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A NEW APPROACH TO LATTICE THERMAL CONDUCTIVITY: APPLICATION TO Ge

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The new expression $\tau_{3\mathrm{ph}}^{-1} = g(\omega)\,(B_{\mathrm{N}} + B_{\mathrm{U}}\mathrm{e}^{-\Theta/\alpha T})T^{m}$ is proposed for the three-phonon scattering relaxation rate, considering contributions due to three-phonon normal and umklapp processes, which give a new approach to the lattice thermal conductivity. With use of the above expression, the lattice thermal conductivity of Ge has been calculated in the entire temperature range 2–1000 K: good agreement is found between the experimental and calculated values of the phonon conductivity in the entire temperature range of investigation. Analytical expressions are also obtained to calculate an approximate value of the lattice thermal conductivity. The role of four-phonon processes is also included in the present study.

Phonon-phonon scattering plays a very important role in the calculation of the lattice thermal conductivity of an insulator, but even at present we lack an exact analytical expression for it. On account of the complex structure of the Brillouin Zone and the strong temperature-dependence of the phonon distribution function, the relaxation times have a complicated dependence on the phonon frequency and the temperature. For practical purpose, it is necessary to express the relaxation rate by simple relations. The phonon-phonon scattering processes can be divided into two groups: normal processes (N-processes), in which momentum is conserved, and umklapp processes (U-processes), in which momentum is not conserved. Several workers [1–8] have studied both processes, to calculate the phonon conductivities of different samples [9–13], and have given simple expressions for the three-phonon scattering relaxation rate for both processes. However, the contributions of both N-processes and U-processes have not been considered in the same integral before, due to their complicated roles. Keeping in view all such expressions and considering the contributions of the N and U-processes, we have proposed an expression for the three-phonon scattering relaxation rate $\tau_{3\mathrm{ph}}^{-1} = g(\omega)\ (B_{\mathrm{N}} + B_{\mathrm{U}}e^{-\Theta/\alpha T})T^{m}$, to calculate the phonon conductivity of an insulator (terms are explained below). For the first time, an expression for $\tau_{3\mathrm{ph}}^{-1}$ is proposed to calculate the phonon conductivity, which includes the contributions of the N and U-processes in the same integral and in the entire temperature range, as well as in the entire range of frequency of the Brillouin Zone.

To examine the applicability of the proposed expression, the lattice thermal conductivity of Ge is calculated in the entire temperature range 2–1000 K. The role of four-phonon processes [14–16] is also incorporated in the present calculations. Good agreement is obtained between the calculated and experimental values of the phonon conductivity in the entire temperature range. Analytical expressions are also obtained to calculate an approximate value of the phonon conductivity using the above expression for $\tau_{3\mathrm{ph}}^{-1}$, first in the absence and then in the presence of four-phonon processes.

Three-phonon scattering relaxation rate

The three-phonon scattering processes dominate over other processes at high temperatures. These processes are not negligibly small at low temperatures and they play an important role even in the region of the conductivity maxima. It is difficult to express $\tau_{3\mathrm{ph}}^{-1}$ by a simple relation, due to the complicated structure of the Brillouin Zone and the strong temperature-dependence of the phonon distribution function. Several workers have calculated approximate expressions for $\tau_{3\mathrm{ph}}^{-1}$. These expressions are listed in Table 1. Their results suggest differences for transverse and longitudinal phonons. From Table 1, it is clear that the frequency-

Table 1

Relaxation times. In these expressions B's are constant, L is the Casimir length of the crystal, V is the atomic volume, f_i is the atomic fraction of the i^{th} impurity whose mass is m_i, m is the mass of the host lattice atom, $\Delta m = m - m_i$, v_s is the average phonon velocity, $q_{\max}$ is the Zone boundary of the crystal lattice

