Additivity of relaxation times and thermal conductivity of nonmetals

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(Received 1 February 1979; revised manuscript received 23 July 1980)

The additivity of the inverse of the relaxation times as assumed by Callaway is observed to be valid to lowest approximation; otherwise, interference terms like Mattheissen's deviation term should be considered. It is also observed that the phonon-scattering processes due to lattice defects and chemical impurities can be included in an approximation with the phonon boundary scattering to analyze the phonon conductivity results at low temperatures.

I. INTRODUCTION

Any deviation from the perfection of crystal leads[1] to a reduction in the phonon conductivity. By a perfect crystal we mean an harmonic crystal whose elementary excitations (phonons[2]) contribute to heat transport. Peierls[3] has shown that for an infinite and perfect crystal, the thermal resistance decreases very rapidly with a decrease in temperature. For a finite crystal, there is a temperature below which the free path of elastic waves becomes comparable to the dimension of the crystal. In this situation, the phonon gets scattered by the boundary of the crystal, reducing the thermal conductivity to a finite value. In these temperature limits, the temperature dependence of the transferred heat mainly depends upon the temperature dependence of the phonon heat capacity, whereas the interaction between elastic waves can be neglected. Casimir[4] has calculated the free path of phonons as

$$\tau_b^{-1}=C_\lambda/L\,, \tag{1}$$

where C_λ is the phonon velocity with λ as polarization index and L is known as Casimir length depending upon the dimension of the crystal. The very presence of temperature dependence[5] introduces deviationlike anharmonicity and nonadiabatic approximation resulting in phonon-phonon interactions and electron-phonon interactions. This endows the bare phonon with a certain lifetime and phonon width. Thermal conductivity analysis shows that at low temperatures the majority of phonons are hardly limited by phonon-phonon scattering, but as the temperature increases it dominates over the phonon boundary scattering. The relaxation times for the different phonon-scattering processes[6] other than phonon boundary scattering are always defined in terms of a transition probability between different states. Herring[7] has therefore analyzed the justification for combining the phonon boundary scattering with other phonon scattering processes to calculate the phonon conductivity and suggested that the phonon conductivity can be expressed as

$$K(T)=(\text{const})_{(T)}+\frac{C_\lambda^2 k_B}{8\pi^3}\int \tau(q)q^2dq\cos^2\theta\,d\Omega\,, \tag{2}$$

where θ is the angle between the phonon group velocity and the phonon wave vector $\vec{q}$. The constant term, which is a function of temperature, corresponds to the phonon conductivity contribution due to size dependence and the integral term combines the contribution due to other phonon scattering processes. Callaway[8] has very successfully calculated the phonon conductivity at low temperatures by

$$K(T)=\frac{1}{2\pi^2 C_\lambda}\int_0^{\omega_D}\tau(\omega_{q\lambda})\frac{\hbar^2\omega_{q\lambda}^4}{k_B T^2}\times\frac{e^{\hbar\omega_{q\lambda}/k_B T}}{(e^{\hbar\omega_{q\lambda}/k_B T}-1)^2}d\omega_{q\lambda}\,, \tag{3}$$

where $\omega_{q\lambda}$ is the phonon frequency. Callaway has, however, assumed (1) the Debye phonon spectrum, (2) no distinction between phonon polarization, and (3) additivity of inverse of the relaxation times. Considering the additivity law, one can define

$$\tau_c^{-1}(\omega_{q\lambda})=\tau_b^{-1}+\tau_I^{-1}(\omega_{q\lambda})+\tau_{pp}^{-1}(\omega_{q\lambda})\,. \tag{4}$$

Knowing that the different phonon scattering processes except boundary scattering depend upon the transition probabilities, we can express the combined relaxation time as[9]

$$\tau_c^{-1}(\omega_{q\lambda})=\frac{C_\lambda}{L}+\sum_i\tau_i^{-1}(\omega_{q\lambda})\,. \tag{4'}$$

Substituting Eq. (4′) into Eq. (3), we find that the Callaway expression reduces to Herring's expression. Casimir has shown that, neglecting phonon-phonon interactions, the combined relaxation time at very low temperature is the first term of Eq. (4′). Many corrections[10-14] have been suggested to improve Callaway's approximation by considering the real situation. These have resulted in the con-

sideration of nonlinear phonon dispersion relations and consideration of separate contributions of longitudinal and transverse phonons to the phonon conductivity. No calculations have been made to study the validity of the additivity of the inverse of the relaxation times as assumed by Callaway. If one attempts to calculate the phonon Boltzmann equation with the boundary conditions applicable to the real situation, the calculations can be done at very low temperatures where phonon-phonon interactions can be neglected, as done by Kazakov and Nageav[15] and Erdös.[16] In another calculation, Kazakov and Nageav[17] have considered the partial reflections at the boundary of the crystal. This results in a parameter F connected with the relaxation time of phonon boundary scattering as

$$\tau_b^{-1}=\frac{C_\lambda}{FL}\,. \tag{5}$$

The parameter F is a measure of acoustic mismatch and has previously been used by Holland[10] and Bhanadari and Verma.[18] These calculations are valid for a single crystallite. Recently there were results which gave evidence of the interface thermal resistance known as Kapitza resistance. We have not considered the effect of this Kapitza resistance[19] in our calculations.

Phonon spectroscopy (phonon conductivity) has now been widely used to study the effect of the presence of impurity and lattice defects in the crystal.[1] The reduction in the phonon conductivity is sometimes followed by indentations or dips in the phonon conductivity curves. Maradudin therefore calculated the phonon conductivity[20] of an isotopically disordered lattice by a more powerful approach suggested by Kubo.[21] Recently we have extended previous calculations[22] to calculate the phonon conductivity of anharmonic crystals containing impurities.[23] Kubo's approach, however, neglects the phonon boundary scattering which can be combined as shown by Eq. (4′).

