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ANALYSIS OF LATTICE THERMAL CONDUCTIVITY AND PHONON-PHONON SCATTERING RELAXATION RATE: APPLICATION TO Ge

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The three-phonon scattering relaxation rates and their temperature exponents have been analysed in the frame of Guthrie's classification of the phonon–phonon scattering events as class I and class II events and as a result of this, a new expression $\tau_{3ph}^{-1} = (B_{N,I} + B_{U,I}\, e^{-\theta/\alpha T})\, g(w) T^m I^{(T)} + (B_{N,II} + B_{U,II}\, e^{-\theta/\alpha T})\, g(w) T^m II^{(T)}$ for the three phonon scattering relaxation rates has been proposed for the first time to calculate the lattice thermal conductivity of a sample. Using the expression proposed above, the lattice thermal conductivity of Ge has been analysed in the temperature range 2 – 1000K and result obtained shows a very good agreement with the experimental data. The percentage contributions due to three-phonon normal and umklapp processes are also reported. The role of four phonon processes is also included at high temperatures. To estimate an approximate value of the scattering strength and the phonon conductivity, the analytical expression is also obtained in the frame of the expression proposed above for τ_{3ph}^{-1}.

The phonon–phonon scattering relaxation rate has been studied by a number of workers [1 – 13] due to its very important role in the lattice thermal conductivity, and it has been found that the three-phonon scattering relaxation rate involves a complicated dependence on the phonon frequency and temperature due to the complicated structure of the Brillouin Zone and the strong temperature dependence of the distribution function. As a result of this, even at present we lack an exact analytical expression for it. However, for practical purposes, it has been expressed by simple relations [1 – 13] as a function of the phonon frequency and temperature. It is also found that 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. The roles of N and U processes have been studied by a number of worker [14 – 22] by calculating the phonon conductivities of different samples.

Recently, Guthrie [7, 8] studied the three-phonon scattering relaxation rate by dividing phonon–phonon scattering events into two classes: class I events, in which the carrier phonon is annihilated by combination and class II events, in which the carrier phonon is annihilated by splitting. Following Guthrie, the three-phonon scattering relaxation rate τ_{3ph}^{-1} can be expressed as

$$\tau_{3ph}^{-1} = \tau_{3ph}^{-1}\,(\text{class I}) + \tau_{3ph}^{-1}\,(\text{class II}) \tag{1}$$

He also pointed out that the three-phonon scattering relaxation rate due to each class of events could be expressed as

$$\tau_{3ph}^{-1} \propto g(w) f(T) \tag{2}$$

where $f(T) = T^{m(T)}$ and $m(T)$ is a continuous function of temperature T. Verma *et al.* [9, 10, 23–25] were the first who attempted to incorporate the Guthrie expression to calculate the phonon conductivity of some insulators, by expressing the three-phonon scattering relaxation rate as

$$\tau_{3ph}^{-1} = B\, g(w)\, T^{m(T)}\, e^{-\theta/\alpha T} \tag{3}$$

Terms are explained in the following section. However they could not include the contribution due to the three-phonon N processes in the calculation of the lattice thermal conductivity. At the same time, the expression used for $m(T)$ includes an empirical factor $(1 + \theta/\alpha T)$.

Following Verma *et al.* considering the Guthrie [7] classification of phonon–phonon scattering events and including the contributions due to three-phonon N and U processes, a new expression for τ_{3ph}^{-1} is proposed as

$$\tau_{3ph}^{-1} = (B_{N,I} + B_{U,I}\, e^{-\theta/\alpha T})\, g(w)\, T^{m\,I(T)} + (B_{N,II} + B_{U,II}\, e^{-\theta/\alpha T})\, g(w)\, T^{m\,II(T)} \tag{4}$$

Terms are explained in the following section. No distinction is made in the value of $m(T)$ for the three-phonon N and U processes, due to the fact that Guthrie obtained the same value of the temperature exponent $m(T)$ for both processes. The value of the temperature exponent $m(T)$ used in the present work differs from that used in the work of Verma *et al.* in that in the present analysis it does not include any empirical relation.

To see the applicability of the expression proposed, the phonon conductivity of Ge has been calculated in the entire temperature range 2–1000 K as an example within the framework of τ_{3ph}^{-1} given in Eq. (4). The values of $m_I(T)$ and $m_{II}(T)$ are also calculated, to permit a comparative study with the temperature exponent $m(T)$ used in the Sharma–Dubey–Verma (SDV) model [9, 10], as well as with the upper limit of $m(T)$ obtained by Guthrie. The percentage contributions of the three-phonon N and U processes towards the three-phonon scattering relaxation rate have been studied for both transverse and longitudinal phonons, and also for class I and for class II events. The percentage contribution of τ_{3ph}^{-1} towards the combined scattering relaxation rate has also been studied for both transverse and longitudinal phonons. To examine the relative contributions of transverse and longitudinal phonons towards the total phonon conductivity, the percentage contributions due to transverse and longitudinal phonons have also been studied in the entire temperature range 2–1000 K.

Analytical expressions have been obtained to estimate approximate values of the scattering strength and the lattice thermal conductivity. A comparative study is made between the results of the present analysis and the earlier reports of other workers.

Three-phonon scattering relaxation rate and its temperature exponents

It is well established that the three-phonon scattering processes dominate over other scattering processes at high temperatures. At the same time, they are not negligibly small at low temperatures. They play a very important role even in the vicinity of conductivity maxima. However, due to the complicated structure of the Brillouin Zone and the strong temperature dependence of the distribution function, even at present we lack an exact expression for it. The phonon–phonon scattering procecces, N and U processes, have been studied widely, their scattering relaxation rates being expressed by simple relations as functions of the phonon frequency and temperature (see Table 1). The lattice thermal conductivities of several samples have been calculated [26–35] in the framework of the combined scattering relaxation rates (see Table 2), at both low and high temperatures, using the expressions reported in Table 1.

Table 1

The scattering relaxation rates. In these expressions, B's are constants and are known as the scattering strengths of the respective processes, L is the Casimir length of the crystal, A is the point-defect scattering strength, v is the average phonon velocity, $q_{\max}$ is the Zone boundary of the first Brillouin Zone, α is a constant and θ is the Debye temperature

Scattering processes	Relaxation rates	
Crystal boundary [39]	$\tau_B^{-1} = v/L$	
Impurities [4]	$\tau_{pt}^{-1} = Aw^4$	
Three-phonon processes τ_{3ph}^{-1}		
Normal processes (N processes) $\tau_{3ph,N}^{-1}$ [1]		
Transverse	$\tau_{TN}^{-1} = B_T w T^4$	at low temperatures
Longitudinal	$\tau_{LN}^{-1} = B_L w^2 T^3$	
Transverse	$\tau_{TN}^{-1} = B'_T w T$	at high temperatures
Longitudinal	$\tau_{LN}^{-1} = B'_L w^2 T$	
Umklapp processes (U processes) $\tau_{3ph,U}^{-1}$		
Klemens [3]	$\tau_U^{-1} = B_U w^2 T^3 e^{-\theta/\alpha T}$	at low temperatures
Klemens [2]	$\tau_U^{-1} = B'_U w T^3 e^{-\theta/\alpha T}$	
Holland [6] (for transverse)	$\tau_U^{-1} = B_{TU} w^2/\sin h(\hbar w/k_B T)$	$\frac{1}{2}q_{\max}$ — $q_{\max}$
	$\tau_U^{-1} = 0$	0 — $\frac{1}{2}q_{\max}$
Callaway [5]	$\tau_U^{-1} = B_U w^2 T^3$	
Klemens [2]	$\tau_U^{-1} = B'_U w^2 T$ at high temperatures	
Four-phonon processes[i]	$\tau_{4ph}^{-1} = B w^2 T^2$	

The combined scattering relaxation rate due to the three-phonon scattering processes can be expressed by Eq. (1). According to Guthrie, the phonon–phonon scattering relaxation rate has the form of

$$\tau_{3ph}^{-1} \alpha\, g(w)\, T^{m(T)}$$

Table 2

The combined scattering relaxation rates. In these expressions, w_D is the Debye frequency, θ is the Debye temperature, w_1 and w_2 are the transverse phonon frequencies at $^1/_2\, q_{\max}$ and $q_{\max}$, respectively, w_3 and w_4 are the same for longitudinal phonons, $q_{\max}$ is the Zone boundary of the first Brillouin Zone, α is a constant depending on the dispersion curve and m is the temperature exponent