Scattering processes		Relaxation rates
Crystal boundary[a] τ_{B}^{-1}		v_s/L
Impurities[b] (mass difference) τ_{pt}^{-1}		$A\,\omega^4,\ A = \dfrac{V}{4\pi v_s^3}\sum_i f_i(\Delta m/m)^2$
Three phonon $\tau_{3\mathrm{ph}}^{-1}$		
Normal process[c] (N-processes) $\tau_{3\mathrm{ph},N}^{-1}$		
Longitudinal	$\tau_{\mathrm{L,N}}^{-1} = B_{\mathrm{L}}\omega^2T^3$	at low temperatures
Transverse	$\tau_{\mathrm{T,N}}^{-1} = B_{\mathrm{T}}\omega T^4$	
Longitudinal	$\tau_{\mathrm{L,N}}^{-1} = B_{\mathrm{L}}'\omega^2T$	at high temperatures
Transverse	$\tau_{\mathrm{T,N}}^{-1} = B_{\mathrm{T}}'\omega T$	
Umklapp processes	(U-processes) $\tau_{3\mathrm{ph,U}}^{-1}$	
Klemens[d]	$\tau_{\mathrm{U}}^{-1} = B_{\mathrm{U}}\omega^2T^3e^{-\Theta/\alpha\mathrm{T}}$	at low temperatures
Klemens[e]	$\tau_{\mathrm{U}}^{-1} = B_{\mathrm{U}}\omega T^3e^{-\Theta/\alpha\mathrm{T}}$	
Holland[f]	$\tau_{\mathrm{U}}^{-1} = B_{\mathrm{TU}}\dfrac{\omega^2}{\sinh(\hbar\omega/k_{\mathrm{B}}T)}$	$\frac{1}{2}q_{\max}$ to $q_{\max}$
	$\tau_{\mathrm{U}}^{-1} = 0$	0 to $q_{\max}$
Callaway[g]	$\tau_{\mathrm{U}}^{-1} = B_{\mathrm{U}}\omega^2T^3$	
Klemens[h]	$\tau_{\mathrm{U}}^{-1} = B_{\mathrm{U}}'\omega^2T$	at high temperatures

[a] Ref. 33
[b] Ref. 4
[c] Ref. 1
[d] Ref. 3
[e] Ref. 2
[f] Ref. 7
[g] Ref. 5
[h] Ref. 2

dependence of τ_{3ph}^{-1} is ω for a transverse phonon, and ω^2 for a longitudinal phonon (an exception is the Callaway expression [5], because he has not considered different modes of phonons; his expression is valid for a longitudinal phonon only). The expression for τ_{3ph}^{-1}-umklapp processes contains an exponential term. It is also found that the Herring [1] relation for τ_{3ph}^{-1} is true at extremum temperatures only.

Several workers [17–23] have incorporated these relaxation rates to calculate the phonon conductivities of insulators, considering that at high temperatures *U*-processes dominate over *N*-processes, and at low temperatures *N*-processes dominate over *U*-processes. Therefore, previous workers have considered only one process, either τ_{3ph}^{-1}-normal or τ_{3ph}^{-1}-umklapp, in one conductivity integral. The combined relaxation rates, τ_c^{-1} used by them are listed in Table 2. It is found that *U*-processes dominate over *N*-processes at high temperatures, while the reverse is true at low temperatures. However, the probability of any one of them does not vanish at any temperature. Keeping in view all such expressions listed in Table 1, we have proposed an expression for τ_{3ph}^{-1} as the combined relaxation rate of *N* and *U*-processes.

Table 2

Combined scattering relaxation rate. In these expressions ω_D is the Debye frequency, Θ is the Debye temperature, ω_1 and ω_2 are transverse phonon frequency at $\frac{1}{2}q_{max}$ and q_{max}, ω_4 and ω_3 are the same for longitudinal phonon and α is a constant

	Combined relaxation rate	Frequency range
Callaway[a]	$\tau_c^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_1 + B_1)\omega^2T^3$	$0 - \omega_D$
Holland[b]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{TN}\omega T^4$	$0 - \omega_1$
	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{TU}\frac{\omega^2}{\sinh(\hbar\omega/k_BT)}$	$\omega_1 - \omega_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{LN}\omega^2T^3$	$0 - \omega_3$
Joshi and Verma[c]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_T\omega T^m$	$0 - \omega_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_L\omega^2T^m$	$0 - \omega_3$
	($m = 1, 2, 3$ and 4 depends on temperature ranges)	
SDV model[d]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{T,I}\omega T^m T^{(T)},I\ e^{-\Theta/\alpha T}$	$0 - \omega_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{L,II}\omega^2T^m L,II^{(T)}e^{-\Theta/\alpha T}$ $+ B_{L,I}\omega^2T^m L,I^{(T)}e^{-\Theta/\alpha T}$	$0 - \omega_3$
Present work	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{TN} + B_{TU}e^{-\Theta/\alpha T})\omega T^m$	$0 - \omega_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{LN} + B_{LU}e^{-\Theta/\alpha T})\omega^2T^m$	$0 - \omega^3$

