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Phonon heat transport in gallium arsenide

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Abstract. The lifetimes of quantum excitations are directly related to the electron and phonon energy linewidths of a particular scattering event. Using the versatile double time thermodynamic Green's function approach based on many-body theory, an *ab-initio* formulation of relaxation times of various contributing processes has been investigated with newer understanding in terms of the linewidths of electrons and phonons. The energy linewidth is found to be an extremely sensitive quantity in the transport phenomena of crystalline solids as a collection of large number of scattering processes, namely, boundary scattering, impurity scattering, multiphonon scattering, interference scattering, electron–phonon processes and resonance scattering. The lattice thermal conductivities of three samples of GaAs have been analysed on the basis of modified Callaway model and a fairly good agreement between theory and experimental observations has been reported.

Keywords. Thermal transport; anharmonic forces; relaxation times; phonon widths.

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1. Introduction

Thermal conductivity of solids being a fundamental transport property, emerged as an indispensable tool for understanding heat carriers and the various scattering processes better [1,2]. Lattice waves (phonons), charged carriers (electrons and holes), electromagnetic waves, spin waves or other excitations participate in thermal transport and their magnitude and temperature dependence dramatically vary from one material to another. In semiconductors and non-metals, the heat is carried by phonons. The study of thermal conductivity in semiconductors is of utmost importance in the field of modern science, micro- and nanoelectronics [3–5]. The exact treatment of the problem of thermal conduction is however greatly hampered by (i) the lack of knowledge of vibrational spectra, (ii) tremendous complexities involved in the treatment of anharmonic forces and (iii) the difficulties in obtaining the exact solution of Boltzmann transport equation, which, in turn, resorts one to consider relaxation time approximation. Callaway [6] proposed a phenomenological and numerically amenable model based on single-mode relaxation time

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approximation which could successfully explain a huge amount of experimental data on thermal conductivity. The Callaway's representative expression of thermal conductivity is given by

$$\kappa = \frac{k_B(\beta\hbar)^2}{2\pi^2 v}\int_0^{\omega_D} \tau(\omega, T)\,\omega^4 e^{\beta\hbar\omega}(e^{\beta\hbar\omega}-1)^{-2}d\omega, \tag{1}$$

where v, ω_D, $\tau(\omega, T)$ stand for phonon velocity, Debye frequency, total relaxation time for all scattering processes, respectively and $\beta = (k_B T)^{-1}$. If one considers the scattering processes as independent, the scattering probabilities may be taken as additives so that

$$\tau^{-1}(\omega, T) = \sum_i \tau_i^{-1}(\omega, T), \tag{2}$$

where there are as many $\tau_i(\omega, T)$ as there are individual scattering processes and are normally obtained by perturbation techniques. This model considers the assumption of isotropy (no distinction between phonon polarization), introduction of Debye density of states (Debye spectrum) and additivity of inverse relaxation times. The simplicity of this model introduced some inadequacies which were addressed by Erdos and Haley [7]. Holland [8,9] and several other condensed matter theorists [10–22] have made significant contributions with newer concepts to improve and repair the original Callaway's phenomenological model. Also, Kubo [23] and his coworkers [24] presented a new approach to calculate the thermal transport coefficients. This approach, based on many-body theory and Green's function technique, has been utilized by several authors [25,26] to study the thermal conductivity. It is noteworthy that some authors [27–30] have made significant contribution to the theory of lattice thermal conductivity based on variational and many-body approaches.

