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ROLE OF THREE PHONON NORMAL PROCESSES IN THE ANALYSIS OF THE THERMAL CONDUCTIVITY OF SOLID HD AT LOW TEMPERATURES

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The role of the three phonon normal processes in the analysis of the lattice thermal conductivity of the solid HD has been studied in the entire temperature range 0.2–4 K. The study is made by calculating the contribution of the correction term due to the three phonon normal processes towards the total lattice thermal conductivity of the pure sample of the solid HD in the temperature range stated above. The study is also made for the different values of the three phonon normal processes scattering relaxation rate. The considerable contribution due to the correction term is found compared to the total phonon conductivity above 1 K.

Inspite of the fact that some refinements [1–5] have been proposed recently, the success of the Callaway [6] theory is excellent for the explanation of the experimental results of the lattice thermal conductivity of insulators at low temperatures. It was Callaway [6], first of all, who distinguished the three phonon normal processes (processes in which momentum is conserved) from the three phonon umklapp processes (those processes in which momentum is not conserved) by introducing the displaced distribution function for it and he obtained an expression for the lattice thermal conductivity of an insulator as a sum of two terms. The first term is due to the combined scattering relaxation rate while the second term is known as the correction term due to the three phonon normal processes which reduces to zero in the absence of the three phonon normal processes. It is usually found that the contribution of the correction term (ΔK) towards the total lattice thermal conductivity is very small [7–13] compared to the contribution due to the first term at low as well as at high temperatures. Solid He [14] and LiF [15] are exception to this.

Recently, Constable and Gaines [16, 17] measured the lattice thermal conductivity of the solid HD at temperatures below 4 K and they tried to explain their measurements on the basis of the contribution due to the correction term (ΔK) alone which creates an interest to have a study of the contribution of the correction term (ΔK) due to the three phonon normal processes towards the total lattice thermal conductivity of the solid HD in the frame of the complete Callaway [6] expression. The aim of the present work is to study the role of the three phonon normal processes in the analysis of the lattice thermal conductivity of the solid HD by calculating the contribution due to the correction term in the entire temperature range 0.2–4 K. The considerable contribution of ΔK towards the total lattice thermal conductivity of the solid HD is found above 1 K which is similar to the previous reports for the solid He [14] and LiF [15]. To study the role of the three phonon normal processes in more detail, ΔK has also been calculated by expressing the three phonon normal processes scattering relaxation rate as a fraction of the three phonon umklapp processes scattering relaxation rate as well as a fraction of the combined scattering relaxation rate due to the all momentum non-conserving processes.

Theory

Considering the special role of the three phonon normal processes and introducing the displaced Planck distribution function for it [6], one can express the Boltzmann transport equation as [6]

$$\left(\frac{\partial N}{\partial t}\right)_{\mathrm{c}} = \frac{N(\lambda) - N_0}{\tau_{\mathrm{N}}} + \frac{N_0 - N}{\tau_R} = \vec{c} \cdot \vec{\nabla} T \frac{\mathrm{d}N}{\mathrm{d}T} \tag{1}$$

where $N(\lambda)$ is the displaced Planck distribution function [6] for the three phonon normal processes as suggested by Callaway [6], N is the Planck distribution function, N_0 is the distribution function at equilibrium, $\left(\frac{\partial N}{\partial t}\right)_{\mathrm{c}}$ represents rate of change of the distribution function with respect to time due to collision, $\frac{\mathrm{d}N}{\mathrm{d}T}$ is the same with respect to temperature, ∇T is the temperature gradient (for detail, see ref. 6), c is the group velocity of phonons, τ_{N} is the three phonon normal processes scattering relaxation time and τ_{R} is the combined scattering relaxation time due to all momentum non-conserving processes. Since our study is confined to low temperatures and for pure sample only, the expression used for the scattering relaxation rate can be expressed as

