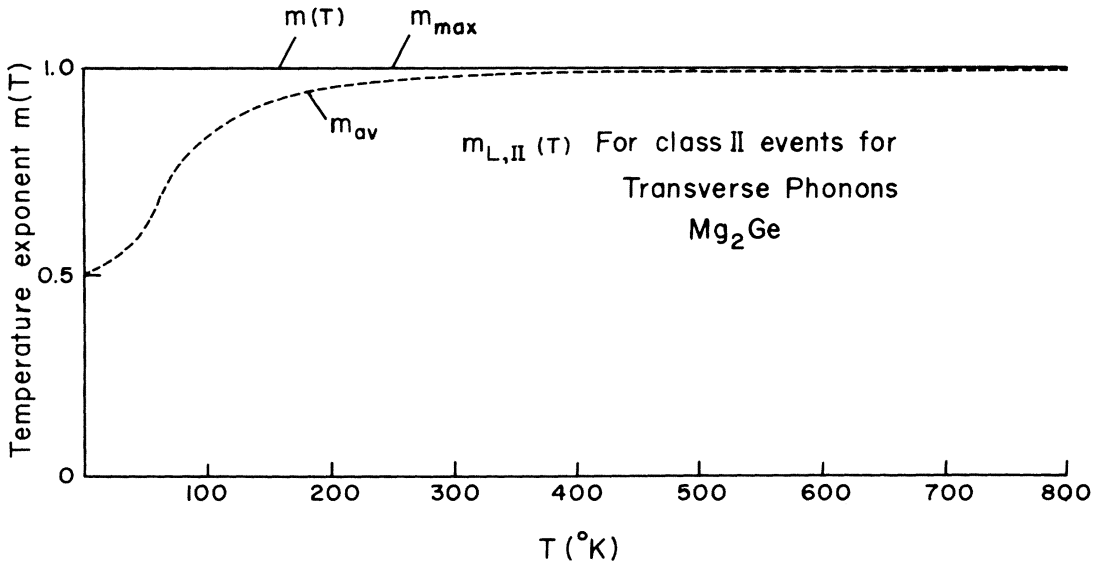

FIG. 3. $m(T)$ for longitudinal phonon for class-II events in Mg_2Ge. m_{max} is the upper bound of Guthrie. m_{av} is the average of upper and lower bounds of Guthrie.

$$\tau_{3ph,L}^{-1} = B_{L,I}\omega^2 T^{m_{L,I}(T)} e^{-\Theta/\alpha T} + B_{L,II}\omega^2 T^{m_{L,II}(T)} e^{-\Theta/\alpha T}, \quad (2)$$

where Θ is the Debye temperature and α is a constant which is the same as in the Klemens expression. The value of the temperature exponent $m(T)$ is given by

$$[m(T)]_I = m_I(T) = \frac{x_{max}}{e^{x_{max}} - 1} + 0.5x_{max} + \frac{\ln(1+\Theta/\alpha T)}{\ln T}, \quad (3)$$

$$[m(T)]_{II} = m_{II}(T) = \frac{0.5x_{max}e^{0.5x_{max}}}{e^{x_{max}} - 1} + 0.5 + \frac{\ln(1+\Theta/\alpha T)}{\ln T}, \quad (4)$$

where

$$x_{max} = \frac{\hbar\omega_{max}}{k_B T}.$$

At high temperature, $e^{-\Theta/\alpha T}$ reduces to unity and the value of $m(T)$ tends to unity for both polarization branches with the result that $\tau_{3ph,T}^{-1} \propto \omega T$ and $\tau_{3ph,L}^{-1} \propto \omega^2 T$, as previously found by Klemens. At low temperature, the exponent $m_{T,I}(T)$ tends to 4 and $m_{L,I}(T)$ tends to 3, which leads to a T^4 dependence for the transverse phonon and a T^3 dependence for the longitudinal phonon, similar to the Herring relations[14] for low-frequency phonons.