Author and reference	Combined scattering relaxation rates	Frequency range
Callaway [5]	$\tau_c^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_1 + B_2)w^2T^3$	$0 - w_D$
Holland [6]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{TN}wT^4$	$0 - w_1$
	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{TU}w^2/\sin h(\hbar w/k_BT)$	$w_1 - w_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_Lw^2T^3$	$0 - w_4$
Joshi and Verma [11]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_TwT^m$	$0 - w_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_Lw^2T^m$	$0 - w_4$
	(m = 1, 2, 3 or 4 depending on temperature range)	
SDV model [9, 10, 23]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{T,1}w\ T^mT,\ \mathrm{I}^{(T)}\, e^{-\theta/\alpha T}$	$0 - w_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + B_{L,1}w^2T^mT,\ \mathrm{I}^{(T)}\, e^{-\theta/\alpha T}$	
	$+ B_{L,II}w^2T^mT,\ \mathrm{II}^{(T)}e^{-\theta/\alpha T}$	$0 - w_4$
Dubey and Misho [12]	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{TN} + B_{TU}\, e^{-\theta/\alpha T})wT^m$	$0 - w_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{LN} + B_{LU}\, e^{-\theta/\alpha T})w^2T^m$	$0 - w_4$
	(m = 1, 2, 3 or 4 for transverse phonons depending on temperature range, and m = 1, 2 or 3 for longitudinal phonons depending on temperature range. $B_{XN} = B_{XU}e^{-\theta/\alpha T}$ at room temperature, i.e. at $T = 300$ K, where $X = T$ or L)	
Present work	$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{TN,I} + B_{TU,I}\, e^{-\theta/\alpha T})wT^mT,\ \mathrm{I}^{(T)}$	$0 - w_2$
	$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + (B_{LN,I} + B_{LU,I}\, e^{-\theta/\alpha T})w^2T^mL,\ \mathrm{I}^{(T)}$	
	$+ (B_{LN,II} + B_{LU,II}\, e^{-\theta/\alpha T})w^2T^mL,\ \mathrm{II}^{(T)}$	

where $g(w)$ is the frequency dependence of the three-phonon scattering relaxation rate; $g(w) = w$ for transverse phonons, and $g(w) = w^2$ for longitudinal phonons, which are the same as obtained by Herring [1]. Guthrie suggested that the temperature exponent $m(T)$ is a continuous function of temperature T. He also pointed out that the temperature dependence of the three-phonon scattering relaxation rates ($\tau_{3ph}^{-1} \propto T^4$ for transverse phonons and $\tau_{3ph}^{-1} \propto T^3$ for longitudinal phonons) obtained by Herring is valid at low temperatures only, and should not be used at high temperatures. At the same time, it is interesting to note that Guthrie could not give any suggestion for the exact value of $m(T)$ which should be used in the calculation of the lattice thermal conductivity, except that he obtained the maximum and minimum values of $m(T)$: For class I events,

$$[m(T)_{\max}] = x_{\max}[2(e^{x_{\max}} - 1)^{-1} + 1.0] - 1.0 \tag{5}$$

$$[m(T)]_{\min} = 1.0 \tag{6}$$

and for class II events, it is given by

$$[m(T)]_{\max} = 1.0 \tag{7}$$

$$[m(T)]_{\min} = x_{\max}(e^{x_{\max}} - 1.0)^{-1}\, e^{0.5\, x_{\max}} \tag{8}$$

where $x_{\max} = \hbar w_{\max, T, L}/k_B T$, $\hbar$ is the Planck constant divided by 2π, k_B is the Boltzmann constant, and $w_{\max}$ is the phonon frequency at the boundary of the first Brillouin Zone. Suffixes T and L represent transverse and longitudinal phonons, respectively. Thus, it is clear that $m(T)$ is different for transverse and longitudinal phonons, due to their frequencies at the Zone boundary. Guthrie also pointed out that the numerical value of $[m(T)]_{\max}$ for class I events should not exceed 4 for transverse phonons and 3 for longitudinal phonons. The maximum value of $m(T)$ at any temperature can be clarified with the help of Table 3, where the Guthrie upper limit of $m(T)$ is reported for Ge, Si, GaAs and InSb.

Thus, there is still large uncertainty in assigning an exact value of $m(T)$. Joshi and Verma [11] used the maximum value of $m(T)$ in the calculation of the lattice thermal conductivity of Si, as reported in Table 3. Thus, they could not use a continuous value of $m(T)$. At the same time, they considered the contribution of the three-phonon N processes only (see Table 1 in ref. [11]). In the lack of an exact value of $m(T)$ and to minimize the uncertainty, it is more realistic to use an average value of its maximum and minimum value in place of its extremum value. Thus, the $m(T)$ used in the present analysis for class I events can be expressed as

$$m_I(T) = x_{\max}(e^{x_{\max}} - 1)^{-1} + 0.5x_{\max} \tag{9}$$

while for class II events it takes the form

$$m_{II}(T) = 0.5x_{\max}(e^{x_{\max}} - 1)\, e^{0.5x_{\max}} + 0.5 \tag{10}$$

Table 3

Guthrie's limits for the temperature exponent m for Ge, Si, GaAs and InSb.[a]

Material	Assumption of $w\,T^4$ relation invalid for $T >$ K	Assumption of $w^2\,T^3$ relation invalid for $T >$ K	$\tau_{3ph}^{-1}\, \alpha T^m$ where $m < 4$ if $T >$ K	$\tau_{3ph}^{-1}\, \alpha T^m$ where $m < 3$ if $T >$ K	$\tau_{3ph}^{-1}\, \alpha T^m$ where $m < 2$ if $T >$ K
Ge	20	26	90	115	167
Si	43	55	149	190	282
GaAs	20	26	85	108	159
InSb	13	16.5	54	69	103

a — see Table 2 of ref. [7]

As stated earlier, the phonon–phonon scattering processes can be grouped into two groups. The N processes are those in which momentum is conserved, and the scattering relaxation rate due to such processes can be expressed as

$$\tau_{3\mathrm{ph,N}}^{-1} = B_{\mathrm{N}}\, g(w)\, T^{\mathrm{m(T)}} \tag{11}$$

where B_{N} is the scattering strength of the three-phonon normal processes, and suffix N is used to represent N processes. The other group of processes are the three-phonon U processes in which momentum is not conserved. According to Klemens [2, 3], the scattering relaxation rate due to these can be expressed as

$$\tau_{3\mathrm{ph,U}}^{-1} = B_{\mathrm{U}}\, g(w)\, T^{\mathrm{m(T)}}\, e^{-\theta/\alpha\mathrm{T}} \tag{12}$$

where B_{U} is the three-phonon U process scattering strength, θ is the Debye temperature of the specimen under study, α is a constant depending on the crystal structure of the sample, and suffix U represents U processes. Thus, the scattering relaxation rates due to class I and class II events can be expressed as

$$\tau_{3\mathrm{ph,I}}^{-1} = (B_{\mathrm{N,I}} + B_{\mathrm{U,I}}\, e^{-\theta/\alpha\mathrm{T}})\, g(w)\, T^{m}\, \mathrm{I}^{(\mathrm{T})} \tag{13}$$

$$\tau_{3\mathrm{ph,II}}^{-1} = (B_{\mathrm{N,II}} + B_{\mathrm{U,II}}\, e^{-\theta/\alpha\mathrm{T}})\, g(w)\, T^{m}\, \mathrm{II}^{(\mathrm{T})} \tag{14}$$

The same frequency dependence is assigned to N and U processes, due to the fact that it depends only on the polarization branches. The same value of $m(T)$ is used for both processes, since Guthrie [7] obtained the same value of $m(T)$ for both N and U processes.

The classification of Guthrie of class I and class II events leads to the participation of transverse phonons alone in class I events, and the participation of longitudinal phonons in both class I and class II events. As a result the three-phonon scattering relaxation rate $\tau_{\mathrm{ph,T}}^{-1}$ due to transverse phonons takes the form

$$\tau_{3\mathrm{ph,T}}^{-1} = (B_{\mathrm{TN,I}} + B_{\mathrm{TU,I}}\, e^{-\theta/\alpha\mathrm{T}})\, w\, T^{m}\, T,\, \mathrm{I}^{(\mathrm{T})} \tag{15}$$

because the contribution due to class II events is not possible for transverse phonons. Similarly, for longitudinal phonons, the three-phonon scattering relaxation rate $\tau_{3\mathrm{ph,L}}^{-1}$ can be expressed as

$$\tau_{3\mathrm{ph,L}}^{-1} = (B_{\mathrm{LN,I}} + B_{\mathrm{LU,I}}\, e^{-\theta/\alpha\mathrm{T}})\, w^{2} T^{\mathrm{m}} L,\, \mathrm{I}^{(\mathrm{T})} + (B_{\mathrm{LN,II}} + B_{\mathrm{LU,II}}\, e^{-\theta/\alpha\mathrm{T}})\, w^{2} T^{\mathrm{m}} L,\, \mathrm{II}^{(\mathrm{T})} \tag{16}$$

Thus, the expression for $\tau_{3\mathrm{ph}}^{-1}$ used in the present analysis is based on the division into class I and class II events, also N and U processes.

Besides the three-phonon scattering processes, four-phonon scattering processes too play an important role in the study of the lattice thermal conductivity at high temperatures. According to Pomeranchuk [36–38], the four-phonon scattering relaxation rate can be expressed as

$$\tau_{4\mathrm{ph}}^{-1} = B_{\mathrm{H}}\, w^{2} T^{2} \tag{17}$$

where B_H is the four-phonon scattering strength. The lattice thermal conductivities of several samples have been calculated [20, 27, 31] by using the above expression for τ_{4ph}^{-1}, and it has been found to give a good response to the phonon conductivity at high temperatures.

It has been shown [39–41] that the lattice thermal conductivity of an insulator at very low temperatures can be explained very well on the basis of boundary scattering alone. According to Casimir [39], the boundary scattering relaxation rate has the form $\tau_B^{-1} = v/L$, where v is the average phonon velocity and L is the characteristic length associated with the crystal under study. Since the boundary scattering is important for low-frequency phonons only, it is immaterial whether v is a phase or a group velocity, because for low-frequency phonons phase and group velocities are almost equal and it is sufficient to use the low-frequency value for v. The characteristic length L is determined by the crystal geometry and is assumed to be the same for all phonons. It is found that the theoretical value of L does not usually agree with the experimentally measured value of the phonon conductivity. It is further found that $L = F$ times the length of the crystal, where F is an adjustable parameter, usually of the order of unity.