$$\begin{aligned} \tau_{\mathrm{N}}^{-1} &= B_{\mathrm{N}}^2 w^2 T^3 \\ \tau_{\mathrm{R}}^{-1} &= \tau_{\mathrm{B}}^{-1} + \tau_{\mathrm{pt}}^{-1} + \tau_{\mathrm{U}}^{-1} \\ \tau_{\mathrm{B}}^{-1} &= v_s/L \\ \tau_{\mathrm{pt}}^{-1} &= A w^4 \\ \tau_{\mathrm{U}}^{-1} &= B_{\mathrm{U}} w^2 T^3 e^{-\Theta/\alpha T} \end{aligned} \tag{2}$$

where τ_{B}^{-1}, τ_{pt}^{-1} and τ_{U}^{-1} are the scattering relaxation rates due to boundary scattering [18], point-defect scattering [19] and three phonon umklapp processes [6] respectively, B_{N} and B_{U} are the scattering strengths due to the three phonon normal and umklapp processes respectively, L is the Casimir [18] length of the crystal, A is the point-defect scattering strength [19], v_s is the average phonon velocity, α is a constant [19] and θ is the Debye temperature of the sample under study.

It is needed to mention here that as stated above, our study is limited to the pure sample of the solid HD (sample having zero concentration of $j = 1$ molecules, i.e. sample does not contain ortho-hydrogen as well as para-deuterium molecules) only, therefore, rotational impurities scattering has been totally ignored in the present study. As a result of it, the rotational impurities scattering relaxation rate has not been stated in the expression for τ_R^{-1}. As we know, for a pure sample, the possible scattering mechanisms are boundary walls of the crystal, point-defects and phonon-phonon processes. Therefore, the expression for τ_R^{-1} includes these three scattering relaxation rates only.

Using Eq. (1) and following Callaway [6], one can have an expression for the lattice thermal conductivity of an insulator as

$$K = CI_1 + \Delta K \tag{3}$$

where

$$C = (k_B/2\pi^2 v_s)(k_B T/\hbar)^3 \tag{4}$$

$$\Delta K = CI_2^2/I_3 \tag{5}$$

$$I_1 = \int_0^{\Theta/T} (\tau_N^{-1} + \tau_R^{-1})^{-1} x^4 e^x (e^x - 1)^{-2} dx \tag{6}$$

$$I_2 = \int_0^{\Theta/T} \tau_N^{-1} (\tau_N^{-1} + \tau_R^{-1})^{-1} x^4 e^x (e^x - 1)^{-2} dx \tag{7}$$

$$I_3 = \int_0^{\Theta/T} \tau_N^{-1} \tau_R^{-1} (\tau_N^{-1} + \tau_R^{-1})^{-1} x^4 e^x (e^x - 1)^{-2} dx \tag{8}$$

where k_B is the Boltzmann constant, and $\hbar$ is the Planck constant divided by 2π.

Contribution of the correction term ΔK for the pure sample of the solid HD

Using the scattering relaxation rates stated above and constants stated in Table I, the contribution of the correction term ΔK towards the total lattice thermal conductivity of the pure sample of the solid HD has been calculated in the entire temperature range 0.2–4 K with the help of Eqs (3–8) and the results obtained are reported in Table 2 in the form of its percentage contribution ($\%\Delta K = 100\ \Delta K/K$).

To study the role of the three phonon normal processes scattering relaxation rate in the calculation of the lattice thermal conductivity of the solid HD in more detail, ΔK has also been calculated for the different values of τ_N^{-1} by expressing the three phonon normal processes scattering relaxation rate as a fraction of τ_U^{-1} as well as a fraction of τ_R^{-1}. Expressing τ_N^{-1} as "a" = 100 τ_N^{-1}/τ_U^{-1}, the percentage contribution of ΔK has been studied for the different value of "a" (in the range 10–90) in the entire temperature range 1–4 K to see the effect of the relative value of the three phonon normal and umklapp processes scattering relaxation rates on ΔK and the results obtained are reported in Table 3. The study is confined