Using the above relaxation rates and applying the dispersion correction,[15] the thermal-conductivity integral can be written

$$\kappa = \kappa_T + \kappa_L \quad (5)$$

and

$$\kappa_T = \frac{2}{3}\frac{k_B}{2\pi^2}\left(\frac{k_B T}{\hbar}\right)^3 \left(\frac{1}{v_{T1}}\int_0^{\Theta_1/T} \frac{x^4 e^x (e^x-1)^{-2}(1+R_1x^2T^2)^2(1+3R_1x^2T^2)^{-1}dx}{(\tau_B^{-1})_T + Dx^4T^4 + \beta_{T,I}xT^{m_{T,I}(T)+1}e^{-\Theta/\alpha T}} + \frac{1}{v_{T2}}\int_{\Theta_1/T}^{\Theta_2/T} \frac{x^4 e^x (e^x-1)^{-2}(1+R_2x^2T^2)^2(1+3R_2x^2T^2)^{-1}dx}{(\tau_B^{-1})_T + Dx^4T^4 + \beta_{T,I}xT^{m_{T,I}(T)+1}e^{-\Theta/\alpha T}}\right), \quad (6)$$

$$\kappa_L = \frac{1}{3}\frac{k_B}{2\pi^2}\left(\frac{k_B T}{\hbar}\right)^3 \left(\frac{1}{v_{L1}}\int_0^{\Theta_4/T} \frac{x^4 e^x (e^x-1)^{-2}(1+R_4x^2T^2)^2(1+3R_4x^2T^2)^{-1}dx}{(\tau_B^{-1})_L + Dx^4T^4 + (\beta_{L,I}T^{m_{L,I}(T)+2} + \beta_{L,II}T^{m_{L,II}(T)+2})x^2e^{-\Theta/\alpha T}} + \frac{1}{v_{L2}}\int_{\Theta_4/T}^{\Theta_3/T} \frac{x^4 e^x (e^x-1)^{-2}(1+R_3x^2T^2)^2(1+3R_3x^2T^2)^{-1}dx}{(\tau_B^{-1})_L + Dx^4T^4 + (\beta_{L,I}T^{m_{L,I}(T)+2} + \beta_{L,II}T^{m_{L,II}(T)+2})x^2e^{-\Theta/\alpha T}}\right), \quad (7)$$

where

$$D = A(k_B/\hbar)^4,$$

$$\beta_{T,I} = B_{T,I}(k_B/\hbar),$$

$$\beta_{L,I} = B_{L,I}(k_B/\hbar)^2,$$

$$\beta_{L,II} = B_{L,II}(k_B/\hbar)^2,$$

$$R_i = r_i(k_B/\hbar)^2, \quad i = 1, 2, 3, 4$$

r_i is dispersion correction constant (as given in Ref. 15), k_B is the Boltzmann constant, $\hbar$ is the Planck constant divided by 2π, τ_B^{-1} is the boundary scattering relaxation rate and is given by $(\tau_B^{-1})_{T,L} = v_{T,L}/L$, where L is the Casmuir[16] length of the crystal. A is a constant, known as the isotropic scattering strength. It is given by

$$A = \frac{V_0}{4\pi v_s^3}\sum_i f_i\left(1 - \frac{m_i}{\bar{m}}\right)^2,$$

where V_0 is the atomic volume, m_i is the mass of the ith species of the atom, f_i is the fractional concentration of the ith species of the atom, $\bar{m}$ is average atomic mass, and v_s is the average phonon velocity.

III. LATTICE THERMAL CONDUCTIVITY OF Mg_2Ge

The constants related to the dispersion curve have been calculated from the dispersion curve of the sample investigated by Chung *et al.*[17] We are interested in explaining the phonon conductivity results of Mg_2Ge sample No. 1 of Martin. Therefore resonant phonon scattering is totally ignored in the calculation owing to the fact that Martin has reported that Mg_2Ge sample No. 1 displayed no neutral donor scattering. The temperature exponents $m_{T,I}(T)$, $m_{L,I}(T)$, and $m_{L,II}(T)$ have been calculated in the temperature range 4–800 °K by using Eqs. (3) and (4) and are shown in Figs. 1–3 together with m_{av} and m_{max}. In Figs. 1–3, m_{max} is the upper bound of the temperature exponent $m(T)$ given by Guthrie and m_{av} is the same as defined in the text. For more clarification the value of the temperature exponent $m(T)$ has also been listed in Table I.