The scatterings due to isotopes, point-defects, etc. are most important scattering mechanisms at temperatures near the conductivity maxima. At such temperatures, high-frequency phonons are not excited to a large extent and it is reasonably good to use the Klemens [2] expression for the point-defect scattering relaxation rate τ_{pt}^{-1} as $\tau_{pt}^{-1} = Aw^4$, which has been obtained for low-frequency phonons, were A is the point-defect scattering strength. A careful analysis of the Klemens expression shows that A depends on the polarization branches and it should be different for longitudinal and transverse phonons. Longitudinal phonons contribute only a little [42] towards the total phonon conductivity as compared to transverse phonons. Thus, it is sufficient to use A for transverse phonons, as was done by earlier workers too. The expression for A, as obtained by Klemens, can be expressed as

$$A = (V_0/4\pi v^3)\, \Sigma_i f_i (1 - m_i/\overline{m})^2 \tag{18}$$

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, $\overline{m}$ is the average atomic mass of the host lattice atom, and v is the average phonon velocity.

Thus, the combined scattering relaxation rates for transverse phonons, $\tau_{c,T}^{-1}$ and for longitudinal phonons, $\tau_{c,L}^{-1}$, used in the present analysis are given by

$$\tau_{c,T}^{-1} = \tau_B^{-1} + Aw^4 + (B_{TN,I} + B_{TU,I}\, e^{-\theta/\alpha T}) w T^m T,\, I^{(T)} + B_{HT} w^2 T^2 \tag{19}$$

$$\begin{aligned} \tau_{c,L}^{-1} = \tau_B^{-1} + Aw^4 + (B_{LN,I} + B_{LU,I}\, e^{-\theta/\alpha T}) w^2 T^m L,\, I^{(T)} + \\ + (B_{LN,II} + B_{LU,II}\, e^{-\theta/\alpha T}) w^2 T^m L,\, II^{(T)} + B_{HL} w^2 T^2 . \end{aligned} \tag{20}$$

Phonon conductivity integral and analytical expressions

To have an expression for the lattice thermal conductivity, one needs to solve the Boltzmann equation which can be expressed as [5]

$$\left(\frac{\partial N}{\partial t}\right)_c - c \cdot \nabla T \left(\frac{\mathrm{d}N}{\mathrm{d}T}\right) = 0 \tag{21}$$

where N is the distribution function, c is the group velocity, $\left(\frac{\partial N}{\partial t}\right)_c$ represents the rate of change of the distribution function due to collisions and $\left(\frac{\mathrm{d}N}{\mathrm{d}T}\right)$ is the same due to the temperature gradient ∇T. Considering the special role of three-phonon N processes, Callaway [5] expressed $\frac{\partial N}{\partial t}$ as

$$\left(\frac{\partial N}{\partial t}\right)_c = \frac{N(\lambda) - N}{\tau_N} + \frac{N_0 - N}{\tau_R} \tag{22}$$

where τ_N and τ_R are the scattering relaxation times due to momentum conserving and momentum non-conserving processes, respectively, N_0 is the Planck distribution function and $N(\lambda)$ is the displaced distribution due to three-phonon N processes. If one solves Eq. (21) in terms of Eq. (22) to have an expression for the lattice thermal conductivity K, it is found that K can be expressed as the sum of two components. The first part is due to the combined scattering relaxation rate and consists of a single integral (in the approach of Callaway), whereas the second part is of a complicated form and is known as the correction term [5] (ΔK) due to the three-phonon N processes. In the absence of N processes, it reduces to zero.

However, Callaway [5, 43] and others [44–48] have studied the contribution of the correction term (ΔK) due to the three-phonon N processes, and have found that this is very small as compared to the contribution due to the first part. Solid He [43], LiF [49] and solid HD [50] are exceptions to this. It has further been reported that the contribution of ΔK towards the total phonon conductivity is negligibly small [51–53] in the generalized Callaway integral [54, 55] at low and at high temperatures. Therefore, the contribution due to ΔK has been neglected in the present analysis.

Considering the spherical symmetry of the Brillouin Zone (i.e. of three polarization branches, two are transverse and one is longitudinal) and the fact that each phonon contributes separately towards the total phonon conductivity, the contribution due to each mode of phonons can be expressed as

$$K_i = (1/6\pi^2) \int \tau_{c,i} v_{gi}^2 (\hbar w/k_B T^2) e^{\hbar w/k_B T} (e^{\hbar w/k_B T} - 1)^{-2} q^2 \mathrm{d}q \tag{23}$$

where the integration is performed over the first Brillouin Zone, v_{gi} is the group velocity corresponding to the polarization branches under study, q is the phonon wave vector corresponding to the phonon frequency w, and suffix i is used to distinguish polarization branches.

Callaway and also Holland [6] expressed q in terms of w inside the conductivity integral in the Debye approximation, i.e. $q = w/v$, which is valid for low-frequency phonons only. Following Verma et al. [56], in the present analysis a better relation

$$q = (w/v)\,(1 + rw^2) \tag{24}$$

has been used to change q into w inside the conductivity integral in Eq. [23], where r is a constant and depends on the dispersion curve of the sample under study. The constant r can be calculated with the help of the dispersion curve. It is interesting to state here that the velocity of the phonons is not the same in the entire range of the first Brillouin Zone. It has a larger value for low-frequency phonons as compared to high-frequency phonons. To be more exact, the entire first Brillouin Zone has been divided into two parts: 0 to $1/2q_{\max}$, and $1/2q_{\max}$ to $q_{\max}$, where $q_{\max}$ is the phonon wave vector corresponding to the Zone boundary of the first Brillouin Zone. At the same time, it should also be noted that the dispersion constant r is calculated separately for these two regions. Thus, the total lattice thermal conductivity can be expressed as

$$K = K_{\mathrm{T}} + K_{\mathrm{L}} \tag{25}$$

where K_{T} and K_{L} are the contributions due to transverse and longitudinal phonons, respectively. These are given by

$$\begin{aligned} K_{\mathrm{T}} = {} & (C/v_{\mathrm{T1}}) \int\limits_{0}^{\theta_1/T} \tau_{\mathrm{c,T}} x^4 e^x (e^x - 1)^{-2} (1 + R_1 x^2)^2 (1 + 3R_1 x^2)^{-1}\, \mathrm{d}x \\ & + (C/v_{\mathrm{T2}}) \int\limits_{\theta_1/T}^{\theta_2/T} \tau_{\mathrm{c,T}} x^4 e^x (e^x - 1)^{-2} (1 + R_2 x^2)^2 (1 + 3R_2 x^2)^{-1}\, \mathrm{d}x \end{aligned} \tag{26}$$

$$\begin{aligned} K_{\mathrm{L}} = {} & (C/2v_{\mathrm{L1}}) \int\limits_{0}^{\theta_3/T} \tau_{\mathrm{c,L}} x^4 e^x (e^x - 1)^{-2} (1 + R_3 x^2)^2 (1 + 3R_3 x^2)^{-1}\, \mathrm{d}x \\ & + (C/2v_{\mathrm{L2}}) \int\limits_{\theta_3/T}^{\theta_4/T} \tau_{\mathrm{c,L}} x^4 e^x (e^x - 1)^{-2} (1 + R_4 x^2)^2 (1 + 3R_4 x^2)^{-1}\, \mathrm{d}x \end{aligned} \tag{27}$$

where $C = (k_{\mathrm{B}}/3\pi^2)\,(k_{\mathrm{B}}T/\hbar)^3$, $R_i = r_i(k_{\mathrm{B}}T/\hbar)^2$; $i = 1, 2, 3$ and 4;
$\tau_{\mathrm{c,X}} = (\tau_{\mathrm{c,X}}^{-1})^{-1}$; $X = T$ and L; $\theta_i = (\hbar w_i/k_{\mathrm{B}})$; $i = 1, 2, 3$ and 4;

v_{T1} and v_{T2} are transverse phonon velocities in the range $0 - 1/2\,q_{\max}$ and $1/2\,q_{\max}$, respectively, v_{L1} and v_{L2} are the same for longitudinal phonons, w_1 and w_2 are transverse phonon frequencies corresponding to wave vectors $1/2\,q_{\max}$, $q_{\max}$, respectively, w_3 and w_4 are the same for longitudinal phonons, r_1 and r_2 are the dispersion constants for transverse phonons in the region $0-1/2\ q_{\max}$ and $1/2\ q_{\max}-$ $-\ q_{\max}$, respectively, r_3 and r_4 are the same for longitudinal phonons, and $\tau_{c,T}^{-1}$ and $\tau_{c,L}^{-1}$ are the combined scattering relaxation rates due to transverse and longitudinal phonons, respectively, as given in Eqs (19) and (20). The above conductivity integrals are evaluated to find analytical expressions for the lattice thermal conductivity at high and at low temperatures.