[a] Ref. 5

[b] Ref. 7

[c] Refs 36 and 40

[d] Refs 31, 37, 38 and 39

The three-phonon normal scattering relaxation rate $\tau_{3ph,N}^{-1}$ is given by

$$\tau_{3ph,N}^{-1} = B_N g(\omega)T^m \tag{1}$$

and for *U*-processes it is given by

$$\tau_{3ph,U}^{-1} = B_U g(\omega)e^{-\Theta/\alpha T}T^m \tag{2}$$

where B_N and B_U are the scattering strengths of three-phonon N and U-processes, respectively, Θ is the Debye temperature, α is a constant depending on the crystal structure, $g(\omega)$ is the frequency-dependence of $\tau_{3\mathrm{ph}}^{-1}$ its value being ω and ω^2 for transverse and longitudinal phonons, respectively, and m is the temperature exponent. We have considered the same value of m for both processes, due to the fact that Guthrie's [8] calculations show equal temperature exponents for N and U-processes. The value of m can be calculated with the help of Guthrie's expression as

$$m = 2x_{\max}\,(e^{x}\max - 1)^{-1} + x_{\max} \tag{3}$$

where $x_{\max} = (\hbar\,\omega_{\max,\mathrm{T,L}})/(k_{\mathrm{B}}T)$, k_{B} is the Boltzmann constant, $\hbar$ is the Planck constant divided by 2 π, and T and L as suffixes refer to transverse and longitudinal phonons, respectively. Therefore, the combined relaxation rate for τ_{3ph}^{-1} can be given as

$$\tau_{3\mathrm{ph}}^{-1} = \tau_{3\mathrm{ph,N}}^{-1} + \tau_{3\mathrm{ph,U}}^{-1} = (B_{\mathrm{N}} + B_{\mathrm{U}}e^{-\Theta/\alpha\mathrm{T}})g(\omega)T^{m} \tag{4}$$

Thus, the combined relaxation rate τ_{c}^{-1} for transverse phonons can be given as

$$\tau_{c}^{-1} = \tau_{\mathrm{b}}^{-1} + \tau_{\mathrm{pt}}^{-1} + (B_{\mathrm{TN}} + B_{\mathrm{TU}}e^{-\Theta/\alpha T})T^{m}\omega \tag{5}$$

Table 3

Combined relaxation rate used in the present calculations

Expression	Temperature range
Combined relaxation rate for transverse phonon $\tau_{\mathrm{c,T}}^{-1}$	
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{TN}} + B_{\mathrm{TU}}\,e^{-\Theta/\alpha\mathrm{T}})\omega T^4 + B_{\mathrm{HT}}\omega^2T^2$	$T < 22$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{TN1}} + B_{\mathrm{TU1}}e^{-\Theta/\alpha\mathrm{T}})\omega T^3 + B_{\mathrm{HT}}\omega^2T^2$	$22 < T < 28$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{TN2}} + B_{\mathrm{TU2}}e^{-\Theta/\alpha\mathrm{T}})\omega T^2 + B_{\mathrm{HT}}\omega^2T^2$	$28 < T < 42$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{TN3}} + B_{\mathrm{TU3}}e^{-\Theta/\alpha\mathrm{T}})\omega T + B_{\mathrm{HT}}\omega^2T^2$	$T > 42$
Combined relaxation rate for longitudinal phonon $\tau_{\mathrm{c,L}}^{-1}$	
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{LN}} + B_{\mathrm{LU}}\,e^{-\Theta/\alpha\mathrm{T}})\omega^2T^4 + B_{\mathrm{HL}}\omega^2T^2$	$T < 65$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{LN1}} + B_{\mathrm{LU1}}e^{-\Theta/\alpha\mathrm{T}})\omega^2T^3\omega + B_{\mathrm{HL}}\omega^2T^2$	$65 < T < 83$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{LN2}} + B_{\mathrm{LU2}}e^{-\Theta/\alpha\mathrm{T}})\omega^2T^2 + B_{\mathrm{HL}}\omega^2T^2$	$83 < T < 124$
$\tau_{\mathrm{B}}^{-1} + A\omega^4 + (B_{\mathrm{LN3}} + B_{\mathrm{LU3}}e^{-\Theta/\alpha\mathrm{T}})\omega^2T + B_{\mathrm{HL}}\omega^2T^2$	$T > 124$
The three phonon scattering strengths are related as[a]	
$B_{\mathrm{TX1}} = 22\,B_{\mathrm{TX}}$, $B_{\mathrm{LX1}} = 65\,B_{\mathrm{LX}}$ $B_{\mathrm{TX2}} = 28\,B_{\mathrm{TX1}}$, $B_{\mathrm{LX2}} = 83\,B_{\mathrm{LX1}}$ $B_{\mathrm{TX3}} = 42\,B_{\mathrm{TX2}}$, $B_{\mathrm{LX3}} = 124\,B_{\mathrm{LX2}}$ X stands for N and U	

[a] Ref. 36

and for longitudinal phonons as

$$\tau_C^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{LN} + B_{LU}e^{-\Theta/\alpha T})T^m\omega^2 \tag{6}$$

where τ_B^{-1} and τ_{pt}^{-1} are the boundary and defect scattering relaxation rates, respectively, and expressions are given in Table 1. In writing the above equations, it is considered that other scatterers of phonons are absent. The values of m are 1, 2, 3 and 4, corresponding to different temperature ranges, as stated in Table 3.