In the present work, we generalize the previous calculations by including the electron-phonon interaction, which is very important in semiconductors and metals. The calculations show that (1) the additivity of the inverse of the relaxation times is valid to a lowest approximation and (2) the relaxation time due to different phonon scattering processes due to lattice defects and chemical impurities can be combined with the phonon boundary scattering relaxation time as an approximation. We therefore modify our previous results[24,25] of low-temperature thermal conductivity of n-Ge considering the fact that the phonon frequency is changed due to interactions by lattice defects and chemical impurities with phonons. We observe that the deviation between theory and experiment obtained previously can be explained by considering the interference term.

II. KUBO'S RELATION AND GREEN'S FUNCTIONS

Kubo's formula for calculating the phonon conductivity[21] can be written as

$$K(T)=\lim_{\epsilon\to 0+}\frac{k_B\beta}{3\Omega}\int_0^\infty dt\,e^{-\epsilon t}\times\int_0^\beta ds\langle Q(0)Q(t+i\hbar s)\rangle\,, \tag{6}$$

where $\beta=(k_BT)^{-1}$, k_B is the Boltzmann factor, Ω is the volume of the crystal, s is the parameter in (energy)$^{-1}$ units with $\hbar$ being the universal Planck (reduced) constant. $Q(t)$ is defined as phonon flux operator and within harmonic approximation; it can be written as

$$\vec{Q}(t)=\sum_{\vec{q}\lambda}\hbar\omega_{\vec{q}\lambda}N_{\vec{q}\lambda}(t)\vec{v}_{\vec{q}\lambda}\,, \tag{7}$$

where $\omega_{\vec{q}\lambda}$ is the phonon frequency with wave vector $\vec{q}$ and polarization index λ. $\vec{v}_{\vec{q}\lambda}$ is the phonon group velocity and $N_{\vec{q}\lambda}$ is the phonon density operator and is written as

$$N_{\vec{q}\lambda}=a^\dagger_{\vec{q}\lambda}a_{\vec{q}\lambda}\,. \tag{8}$$

Substituting Eqs. (7) and (8) into Eq. (6), we obtain the thermal conductivity

$$K(T)=\lim_{\epsilon\to 0+}\frac{\hbar^2k_B\beta}{3}\sum_{\vec{q}\lambda}\sum_{\vec{q}'\lambda'}\omega_{\vec{q}\lambda}\omega_{\vec{q}'\lambda}\vec{v}_{\vec{q}\lambda}\vec{v}_{\vec{q}'\lambda'}\int_0^\infty dt\,e^{-\epsilon t}\int_0^\beta ds\langle a^\dagger_{\vec{q}\lambda}(0)a_{\vec{q}\lambda}(0)a^\dagger_{\vec{q}\lambda}(t+i\hbar s)a_{\vec{q}\lambda}(t+i\hbar s)\rangle\,. \tag{9}$$

The correlation function can be calculated by several techniques. Zubarev's double-time Green's functions[26] are, however, widely used to calculate the thermodynamic properties of the crystals. The correlation functions are related to the corresponding Green's functions as

$$\langle a^\dagger_{\vec{q}'\lambda'}(0)a_{\vec{q}\lambda}(t)\rangle=\int_{-\infty}^{\infty}d\omega\,e^{-i\omega t}\frac{[G_{\vec{q}\lambda\vec{q}'\lambda'}(\omega+i\epsilon)-G_{\vec{q}\lambda\vec{q}'\lambda'}(\omega-i\epsilon)]}{e^{-\beta\hbar\omega}-1}\,. \tag{10}$$

However, the correlation function in Eq. (9) can be simplified by using the following simple decoupling scheme:

$$\langle a^{\dagger}_{\vec{q}\lambda}(0)a_{\vec{q}\lambda}(0)a^{\dagger}_{\vec{q}\lambda}(t+i\hbar s)a_{\vec{q}\lambda}(t+i\hbar s)\rangle=\langle a^{\dagger}_{\vec{q}\lambda}(0)a_{\vec{q}\lambda}(t+i\hbar s)\rangle\langle a_{\vec{q}\lambda}(0)a^{\dagger}_{\vec{q}\lambda}(t+i\hbar s)\rangle\,. \tag{11}$$

In Eq. (10), we define the Green's functions

$$G_{\vec{q}\lambda\vec{q}'\lambda'}(t)=\langle\langle a_{\vec{q}\lambda}(t)\,|\,a^{\dagger}_{\vec{q}'\lambda'}(0)\rangle\rangle=-i\Theta(t)\langle[a_{\vec{q}\lambda}(t),a^{\dagger}_{\vec{q}'\lambda'}(0)]\rangle\,. \tag{12}$$

The solution of the Green's function can be obtained by writing down the equation of motion of the Green's function as

$$\omega\langle\langle a_{\vec{q}\lambda}(t)\,|\,a^{\dagger}_{\vec{q}'\lambda'}(0)\rangle\rangle_{\omega}=\frac{1}{2\pi}\langle[a_{\vec{q}\lambda},a^{\dagger}_{\vec{q}'\lambda'}]_{-}\rangle+\left\langle\!\!\left\langle\frac{1}{\hbar}[a_{\vec{q}\lambda}(t),H(t)]_{-}\,\middle|\,a^{\dagger}_{\vec{q}'\lambda'}(0)\right\rangle\!\!\right\rangle_{\omega}\,. \tag{13}$$

Here H is the Hamiltonian of the system. The generalized Hamiltonian can be expressed as