At high temperatures, the upper limit of all the four integrals, θ_i/T (i = 1, 2, 3 and 4), is a small quantity due to the large value of T, which results in $x \ll 1$, and $x^2 e^x (e^x - 1)^{-2}$ reduces to unity. Thus, the conductivity integrals can be evaluated analytically as below:

(A) If $\tau_{3\mathrm{ph}}^{-1} \gg \tau_{4\mathrm{ph}}^{-1} \gg \tau_{\mathrm{pt}}^{-1}$

$$\begin{aligned} K_T = {} & (C/v_{T1} b_T)\,[X^2/2 - (R_1/4)X^4 - (5R_1^2/6)X^6 - (D/b_T)\,\{X^5/5 - (R_1/7)X^7 - \\ & - (5R_1^2/9)X^9\} - (b_{HT}/b_T)\,\{X^3/3 - (R_1/5)X^5 - (5R_1^2/7)X^7\}] + \\ & + C/(3R_2 v_{T2} b_T)\,[1/3 \ln(\theta_2/\theta_1) + (5R_2/6)Y^2 + (R_2^2/4)Y^4 + (R_2/6)Y^{-2} - \\ & - (D/b_T)\,\{Y^3/9 + (R_2/3)Y^5 + (R_2^2/7)Y^7 - (R_2/3)Y\} - (b_{HT}/b_T)\,\{Y/3 + \\ & + (5R_2/9)Y^3 + R_2^2 Y^5 + (R_2/3)Y^{-1}\}] \end{aligned} \tag{28}$$

$$\begin{aligned} K_L = {} & C/(2v_{L1} b_L)\,[X_1 - D/3b_L)X_1^3 - (b_{HL}/b_L)X_1] + C/(2v_{L2} b_L)\,[Y_1 - (R_4/3)Y_1^3 - \\ & - R_4^2 Y_1^5 - (D/b_L)\,\{Y_1^3/3 - (R_4/5)Y_1^5 - (5R_4^2/7)Y_1^7\} - (b_{HL}/b_L)\,\{Y_1 - \\ & - (R_4/3)Y_1^3 - R_4^2 Y_1^5\}] \end{aligned} \tag{29}$$

(B) If $\tau_{4\mathrm{ph}}^{-1} \gg \tau_{3\mathrm{ph}}^{-1} \gg \tau_{\mathrm{pt}}^{-1}$

$$\begin{aligned} K_T = {} & C/(v_{T1} b_{HT})\,[X - (R_1/3)X^3 - R_1^2 X^5 - (D/b_{HT})\,\{X^3/3 - (R_1/5)X^5 - \\ & - (5R_1^2/7)X^7\} - (b_T/b_{HT})\,\{\ln(X) - (R_1/2)X^2 - (5R_1^2/4)X^4\}] + \\ & + C/(3R_2 v_{T2} b_{HT})\,[(5R_2/3)Y + (R_2^2/3)Y^3 - (\mathrm{R}_2/9)Y^{-3} - Y^{-1}/3 - \\ & - (D/b_{HT})\,\{Y/3 + (5R_2/9)Y^3 + (R_2^2/5)Y^5 - (R_2/3)Y^{-1}\} - \\ & - (b_\Gamma/b_{HT})\,\{(5R_2/3)\ln(\theta_2/\theta_1) + (R_2^2/2)Y^2 + (R_2/12)Y^{-4} - Y^{-2}/6\}] \end{aligned} \tag{30}$$

$$\begin{aligned} K_L = {} & C/(2v_{L1} b_{HL})[X_1(1 - b_L/b_{HL}) - (D/3b_{HL})X_1^3] + C/(2v_{L2} b_{HL})]\,\{Y_1 - \\ & - (R_4/3)Y_1^3 - R_4^2 Y_1^5\}\,(1 - b_L/b_{HL}) - (D/b_{HL})\,\{Y_1^3/3 - \\ & - (R_4/5)Y_1^5 - (5R_4^2/7)Y_1^7\}] \end{aligned} \tag{31}$$

where

$X = \theta_1/T$, $X_1 = \theta_3/T$, $Y^n = [(\theta_2/T)^n - (\theta_1/T)^n]$; $n = 1, 2, 3, \ldots$

$Y_1^n = [(\theta_4/T)^n - (\theta_3/T)^n]$; $n = 1, 2, 3, \ldots$

$$D = A(k_B T/\hbar)^4, \quad b_{HT} = B_{HT}(k_B/\hbar)^2 T^4, \quad b_{HL} = B_{HL}(k_B/\hbar)^2 T^4,$$

$$b_T = (B_{TN,\,I} + B_{TU,\,I}\, e^{-\theta/\alpha T})(k_B/\hbar)\; T^m T, \;\; I^{(T)+1}$$

$$b_L = (B_{LN,\,I} + B_{LU,\,I}\, e^{-\theta/\alpha T})(k_B/\hbar)^2 T^m L, \;\; I^{(T)+2}$$

$$+ (B_{LN,\,II} + B_{LU,\,II}\, e^{-\theta/\alpha T})(k_B/\hbar)^2 T^m L, \; II^{(T)+2}$$

At low temperatures, the contributions of the second integrals in Eqs (26) and (27) are very small as compared to the contributions due to the first integrals and can be ignored. At the same time, due to the small value of T, the upper limits θ_1/T and θ_3/T of the first integral in Eqs (26) and (27) are large enough. Thus, the factor $(e^x - 1)$ can be approximated to e^x. With these approximations, one has the following analytical expression for the lattice thermal conductivity:

(C) If $\tau_{3ph}^{-1} \gg \tau_{pt}^{-1} \gg \tau_B^{-1}$

$$K_T = C/(v_{T1} b_T)\,[(Z^5 - R_1 Z^5 - 5R_1^2 Z^7) - (D/b_T)(Z^6 - R_1 Z^8 - 5R_1^2 Z^{10}) - - (\tau_B^{-1}/b_T)(Z^2 - R_1 Z^4 - 5R_1^2 Z^6) - (b_{HT}/b_T)(Z^4 - R_1 Z^6 - 5R_1^2 Z^8)] \tag{32}$$

$$K_L = C/(2v_{L1} b_L)\,[Z_1^2 - (D/b_L) Z_1^4 - (\tau_B^{-1}/b_L) Z_1^0 - (b_{HL}/b_L) Z_1^2] \tag{33}$$

where

$$Z^n = 1 - n!\, e^{-X}\, 2(X_2^n/(n-1)! + X_2^{n-1}/(n-2)! + \ldots + X_2^2/3! + X_2/2! - 1)$$

$$Z_1^n = 1 - n!\, e^{-X}\, 3(X_3^n/(n-1)! + X_3^{n-1}/(n-2)! + \ldots + X_3^2/3! + X_3/2! - 1)$$

$$X_2 = \theta_1/T \quad \text{and} \quad X_3 = \theta_3/T$$

(D) If $\tau_B^{-1} \gg \tau_{pt}^{-1} \gg \tau_{3ph}^{-1}$

$$K_T = 24C\tau_B/(v_{T1})[1 - 5\tau_B b_T - 1680\tau_B D] \tag{34}$$

$$K_L = 24C\tau_B/(v_{L1})[1 - 30\tau_B b_L - 1680 D \tau_B] \tag{35}$$

It should be noted that in obtaining Eqs (34) and (35), the upper limits of the first integrals in Eqs (26) and (27) are taken as infinity due to the very low value of temperature T. It should also be noted that the analytical expressions are obtained in terms of the numerical values of the constants for Ge.

Phonon conductivity of Ge

To see the applicability of the expression proposed for τ_{3ph}^{-1} through Eqs (15) and (16), the lattice thermal conductivity of Ge has been calculated in the entire temperature range 2–1000 K via Eqs (26) and (27) as an example. The dispersion constants r are calculated with the help of the equation

$$r = (1/w^2)(qv/w - 1) \tag{36}$$

and using the experimental dispersion curve (57) of Ge. The values of these constants are found to be the same as reported by Verma *et al.* (56). The values obtained are reported in Table 4. As far as the Casimir length of the crystal and the point-defect scattering strength are concerned, these two constants do not need any adjustment for Ge, due to the fact that they have been studied by several workers and the values obtained are nearly the same. In the present analysis, the values of L and A are taken from the earlier report of Holland [6]. The temperature exponents $m_{T,I}(T)$, $m_{L,I}(T)$ and $m_{L,II}(T)$ for the three-phonon scattering relaxation rate have been calculated in the entire temperature range 2 – 1000 K with the help of Eqs (9) and (10), and the values obtained are listed in Tables 5 – 7, respectively. To permit a comparative study of the temperature exponent $m(T)$ with that used by Verma *et al.* [9] and also with the upper limit found by Guthrie, values of $m(T)$ have also been calculated by the different methods and are given in Tables 5 – 7. The variations of the various $m(T)$ values with temperature are shown in Figs 1 – 3.

The difficulties lie in estimating the three-phonon N and U process scattering strengths. This can be done with the analytical expressions reported in the preceding section. At low temperatures, $\tau_{3ph,N}^{-1} \gg \tau_{3ph,U}^{-1}$. Thus rough values of $B_{TN,I}$, $B_{LN,I}$ and $B_{LN,II}$ at 15 K have been estimated with the analytical expressions in Eqs (32) and (33), ignoring the contribution due to the three-phonon U processes. Similarly, at high temperatures $\tau_{3ph,U}^{-1} \gg \tau_{3ph,N}^{-1}$ and one can neglect $\tau_{3ph,N}^{-1}$ at these temperatures. $B_{TU,I}$, $B_{LU,I}$ and $B_{LU,II}$ at 400 K have been estimated with the analytical expressions in Eqs (28) and (29). From the approximate values of these constants, better values have been obtained by numerical integration of the con-

Table 4

The constants and parameters used in the calculation of the lattice thermal conductivity of Ge in the temperature range 2 – 1000 K

$v_{T1} = 3.55\ 10^5$ cm/sec	$r_3 = 0$
$v_{L1} = 4.92\ 10^5$ cm/sec	$r_4 = 1.13\ 10^{-27}\ \sec^2$
$v_{T2} = 1.30\ 10^5$ cm/sec	$\tau_B^{-1} = 1.96\ 10^6\ \sec^{-1}$
$v_{L2} = 2.46\ 10^5$ cm/sec	$A = 2.4\ 10^{-44}\ \sec^3$
$\theta_1 = 90$ K	$B_{TN,I} = 1.0\ 10^{-12}$ deg^{-m}
$\theta_2 = 118$ K	$B_{TU,I} = 1.95\ 10^{-6}$ deg^{-m}
$\theta_3 = 208$ K	$B_{LN,I} = 1.2\ 10^{-23}$ sec deg^{-m}
$\theta_4 = 319$ K	$B_{LU,I} = 1.0\ 10^{-20}$ sec deg^{-m}
$\theta = 376$ K	$B_{LN,II} = 2.0\ 10^{-21}$ sec deg^{-m}
$\alpha = 2.0$	$B_{LU,II} = 5.0\ 10^{-18}$ sec deg^{-m}
$r_1 = 2.95\ 10^{-27}\ \sec^2$	$B_{HT} = 1.0\ 10^{-23}$ sec deg^{-2}
$r_2 = 8.28\ 10^{-27}\ \sec^2$	$B_{HL} = 1.0\ 10^{-23}$ sec deg^{-2}