Assuming that the entire phonon conductivity at 4 °K is due to the boundary scattering alone, $(\tau_B^{-1})_T$ and $(\tau_B^{-1})_L$ are calculated by adjusting the Casimir length of the crystal. The point-defect scattering

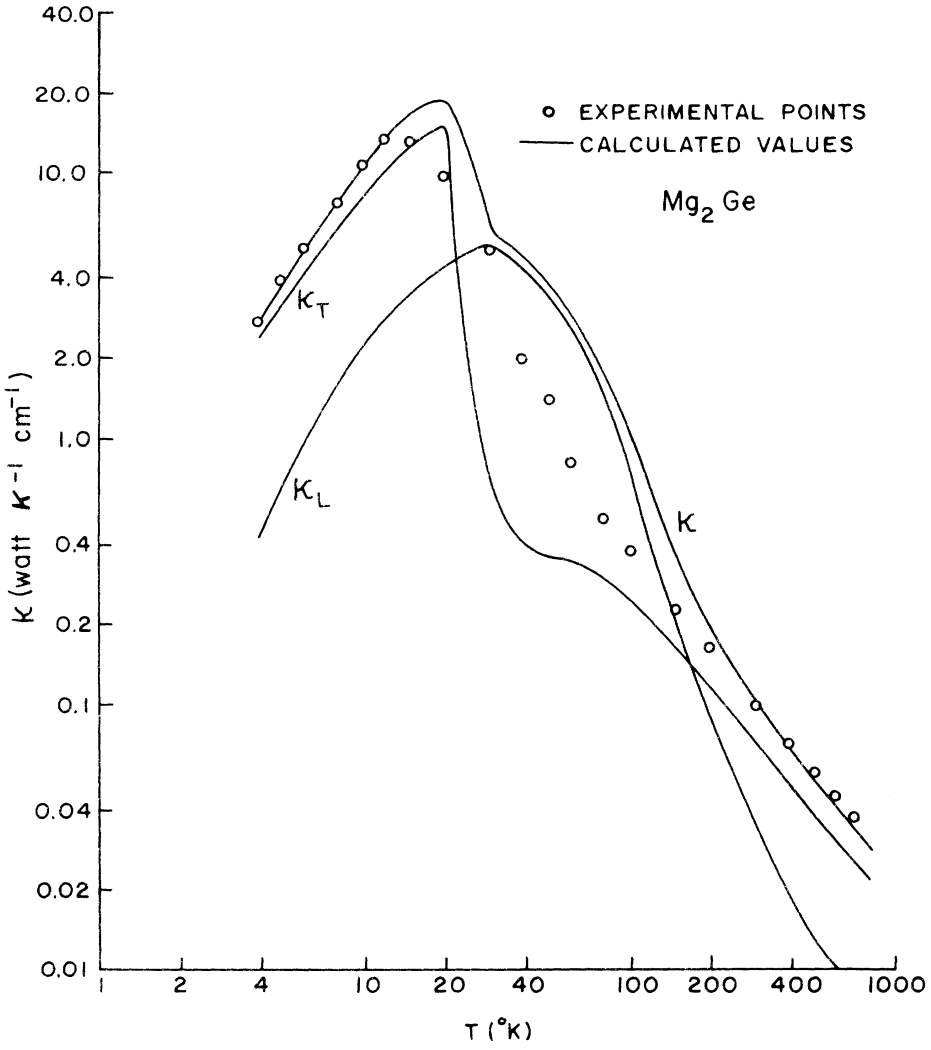

FIG. 4. Phonon conductivity of Mg_2Ge in the temperature range 4–800 °K. Solid lines are calculated values based on the assumption that each phonon contributes equally at room temperature, i.e., at 300 °K. Circles are experimental values.

strength A is adjusted at 10 °K ignoring the phonon-phonon scattering relaxation rate. Knowing the above constants and parameters, the phonon conductivity of Mg_2Ge has been calculated by two different approaches. In the first approach it is assumed that at room temperature, i.e., at 300 °K each phonon contributes equally. Therefore, the contributions of the transverse and longitudinal phonons towards the total phonon conductivity are 66.7% and 33.3% at 300 °K. Assuming these contributions, $B_{T,I}$, $B_{L,I}$, and $B_{L,II}$ have been adjusted, and it is found that the factor $\beta_{L,I}x^2 \times T^{m_{L,I}(T)+2} e^{-\Theta/\alpha T}$ contributes very little. Therefore this factor has been neglected in actual calculations. Adjusting these constants, the phonon conductivity of Mg_2Ge has been calculated.