The importance of four-phonon processes is studied by Pomeranchuk [14–16], and it is found that at high temperature these play an important role [19, 24] in the calculation of the phonon conductivity. The scattering relaxation rate for them is given by

$$\tau_{4ph}^{-1} = B_H\omega^2T^2 \tag{7}$$

where B_H is the four-phonon scattering strength. Therefore, the combined scattering relaxation rate used in the present study is given by

$$\tau_C^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + \tau_{3ph}^{-1} + \tau_{4ph}^{-1} \tag{8}$$

The complete expression for the combined scattering relaxation rate is stated in Table 3.

Phonon conductivity integral

Considering the role of N-processes, Callaway [5] expressed the lattice thermal conductivity as the sum of two terms. One term (which we will refer to as the first term) consists of a single integral containing the combined scattering relaxation rate, while the second term (usually referred to as a correction term due to the N-processes) has a much more complicated form. However, Callaway and others [25, 26] have shown that the contribution of the correction term towards the total phonon conductivity is usually very small (exceptions are solid He [6] and LiF [27]) compared to the first term at low temperatures. It is also found that the contribution of the correction term is very small at high temperatures [28] even in the frame of the generalized Callaway integral [29, 30]. Therefore, we can neglect the contribution of the correction term in the entire temperature range. Considering the spherical symmetry of the Brillouin Zone (i.e. out of three polarization branches, one is longitudinal and two are transverse) and the fact that each phonon contributes separately towards the total phonon conductivity, the contribution of each branch can be expressed as

$$K_i = (1/6\pi^2) \int \tau_c v_{gi}^2(\hbar^2\omega^2/k_BT^2)(e^{\hbar\omega/k_bT} - 1)^{-2}e^{(n\omega/k_BT)}q^2\,dq \tag{9}$$

where the integral is performed over the first Brillouin Zone. v_{gi} is the group velocity corresponding to the polarization branch under study. q is the phonon wave vector corresponding to the phonon frequency ω. Callaway has taken $q =$

$= \omega/v$ to express the above integral as a phonon frequency term, which is a crude approximation.

Following Verma et al. [24, 31] we have taken the better dispersion relation $q = (1 + r\omega^2)(\omega/v)$ to express q in terms of ω in Eq. (9), where r is a constant calculated with the help of the dispersion curve. It is also found that the velocity of phonons in the entire Brillouin Zone does not remain constant. Therefore, we have taken different velocities in the ranges 0 to $q_{\max}/2$, and $q_{\max}/2$ to $q_{\max}$. $q_{\max}$ is the phonon wave vector at the Brillouin Zone. Thus, one can express the phonon conductivity integral as

$$K = K_T + K_L \tag{10}$$

where K_T and K_L are the contributions of transverse and longitudinal phonons, respectively, and are given by

$$\begin{aligned} K_T = {} & (C/v_{T1}) \int_0^{\frac{\theta_1}{T}} \tau_{c,T} x^4 e^x (e^x - 1)^{-2} (1 + R_1 x^2 T^2)^2 (1 + 3\, R_1 x^2 T^2)^{-1} \mathrm{d}x + \\ & + (C/v_{T2}) \int_{\frac{\theta_1}{T}}^{\frac{\theta_2}{T}} \tau_{c,T} x^4 e^x (e^x - 1)^{-2} (1 + R_2 x^2 T^2)^2 (1 + 3\, R_2 x^2 T^2)^{-1} \mathrm{d}x \end{aligned} \tag{11}$$

$$\begin{aligned} K_L = {} & (C/2v_{L1}) \int_0^{\frac{\theta_4}{T}} \tau_{c,L} x^4 e^x (e^x - 1)^{-2} (1 + R_4 x^2 T^2)^2 (1 + 3\, R_4 x^2 T^2)^{-1} \mathrm{d}x + \\ & + (C/2v_{L2}) \int_{\frac{\theta_4}{T}}^{\frac{\theta_3}{T}} \tau_{c,L} x^4 e^x (e^x - 1)^{-2} (1 + \mathrm{R}_3 x^2 T^2)^2 (1 + 3\, R_3 x^2 T^2)^{-1} \mathrm{d}x \end{aligned} \tag{12}$$