Table 5

The temperature exponent $m_{\mathrm{T,I}}(T)$ used for class I events for transverse phonons. m(SDV represents the value used in the SDV model and m(Guthrie) represents the upper limit of the temperature exponent obtained by Guthrie

Temperature T, K	m(Present)	m(SDV)	m(Guthrie)
1000	1.00116	1.02610	1.00232
900	1.00143	1.02932	1.00286
800	1.00181	1.03339	1.00362
700	1.00237	1.03868	1.00473
600	1.00322	1.04583	1.00644
500	1.00464	1.05600	1.00927
400	1.00724	1.07154	1.01448
300	1.0186	1.09816	1.02572
200	1.02884	1.15392	1.05768
100	1.11343	1.34312	1.22685
90	1.13931	1.38994	1.27862
80	1.17505	1.45094	1.35010
70	1.22630	1.53334	1.45259
60	1.30328	1.64988	1.60656
50	1.42506	1.82490	1.85213
40	1.63793	2.10974	2.27586
30	2.04521	2.63833	3.09042
20	2.96621	3.74792	4.0
10	4.0	4.0	4.0
8	4.0	4.0	4.0
6	4.0	4.0	4.0
4	4.0	4.0	4.0
2	4.0	4.0	4.0

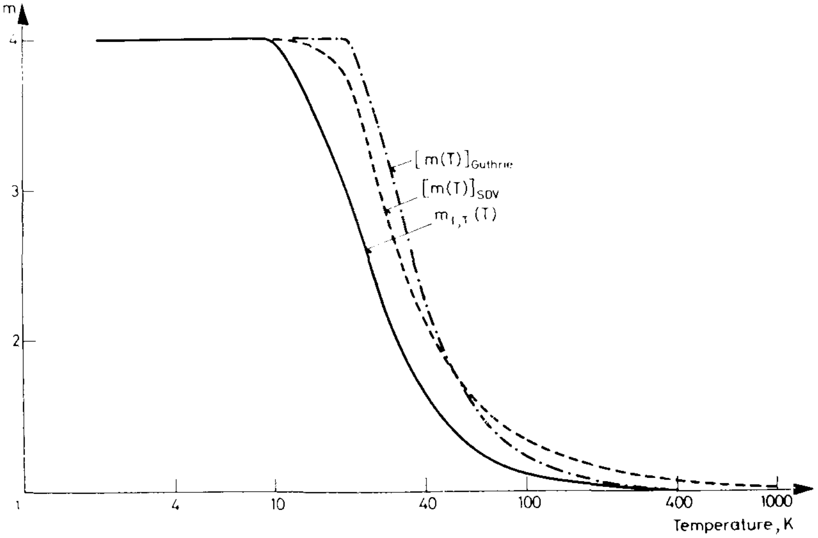

Fig. 1. The temperature exponent $m_{T,I}(T)$ for class I events for transverse phonons for Ge, —— present work; ——— SDV model; - . - . - upper limit obtained by Guthrie

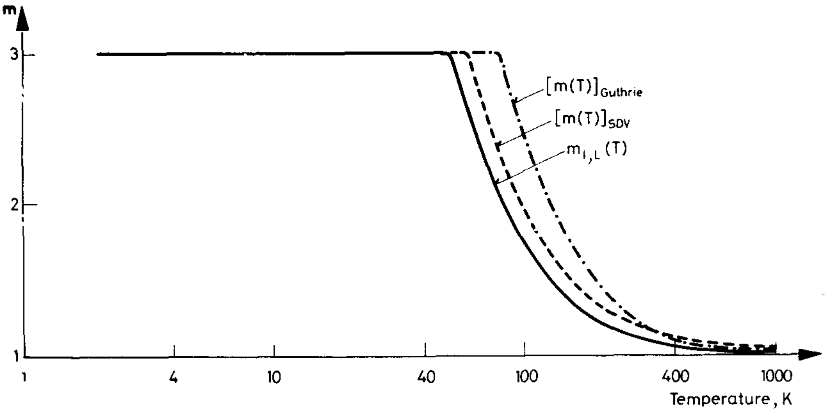

Fig. 2. The temperature exponent $m_{L,I}(T)$ for class I events for longitudinal phonons for Ge, —— present work; — — — SDV model; - . - . - upper limit obtained by Guthrie

Table 6

The temperature exponent $m_{L,I}(T)$ used for class I events for longitudinal phonons. m(SDV) represents the value used in the SDV model and m(Guthrie) represents the upper limit of the temperature exponent obtained by Guthrie

Temperature, T, K	m(Present)	m(SDV)	m(Guthrie)
1000	1.00847	1.03340	1.01693
900	1.01045	1.03833	1.02089
800	1.01322	1.04479	1.02643
700	1.01725	1.05356	1.03449
600	1.02345	1.06605	1.04689
500	1.03369	1.08505	1.06738
400	1.05245	1.11675	1.10489
300	1.09249	1.17779	1.18499
200	1.20352	1.32860	1.40705
100	1.73198	1.96167	2.46396
90	1.87765	2.12828	2.75529
80	2.06910	2.34499	3.0
70	2.32689	2.63393	3.0
60	2.68456	3.0	3.0
50	3.0	3.0	3.0
40	3.0	3.0	3.0
30	3.0	3.0	3.0
20	3.0	3.0	3.0
10	3.0	3.0	3.0
8	3.0	3.0	3.0
6	3.0	3.0	3.0
4	3.0	3.0	3.0
2	3.0	3.0	3.0

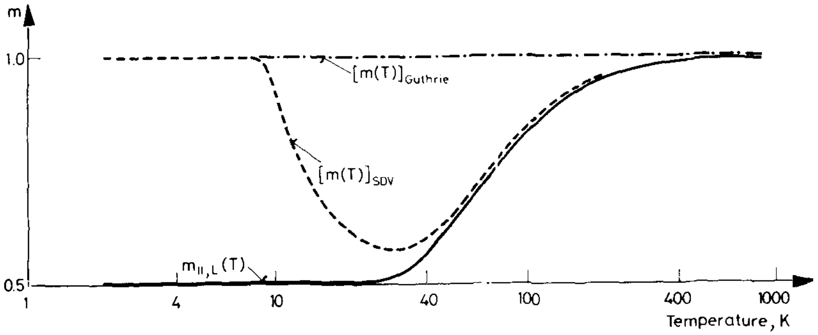

Fig. 3. The temperature exponent $m_{L,II}(T)$ for class II events for longitudinal phonons for Ge, —— present work; — — — SDV model; - . - . - upper limit obtained by Guthrie

Table 7

The temperature exponent $m_{L,II}(T)$ used for class II events for longitudinal phonons. m(SDV) represents the value used in the SDV model and m(Guthrie) represents the upper limit of the temperature exponent obtained by Guthrie

Temperature, T, K	m(Present)	m(SDV)	m(Guthrie)
1000	0.99789	0.99791	1.0
900	0.99739	0.99742	1.0
800	0.99670	0.99674	1.0
700	0.99570	0.99575	1.0
600	0.99416	0.99423	1.0
500	0.99162	0.99172	1.0
400	0.98699	0.98716	1.0
300	0.97720	0.97751	1.0
200	0.95068	0.95144	1.0
100	0.83754	0.84097	1.0
90	0.81016	0.81449	1.0
80	0.77665	0.78226	1.0
70	0.73587	0.74341	1.0
60	0.68717	0.69776	1.0
50	0.63156	0.64737	1.0
40	0.57398	0.59974	1.0
30	0.52610	0.57407	1.0
20	0.50274	0.61476	1.0
10	0.50	0.91414	1.0
8	0.50	1.0	1.0
6	.050	1.0	1.0
4	.050	1.0	1.0
2	.050	1.0	1.0

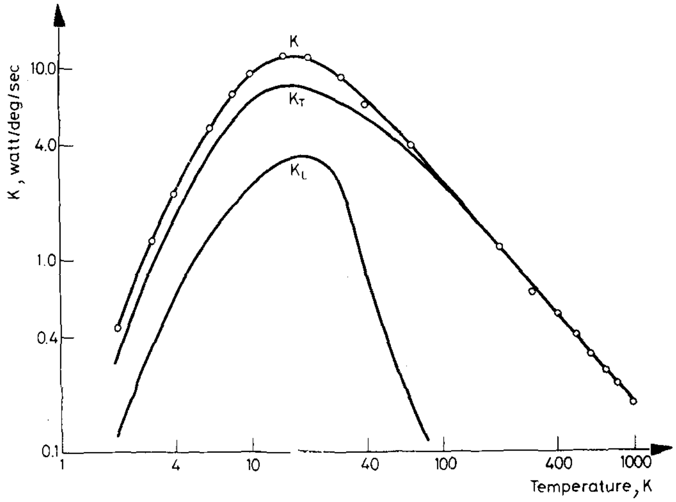

Fig. 4. The total lattice thermal conductivity of Ge in the temperature range 2—1000 K. K_T and K_L are the separate contributions due to transverse and longitudinal phonons, respectively. Solid line: calculated values, circles: experimental points

Table 8

The percentage contributions of the three-phonon scattering relaxation rate $\tau_{3ph,T}^{-1}$ towards the combined scattering relaxation rate $\tau_{c,T}^{-1}$ due to transverse phonons due to class I events in the absence of the four-phonon scattering processes for four different values of the phonon frequency. w_{max} represents the maximum frequency of the transverse phonons