Table 1

The values of the constants used in the calculations of the contribution of the correction term (ΔK) towards the total lattice thermal conductivity of solid HD

0	$= 112$ K
α	$= 2.0$
τ_B^{-1}	$= 1.37 \cdot 10^{5}$ sec^{-1}
A	$= 2.20 \cdot 10^{-44}$ sec^{3}
B_N	$= 3.42 \cdot 10^{-20}$ sec deg^{-3}
B_U	$= 1.0 \cdot 10^{-15}$ sec deg^{-3}

Table 2

The percentage contribution of the correction term (ΔK) due to the three phonon normal processes towards total lattice thermal conductivity of the solid HD in the temperature range 0.2–4 K

Temperature, K	% ΔK
0.2	$1.13 \cdot 10^{-3}$
0.4	$3.58 \cdot 10^{-2}$
0.6	0.26
0.8	1.0
1.0	8.23
1.5	28.69
2.0	47.15
2.5	57.15
3.0	63.30
3.5	66.0
4.0	66.20

to 1 K due to the fact that Table 2 shows very small contribution of ΔK below 1 K. To see the effect of the relative value of τ_N^{-1} and τ_R^{-1} on the correction term, the percentage contribution of the correction term ($\%\Delta K$) has been calculated in the entire temperature range 1–4 K for the different value of "b" which gives the relative value of τ_N^{-1} and τ_R^{-1} i.e. "b" $= 100\, \tau_N^{-1}/\tau_R^{-1}$. The result obtained is reported in Table 4.

Table 3

The percentage contribution of the correction term $\%\Delta K$ towards the total lattice thermal conductivity of the solid HD in the temperature range 1—4 K for the different values of "a" which is defined as $a = 100\, \tau_N^{-1}/\tau_U^{-1}$

T, K	Percentage contribution of the correction term $\%\Delta K$								
	$a = 10$	$a = 20$	$a = 30$	$a = 40$	$a = 50$	$a = 60$	$a = 70$	$a = 80$	$a = 90$
1.0	0.78	1.56	2.36	3.17	3.99	4.81	5.66	6.57	7.38
1.5	2.31	4.73	7.26	9.90	12.67	15.57	18.64	21.81	25.18
2.0	3.19	6.61	10.30	14.29	18.60	23.30	28.42	34.03	40.21
2.5	3.38	7.08	11.15	15.67	20.69	26.31	32.64	39.83	48.07
3.0	3.32	7.01	11.14	15.79	21.06	27.09	34.05	42.18	51.79
3.5	3.22	6.84	10.93	15.59	20.94	27.16	34.47	43.20	53.78
4.0	3.14	6.69	10.73	15.38	20.77	27.12	34.69	43.88	55.27

Table 4

The percentage contribution of the correction term $\%\Delta K$ towards the total lattice thermal conductivity of the solid HD in the temperature range 1—4 K for the different values of "b" which is defined as $b = 100\, \tau_N^{-1}/\tau_R^{-1}$

T, K	Percentage contribution of the correction term $\%\Delta K$								
	$b = 10$	$b = 20$	$b = 30$	$b = 40$	$b = 50$	$b = 60$	$b = 70$	$b = 80$	$b = 90$
1.0	9.71	19.48	29.31	39.21	49.18	59.21	69.31	79.47	89.70
1.5	7.90	16.17	24.85	33.97	43.55	53.65	64.29	79.23	87.41
2.0	5.77	12.10	19.10	26.86	35.52	45.24	56.24	68.73	83.22
2.5	4.52	9.62	15.45	22.12	29.87	38.98	49.84	63.01	79.31
3.0	3.87	8.30	13.43	19.45	26.59	35.20	45.80	59.16	76.52
3.5	3.51	7.57	12.31	17.93	24.69	32.95	43.33	56.72	74.68
4.0	3.31	7.14	11.65	17.03	23.53	31.59	41.80	55.18	73.48

Results and discussions

With the help of Table 2, it is very clear that the percentage contribution of the correction term ($\%\Delta K$) due to the three phonon normal processes towards the total lattice thermal conductivity of the pure sample of the solid HD is very small below 1 K. But its contribution is considerably large above 1 K and it is as high as 66.2% at 4 K. The similar conclusion is also drawn by previous workers for solid He [14] and LiF [15].