where $C = (K_B/3\pi^2)(K_B T/\hbar)^3$, $R_i = r_i(K_B/\hbar)^2$, $i = 1, 2, 3$ and 4, $\tau_{c,j} = (\tau_{c,j}^{-1})^{-1}$, $j = T$ and L. v_{T1} and v_{T2} are the transverse phonon velocities in the range $0 - 1/2q_{\max}$ and $1/2q_{\max} - q_{\max}$, respectively, v_{L1} and v_{L2} are the same for longitudinal phonons, $\theta_i = (\hbar\omega_i/k_B)$, $i = 1, 2, 3$ and 4, ω_1 and ω_4 are the frequencies of transverse and longitudinal phonons, respectively, at $1/2q_{\max}$ and ω_2 and ω_3 are the same at $q_{\max}$, $\tau_{c,T}^{-1}$ and $\tau_{c,L}^{-1}$ are the combined relaxation rates for transverse and longitudinal phonons, as given in Table 3. The above conductivity integral is evaluated to find out analytical expressions in the high temperature approximations.

At high temperatures, θ_2/T and θ_3/T are very small quantities. Therefore, $x \ll 1$ and $x^2 e^x (e^x - 1)^{-2}$ reduces to unity, and the conductivity integral can be evaluated in the above approximations. The obtained expressions are as follows:

A) If $\tau_{3ph}^{-1} > \tau_{4ph}^{-1} > \tau_{pt}^{-1}$

$$\begin{aligned}K_{\mathrm{T}} = {} & (C/v_{\mathrm{T1}}{}^{\beta}_{\mathrm{T}})[X^2 - (R_1/4)X^4 - (5\,R_1^2/6)X^6 - (D/{}^{\beta}_{\mathrm{T}})\{X^5 - (R_1/7)X^7 - (5\,\mathrm{R}_1^2/9)X^9\} \\ & - ({}^{\beta}_{\mathrm{H}}/{}^{\beta}_{\mathrm{T}})\{X^3 - (R_1/5)X^5 - (5\,R_1^2/7)X^7\}] + (C/3\,R_2 v_{\mathrm{T2}}{}^{\beta}_{\mathrm{T}})[1/3\ln(\theta_2/\theta_1) + (5\,R_2/6)Y^2 + \\ & + (R_2^2/4)Y^4 + (R_2/6)Y^{-2} - (D/{}^{\beta}_{\mathrm{T}})\{(1/9)Y^3 + (R_2/3)Y^5 + (R_2/7)Y^7 - (R_2/3)Y\} - \\ & - ({}^{\beta}_{\mathrm{H}}/{}^{\beta}_{\mathrm{T}})\{Y/3 + (5\,R_2/9)Y^3_{+} + (5\,R_2/5)Y^5 + (R_2/3)Y^{-1}\}]\end{aligned} \tag{13}$$

$$\begin{aligned}K_{\mathrm{L}} = {} & (C/2\,v_{\mathrm{L1}}{}^{\beta}_{\mathrm{L}})[X_1 - (D/3\,{}^{\beta}_{\mathrm{L}})X_1^3 - ({}^{\beta}_{\mathrm{H}}/{}^{\beta}_{\mathrm{L}})X_1] + (C/2\,v_{\mathrm{L2}}{}^{\beta}_{\mathrm{L}})[Y_1 - (R_3/3)Y_1^3 - R_3^2Y_1^5 - \\ & - (D/{}^{\beta}_{\mathrm{L}})\{Y_1^3/3 - (R_3/5)Y_1^5 - (5\,R_3^2/7)Y_1^7\} - ({}^{\beta}_{\mathrm{H}}/{}^{\beta}_{\mathrm{L}})\{Y_1 - (R_3/3)Y_1^3 - R_3^2Y_1^5\}]\end{aligned} \tag{14}$$

(B) If $\tau_{4\mathrm{ph}}^{-1} > T_{3\mathrm{ph}}^{-1} > \tau_{\mathrm{pt}}^{-1}$, the above expression reduces to

$$\begin{aligned}K_{\mathrm{T}} = {} & (C/v_{\mathrm{T1}})(1/{}^{\beta}_{\mathrm{H}})[X - (R_1/3)X^3 - R_1^2X^5 - (D/{}^{\beta}_{\mathrm{H}}\{X^3/3 - (R_1/5)X^5 - \\ & - (5R_1^2/7)X^7\} - ({}^{\beta}_{\mathrm{T}}/{}^{\beta}_{\mathrm{H}})\{\ln X - (R_1/2)X^2 - (5\,R_1^2/4)X^4\}] + (C/3\,R_2 v_{\mathrm{T2}}{}^{\beta}_{\mathrm{H}})[(5\,R_2/3)Y + \\ & + (R_2^2/3)Y^3 - (R_2/9)Y^{-3} - Y^{-1}/3 - (D/{}^{\beta}_{\mathrm{H}})\{Y/3 + (5\,R_2/9)Y^3 + (R_2^2/5)Y^5 + \\ & + (R_2/3)Y^{-1}\} - ({}^{\beta}_{\mathrm{T}}/{}^{\beta}_{\mathrm{H}})\{(5\,R_2/3)\ln(\theta_2/\theta_1) + (R_2^2/2)Y^2 + (R_2/12)Y^{-4} - Y^{-2}/6\}]\end{aligned} \tag{15}$$