$$\begin{aligned}H=&\sum_{\vec{q}\lambda}\hbar\omega_{\vec{q}\lambda}(a^{\dagger}_{\vec{q}\lambda}a_{\vec{q}\lambda}+\tfrac{1}{2})+\sum_{\substack{\vec{q}_1\vec{q}_2\vec{q}_3\\ \lambda_1\lambda_2\lambda_3}}\hbar V^{(3)}(\vec{q}_1\lambda_1,\vec{q}_2\lambda_2,\vec{q}_3\lambda_3)A_{\vec{q}_1\lambda_1}A_{\vec{q}_2\lambda_2}A_{\vec{q}_3\lambda_3}\\&+\sum_{\substack{\vec{q}_1\vec{q}_2\vec{q}_3\vec{q}_4\\ \lambda_1\lambda_2\lambda_3\lambda_4}}\hbar V^{(4)}(\vec{q}_1\lambda_1,\vec{q}_2\lambda_2,\vec{q}_3\lambda_3,\vec{q}_4\lambda_4)A_{\vec{q}_1\lambda_1}A_{\vec{q}_2\lambda_2}A_{\vec{q}_3\lambda_3}A_{\vec{q}_4\lambda_4}-\sum_{\substack{\vec{q}_1\vec{q}_2\\ \lambda_1\lambda_2}}\hbar C(q_1\lambda_1,q_2\lambda_2)B_{\vec{q}_1\lambda_1}B_{\vec{q}_2\lambda_2}\\&+\sum_{\vec{k}}E_k C^{\dagger}_{k}C_{k}+\sum_{\vec{k}\vec{q}\lambda}F_q C^{\dagger}_{\vec{k}}C_{\vec{k}-\vec{q}}(a_{\vec{q}\lambda}+a^{\dagger}_{-\vec{q}\lambda})\,,\end{aligned} \tag{14}$$

where $a_{\vec{q}\lambda}$ and $a^{\dagger}_{\vec{q}\lambda}$ are the phonon annihilation and creation operators, respectively, with phonon wave vector q and polarization index λ, and obey the following commutation relations:

$$[a_{\vec{q}\lambda},a^{\dagger}_{\vec{q}'\lambda'}]_{-}=\delta_{\vec{q}\vec{q}'}\delta_{\lambda\lambda'},\quad [a_{\vec{q}\lambda},a_{\vec{q}'\lambda'}]_{-}=0=[a^{\dagger}_{\vec{q}\lambda},a^{\dagger}_{\vec{q}'\lambda'}]_{-}\,. \tag{15}$$

We also substitute

$$A_{\vec{q}\lambda}=A^{\dagger}_{-\vec{q}\lambda}=(a_{\vec{q}\lambda}+a^{\dagger}_{-\vec{q}\lambda}),\quad B_{\vec{q}\lambda}=-B^{\dagger}_{-\vec{q}\lambda}=(a^{\dagger}_{-\vec{q}\lambda}-a_{\vec{q}\lambda})\,, \tag{16}$$

obeying the commutation relations

$$[A_{\vec{q}\lambda},A_{\vec{q}'\lambda'}]_{-}=0,\quad [A_{\vec{q}\lambda},B_{\vec{q}'\lambda'}]_{-}=-2\delta_{\vec{q}\vec{q}'}\delta_{\lambda\lambda'}\,. \tag{17}$$

The different coupling coefficients are expressed as

$$V^{(3)}(\vec{q}_1\lambda_1,\vec{q}_2\lambda_2,\vec{q}_3\lambda_3)=\frac{\hbar^{1/2}}{6(2)^{3/2}N^{1/2}(\omega_{q_1\lambda_1}\omega_{q_2\lambda_2}\omega_{q_3\lambda_3})^{1/2}}\phi(q_1\lambda_1,q_2\lambda_2,q_3\lambda_3)\Delta(\vec{q}_1+\vec{q}_2+\vec{q}_3)\,, \tag{18}$$

$$V^{(4)}(\vec{q}_1\lambda_1,\vec{q}_2\lambda_2,\vec{q}_3\lambda_3,\vec{q}_4\lambda_4)=\frac{\hbar^{1/2}}{96N(\omega_{q_1\lambda_1}\omega_{q_2\lambda_2}\omega_{q_3\lambda_3}\omega_{q_4\lambda_4})^{1/2}}\phi(q_1\lambda_1,q_2\lambda_2,q_3\lambda_3,q_4\lambda_4)\Delta(\vec{q}_1+\vec{q}_2+\vec{q}_3+\vec{q}_4)\,, \tag{19}$$

$$C(q_1\lambda_1,q_2\lambda_2)=\frac{1}{2\mu}\frac{M_0}{2N}(\omega_{q_1\lambda_1}\omega_{q_2\lambda_2})^{1/2}(\vec{e}_{\vec{q}_1\lambda_1}\cdot\vec{e}_{\vec{q}_2\lambda_2})\left(\sum_{1}^{N}fe^{i(\vec{q}_1+\vec{q}_2)\cdot\vec{R}(i)}-\sum_{i}^{n}e^{i(\vec{q}_1+\vec{q}_2)\cdot\vec{R}(i)}\right)\,, \tag{20}$$

with

$$\frac{1}{M_0}=\frac{f}{M'}+\frac{1-f}{M},\quad \mu=\frac{MM'}{M-M'}\,. \tag{21}$$

Here M and M' are the masses of host and impurity atoms, respectively. The electron-phonon coupling coefficient F_q is defined as

$$F_q=F(\omega_{q\lambda}/\Omega)^{1/2}\,. \tag{22}$$

F is the coupling constant depending upon the electron-phonon matrix elements. $C_{\vec{k}}$ and $C^{\dagger}_{\vec{k}}$ are the electron annihilation and creation operators, respectively, with wave vector $\vec{k}$ and energy $E_k=k^2/2m-\xi$. m is the electron mass and ξ is the chemical potential.

Substituting Eq. (14) into Eq. (13), the equation of motion for the Green's function $G_{\vec{q}\lambda\vec{q}'\lambda'}(\omega)$ is written as

$$\omega G_{\vec{q}\lambda\vec{q}'\lambda'}(\omega)=\frac{1}{2\pi}\delta_{\vec{q}\,\vec{q}'}\delta_{\lambda\lambda'}+3\sum_{\vec{q}_1\lambda_1\vec{q}_2\lambda_2}V^{(3)}(-\vec{q}\lambda,\vec{q}_1\lambda_1,\vec{q}_2\lambda_2)\langle\langle A_{\vec{q}_1\lambda_1}A_{\vec{q}_2\lambda_2}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}$$
$$+4\sum_{\vec{q}_1\lambda_1\vec{q}_2\lambda_2\vec{q}_3\lambda_3}V^{(4)}(-\vec{q}\lambda,\vec{q}_1\lambda_1,\vec{q}_2\lambda_2,\vec{q}_3\lambda_3)\langle\langle A_{\vec{q}_1\lambda_1}A_{\vec{q}_2\lambda_2}A_{\vec{q}_3\lambda_3}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}$$
$$+2\sum_{\vec{q}_1\lambda_1}C(-\vec{q}\lambda,\vec{q}_1\lambda_1)\langle\langle B_{\vec{q}_1\lambda_1}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}+\omega_{q\lambda}\langle\langle a_{\vec{q}\lambda}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}+\sum_{\vec{k}}F_q\langle\langle C^{\dagger}_{\vec{k}}C_{\vec{k}+\vec{q}}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}\,. \tag{23}$$