The effect of the relative scattering relaxation rates of the three phonon normal and umklapp processes can be seen with the help of the Table 3. From this table, it is clear that the percentage contribution of the correction term $\%\Delta K$ increases with increase of "a" at each temperature which shows that $\%\Delta K$ increases with increase of the three phonon normal processes scattering relaxation rate, or $\%\Delta K$ increases with decrease of the three phonon umklapp processes scattering relaxation rate, provided the boundary and point-defect scattering relaxation rates are remain unchanged. The effect of the relative value of τ_N^{-1} and τ_R^{-1} (combined scattering relaxation rate due to all momentum non-conserving processes) can be seen in Table 4. From this table, one can see that $\%\Delta K$ increases with increase of "b". As a result of it, one can say that $\%\Delta K$ increases with increase of τ_N^{-1}. In other words, one can also say that $\%\Delta K$ decreases with decrease of the scattering relaxation rate due to any momentum non-conserving processes.

Thus, the entire study can be summarized as following: (a), the role of the three phonon normal processes scattering relaxation rate in the calculation of the lattice thermal conductivity of the pure sample of the solid HD has been studied in the entire temperature range 0.2 – 4 K by calculating the contribution of the correction term. (b), The contribution of the correction term ΔK towards the total lattice thermal conductivity is found considerably large above 1 K and it should not be ignored in the calculation of the lattice thermal conductivity of the solid HD. (c), the percentage contribution of the correction term increases with increase of the three phonon normal processes scattering relaxation rate.

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Résumé — On a étudié, dans tout l'intervalle des températures comprises entre 0.2 et 4 K, le rôle des processus normaux à trois phonons dans l'analyse de la conductivité thermique du réseau de HD solide. L'étude s'est effectuée en calculant la contribution du terme de correction dû aux processus normaux à trois phonons, à la conductivité thermique totale du réseau de l'échantillon pur de HD solide, dans l'intervalle de températures considéré. On a, de même, effectué l'étude pour les différentes valeurs de la vitesse de relaxation de dispersion des processus normaux à trois phonons. On a trouvé une contribution considérable due au terme de correction comparé à la conductivité totale de phonon au-dessus d'1 K.

Zusammenfassung — Die Rolle der Drei-Phonon-Normalprozesse in der Analyse der Gitter-Wärmeleitung von festem HD wurde im Temperaturbereich von 0.2 bis 4 K untersucht. Die Untersuchung wurde durch Berechnung des Beitrages des den Drei-Phonon-Normalvorgängen zuzuschreibenden Korrekturterms an der totalen Gitter-Wärmeleitung der reinen Probe des festen HD im o.a. Temperaturbereich durchgeführt. Diese Betrachtung wurde auch für die verschiedenen Werte der Streuungsrelaxationsgeschwindigkeit der Drei-Phonon-Normalvorgänge durchgeführt. Ein wesentlicher Beitrag des Korrekturglieds wurde im Vergleich zu der totalen Phononleitfähigkeit oberhalb von 1 K gefunden.

Резюме — Изучена роль трехфононовых нормальных процессов при анализе решеточной термической проводимости твердого НД в области температур 0.2—4 К. Исследование было проведено путем вычисления поправочного члена, обусловленного трехфононовыми нормальными процессами, относительно обшей решеточной термической проводимости чистого твердого НД в вышеуказанной области температур. Изучение было также проведено для различных значений релаксационной скорости рассеивания трехфононовых нормальных процессов. Установлен значительный вклад поправочного члена, который сравним с общей фононовой проводимостью выше 1 К.