$$\begin{aligned}K_{\mathrm{L}} = {} & (C/2v_{\mathrm{L1}}{}^{\beta}_{\mathrm{H}})[X_1(1 - {}^{\beta}_{\mathrm{L}}/{}^{\beta}_{\mathrm{H}}) - (D/3\,{}^{\beta}_{\mathrm{H}})X_1^3] + (C/2\,v_{\mathrm{L2}}{}^{\beta}_{\mathrm{H}})[\{Y_1 - (R_3/3)Y_1^3 - \\ & - (R_3^2/5)Y_1^5\}\{(1 - {}^{\beta}_{\mathrm{L}}/{}^{\beta}_{\mathrm{H}})\} - (D/{}^{\beta}_{\mathrm{H}})\{Y_1^3/3 - (R_3/5)Y_1^5 - (5\,R_3^2/7)Y_1^7\}]\end{aligned} \tag{16}$$

where

$$X = \theta_1/T, \quad X_1 = \theta_4/T, \quad Y^n = \{(\theta_2/T)^n - (\theta_1/T)^n\}, \quad Y_1^n = \{(\theta_3/T)^n - (\theta_4/T)'\}$$

$$D = A(k_{\mathrm{B}}T/\hbar)^4, \quad {}^{\beta}_{\mathrm{H}} = B_{\mathrm{H}}(k_{\mathrm{B}}T^2/\hbar)^2$$

$${}^{\beta}_{\mathrm{T}} = (B_{\mathrm{TN}} + B_{\mathrm{TU}}e^{-\Theta/\alpha \mathrm{T}})(k_{\mathrm{B}}/\hbar)T^{m+1}, \quad {}^{\beta}_{\mathrm{L}} = (B_{\mathrm{LN}} + B_{\mathrm{LU}}e^{-\theta/\alpha T})(k_{\mathrm{B}}/\hbar)^2T^{m+2}$$

where n is any integer and $m = 1, 2, 3$ and 4.

Application to Ge

To see the result of the above-proposed expression for $\tau_{3\mathrm{ph}}^{-1}$, the entire calculation is made for Ge. The constant regarding the dispersion curve is calculated with the help of the experimental curve [32]. As far as boundary and point defect scattering strengths are concerned, they do not need any adjustment for Ge. The value of Casimir's length [33] of the crystal L and the point defect scattering strength A are taken from the previous report of Holland [7]. The temperature exponent m is calculated with the help of Eq. (3), separately for both modes, whereas Tiwari and Agrawal [37] have taken equal values of m for both modes. As far as the three-phonon scattering strength is concerned, this involves complications due to the fact that we have included both N and U-processes. Previous workers have considered only one type of process in one integral. As we know, at low temperatures $\tau_{3\mathrm{ph,N}}^{-1}$ dominates over $\tau_{3\mathrm{ph,U}}^{-1}$. Keeping in view the above ideas, B_{TN} and B_{LN} are adjusted at 20 K (a temperature near the conductivity maxima), ignoring the contribution of three-phonon U-processes. The three-phonon U-processes dominate over N-processes at high temperature. Therefore, one can assume that the probabilities of N and U-processes are equal at room temperature. Thus, B_{TU} and B_{LU} are calculated at 300 K with the help of the relation $\tau_{3\mathrm{ph,U}}^{-1} = \tau_{3\mathrm{ph,N}}^{-1}$. Considering the role of U-processes also, B_{TN} and B_{LN} are finally adjusted at 20 K. We can say that in the present calculation too we have only one adjusting parameter for τ_{3ph}^{-1} for both modes. Knowing the value of the three-phonon scattering strength, the phonon conductivity of Ge is calculated: some discrepancies are

Table 4

The constant and parameters used in the analysis of phonon conductivity of Ge in the temperature range 2—1000 K