Similarly we can obtain the equations of motion for the Green's function, $\langle\langle a^{\dagger}_{-\vec{q}\lambda}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}$, as

$$\omega\langle\langle a^{\dagger}_{-\vec{q}\lambda}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}=-\omega_{q\lambda}\langle\langle a^{\dagger}_{-\vec{q}\lambda}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}-\sum_{k}F_q\langle\langle C^{\dagger}_{\vec{k}}C_{\vec{k}+\vec{q}}\,|\,a^{\dagger}_{\vec{q}'\lambda'}\rangle\rangle_{\omega}$$
$$-3\sum_{q_1q_2\lambda_1\lambda_2}V^{(3)}(q_1\lambda_1,q_2\lambda_2,-q\lambda)\langle\langle A_{q_1\lambda_1}A_{q_2\lambda_2}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$-4\sum_{q_1q_2q_3\lambda_1\lambda_2\lambda_3}V^{(4)}(q_1\lambda_1,q_2\lambda_2,q_3\lambda_3,-q\lambda)\langle\langle A_{q_1\lambda_1}A_{q_2\lambda_2}A_{q_3\lambda_3}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)\langle\langle B_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,. \tag{24}$$

Equations (23) and (24) can be simplified only after writing the equation of motion of higher-electron Green's functions as written below:

$$\omega\langle\langle C^{\dagger}_{k}C_{k+q}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=(E_{k+q}-E_k)\langle\langle C^{\dagger}_{k}C_{k+q}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$+\sum_{q_1\lambda_1}F_{q_1}(\langle\langle C_kC_{k-q_1+q}A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}-\langle\langle C_{k+q_1}C_{k+q}A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega})\,. \tag{25}$$

We can decouple higher phonon and electron Green's functions by using the simple decoupling scheme

$$\langle\langle A_{q_1\lambda_1}A_{q_2\lambda_2}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=\frac{1}{\hbar}F(q_1\lambda_1,q_2\lambda_2,\omega)\sum_{q\lambda}V^{(3)}(-q_1\lambda_1,-q_2\lambda_2,q\lambda)\langle\langle a_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,, \tag{26}$$

with

$$F(q_1\lambda_1,q_2\lambda_2,\omega)=6(N_{q_1\lambda_1}+N_{q_2\lambda_2})\frac{\omega_{q_1\lambda_1}+\omega_{q_2\lambda_2}}{\omega^2-(\omega_{q_1\lambda_1}+\omega_{q_2\lambda_2})^2}+6(N_{q_2\lambda_2}-N_{q_1\lambda_1})\frac{\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2}}{\omega^2-(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2})^2}$$
$$+6(N'_{q_1\lambda_1}+N'_{q_2\lambda_2})\left[\frac{\omega}{\omega^2-(\omega_{q_1\lambda_1}+\omega_{q_2\lambda_2})^2}-\frac{\omega}{\omega^2-(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2})^2}\right],$$
$$\langle\langle A_{q_1\lambda_1}A_{q_2\lambda_2}A_{q_3\lambda_3}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\Rightarrow N_{q_2\lambda_2}\langle\langle a_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,, \tag{27}$$

and

$$\langle\langle C^{\dagger}_{k}C_{k-q_1-q}A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\Rightarrow\delta_{qq_1}\langle n_k\rangle\langle\langle A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,, \tag{28}$$
$$\langle\langle C^{\dagger}_{k+q_1}C_{k+q}A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\Rightarrow\delta_{qq_1}\langle n_{k+q}\rangle\langle\langle A_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,. \tag{29}$$

Here we set:

$$N_{q\lambda}=\langle A_{q\lambda}A^{\dagger}_{q\lambda}\rangle,\quad N_{q_2\lambda_2}=\langle A^{\dagger}_{q_2\lambda_2}B_{q_2\lambda_2}\rangle,\quad N'_{q_1\lambda_1}=\langle B_{q_1\lambda_1}A^{\dagger}_{q_1\lambda_1}\rangle\,. \tag{30}$$

After decoupling higher-order Green's functions and doing simple mathematics, we find that

$$(\omega-\omega_{q\lambda}-M_{q\lambda})\langle\langle a_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\sum_{k}|F_q|^2\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}\langle\langle A_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)(\langle\langle a_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}-\langle\langle a^{\dagger}_{-q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega})\,. \tag{31}$$

and

$$(\omega+\omega_{q\lambda}+M_{q\lambda})\langle\langle a^{\dagger}_{-q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=-\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}\langle\langle A_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)(\langle\langle a_{q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}-\langle\langle a^{\dagger}_{-q_1\lambda_1}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}), \tag{32}$$

where

$$M_{q\lambda}=3\sum_{q_1q_2\lambda_1\lambda_2}|V^{(3)}(q_1\lambda_1,q_2\lambda_2,-q\lambda)|^{2}F(q_1\lambda_1,q_2\lambda_2,\omega)+12\sum_{q_1\lambda_1}V^{(4)}(q_1\lambda_1,q_2\lambda_2,q_3\lambda_3,-q\lambda)N_{q_1\lambda_1} \tag{33}$$

and

$$n_{k}=\langle C^{\dagger}_{k}C_{k}\rangle\,. \tag{34}$$

Comparing Eqs. (31) and (32), we get

$$(\omega-\omega_{q\lambda}-M_{q\lambda})\langle\langle a_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+2\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}-E_{k+q}}\langle\langle A_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}$$
$$+(\omega+\omega_{q\lambda}+M_{q\lambda})\langle\langle a^{\dagger}_{-q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}\,. \tag{35}$$