The size of the specimen, impurities present in the crystals, anharmonic forces [31,32], concentration of carriers, interaction between the carriers and the lattice waves, interactions between magnetic ions and the lattice waves, etc. [28,33,34] are the major factors which greatly affect the thermal conductivity of a crystal which can be systematically studied via relaxation time $\tau(\omega, T)$. Of various scattering mechanisms appearing in $\tau(\omega, T)$ the scattering from external and internal microboundaries [35,36], impurities, imperfections [2] and crystal anharmonicities [31] limit the free mean free path of heat carriers and all of these factors significantly influence the thermal conductivity of solids. Based on Callaway's model, several authors [1,2,8,10,11,31,32,37,38] analysed the lattice thermal conductivity of a large number of solids and obtained excellent fits. However, adequate justifications to some issues are still missing. The comments and investigations on the additivity of relaxation time (the Matthiessen's rule is applicable to independent scattering processes only) were further made by Altukov and Zavt [42] but successful attempts to resolve this difficulty were made by Gairola [43] and Bahuguna *et al* [41] with a sound justification. The inadequacies which could not be removed by the Callaway and modified models have been removed by introducing the equivalence of relaxation times and electron (phonon) linewidths [39–41].

In the present paper, this new approach of equivalence of relaxation time and electron (phonon) linewidths has been utilized to analyse the lattice thermal conductivity of GaAs samples with some additional findings and an excellent agreement between theory and experimental data has been reported.

2. Theory

Following the new approach (quantum dynamical many-body theory) of linewidths, the relaxation times for various scattering processes can be given by [41,43]

$$\tau^{-1}(\omega, T) = \Gamma_k(\omega, T) + \tau_{\mathrm{CB}}^{-1}, \tag{3}$$

where $\Gamma_k(\omega, T)$ is the phonon frequency linewidth and is given by

$$\begin{aligned}\tau^{-1}(\omega, T) \;=\; & \Gamma_k^{\mathrm{D}}(\omega) + \Gamma_k^{\mathrm{3A}}(\omega, T) + \Gamma_k^{\mathrm{4A}}(\omega, T) + \Gamma_k^{\mathrm{AD}}(\omega, T) + \Gamma_k^{\mathrm{ep}}(\omega, T) \\ & + \tau_{\mathrm{CB}}^{-1}.\end{aligned} \tag{4}$$

In above equation various superscripts D, 3A, 4A, AD and ep stand for the contribution due to defect scattering, cubic and quartic anharmonic phonon scattering, anharmonicity-defect interference scattering and electron–phonon scattering. τ_{CB}^{-1} describes combined boundary scattering relaxation time. At very low temperatures the first and the foremost scattering phenomenon which dominantly contributes to the thermal conductivity is the combined boundary scattering which can be described by [35,36]

$$\tau_{\mathrm{CB}}^{-1} = \frac{v}{L(B)}, \tag{5}$$

where $L(B) = 1.12 l B_{\mathrm{I}}$, $L(B)^{-1} = 1/2v(t_1^{-1} + t_2^{-1})$, $B_{\mathrm{I}} = 1.7858 t^{-1}(t_1^{-1} + t_2^{-1})^{-1}$ is the internal boundary parameter which is related to large scale fluctuations in the crystal microboundaries and t is the time taken to traverse the path l in the absence of internal boundaries. The phonon is traversing the free path in time t_1 with average phonon velocity v and scatters from the microboundary. After scattering, it will not travel exactly through the opposite path, but we assume that the phonon apparently reaches the previous position after time t_2 to repeat scattering. Hence, $L(B)$ is always greater than Casimir length L and will offer higher thermal resistance than L and hence the conductivity curve will fall considerably.

At very low temperatures below the conductivity maximum, the scattering due to isotopic point impurities etc. are the next dominant relaxation processes as described by Klemens using a perturbation theoretic technique [2,10]

$$\tau_{\mathrm{D}}^{-1}(\omega) = A\omega^4, \tag{6}$$

where $A = v_0\Gamma/4\pi v^3$ and $\Gamma = \sum_i f_i(1 - (m_i/\bar{m}))^2$. When it is expressed in terms of frequency linewidth, a new form occurs similar to Klemens expression [39,40] with additional information

$$\begin{aligned}\Gamma_k^{\mathrm{D}}(\omega) \;&=\; 8\pi\epsilon(\omega) \sum_{k_1} R(-k, k_1) R^*(-k, k_1) \omega_{k_1} \delta(\omega^2 - \tilde{\omega}_{k_1}^2) \\ &\approx\; A_1\omega^4 + A_2\omega^2.\end{aligned} \tag{7}$$

The coefficients A_1 and A_2 respectively occur due to the change in mass and force constant because of the presence of isotopic defects in the host crystal.