Temperature, T, K	% $\tau_{3ph,T}^{-1}$ for $w = 1/4\, w_{max}$	% $\tau_{3ph,T}^{-1}$ for $w = {}^1/_2\, w_{max}$	% $\tau_{3ph,T}^{-1}$ for $w = 3/4\, w_{max}$	% $\tau_{3ph,T}^{-1}$ for $w = w_{max}$
1000	76.24	86.52	90.59	92.77
900	75.89	86.30	90.43	92.64
800	75.46	86.01	90.22	92.48
700	74.89	85.65	89.94	92.27
600	74.14	85.15	89.59	91.98
500	73.08	84.45	89.07	91.57
400	71.49	83.38	88.27	90.94
300	68.84	81.55	86.89	89.84
200	63.62	77.77	83.99	87.49
100	49.71	66.41	74.78	79.81
90	47.10	64.04	72.26	78.08
80	44.11	61.21	70.30	75.94
70	40.66	57.81	67.27	73.26
60	36.69	53.68	63.49	69.86
50	32.14	48.65	58.70	65.45
40	26.88	42.37	52.45	59.52
30	20.34	33.80	43.36	50.50
20	10.36	18.78	25.73	31.53
10	0.20	0.40	0.58	0.74
8	0.10	0.20	0.28	0.34
6	0.04	0.08	0.11	0.11
4	0.01	0.02	0.02	0.02
2	0	0	0	0

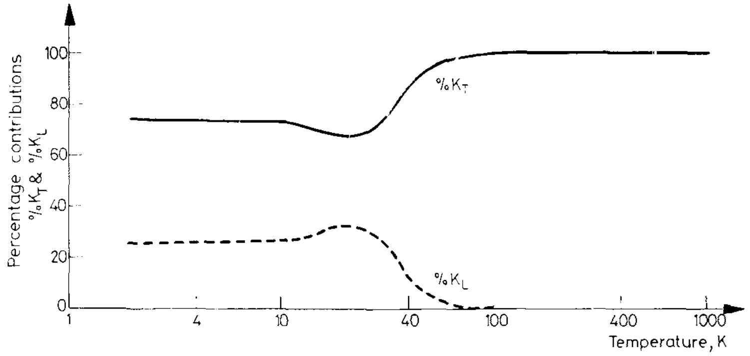

Fig. 5. The percentage contributions of transverse and longitudinal phonons towards the total lattice thermal conductivity of Ge. Solid line: % K_T; dotted line: % K_L

Table 9

The percentage contributions of the three-phonon scattering relaxation rate $\tau_{3ph,L}^{-1}$ towards the combined scattering relaxation rate $\tau_{c,L}^{-1}$ due to longitudinal phonons due to the combined effect of class II events in the absence of four-phonon processes for four different values of the phonon frequency. w_{max} represents the maximum frequency of longitudinal phonons

Temperature, T, K	% $\tau_{3ph,L}^{-1}$ for $w = 1/4\ w_{max}$	% $\tau_{3ph,L}^{-1}$ for $w = 1/2\ w_{max}$	% $\tau_{3ph,L}^{-1}$ for $w = 3/4\ w_{max}$	% $\tau_{3ph,L}^{-1}$ for $w = w_{max}$
1000	99.94	99.74	99.43	98.99
900	99.93	99.71	99.35	98.85
800	99.92	99.66	99.25	98.67
700	99.90	99.60	99.11	98.42
600	99.88	99.51	98.90	98.06
500	99.84	99.36	98.58	97.50
400	99.77	99.11	98.01	96.52
300	99.63	98.54	96.78	94.41
200	99.14	96.66	92.80	87.88
100	93.92	79.53	63.34	49.28
90	91.84	73.89	55.72	41.45
80	89.63	68.50	49.15	35.22
70	89.06	67.20	47.66	33.88
60	91.99	74.29	56.23	41.95
50	92.40	75.35	57.61	43.33
40	72.62	40.01	22.87	14.30
30	25.58	7.96	3.70	2.12
20	4.13	1.07	0.48	0.27
10	0.69	0.17	0.08	0.04
8	0.44	0.11	0.05	0.02
6	0.28	0.07	0.03	0.02
4	0.18	0.04	0.02	0.01
2	0.11	0.03	0.01	0

ductivity integrals in Eqs (26) and (27) at 15 K (near the conductivity maxima) and at 300 K (near room temperature). The four-phonon scattering strengths B_{HT} and B_{HL} are estimated with the analytical expressions in Eqs (30) and (31), which are further corrected at 500 K by numerical integration of the conductivity integrals. The values obtained for these contents are listed in Table 4. From all the constants is Table 4 the lattice thermal conductivity of Ge has been obtained by calculating the separate contributions due to transverse and longitudinal phonons in the entire temperature range 2—1000 K; the result is shown in Fig. 4. Each contribution has been estimated by numerical integration of the conductivity integrals with an HP-9830A mini computer. The separate percentage contributions of transverse and longitudinal phonons have also been studied in the entire temperature range of study and the results are shown in Fig. 5.

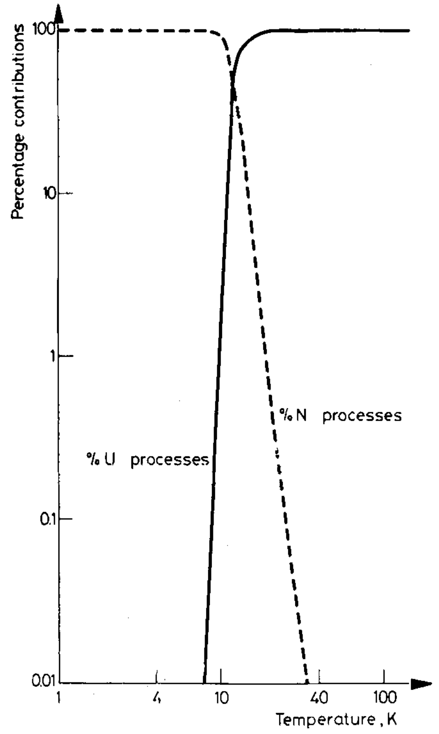

Fig. 6. The percentage contributions of $\tau^{-1}_{3ph,N}$ and $\tau^{-1}_{3ph,U}$ towards $\tau^{-1}_{3,ph,T}$ for class I events for transverse phonons for Ge in the temperature range 2—1000 K. Solid line: percentage contribution due to $\tau^{-1}_{3ph,U}$; dotted line: percentage contribution due to $\tau^{-1}_{3ph,N}$

To study the roles of three-phonon N and U processes, the percentage contributions of $\tau^{-1}_{3ph,N}$ and $\tau^{-1}_{3ph,U}$ towards τ^{-1}_{3ph} have been calculated for class I events for transverse phonons, and for class I and class II events for longitudinal phonons. The results are shown in Figs 6—8. The percentage contributions of $\tau^{-1}_{3ph,T}$ (class I) for transverse phonons and $\tau_{3ph,L}^{-1}$ (class I + class II) for longitudinal phonons towards the combined scattering relaxation rate of the respective modes $\tau_{c,T}^{-1}$ and $\tau_{c,L}^{-1}$ are listed in Tables 8 and 9. The percentage contributions of $\tau_{3ph,L}^{-1}$ (class I) and $\tau_{3ph,L}^{-1}$ (class II) towards $\tau_{c,L}^{-1}$ have also been calculated and are reported in Tables 10 and 11.

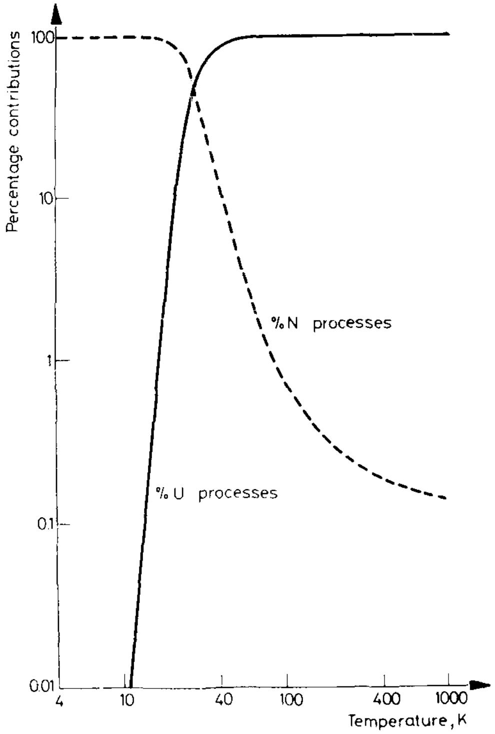

Fig. 7. The percentage contributions of $\tau_{3ph,N}^{-1}$ and $\tau_{3ph,U}^{-1}$ towards $\tau_{3ph,L,I}^{-1}$ for class I events for longitudinal phonons for Ge in the temperature range 2—1000 K. Solid line: percentage contribution due to $\tau_{3ph,U}^{-1}$; dotted line: percentage contribution due to $\tau_{3ph,N}^{-1}$