$(v_{T1})_{0<\omega<\omega_1} = 3.55\times 10^5$ cm/sec
$(v_{L1})_{0<\omega<\omega_4} = 4.92\times 10^5$ cm/sec
$r_1 = 2.95\times 10^{-27}$ sec^2
$r_4 = 0$
$\theta_1 = 90$ K
$\theta_4 = 208$ K
$\theta = 376$ K
$\alpha = 2.0$
$\tau_B^{-1} = 1.96\times 10^6$ sec^{-1}
$A = 2.40\times 10^{-44}$ sec^3
$B_{TN} = 1.12\times 10^{-11}$ deg^{-4}
$B_{TU} = 2.10\times 10^{-11}$ deg^{-4}
$B_{LN} = 2.50\times 10^{-24}$ sec deg^{-4}
$B_{LU} = 4.69\times 10^{-24}$ sec deg^{-4}
$B_{HT} = 6.88\times 10^{-24}$ sec deg^{-2}
$B_{HL} = 6.88\times 10^{-24}$ sec deg^{-2}

$(v_{T2})_{\omega_1<\omega<\omega_2} = 1.30\times 10^5$ cm/sec
$(v_{L2})_{\omega_4<\omega<\omega_3} = 2.46\times 10^5$ cm/sec
$r_2 = 8.28\times 10^{-27}$ sec^2
$r_3 = 1.13\times 10^{-27}$ sec^2
$\theta_2 = 108$ K
$\theta_3 = 319$ K

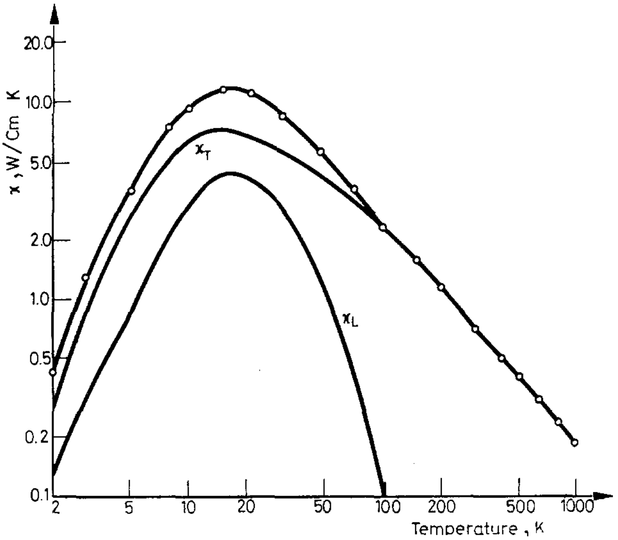

Fig. 1. Phonon conductivity of Ge in the temperature range 2—1000 K. K_T is the transverse phonon contribution and K_L the longitudinal phonon contribution. Solid line indicates calculated values. Circles are experimental points

found at temperatures beyond 500 K. These discrepancies are balanced by including the role of four-phonon processes and the four-phonon scattering strength is adjusted at 500 K. The analytical expressions reported in the present paper are very useful to calculate an approximate value of the scattering strength, as well as the phonon conductivity.

The constants and parameters used in the present analysis are listed in Table 4. Using these constants and parameters, the phonon conductivity of Ge is calculated in the entire temperature range 2 – 1000 K by calculating the separate contributions of the transverse and longitudinal phonons, and is shown in Fig. 1. The percentage contributions of the three-phonon N and U scattering relaxation rates towards the three-phonon scattering relaxation rate are also calculated, to study the dominating nature of one over the other in the temperature range 10 – 1000 K, and they are shown in Fig. 2.

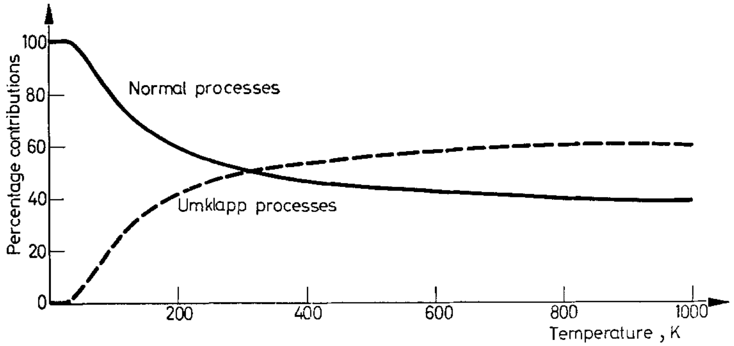

Fig. 2. Percentage contributions of three-phonon N and U-processes towards three-phonon scattering relaxation rate. Solid line is percentage of $\tau_{3ph,N}^{-1}$ and dotted line is percentage of $\tau_{3ph,U}^{-1}$

From Fig. 2, it can be concluded that at low temperatures the contribution of U-processes is negligibly small compared to that of N-processes, whereas the U dominate over the N-processes at high temperatures. However, the contribution of N-processes is not very small at high temperature. It is nearly 35% at 1000 K. From Fig. 1, one can see that the agreement between the experimental and calculated values of the phonon conductivity is excellent in the entire temperature range 2 – 1000 K, whereas Verma et al. [30] could not get good agreement in the vicinity of the conductivity maxima.

