Lattice Thermal Conductivity of Solid Helium

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Callaway's theory is applied to explain the thermal conductivity of solid He[4] at different densities, i.e., $\rho=0.194$, 0.262, and 0.282 g/cm[3], where the data are available on either side of the conductivity maximum. Good agreement between theory and experiment is obtained by taking $\alpha=2.4$ for all the three cases for the exponential temperature dependence of umklapp processes given by $\propto e^{-\theta/\alpha T}$, which verifies the θ dependence of umklapp processes. The present calculations also reveal the importance of normal processes near the conductivity maximum where the correction term in Callaway's expression for thermal conductivity dominates. This also shows that the additivity of reciprocal relaxation times is not valid for single isotope pure solid helium. The calculations also suggest the presence of internal boundaries in the composition of the solid.

INTRODUCTION

A THEORY of lattice thermal conductivity which was applicable to low temperatures was developed by Peierls.[1] He considered the effect of anharmonicity and quantized the lattice waves. He studied the thermal equilibrium in the crystal and established two types of interactions of the phonons with the lattice. Interactions of one type, in which the quasi-momentum of the lattice waves is conserved, are called normal processes, while those of the other type, in which the quasi-momentum is not conserved, are called umklapp processes. Umklapp processes contribute to the thermal resistance a term which decreases exponentially at low temperatures. This exponential dependence of conductivity upon temperature, as predicted by Peierls, was confirmed in the case of quartz,[2] sapphire,[2] solid helium,[3] and lithium fluoride.[4] Peierls also predicted that lattice imperfections and external boundaries increase the resistance.

A number of papers have been presented in the past in which attempts were made to develop Peierls' picture. It can be shown that if terms of higher than the third order are neglected in the elastic potential energy, the expression for the thermal conductivity of an infinite perfect crystal diverges. To make the conductivity finite or to remove the sensitive size dependence, Pomeranchuk[5] invoked fourth-order terms in the elastic potential energy; this is rather unsatisfactory as such terms must have an extremely small effect at moderately low temperatures. Herpin's[6] suggestion that imperfections in the crystal such as isolated impurities, dislocations, and stacking faults can lower the conductivity at low temperatures, could not stand, since the number of such defects in a reasonably good crystal is too small to have an appreciable effect. Herring[7] has shown that in many crystals the elastic

anisotropy removes the divergence of the thermal conductivity.

The role of normal processes, which by themselves conserve wave vector, and do not lead to thermal resistance, has been emphasized by Klemens[8] and Callaway.[9] According to Klemens' cutoff mechanism low-frequency phonons, which cause the integral to diverge at low temperatures, are scattered by normal three-phonon processes. This scattering annihilates long-wavelength phonons and creates phonons with the typical frequency $\omega_I=kT/\hbar$. In Callaway's theory a natural cutoff for long-wavelength phonons is provided by boundary scattering. Normal processes play a very important role in determining the thermal resistance in the sense that they can change the distribution of phonons among the various states. For example, if due to a certain frequency-dependent scattering mechanism, certain states are more depleted in population compared to others, then normal processes would tend to refill these states.

In the present paper, we have applied Callaway's theory to explain the thermal conductivity of solid helium at different densities. Solid helium is very interesting in the sense that it is possible to vary the Debye temperature by varying the density and hence it is possible to study the θ dependence of umklapp processes in the same substance within the framework of Callaway's theory.

The thermal conductivity K according to Callaway is given by

$$K = (k/2\pi^2 c)(I_1 + \beta I_2),$$

TABLE I. Values of the Debye temperature θ_D, the phonon velocity c, and the characteristic length L at different densities.

Density, ρ (g/cm[3])	θ_D[a] (°K)	c (10^4 cm/sec)	L (mm)
0.194	24	2.62	0.5
0.262	48	4.73	0.5
0.282	57	5.50	0.5

[a] Values of θ_D are from C. Domb and J. S. Dugdale [*Progress in Low-Temperature Physics*, edited by J. C. Gorter (North-Holland Publishing Company, Amsterdam, 1957), Vol. 2, p 338].

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[2] R. Berman, Proc. Roy. Soc. (London) A208, 90 (1951).
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[6] A. Herpin, Ann. Phys. (New York) 7, 91 (1952).
[7] C. Herring, Phys. Rev. 95, 954 (1954).
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[9] J. Callaway, Phys. Rev. 113, 1046 (1959).

TABLE II. Values of the relaxation parameters which give the best fit to experimental results at different densities.

Density (g/cm^3)	B_N (10^{-16} sec deg^{-3})	$B_U e^{-\theta/\alpha T}$ (10^{-16} sec deg^{-3})	$(c/L)_{exp}$ (10^6 sec^{-1})	$(c/L)_{exp}/(c/L)_{cal}$	α
0.194	2	2400 $e^{-10/T}$	1.22	2.33	2.4
0.262	0.5	5700 $e^{-20/T}$	6.3	6.67	2.4
0.282	1	1735 $e^{-24/T}$	5.8	5.30	2.4

where

$$I_1 = (kT/\hbar)^3 \int_0^{\theta/T} \tau_c \frac{x^4 e^x}{(e^x-1)^2} dx$$

and

$$\beta I_2 = (kT/\hbar)^3 \left[\int_0^{\theta/T} \frac{\tau_c}{\tau_N} \frac{x^4 e^x}{(e^x-1)^2} dx \right]^2$$

$$\div \int_0^{\theta/T} \frac{1}{\tau_N} \left(1 - \frac{\tau_c}{\tau_N} \right) \frac{x^4 e^x}{(e^x-1)^2} dx.$$

The combined relaxation time τ_c is given by

$$\tau_c^{-1} = \tau_N^{-1} + \tau_U^{-1} + \tau_B^{-1} + \tau_{pt}^{-1}$$

$$= B_N T^3 \omega^2 + B_U T^3 e^{-\theta/\alpha T} \omega^2 + c/L + A\omega^4,$$

where τ_N, τ_U, τ_B, and τ_{pt} are the relaxation times for the normal processes, umklapp processes, boundary scattering, and point-defect scattering, respectively.