A comparative study of the present analysis with previous analysis

In this section, a comparative study between the present analysis and the previous analyses of other workers has been made. From Tables 1 and 2 it is clear that Callaway used $\tau_{3ph}^{-1} \alpha w^2 T^3$ in the entire temperature range 2—100 K, although this is valid at low temperatures only. Callaway did not make any distinction between transverse and longitudinal phonons and his entire analysis is valid for longitudinal phonons, except for the average phonon velocity. Due to this, Callaway could not get good agreement at high temperature (see Fig. 2 of ref. (6)). Holland [6] and Verma *et al.* [56] calculated the phonon conductivity of Ge in the temperature range 2—1000 K using two-mode conduction of phonons. However, from Tables 1 and 2 it is clear that they used $\tau^{-1}_{3ph,N} \propto wT^4$ for transverse phonons and $\tau^{-1}_{3ph,N} \propto w^2T^3$ for longitudinal phonons in the entire temperature range of study, which are valid for low-frequency phonons and at low temperature only. At the same time, in calculating the contribution due to transverse phonons, they could not include $\tau^{-1}_{3ph,U}$ in the range $0-1/2\ q_{max}$ and $\tau^{-1}_{3ph,N}$ in the range $1/2\ q_{max} - q_{max}$. They totally ignored $\tau^{-1}_{3ph,U}$ in calculating the contribution due to longitudinal phonons. The expression $\tau^{-1}_{3ph,U} \propto w^2/\sin h(hw/k_BT)$ which they used for transverse phonons gives a T-dependence for the three-phonon scattering relaxation rate which is valid at high temperatures only. Tiwari and Agrawal [58] also calculated the phonon conducitivity of Ge in the temperature range 2–1000 K by using the expressions $\tau^{-1}_{3ph,T} \propto wT^m$ and $\tau^{-1}_{3ph,L} \propto w^2T^m$, which reveals that they employed the same temperature exponent value for both modes, i.e. they used the same temperature-dependence for transverse and longitudinal phonons, though these should be different. It is clear from Table 2 that they considered only the contribution due to the three-phonon N processes in their analysis (for details, see Table 1 of ref. (58)).

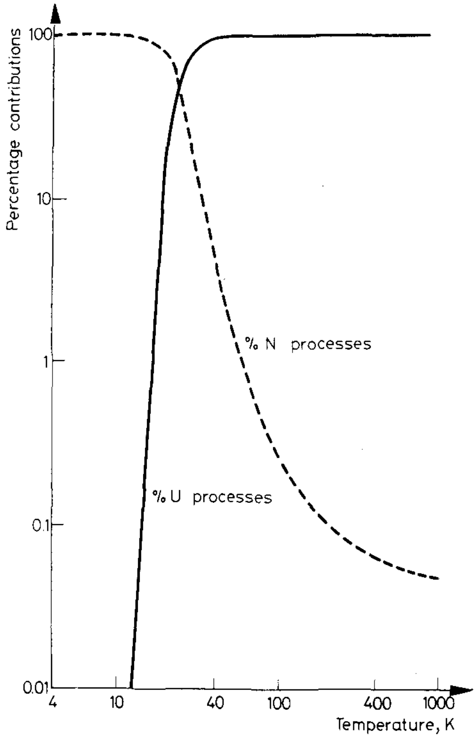

Fig. 8. The percentage contributions of $\tau^{-1}_{3ph,N}$ and $\tau^{-1}_{3ph,U}$ towards $\tau^{-1}_{3ph,L,II}$ for class II events for longitudinal phonons for Ge in the temperature range 2–1000 K. Solid line: percentage contribution due to $\tau^{-1}_{3ph,U}$; dotted line: percentage contribution due to $\tau^{-1}_{3ph,N}$

However, they used a better temperature dependence for $\tau_{3\text{ph}}^{-1}$ than the previous workers.

The first attempt to incorporate the Guthrie expression for $m(T)$ for $\tau_{3\text{ph}}^{-1}$ in the calculation of the lattice thermal conductivity was made by Verma *et al.*, who proposed the new Sharma–Dubey–Verma (SDV) model [9, 10, 23–25]. In the SDV model, a continuous temperature exponent is used for the first time for the three-phonon scattering relaxation rate. However, the expression for $m(T)$ in the SDV model (see Eqs (5), (6) and (22) of ref. [9]) contains an empirical factor $(1 + \theta/\alpha T)$. At the same time, a careful analysis of Figs 1–3 and Tables 5–7 shows that at high temperatures the value of $m(T)$ used in the SDV model is larger than the upper Guthrie limit. (This can also be seen in Figs 2–4 of ref. [9].) From Tables 1 and 2 (and also from Eqs (20) and (21) of ref. [9]), it is clear that the expression used for $\tau_{3\text{ph}}^{-1}$ in the SDV model does not include the contribution due to the three-phonon N processes. As a result of this, they could not get good agreement at high temperatures (see Fig. 1 of ref. [9]).

Table 10

The percentage contributions of the three-phonon scattering relaxation rate $\tau_{3\text{ph},\text{L},\text{I}}^{-1}$ towards the combined scattering relaxation rate $\tau_{\text{c},\text{L}}^{-1}$ due to longitudinal phonons due to the class I events alone in the absence of four-phonon processes for four different values of the phonon frequency. $w_{\max}$ represents the maximum frequency of longitudinal phonons

Temperature, T, K	% $\tau_{3\text{ph},\text{L},\text{I}}^{-1}$ for $w = 1/4\ w_{\max}$	% $\tau_{3\text{ph},\text{L},\text{I}}^{-1}$ for $w = 1/2\ w_{\max}$	% $\tau_{3\text{ph},\text{L},\text{I}}^{-1}$ for $w = 3/4\ w_{\max}$	% $\tau_{3\text{ph},\text{L},\text{I}}^{-1}$ for $w = w_{\max}$
1000	0.21	0.21	0.21	0.21
900	0.22	0.22	0.22	0.22
800	0.22	0.22	0.22	0.22
700	0.23	0.23	0.23	0.23
600	0.24	0.24	0.24	0.24
500	0.26	0.26	0.26	0.25
400	0.30	0.30	0.29	0.28
300	0.38	0.38	0.37	0.36
200	0.75	0.73	0.70	0.67
100	10.33	8.73	6.97	5.42
90	18.10	14.56	10.98	8.17
80	32.84	25.18	18.07	12.95
70	56.61	42.71	30.30	21.53
60	80.84	65.29	49.41	36.86
59	88.35	72.05	55.09	41.43
40	68.51	37.75	21.58	13.49
30	23.64	7.36	3142	1.96
20	3.73	0.97	0.43	0.21
10	0.45	0.11	0.05	0.03
8	0.23	0.06	0.03	0.01
6	0.11	0.02	0.01	0
4	0.03	0	0	0
2	0	0	0	0

Thus, for the first time, the Guthrie expression for the temperature exponent $m(T)$ for τ_{3ph}^{-1} has been incorporated in the calculation of the lattice thermal conductivity without any empirical factor. At the same time, the contributions due to three-phonon N and U processes are also included for both of the modes in the entire temperature range 2–10000 K. The contributions due to $\tau_{3ph,N}^{-1}$ and $\tau_{3ph,U}^{-1}$ have also been included in the entire range of the first Brillouin Zone, i.e. both N and U processes have been considered in the same conductivity integral. The expression proposed for the three-phonon scattering relaxation rate is based on the division into class I and class II events, and also N and U processes. The role of four-phonon processes also is considered in the present analysis.

Table 11

The percentage contribution of the three-phonon scattering relaxation rate $\tau_{3ph,L,II}^{-1}$ towards the combined scattering relaxation rate $\tau_{c,L}^{-1}$ due to longitudinal phonons due to class II events alone in the absence of four-phonon processes for four different values of the phonon frequency w_{max} represents the maximum frequency of longitudinal phonon

Temperature, T, K	% $\tau_{3ph,L,II}^{-1}$ for $w = 1/4\ w_{max}$	% $\tau_{3ph,L,II}^{-1}$ for $w = 1/2\ w_{max}$	% $\tau_{3ph,L,II}^{-1}$ for $w = 3/4\ w_{max}$	% $\tau_{3ph,L,II}^{-1}$ for $w = w_{max}$
1000	99.72	99.53	99.21	98.78
900	99.71	99.49	99.13	98.63
800	99.69	99.44	99.02	98.45
700	99.67	99.37	98.88	98.19
600	99.64	99.27	98.66	97.83
500	99.58	99.11	98.32	97.25
400	99.48	98.81	97.72	96.23
300	99.25	98.16	96.40	94.05
200	98.39	95.93	92.09	87.21
100	83.58	70.78	56.37	43.86
90	73.74	59.33	44.74	33.28
80	56.68	43.30	31.09	22.29
70	32.45	24.48	17.37	12.34
60	11.15	9.00	6.81	5.08
50	4.04	3.30	2.52	1.90
40	4.10	2.26	1.29	0.81
30	1.94	0.60	0.28	0.16
20	0.40	0.10	0.05	0.03
10	0.24	0.06	0.03	0.02
8	0.21	0.05	0.02	0.01
8	0.19	0.04	0.02	0.01
4	0.15	0.03	0.01	0.01
2	0.11	0.03	0.01	0

Results and discussion

We shall first discuss the expression proposed for $\tau_{3\mathrm{ph}}^{-1}$ through Eqs (15) and (16). At low temperatures, the temperature exponents for transverse phonons, $m_{\mathrm{T,I}}(T)$, and for longitudinal phonons, $m_{\mathrm{L,I}}(T)$, reduce to 4 and 3, respectively, i.e. at low temperatures $m_{\mathrm{T,I}}(T) \rightarrow 4$ and $m_{\mathrm{L,I}}(T) \rightarrow 3$. Through numerical analysis of K_{L}, it has been found that at low temperatures the scattering relaxation rate due to class I events dominates over the scattering relaxation rate due to class II events. At the same time, due to the low value of temperature T, the factor $e^{-\theta/\alpha T}$ is very small. As a result, at low temperatures the expression proposed for $\tau_{3\mathrm{ph}}^{-1}$ reduces to $\tau_{3\mathrm{ph,T}}^{-1} = B_{\mathrm{TN,I}}\, wT^4$ for transverse phonons, and to $\tau_{3\mathrm{ph,L}}^{-1} = B_{\mathrm{LN,I}}\, w^2T^3$ for longitudinal phonons, which are similar to the earlier findings of Herring [1]. At the same time, they are similar to the expressions used by Joshi and Verma [11].