At comparatively higher temperatures near and above the conductivity maximum, the three-phonon processes are observed as the dominant collision processes which are described by [1,11,37]

$$\tau_{\mathrm{ph}}^{-1}(\omega, T) = \begin{cases} B\omega^2 T^3 & \text{at low } T, \\ B'\omega^2 T^2 & \text{at high } T, \end{cases} \tag{8}$$

in which the ω and T dependences have been varied without any sound justification, but, the use of frequency linewidth enables one to fix the problem via the expression [39,40]

$$\begin{aligned}\Gamma_k^{3A}(\omega, T) &= 18\pi\epsilon(x)\sum_{k_1,k_2}|V_3(k_1,k_2,-k)|^2\eta_1[S_{\alpha_1}\epsilon_{\alpha_1}\delta(\epsilon^2-\epsilon_{\alpha_1}^2)\\ &\quad\times S_{\beta_2}\epsilon_{\beta_2}\delta(\epsilon^2-\epsilon_{\beta_2}^2)\\ &\approx \lambda V\omega_k^2\theta(\omega_L-\omega_k)/16\pi N\beta\hbar a_0 v_p^2\\ &\approx B\omega^2 T \qquad (9)\\ \Gamma_k^{4A}(\omega, T) &= \left(\frac{\hbar}{48M}\right)\left(\frac{a_0\hbar\eta\phi^{IV}V}{4\pi^2\beta\phi^{II}v^3}\right)^2(\tilde{\omega}_k^2+\tilde{\omega}_{k_1}^2-\tilde{\omega}_{k+k_1}^2)\\ &\approx B_H\omega^2T^2, \qquad (10)\end{aligned}$$

where λ is a dimensionless quantity and $\omega_k = \omega_L \sin(\pi k a_0)$ which shows the exact frequency and temperature dependence. At conductivity maximum, the Umklapp processes contribute more effectively compared to normal processes. The relaxation time for normal three-phonon processes is taken to be proportional to $(\omega^2T^3)^{-1}$ while for the Umklapp processes the relaxation time is proportional to $(e^{\theta/aT}\omega^2T^3)^{-1}$ [6]. The impurity and anharmonicity modes give rise to the interference scattering of phonons of localized and anharmonic fields. This scattering dominates near and above the thermal conductivity maximum where conventional impurity scattering starts losing its influence. The phonon linewidth for this scattering is given by [40]

$$\begin{aligned}\Gamma_k^{3D}(\omega, T) &= 16\sum_{k_1}|C(-k,k_1)|^2\epsilon^{-2}(\Gamma_{3p}^{(p)})_{k\to k_1}\\ &\approx (3\lambda V\mu_{-2}/\pi\beta\hbar a_0)[(M_0^2c(1-c)/4N\mu V)^2]\omega_k^4\\ &\approx D\omega^4T. \qquad (11)\end{aligned}$$

Here μ_{-2} is the second negative moment.

The possibility of electrons carrying heat as well as acting as scatterers was discussed by Ziman [44] by considering the parabolic bands and derived the relaxation time as

$$\tau_{eph}^{-1} = DT\ln\left(\frac{1+\exp(\eta^*-N/T-PTx^2+x/2)}{1+\exp(\eta^*-N/T-PTx^2-x/2)}\right). \qquad (12)$$

But on the basis of quantum dynamical approach, the linewidth for electron–phonon collision processes can be described in the form

$$\Gamma_{eph}(\omega, T) = \frac{2g^2}{\pi}\frac{(\hbar^2\omega^2k_B^{-4}+4g^2\coth(\beta\hbar\omega/2))}{(e^{\beta\hbar\omega/2}+1)}, \qquad (13)$$

where g is the electron–phonon coupling constant.