Setting

$$\Omega_{q\lambda}=\omega_{q\lambda}+M_{q\lambda}+2\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}\,,$$
$$G^{\lambda\lambda'}_{qq'}(\omega)=\langle\langle a_{q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=G^{(1)}(\omega)\,, \tag{36}$$
$$G^{\lambda\lambda'}_{-qq'}(\omega)=\langle\langle a^{\dagger}_{-q\lambda}\,|\,a^{\dagger}_{q'\lambda'}\rangle\rangle_{\omega}=G^{(2)}(\omega)\,,$$

Eq. (35) reduces to the form

$$(\omega-\Omega_{q\lambda})G^{\lambda\lambda'}_{qq'}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+(\omega+\Omega_{q\lambda})G^{\lambda\lambda'}_{-qq'}(\omega)\,. \tag{35'}$$

Equations (31) and (32) can be rewritten as

$$(\omega-\omega^{(1)}_{q\lambda})G^{\lambda\lambda'}_{qq'}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}G^{\lambda\lambda'}_{-qq'}(\omega)+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)[G^{\lambda_1\lambda'}_{q_1q'}(\omega)-G^{\lambda_1\lambda'}_{-q_1q'}(\omega)]\,, \tag{31'}$$

$$(\omega+\omega^{(1)}_{q\lambda})G^{\lambda\lambda'}_{-qq'}(\omega)=-\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}G^{\lambda\lambda'}_{qq'}(\omega)+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)[G^{\lambda_1\lambda'}_{q_1q'}(\omega)-G^{\lambda_1\lambda'}_{-q_1q'}(\omega)]\,, \tag{32'}$$

with

$$\omega^{(1)}_{q\lambda}=\left(\omega_{q\lambda}+M_{q\lambda}+\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}\right). \tag{37}$$

Substituting Eq. (35′) into Eq. (31′), we get

$$(\omega-\omega^{(1)}_{q\lambda})G^{(1)}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}-E_{k+q}}G^{(2)}(\omega)$$
$$+2\sum_{q_1\lambda_1}C(q_1\lambda_1,-q\lambda)\left(G^{(1)}(\omega)-\frac{\omega-\Omega_{q_1\lambda_1}}{\omega+\Omega_{q_1\lambda_1}}G^{(1)}(\omega)+\frac{1}{2\pi}\frac{\delta_{qq'}\delta_{\lambda\lambda'}}{\omega+\Omega_{q_1\lambda_1}}\right)$$
$$=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\frac{C(q_1\lambda_1,-q\lambda)}{\pi(\omega+\Omega_{q_1\lambda_1})}+4\sum_{q'\lambda'}\frac{C(q_1\lambda_1,-q\lambda)}{(\omega+\Omega_{q_1\lambda_1})}\Omega_{q_1\lambda_1}G^{(1)}(\omega)+\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega-E_{k}+E_{k+q}}G^{(2)}(\omega)\,. \tag{38}$$

Similarly we obtain

$$(\omega+\omega^{(1)}_{q\lambda})G^{(2)}(\omega)=\frac{C(q'\lambda',-q\lambda)}{\pi(\omega+\Omega_{q\lambda})}+4\sum_{q_1\lambda_1}\frac{C(q_1\lambda_1,-q\lambda)}{\omega+\Omega_{q\lambda}}\Omega_{q_1\lambda_1}G^{(1)}(\omega)-\sum_{k}|F_{q}|^{2}\frac{n_{k}-n_{k+q}}{\omega\rho-E_{k}+E_{k+q}}G^{(1)}(\omega)\,. \tag{39}$$

The value of $G^{(1)}(\omega)$ from Eq. (39) can be substituted into Eq. (38) to get

$$(\omega-\omega_{q\lambda}^{(1)})G^{(1)}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\frac{C(q'\lambda',-q\lambda)}{\pi(\omega+\Omega_{q'\lambda'})}+\sum_k|F_q|^2\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}\frac{C(q'\lambda',-q\lambda)}{\pi(\omega+\Omega_{q'\lambda'}}$$

$$+4\sum_{q_1\lambda_1}\frac{C(q_1\lambda_1,-q\lambda)}{\omega+\Omega_{q_1\lambda_1}}\Omega_{q_1\lambda_1}G^{(1)}(\omega)+4\sum_k|F_q|^2\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}\sum_{q_1\lambda_1}\frac{C(q_1\lambda_1,-q\lambda)}{\omega+\Omega_{q_1\lambda_1}}\Omega_{q_1\lambda_1}G^{(1)}(\omega)$$

$$-\sum_{kk'}|F_q|^4\frac{(n_k-n_{k+q})(n_{k'}-n_{k'+q})}{(\omega+\Omega_{q\lambda})(\omega-E_k+E_{k+q})(\omega-E_{k'}+E_{k'+q})}G^{(1)}(\omega)\,. \tag{40}$$

To simplify Eq. (40), we neglect the last term and we can express Eq. (40) as

$$(\omega-\omega_{q\lambda}^{(1)})G^{(1)}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\Delta_1+4\sum_{q_1\lambda_1}\psi\frac{C(q_1\lambda_1,-q\lambda)}{\omega+\Omega_{q_1\lambda_1}}\Omega_{q_1\lambda_1}G_1^{(1)}(\omega)\,, \tag{40'}$$

where

$$\Delta_1=\psi_k\frac{C(q'\lambda',-q\lambda)}{\pi(\omega+\Omega_{q'\lambda'})},\quad \psi_k=\left(\sum_k|F_q|^2\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}+1\right). \tag{41}$$