At high temperatures, the temperature exponents $m_{\mathrm{T,I}}(T)$, $m_{\mathrm{L,I}}(T)$ and $m_{\mathrm{L,II}}(T)$ all tend to unity. It is well known that at high temperatures the scattering relaxation rate due to three-phonon U process dominates over the scattering relaxation rate due to three-phonon N processes. Thus, the expression for $\tau_{3\mathrm{ph}}^{-1}$ used in the present analysis reduces to $\tau_{3\mathrm{ph,T}}^{-1} = BwTe^{-\theta/\alpha T}$ for transverse phonons, and to $\tau_{3\mathrm{ph,L}}^{-1} = B'w^2Te^{-\theta/\alpha T}$ for longitudinal phonons. These expressions are similar to the findings of Klemens. It is interesting to point out that, in the absence of three-phonon N processes, the expression for $\tau_{3\mathrm{ph}}^{-1}$ proposed in the present work reduces to the expression used in the SDV model.

The values of the temperature exponents $m_{\mathrm{T,I}}(T)$, $m_{\mathrm{L,I}}(T)$ and $m_{\mathrm{L,II}}(T)$ (as obtained from Eqs (9) and (10) used in the present analysis are reported in Tables 5–7. Their continuous nature with temperature can be seen in Figs 1–3. With the help of Tables 5–7 and Figs 1–3, it can be seen that at low temperatures $m_{\mathrm{T,I}}(T)$ and $m_{\mathrm{L,I}}(T)$ tend to 4 and 3, respectively, which is the same as obtained by Herring [1] at low temperatures. It can also be seen that at high temperatures $m_{\mathrm{T,I}}(T)$, $m_{\mathrm{L,I}}(T)$ and $m_{\mathrm{L,II}}(T)$ all tend to unity, which results in $\tau_{3\mathrm{ph}}^{-1} \alpha\, T$, similar to the earlier findings of Herring at high temperatures. It also results in $K \alpha\, 1/T$ at high temperatures, which is similar to the previous findings. From these Figures, it is very clear that the value of $m(T)$ used in the present analysis lies between 1 and 4 for transverse phonons, and between 1 and 3 for longitudinal phonons, and it does not exceed the upper Guthrie limit any temperature. Thus, it is free from Guthrie's comments [8] too. Therefore, one can say that the value of $m(T)$ used in the present analysis is more realistic than those used by previous workers.

With the help of Fig. 4, it can be seen that the agreement between the calculated and experimental values of the lattice thermal conductivity is very good in the entire temperature range 2–1000 K, which tells that the expression proposed for $\tau_{3\mathrm{ph}}^{-1}$ in the present analysis gives a very good response to the experimental data of the lattice thermal conductivity at high as well as at low temperatures. The separate percentage contributions due to transverse and longitudinal phonons can be studied with the help of Fig. 5. From Figs 4 and 5 it can be concluded that at high temperatures most of the heat is carried by the transverse phonons alone, which is in agreement with the findings of previous workers [6, 9–13]. It is also similar to the findings of Hamilton and Parrott [42] and Srivastava [59, 60] based on the variational approach.

The separate contributions due to three-phonon N and U processes can be studied via Figs 6–8. From these Figures it is clear that at low temperatures $\tau^{-1}_{3\mathrm{ph,N}}$ dominates over $\tau^{-1}_{3\mathrm{ph,U}}$ for both transverse and longitudinal phonons, and also in both class I and class II events. This results in the dominating role of the three-phonon N processes in the lattice thermal conductivity at low temperatures. At high temperatures, the domination of $\tau^{-1}_{3\mathrm{ph,U}}$ over $\tau^{-1}_{3\mathrm{ph,N}}$ can be seen from these Figures; this shows that at high temperatures the lattice thermal resistivity is mainly due to the three-phonon U processes. These conclusions are in agreement with the findings of the previous workers.

The percentage contribution of $\tau^{-1}_{3\mathrm{ph}}$ towards the combined scattering relaxation rate τ_c^{-1} in the absence of the four-phonon scattering relaxation rate $\tau^{-1}_{4\mathrm{ph}}$ can be seen in Tables 8–11. It is clear that at high temperatures $\tau^{-1}_{3\mathrm{ph}}$ dominates over the boundary and point-defect scattering relaxation rates, which has the result that at high temperatures the lattice thermal resistivity is mainly due to the three-phonon scattering relaxation rates. This is similar to the findings of Hamilton and Parrott based on the variational techniques, and also to those of Dubey [13] based on the relaxation time approach for Si. From Tables 10 and 11 it is obvious that $\tau^{-1}_{3\mathrm{ph,L}}$ (class II events) is much larger than $\tau^{-1}_{3\mathrm{ph,L}}$ (class I events) at high temperatures, which is similar to the results obtained by Verma *et al.* [9, 10]. From these two Tables it can also be seen that $\tau^{-1}_{3\mathrm{ph,L}}$ (class I events) is larger than $\tau^{-1}_{3\mathrm{ph,L}}$ (class II events) at low temperatures.

*

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RÉSUMÉ — On a analysé, dans le cadre de la classification de Guthrie des phénomènes de diffusion phonon-phonon en deux classes, I et II, les vitesses de relaxation de la diffusion à 3 phonons et l'on propose pour la première fois, comme résultat de ce travail, une nouvelle expression:

$$\tau_{3\mathrm{ph}}^{-1} = (B_{\mathrm{N,1}} + B_{\mathrm{U,I}} e^{-\theta/\alpha \mathrm{T}}) g(w) T^{\mathrm{m}_{\mathrm{I}}(\mathrm{T})} + (B_{\mathrm{N,II}} e^{-\theta/\alpha \mathrm{T}}) g(w) T^{\mathrm{m}_{\mathrm{II}}(\mathrm{T})}$$

pour les vitesses de relaxation de la diffusion à trois phonons, permettant de calculer la conductivité thermique du réseau d'un prélèvement. En se servant de l'expression proposée ci-dessus, on a analysé la conductivité thermique du réseau de Ge dans l'intervalle de températures allant de 2 à 1000K et le résultat obtenu montre un très bon accord avec les données expérimentales. On décrit de même le pourcentage des contributions dues aux processus normaux et inversés à trois phonons. Le rôle de processus à quatre phonons est aussi inclus aux températures élevées. Afin d'estimer une valeur approchée de la force diffusante et de la conductivité du phonon, l'expression analytique peut aussi être obtenue dans le cadre de l'expression proposée ci-dessus pour τ_{3ph}^{-1}.

ZUSAMMENFASSUNG — Die Drei-Phonon-Streuungsrelaxationsgeschwindigkeiten und ihre Temperaturexponenten wurden, im Rahmen von Guthries Klassifizierung der Phonon-Phonon-Streuungsergebnisse als solche der Klasse I und Klasse II, analysiert und als Ergebnis dieser Arbeit wurde zum ersten Mal ein neuer Ausdruck

$$\tau_{3ph}^{-1} = (B_{N,I} + B_{U,I}e^{-\theta/\alpha T})g(w)T^{m_{I}(T)} + (B_{N,II} + B_{U,II}e^{-\theta/\alpha T})g(w)T^{m_{II}(T)}$$

für die Drei-Phonon-Streuungsrelaxationsgeschwindigkeiten vorgeschlagen um die Gitter-Wärmeleitfähigkeit einer Probe zu berechnen. Unter Anwendung des oben vorgeschlagenen Ausdrucks wurde die Gitter-Wärmeleitfähigkeit von Ge im Temperaturbereich von 2 bis 1000K analysiert und das erhaltene Ergebnis zeigt eine sehr gute Übereinstimmung mit den Versuchsangaben. Die den Drei-Phonon Normal- und Umklapp-Prozessen zuzuschreibenden prozentualen Beiträge werden ebenfalls mitgeteilt. Die Rolle der Vier-Phonon-Vorgänge ist bei hohen Temperaturen ebenfalls mit inbegriffen. Um einen annähernden Wert der Streuungsstärke und der Phonon-Leitfähigkeit zu schätzen, wird der analytische Ausdruck auch im Rahmen des für τ_{3ph}^{-1} oben vorgeschlagenen Ausdrucks erhalten.

Резюме — Изучена релаксационная скорость трехфононового рассеяния и ее температурная экспонента в рамках выражения, предложенного Гутрье. Случаи фонон-фононового рассеяния разделены на два класса: события I класса, в которых носитель фонона аннигилирован комбинацией, и события II класса — где носитель фонона аннигилирован расщеплением. В результате этого предложено новое выражение

$$\tau_{3ph}^{-1} = (B_{N,I} + B_{U,I}e^{-\theta/\alpha T})g(w)T^{m_{I}(T)} + (B_{N,II} + B_{U,II}e^{-\theta/\alpha T})g(w)T^{m_{II}(T)}$$

для скорости релаксации трехфоновнового рассеяния с целью вычисления решеточной теплопроводности образца. Температурная экспонента $m(T)$ была изучена для обоих классов событий. В качестве примера была проанализирована решеточная теплопроводность германия в области температур 2—1000 К. Полйченные результаты хорошо совпадали с экспериментальными данными. При высоких температурах была принята во внимание роль четырехфононовых процессов. Для установления приближенного значени силы рассеяния и фононной проводимости, было чолучено аналитическое выражение на основе выражения, предложенного для $\tau_{3\text{фон}}^{-1}$.