The results of Pohl and Walker [45,46] on resonance scattering mechanisms indicate that the dips present in the data at temperatures just above the maximum in κ are also associated with impurities. In fact, if one examines the older data on Si [47], a similar dip can be noted. It was only when the oxygen was removed from the Si that the dip disappeared from the data and the isotope scattering could be used to obtain a good fit.

Table 1. Constants and parameters used in the analysis of thermal conductivity of GaAs.

Samples	$L(B)$ (cm)	θ_D (K)	A_1 (s^3) $\times 10^{-44}$	B ($s\ K^{-1}$) $\times 10^{-23}$	D ($s^3\ K^{-1}$) $\times 10^{-44}$	v ($cm\ s^{-1}$) $\times 10^5$
I	0.0070	345	4.710	3.630	0.76	2.54
II	0.00021	345	0.00121	1.0110	0.69	2.54
III	0.00041	345	0.00015	4.0025	0.67	2.54

Data which can be characterized as having a bump or dip can often be fitted by using a resonance-type relaxation time of the form first used by Pohl [45,48] which is given by

$$\tau_R^{-1}(\omega, T) = \frac{R\omega^2 T^n}{[(\omega_0^2 - \omega^2)^2 + (\Omega/\pi)^2\omega_0^2\omega^2]}, \tag{14}$$

where R is a proportionality constant containing the concentration of impurities causing the resonance scattering, ω_0 is the resonance frequency, $n = 0$ (for polyatomic impurity centres [49]) and Ω describes the damping of the resonance.

3. Analysis of thermal conductivity

In order to examine this model, we have taken the experimental data of Carlson *et al* [50] and Blakemore [51] for samples I, II and III (given in table 1), respectively. The lattice thermal conductivity of GaAs has been analysed by numerically integrating eq. (1) after using various constants and parameters furnished in table 1. It is observed that at low temperatures, below the thermal conductivity maximum, major contribution to thermal

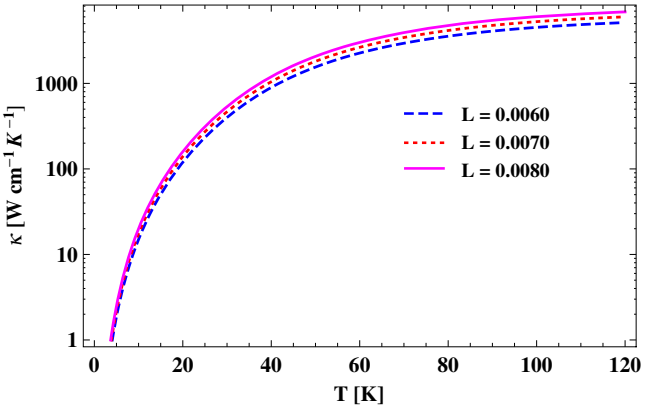

Figure 1. Effect of the scattering length of combined boundaries on lattice thermal conductivity.

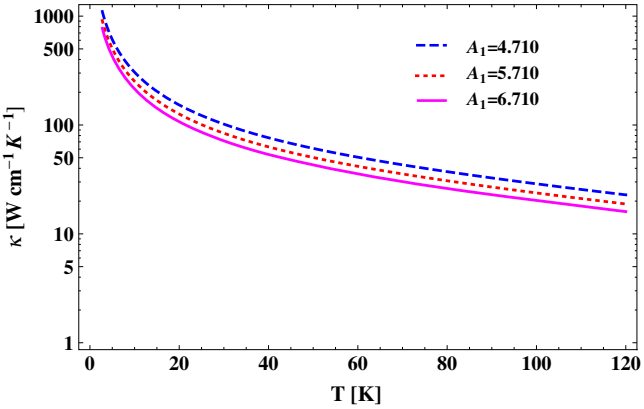

Figure 2. Variation of conductivity at different impurity concentrations.