Iterating Eq. (40′), we get

$$(\omega-\omega_{q\lambda}^{(1)})G^{(1)}(\omega)=\frac{1}{2\pi}\delta_{qq'}\delta_{\lambda\lambda'}+\Delta_{(1)}^{q'\lambda'}+\frac{2\psi C(q\lambda,-q\lambda)\Omega_{q\lambda}}{\pi(\omega-\omega_{q\lambda}^{(1)})(\omega+\Omega_{q\lambda})}+4\sum_{q_1\lambda_1}\frac{\psi C(q_1\lambda_1,-q\lambda)\Delta_{(1)}^{q_1\lambda_1}\Omega_{q_1\lambda_1}}{(\omega-\omega_{q_1\lambda_1}^{(1)})(\omega+\Omega_{q_1\lambda_1})}$$

$$+16\sum_{q_1\lambda_1q_2\lambda_2}\Omega_{q_1\lambda_1}\Omega_{q_2\lambda_2}G_2^{(1)}(\omega)\frac{\psi\psi_1C(q_1\lambda_1,-q\lambda)C(q_2\lambda_2,-q\lambda)}{(\omega+\Omega_{q_1\lambda_1})(\omega-\omega_{q_1\lambda_1}^{(1)})(\omega+\Omega_{q_2\lambda_2})}+\cdots\,. \tag{42}$$

To terminate the series, we find a suitable substitution such as $q_2\lambda_2=q\lambda$. We get the solution of the Green's function as

$$G_{qq'}^{\lambda\lambda'}(\omega)=\frac{(1/2\pi)\delta_{qq'}\delta_{\lambda\lambda'}}{\omega-\omega_{q\lambda}^{(1)}-\Sigma_{q\lambda}}\,, \tag{43}$$

where

$$\Sigma_{q\lambda}=16\sum_{q_1\lambda_1}\frac{C(q_1\lambda_1,-q\lambda)C(q\lambda,-q\lambda)\Omega_{q\lambda}\Omega_{q_1\lambda_1}}{(\omega+\Omega_{q\lambda})(\omega+\Omega_{q_1\lambda_1})(\omega-\omega_{q_1\lambda_1}^{(1)})}\psi\psi_1\,. \tag{44}$$

We neglect other correction terms; we can now explicitly write

$$\omega_{q\lambda}^{(1)}+\Sigma_{q\lambda}=\Delta_{q\lambda}^{(\omega)}\mp i\Gamma_{q\lambda}(\omega)\,, \tag{45}$$

where $\Delta_{q\lambda}(\omega)$ and $\Gamma_{q\lambda}(\omega)$ are the phonon widths and phonon lifetimes which result due to different phonon scattering processes in the presence of lattice defects and chemical impurities. One can show that

$$\Delta_{q\lambda}(\omega)=16\Omega_{q\lambda}P\left(\frac{1}{\omega+\Omega_{q\lambda}}\right)\sum_{q_1\lambda_1}\frac{\Omega_{q_1\lambda_1}}{\omega_{q_1\lambda_1}^{(1)}+\Omega_{q_1\lambda_1}}C(q_1\lambda_1,-q\lambda)C(q\lambda,-q_1\lambda_1)\left[P\left(\frac{1}{\omega-\omega_{q_1\lambda_1}^{(1)}}\right)-P\left(\frac{1}{\omega+\Omega_{q_1\lambda_1}}\right)\right]$$

$$-16\pi^2\Omega_{q\lambda}\delta(\omega+\Omega_{q\lambda})\sum_{q_1\lambda_1}\frac{\Omega_{q_1\lambda_1}}{\omega_{q_1\lambda_1}^{(1)}+\Omega_{q_1\lambda_1}}C(q_1\lambda_1,-q\lambda)C(-q_1\lambda_1,q\lambda)[\delta(\omega-\omega_{q_1\lambda_1}^{(1)})-\delta(\omega+\Omega_{q_1\lambda_1})]\,, \tag{46}$$

$$\Gamma_{q\lambda}(\omega)=16\pi\Omega_{q\lambda}\sum_{q_1\lambda_1}\frac{\Omega_{q_1\lambda_1}}{\omega_{q_1\lambda_1}^{(1)}+\Omega_{q_1\lambda_1}}C(q_1\lambda_1,-q\lambda)C(-q_1\lambda_1,q\lambda)$$

$$\times\left\{\delta(\omega+\Omega_{q\lambda})\left[P\left(\frac{1}{\omega-\omega_{q_1\lambda_1}^{(1)}}\right)-P\left(\frac{1}{\omega+\Omega_{q_1\lambda_1}}\right)\right]\right.$$

$$\left.+P\left(\frac{1}{\omega+\Omega_{q\lambda}}\right)[\delta(\omega-\omega_{q_1\lambda_1}^{(1)})-\delta(\omega+\Omega_{q_1\lambda_1})]\right\}. \tag{47}$$

Using the Green's functions expressed by Eq. (43), we can obtain the correlation function by Eq. (10). Substituting Eq. (10) into Eq. (9), the thermal conductivity can be obtained as

$$K(T)=\lim_{\epsilon\to 0+}\frac{i\hbar k_B\beta}{3\Omega}\sum_{q\lambda q'\lambda'}\omega_{q\lambda}\omega_{q'\lambda'}\vec{v}_{q\lambda}\cdot\vec{v}_{q'\lambda'}\times\int\int d\omega_{q_1\lambda_1}d\omega_{q_2\lambda_2}\times\frac{e^{\beta\hbar\omega_{q_1\lambda_1}}-e^{\beta\hbar\omega_{q_2\lambda_2}}}{(e^{\beta\hbar\omega_{q_1\lambda_1}}-1)(e^{\beta\hbar\omega_{q_2\lambda_2}}-1)(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2})(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2}-i\epsilon)}\times[G^{\lambda_1\lambda'}_{q_1q'}(\omega+i\epsilon)-G^{\lambda_1\lambda'}_{q_1q'}(\omega-i\epsilon)][G^{\lambda_2\lambda'}_{q_2q'}(\omega+i\epsilon)-G^{\lambda_2\lambda'}_{q_2q'}(\omega-i\epsilon)]\,. \tag{48}$$

Following Deo and Behra, Eq. (48) can be further simplified by interchanging $\omega_{q_1\lambda_1}$ with $\omega_{q_2\lambda_2}$ and using the identity

$$[(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2}-i\epsilon)^{-1}-(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2}+i\epsilon)^{-1}]=2\pi i\delta(\omega_{q_1\lambda_1}-\omega_{q_2\lambda_2})\,. \tag{49}$$