In the second approach, $B_{T,I}$ is adjusted at 700 °K using the assumption that heat conduction is due to transverse phonons. Adjusting $B_{T,I}$, the phonon conductivity due to the transverse phonon has been calculated in the temperature range 4–800 °K. At lower temperatures, the transverse-phonon contribution is noticeably smaller than the observed phonon conductivity of the sample. This amount $(\kappa - \kappa_T)$ is attributed to the contribution of longitudinal phonons, and $B_{L,II}$ has been adjusted by this amount at 80 °K by numerical integration of Eq. (7).

Results of both approaches are shown in Figs. 4 and 5. Separate percentage contributions of the transverse and longitudinal phonons are also calculated in both approaches and are shown in Figs. 6 and 7. The constants and parameters used in the present investigation are given in Table II.

IV. RESULTS AND DISCUSSIONS

From Fig. 4, it can be observed that the agreement between calculated and experimental data is

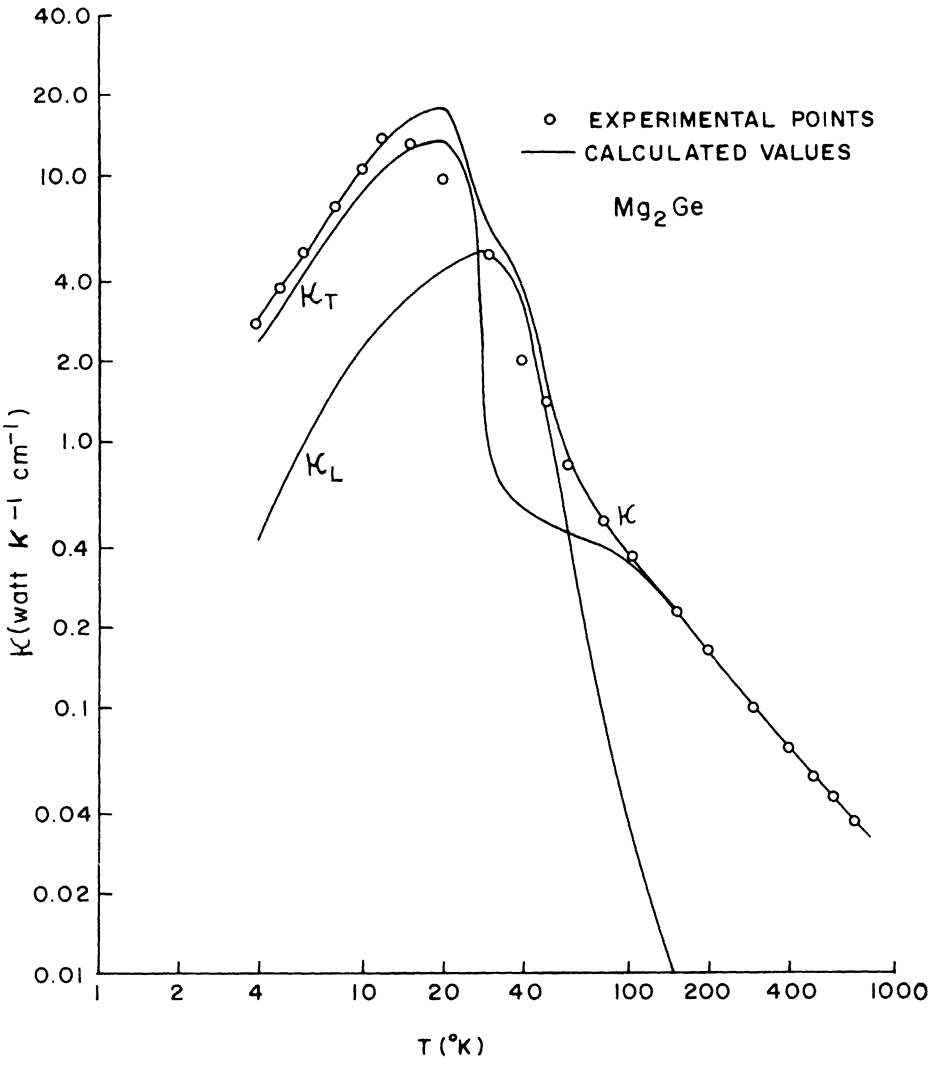

FIG. 5. Phonon conductivity of Mg_2Ge in the temperature range 4–800 °K. Solid lines are calculated values based on the assumption that at high temperature all the heat is carried by the transverse phonon alone. Circles are experimental points.