Discussion

The expression proposed for $\tau_{3ph}^{-1} = (B_L + B_U e^{-\theta/\alpha T}) \cdot g(\omega)T^m$ shows very good results at high as well as at low temperatures. At low temperatures $\theta/\alpha T \gg 1$, which results in $\tau_{3ph}^{-1} = \tau_{3ph,N}^{-1} = B_N g(\omega)T^m$ due to very small value of $e^{-\theta/\alpha T}$.

This shows that our proposed expression for $\tau_{3\text{ph}}^{-1}$ reduces at low temperature to an expression for the three-phonon *N*-processes scattering relaxation rate $\tau_{3\text{ph,N}}^{-1}$. Thus, the proposed new expression indicates the dominating nature of *N*-processes at low temperatures. At high temperatures, the percentage contribution of $\tau_{3\text{ph,U}}^{-1}$ is as high as 65% (at 1000 K), which reveals the dominating nature of *U*-processes over *N*-processes. As far as intermediate temperatures are concerned, these can be studied with the help of Fig. 2. Thus, for the first time, we have incorporated the contribution of both *N* and *U*-processes in the entire temperature range, as well as in the entire frequency range of the Brillouin Zone (0 to $\omega_{\max}$), to calculate the phenon conductivity of an insulator. The value of the temperature exponent *m* is calculated for both modes, whereas Tiwari and Agrawal used equal values of *m* for both polarization branches. From Fig. 1, it can be seen that at high temperatures the entire heat is transported by transverse phonons alone, which is similar to the previous findings of other workers. At the same time, it can also be seen that the agreement between calculated and experimental values is excellent at high as well as at low temperatures. The temperature exponent used in the proposed expression also follows Guthrie's limit [8, 35] for three-phonon scattering relaxation rates.

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Résumé — On a proposé la nouvelle expression $\tau_{3\mathrm{ph}}^{-1} = g(\omega)(B_{\mathrm{N}} + B_{\mathrm{U}}e^{-\theta/\alpha T})T^m$ qui constitue une approximation nouvelle à la conductivité thermique des réseaux. On a calculé ainsi, en incorporant cette expression, la conductivité thermique du réseau de germanium dans tout l'intervalle de températures compris entre 2 et 1000 K. Un bon accord a été observé entre les valeurs calculées et expérimentales pour la conductivité des phonons dans tout l'intervalle de températures étudié. On a également obtenu des expressions analytiques pour le calcul de la valeur approchée de la conductivité thermique du réseau. Le rôle des processus à quatre phonons est également inclus dans la présente étude.

Zusammenfassung — Der neue Ausdruck $\tau_{3\mathrm{ph}}^{-1} = g(\omega)(B_{\mathrm{N}} + B_{\mathrm{U}}e^{-\theta/\alpha T})T^m$ wird für die Drei-Phonon-Streuungs-Relaxationsgeschwindigkeit vorgeschlagen, welche Drei-Phonon-Normal- und Umklapp-Prozesse berücksichtigt. Daraus ergibt sich eine neue Annäherung zur Wärmeleitfähigkeit von Gittern. Durch obigen Ausdruck wurde die Wärmeleitfähigkeit von Ge im Temperaturbereich von 2 bis 1000 K berechnet und eine gute Übereinstimmung der berechneten und Versuchswerte der Phononleitfähigkeit im untersuchten Temperaturbereich gefunden. Analytische Ausdrücke können auch für die Berechnung eines Näherungswertes der Wärmeleitfähigkeit des Gitters erhalten werden. Die Rolle des Vier-Phonon-Vorganges wird ebenfalls mit behandelt.

Резюме — Предложено новое выражение $\tau_{3\mathrm{ph}}^{-1} = g(\omega)\ (B_{\mathrm{N}} + B_{\mathrm{U}}e^{-\theta/\alpha T})T^m$ для скорости релаксации трехфононного рассеяния, учитывая вклады, обусловленные тремя нормальными фононными процессами и процессами переброса, что дает новое приближение для решеточной термической проводимости. С помощью этого выражения была вычислена решеточная термическая проводимость германия во всей температурной области 2—1000К и найдено хорошее совпадение между вычисленными и экспериментальными значениями термической проводимости во всей исследованной температурной области. Получены также аналитические выражения для вычисления приближенного значения решеточной термической проводимости. В настоящее изучение включена также роль четырехфононных процессов.