Finally we get

$$K(T)=\frac{\hbar^2k_B\beta^2}{3\Omega}\sum_{q\lambda}\omega^2_{q\lambda}v^2_{q\lambda}\int_{-\infty}^{\infty}d\omega\,\frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)^2}\,\frac{\Gamma^2_{q\lambda}(\omega)}{\{[\omega^2-\epsilon_{q\lambda}(\omega)]^2+\Gamma^2_{q\lambda}(\omega)\}^2}\,, \tag{50}$$

where

$$\omega_{q\lambda}+\Delta_{q\lambda}(\omega)=\epsilon_{q\lambda}(\omega)\,. \tag{51}$$

Considering the small values of $\Gamma_{q\lambda}(\omega)$, the integrand is peaked around the value $\omega=\epsilon_{q\lambda}(\omega)$, the thermal conductivity is then approximated as

$$K(T)=\frac{\hbar^2k_B\beta^2}{3\Omega}\sum_{q\lambda}\omega^2_{q\lambda}v^2_{q\lambda}\,\frac{e^{\beta\hbar\epsilon_{q\lambda}}}{(e^{\beta\hbar\epsilon_{q\lambda}}-1)^2}\left(\frac{1}{2\Gamma_{q\lambda}(\omega)}\right). \tag{52}$$

If we assume that

$$\tau_{q\lambda}(\omega)=1/2\Gamma_{q\lambda}(\omega)\,, \tag{53}$$

the thermal conductivity $K(T)$ can be reexpressed as

$$K(T)=\frac{\hbar^2k_B\beta^2}{3\Omega}\sum_{q\lambda}\omega^2_{q\lambda}v^2_{q\lambda}\,\frac{e^{\beta\hbar\epsilon_{q\lambda}}}{(e^{\beta\hbar\epsilon_{q\lambda}}-1)^2}\,\tau_{q\lambda}(\omega)\,, \tag{54}$$

which is the same as that obtained previously by solving the phonon Boltzmann equation (Callaway).

III. RELAXATION TIMES

Equation (53) clearly shows that the thermal conductivity depends upon the relaxation time, which is nothing but the measure of the phonon lifetime as given by

$$2\Gamma_{q\lambda}(\omega)=\tau^{-1}_{q\lambda}(\omega)\,. \tag{53'}$$

From Eq. (45) we observe that $\Gamma_{q\lambda}(\omega)$ is the imaginary part of $\omega^{(1)}_{q\lambda}+\Sigma_{q\lambda}$ and the phonon frequency is finally changed to $(\omega_{q\lambda}+\Delta\omega_{q\lambda})$. We now calculate the shift in the phonon frequency $\Delta\omega_{q\lambda}$ and the phonon lifetime from the self-energy $\Sigma^{(1)}_{q\lambda}$ as

$$\Sigma^{(1)}_{q\lambda}=\Sigma^{\rm anh}_{q\lambda}+\Sigma^{ep}_{q\lambda}+\Sigma^{\rm imp}_{q\lambda}=M_{q\lambda}+\sum_k|F_q|^2\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}+16\sum_{q_1\lambda_1}\psi\psi_1\frac{C(q_1\lambda_1,-q\lambda)C(-q_1\lambda_1,q\lambda)\Omega_{q\lambda}\Omega_{q_1\lambda_1}}{(\omega+\Omega_{q\lambda})(\omega+\Omega_{q_1\lambda_1})(\omega-\omega^{(1)}_{q_1\lambda_1})}\,. \tag{55}$$

To the lowest approximation, i.e., $\psi\cong 1$, if we assume that

$$M_{q\lambda}=\Delta^{\rm anh}_{q\lambda}\mp i\Gamma^{\rm anh}_{q\lambda}\,,$$

$$\sum_k|F^2_q|\,\frac{n_k-n_{k+q}}{\omega-E_k+E_{k+q}}=\Delta^{ep}_{q\lambda}\mp i\Gamma^{ep}_{q\lambda}\,,$$

$$\Sigma^{\rm imp}_{q\lambda}=\Delta^{\rm imp}_{q\lambda}\mp i\Gamma^{\rm imp}_{q\lambda}\,, \tag{56}$$

we find that

$$\Sigma_{q\lambda}^{(1)} = (\Delta_{q\lambda}^{anh} + \Delta_{q\lambda}^{ep} + \Delta_{q\lambda}^{imp}) \mp i(\Gamma_{q\lambda}^{anh} + \Gamma_{q\lambda}^{ep} + \Gamma_{q\lambda}^{imp}) . \tag{57}$$

$\Delta_{q\lambda}^{anh}$, $\Delta_{q\lambda}^{ep}$, $\Gamma_{q\lambda}^{anh}$, and $\Gamma_{q\lambda}^{ep}$ have been previously calculated by many workers; therefore we calculate the phonon width and phonon lifetime in the presence of other phonon scattering processes such as electron-phonon interaction and anharmonic interactions. Equation (46) gives the value of the phonon broadening depending upon the changed frequency $\Omega_{q\lambda}$ or $\omega_{q\lambda}^{(1)}$ and lifetime can be calculated from Eq. (47). However, for simplicity we consider $(\omega_{q\lambda}^{(1)} \sim \Omega_{q\lambda})$

$$\Gamma_{q\lambda}^{imp} = 8\pi\Omega_{q\lambda}\frac{V}{8\pi^3}\int \sin\theta\, d\theta\, d\phi\, |C(-q\lambda, q_1\lambda_1)|^2 q_1^2 dq_1 P\left(\frac{1}{\omega+\Omega_{q\lambda}}\right)\delta(\omega-\Omega_{q_1\lambda_1}) , \tag{58}$$

where θ and ϕ measure the direction of $\vec{q}_1$ with respect to $\vec{q}$. After doing the integration, we find that

$$\Gamma_{q\lambda}^{imp} = \frac{1}{24N\pi C_\lambda^3}\left(\frac{M_0}{\mu}\right)^2 f(1-f)\tilde{\epsilon}_{q\lambda}^4$$

$$= A(1+a)^4\omega_{q\lambda}^4 , \tag{59}$$

where

$$A = \frac{1}{24N\pi C_\lambda^3}\left(\frac{M_0}{\mu}\right)^2 f(1-f) ,$$
$$\tilde{\epsilon}_{q\lambda} = \omega_{q\lambda}(1+a) . \tag{60}$$