good at low temperatures. At high temperature, the agreement between calculated and experimental data is considerably good except in the temperature range 20–200 °K. At the same time, from Fig. 6, it can be seen that the transverse-phonon contribution is as high as 80% at 800 °K and apparently even under this set of starting assumptions, we find that at high temperatures, most of the heat is transported by transverse phonons. From Fig. 5, one can observe that the agreement between calculated values and experimental data is excellent in the entire temperature range except at some temperature near the conductivity maximum, i.e., 15–40 °K. These discrepancies are due to point-defect scattering. At high temperature all the heat is carried by transverse phonons alone. Thus in both approaches agreement between calculated and experimental data is found to be good except at some temperatures near conductivity maxima which are due to point-defect scattering. From Figs. 6 and 7, one concludes that in the low-temperature region, the ratio of the percentage contributions of the

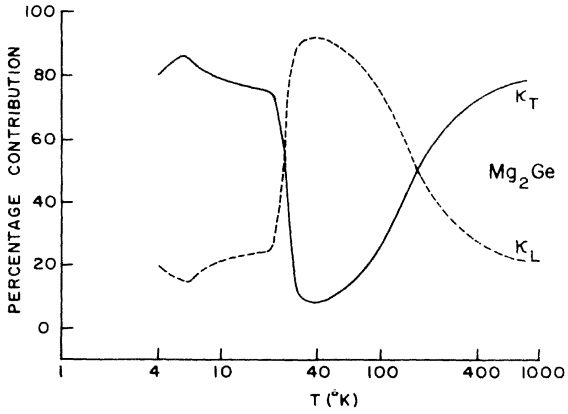

FIG. 6. Separate percentage contributions of the transverse and longitudinal phonons of Fig. 4.

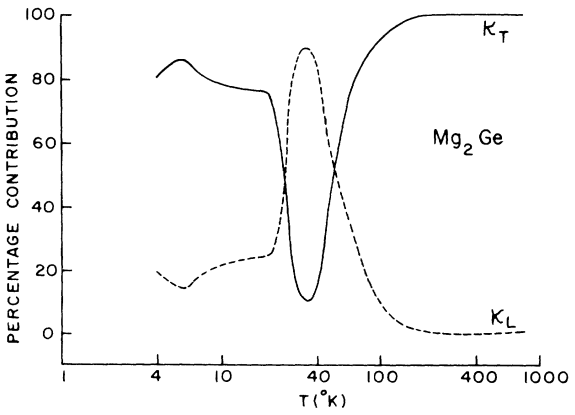

FIG. 7. Separate percentage contributions of the transverse and longitudinal phonons of Fig. 5.

transverse and longitudinal phonon, $\%\kappa_T/\%\kappa_L$, depends upon the ratio of

$$2(v_L/v_T)(\tau_{B,L}^{-1}/\tau_{B,T}^{-1}) = 2(v_L/v_T)^2 ,$$

where v_T and v_L are the velocities of the transverse and longitudinal phonon. This ratio found by the above calculation is nearly the same as shown in Figs. 6 and 7.