conductivity comes from the combined boundary scattering while the point defect scattering invokes at relatively elevated temperatures. Figure 1 shows that the internal boundary parameter shows its high sensitivity below 10 K. The experimental results of Carlson *et al* [50] revealed that the samples of GaAs are either of high purity ($\leq 10^{16}$) or have low impurity concentration ($\approx 10^{18}$). For such samples, at very low temperatures, the impurity scattering cannot offer sufficient thermal resistance to reduce the thermal conductivity compared to that offered by the boundary scattering. This is also due to the fact that at very low temperatures the low-frequency phonons are excited which get scattered from crystal boundaries and the probability of their scattering from low concentration defects is meagre. But as the temperature rises, higher and higher frequency phonons with comparatively lower wavelengths are excited and start scattering from impurity centres and other phonons giving rise to defect scattering and phonon–phonon scattering events. A marked decrease in thermal conductivity is observed at temperatures above the thermal

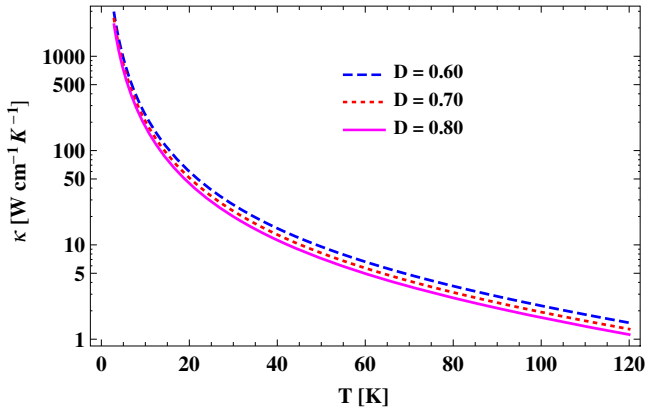

Figure 3. Influence of interference scattering on thermal conductivity.

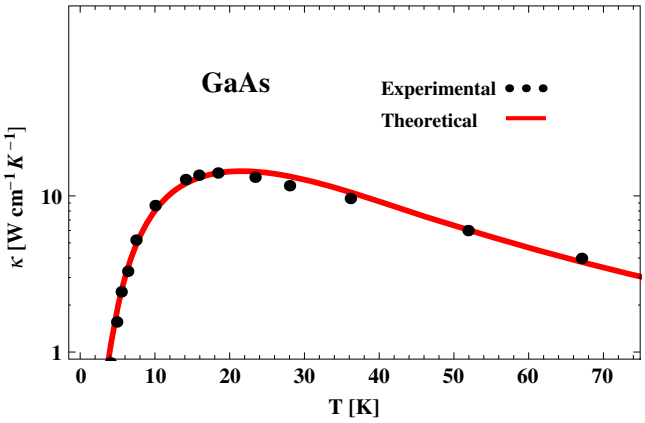

Figure 4. Analysis of thermal conductivity of GaAs (sample I).

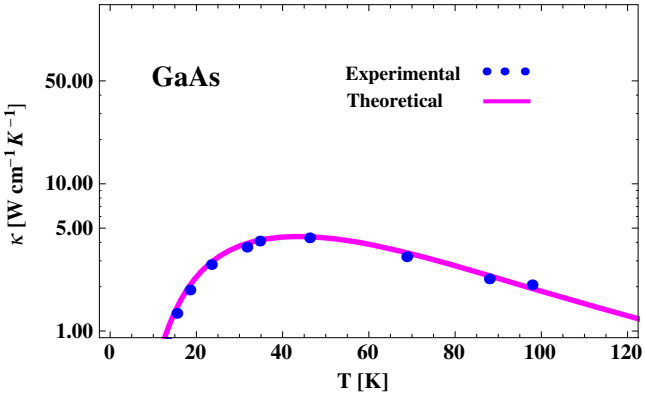

Figure 5. Analysis of thermal conductivity of GaAs (sample II).