Here a is a constant which depends upon the anharmonic constants and electron-phonon interaction constants. Comparing Eqs. (45) and (57), we find that the total lifetime is

$$\Gamma_{q\lambda}^{tot} = \Gamma_{q\lambda}^{anh} + \Gamma_{q\lambda}^{ep} + \Gamma_{q\lambda}^{imp} . \tag{61}$$

Equation (61) can be reexpressed in terms of the relaxation times as

$$(\tau_{q\lambda}^{-1})^{tot} = (\tau_{q\lambda}^{-1})^{anh} + (\tau_{q\lambda}^{-1})^{ep} + (\tau_{q\lambda}^{-1})^{imp} . \tag{61'}$$

It is thus observed that the inverse of the relaxation times should be added to get the effective relaxation time—this is nothing but what had been assumed by Callaway before. However, there is one important difference in not finding the addition of the inverse of the relaxation time for the phonon boundary processes. It should be noted that this difference is completely artificial and results simply because heat transfer across the crystal boundary was not included in the formulation of the problem in our treatment. We can partially consider the effect of the finite size by using it as boundary conditions to calculate the phonon Boltzmann equations as done previously by Erdos, Kazakov and Negeav, and Kumar and Joshi. If we do not assume $\psi \cong 1$, we find that in our calculations we must consider the interference terms such as Mattheissen's deviation term.

IV. MODIFIED CALCULATIONS OF KUMAR AND JOSHI (REF. 23)

Previously Kumar and Joshi have extended the calculations by Kazakov and Nageav to calculate the phonon conductivity of n-Ge at temperatures between 0.5 and 4.2 K. A deviation was observed between experimental and theoretical results above 2 K, which was supposed to be due to the neglect of phonon-phonon interactions. We, in the present calculations, find that the phonon frequency becomes modified due to other scattering processes, such as electron-phonon interaction and phonon-phonon interaction. Within the assumption of the validity of Kazakov and Nageav's calculations, we can recalculate these results by considering the modified relaxation time for mass difference scattering as

$$(\tau_{q\lambda}^{-1})_{imp} = A(1+a)^4\omega_{q\lambda}^4 .$$

Following Kumar and Joshi, we get the phonon conductivity as

$$K(T) = K_0(T)\left[1 - A(1+a)^4\frac{L}{C_\lambda}\left(\frac{k_B T}{\hbar}\right)^4\frac{g_2}{g_1}\right], \tag{62}$$

where

$$K_0(T) = 3\left(\frac{k_B}{2\pi^2 C_\lambda}\right)\left(\frac{k_B T}{\hbar}\right)^3\frac{L}{C_\lambda}g_1 ,$$
$$g_1 = \int_0^\infty \frac{x^3 dx}{e^x - 1} = 6.5 , \tag{63}$$
$$g_2 = \int_0^\infty \frac{x^7 dx}{e^x - 1} = 7.2\times 10^2 .$$

In previous calculations by the author and Joshi, Eq. (62) had been used with $a = 0$ to analyze the phonon conductivity of a sample of Ge. A deviation has been observed between theoretical and experimental results above 2 K, which is now to be analyzed. Using Eq. (62) we have calculated the phonon conductivity for Ge for $a = 0$, whose experimental data has been given by Bird and Pearlmann.[27] Now we take $a = 0.2$ in Eq. (62) and find

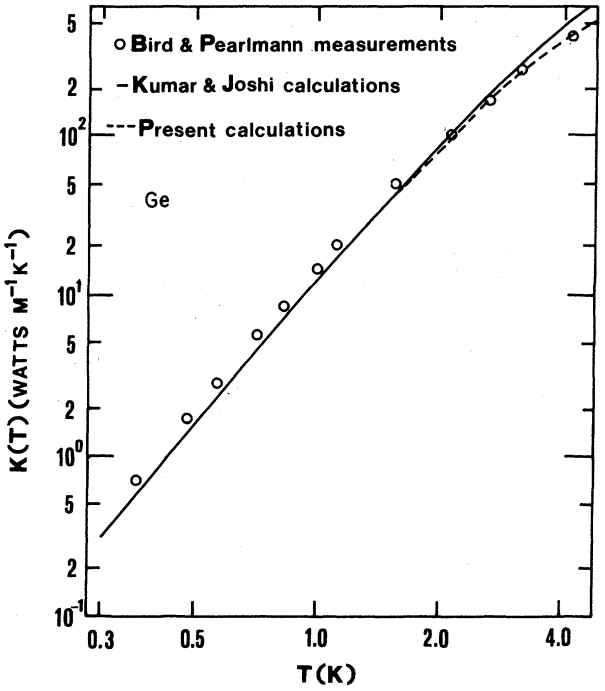

FIG. 1. Phonon conductivity versus temperature curve for Ge with $N_{ex} = 5.4 \times 10^{13}$ cm^{-3} (see Ref. 19). Solid line stands for $a = 0$, and dotted line represents $a = 0.2$.

that the agreement between theory and experiment is very good. This suggests that the deviation was due to the interference of the other scattering processes and hence it is advisable to consider the interference term a for more accurate calculations. Figure 1 shows the agreement between the theoretical calculations and the experimental data.

V. CONCLUSIONS

We find that the additivity of the inverse of the relaxation times is valid to a lowest approximation; otherwise, we must consider an interference term such as Mattheissen's deviation term. It is also observed that one can partially consider the effect of finite size of the crystal through the boundary conditions while calculating the phonon Boltzmann equations as done by Kumar and Joshi instead of adding it to obtain the total relaxation time as done by Callaway.

ACKNOWLEDGMENTS

The author is grateful to Professor Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the International Centre for Theoretical Physics, Trieste, where this work was carried out. Financial support from the Swedish International Development Authority is gratefully acknowledged. He is also grateful to the University Grants Commission, New Delhi, for financial support towards a travel grant. He also gratefully acknowledges the interest of Professor M. P. Tosi and Professor M. M. Joshi in the present work.

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