At a little higher temperature, towards the conductivity maximum, the contribution of the transverse phonon begins to decrease while the reverse is true for the longitudinal-phonon contribution. At a certain temperature (nearly 35 °K for Mg_2Ge) the transverse-phonon contribution begins to increase again and tends to 80% in the first approach and to 100% in the second approach at higher temperature. The same behavior is also observed by Sharma *et al.*[15] in the phonon conductivity of Ge, based on the Holland model. The original cause of this kind of curve resides in the role of point-defect scattering at these temperatures. (This was already stated in the previous paper by the author, Ref. 15.) From Figs. 1–3, and Table I, one concludes that $m(T)$ tends to unity at higher temperatures, which shows the T dependence of the phonon-phonon scattering relaxation rate for both modes, longitudinal and transverse phonons, as previously found by Klemens and other workers. At lower temperatures, the values of $m_{T,\mathrm{I}}(T)$ and $m_{L,\mathrm{I}}(T)$ tend to 4 and 3, respectively, as previously found by Herring. Thus one can conclude that the relations $\tau_{3\mathrm{ph},T}^{-1} \propto T$, $\tau_{3\mathrm{ph},L}^{-1} \propto T$ and $\tau_{3\mathrm{ph},T}^{-1} \propto T^4$, $\tau_{3\mathrm{ph},L}^{-1} \propto T^3$ are used at high and low temperatures, respectively, in the present analysis. At the same time, it can also be concluded that at intermediate temperatures, the values of the temperature exponent $m(T)$ lie between 1 and 4 for transverse phonons and between 1 and 3 for longitudinal phonons, which is similar to the earlier prediction of Guthrie. It should be noted that the distribution of the heat transport (assignment to separate modes) is indeed sensitive to the input assumptions. It is to be expected that a set of original assumptions taking account of the more favorable group velocity of the longitudinal phonons would produce results assigning larger fractions of the conduction to that branch at all temperatures.

TABLE I. The values of the temperature exponents $m(T)$ which are obtained from the calculation of Eqs. (3) and (4) and are used in the present investigation.

T	$m_{T,\mathrm{I}}(T)$	$m_{L,\mathrm{I}}(T)$	$m_{L,\mathrm{II}}(T)$
800	1.0638	1.0698	1.0
700	1.0736	1.0825	1.0
600	1.0769	1.0883	1.0
500	1.1056	1.1180	1.0
400	1.1345	1.1592	1.0
300	1.1840	1.2288	1.0
200	1.1912	1.3888	1.0
150	1.4102	1.5768	1.0
100	1.6836	2.0211	1.0
80	1.9167	2.3965	1.0
60	2.3455	3.0	1.0
40	3.2901	310	1.0
30	4.0	3.0	1.0
20	4.0	3.0	1.0
15	4.0	3.0	1.0
10	4.0	3.0	1.0
8	4.0	3.0	1.0
6	4.0	3.0	1.0
5	4.0	3.0	1.0
4	4.0	3.0	1.0

TABLE II. Constants and parameters used in the calculation of the phonon conductivity of Mg_2Ge in the temperature range 4–800 °K.

$V_{T_1} = 3.9\times10^5$ cm/sec, $V_{T_2} = 1.8\times10^5$ cm/sec	
$V_{L_1} = 5.8\times10^5$ cm/sec, $V_{L_2} = 2.4\times10^5$ cm/sec	
$\Theta_1 = 140$ °K, $\Theta_4 = 210$ °K	
$\Theta_2 = 210$ °K, $\Theta_3 = 306$ °K	
$\gamma_1 = 1.24\times10^{-28}$ sec^2, $\gamma_2 = 5.534\times10^{-28}$ sec^2	
$\gamma_4 = 6.518\times10^{-29}$ sec^2, $\gamma_3 = 2.660\times10^{-28}$ sec^2	
$\Theta/\alpha = 380$ °K, $a = 6.387\times10^{-8}$ cm	
$(\tau_B^{-1})_T = 1.77\times10^6$ sec^{-1}, $(\tau_B^{-1})_L = 2.63\times10^6$ sec^{-1}	
$A = 1.4\times10^{-44}$ sec^3	
$B_{T,\mathrm{I}} = 2.61\times10^{-5}$ K^{-m}	In the second approach, i.e., all heat is carried by the transverse phonon.
$B_{L,\mathrm{II}} = 1.92\times10^{-16}$ sec K^{-m}	
$B_{T,\mathrm{I}} = 3.84\times10^{-5}$ K^{-m}	In the first approach, i.e., at 300 °K each phonon contributes equally.
$B_{L,\mathrm{II}} = 7.0\times10^{-18}$ sec K^{-m}	

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