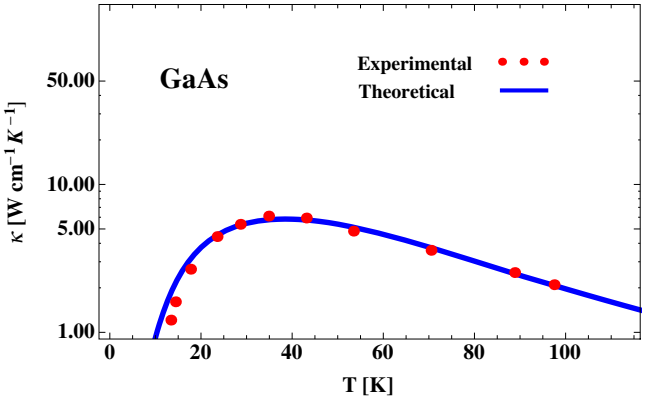

Figure 6. Analysis of thermal conductivity of GaAs (sample III).

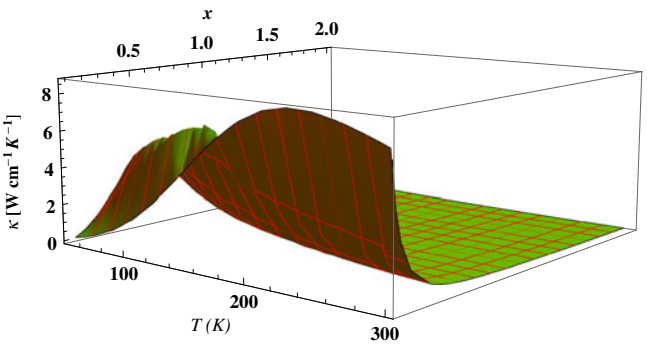

Figure 7. Variation of thermal conductivity function with x and T.

conductivity maximum. In the vicinity of maximum region the anharmonic phonons start interacting with the phonons of localized fields giving rise to interference scattering. The variation of impurity scattering contribution to thermal conductivity is depicted in figure 2 and that of the interference scattering is shown in figure 3. The resonance and electron–phonon scattering events are also found effective in this region but we have not considered them in the present analysis. The phonon–phonon and interference scattering processes dominate in the close vicinity of thermal conductivity maximum and above. The present calculations depicted in figures 4–6 show an excellent agreement with experiments. The variation of thermal conductivity function $(k_{\mathrm{B}}/2\pi^2 v)(k_{\mathrm{B}}T/\hbar)^3[x^4 e^x/\Gamma(x, T)(e^x - 1)^2]$ with reduced frequency $x = (\hbar\omega/k_B T)$ and temperature T dipicted in figure 7 reveals that $\Gamma(x, T)$ is a very sensitive quantity to frequency and/or temperature variations. The sensitivity of thermal conductivity function further increases if the electron–phonon interaction $\Gamma_{\mathrm{eph}}(\omega, T)$ is also included in $\Gamma(\omega, T)$.

4. Conclusions

From the present work it is emerged that the thermal conductivity of GaAs–type crystals can be successfully studied with the help of present formulation which includes the replacement of phonon relaxation times by phonon linewidths. Also, it has been reported that, the phonon linewidth provides a detailed scattering mechanism for the crystalline solids and is a very sensitive quantity for studying thermal transport in semiconductors. The objections and deficiencies occurred in Callaway model have been removed with the help of this formulation and the theory is equally applicable to analyse the thermal conductivity data of other crystals.

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