RESULTS AND DISCUSSION

The thermal conductivity of solid helium of different densities at low temperatures has been investigated by Webb et al. Measurements for the thermal conductivity of solid helium at density 0.194 g/cm^3 were given by Webb, Wilkinson, and Wilks.[3] Further results for higher
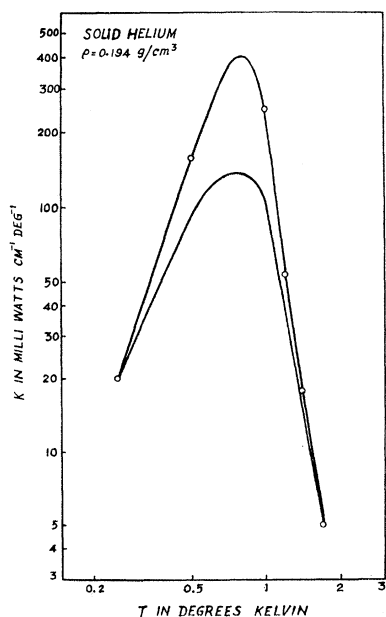
densities were obtained by Webb and Wilks.[10] The results for densities higher than 0.3 g/cm^3 were unreliable due to the size dependence of thermal conductivity, as was pointed out by Webb and Wilks. Therefore, theoretical curves are obtained only for densities 0.194, 0.262, and 0.282 g/cm^3. Also for these densities, the experimental results are available on either side of the conductivity maximum.

The results of calculations based on Callaway's theory are shown in Figs. 1 to 3, and the various

TABLE III. Contribution of the correction term $(k/2\pi^2 c)\beta I_2$ to the total thermal conductivity at different temperatures for $\rho=0.194$ g/cm^3.

T (°K)	$(k/2\pi^2 c)I_1$ (W cm^{-1} deg^{-1})	$(k/2\pi^2 c)\beta I_2$ (W cm^{-1} deg^{-1})	$K=(k/2\pi^2 c)(I_1+\beta I_2)$ (W cm^{-1} deg^{-1})
0.25	0.019	0.001	0.020
0.5	0.060	0.094	0.154
1.0	0.042	0.208	0.250
1.2	0.028	0.025	0.053
1.4	0.014	0.004	0.018
1.7	0.0047	0.0004	0.0051

FIG. 1. Thermal conductivity of solid He^4 for density $\rho=0.194$ g/cm^3. The simple curve is the experimental curve given by Webb et al. The curve marked by circles is the theoretical curve based on Callaway's model, taking $\alpha=2.4$. $\theta_D=24$°K.
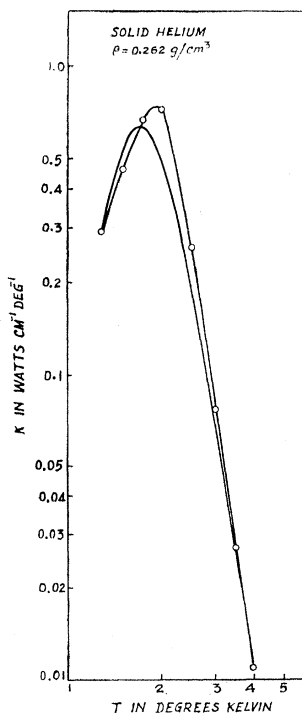

FIG. 2. Thermal conductivity of solid He^4 for density $\rho=0.262$ g/cm^3. The simple curve is the experimental one given by Webb and Wilks. The circles denote the theoretical curve calculated on the basis of Callaway's model, taking $\alpha=2.4$. $\theta_D=48$°K.

[10] F. J. Webb and J. Wilks, Phil. Mag. 44, 644 (1953).

TABLE IV. Contribution of the correction term $(k/2\pi^2 c)\beta I_2$ to the total thermal conductivity at different temperatures for density $\rho = 0.262$ g/cm^3.

T (°K)	$(k/2\pi^2 c)I_1$ (W cm^{-1} deg^{-1})	$(k/2\pi^2 c)\beta I_2$ (W cm^{-1} deg^{-1})	$K = (k/2\pi^2 c)(I_1 + \beta I_2)$ (W cm^{-1} deg^{-1})
1.28	0.26	0.03	0.29
1.5	0.37	0.09	0.46
1.75	0.48	0.19	0.67
2.0	0.46	0.22	0.68
2.5	0.24	0.02	0.26
3.0	0.076	0.001	0.077
3.5	0.026	$\cdots$	0.026
4.0	0.011	$\cdots$	0.011

quantities used in these calculations are given in Table I. For the evaluation of basic parameters of the Callaway's model the reader is referred to our earlier paper.[11]

The thermal conductivity, in the absence of isotope scattering, can be expressed as

$$K = (k/2\pi^2 c)(kT/\hbar)^3 \left\{ \int_0^{\theta/T} \frac{G(x)}{Ex^2 + c/L} dx \right.$$

$$+ \left[\int_0^{\theta/T} \frac{E_N H(x)}{Ex^2 + c/L} dx \right]^2 \Bigg/$$

$$\left. \int_0^{\theta/T} \frac{E_N(E_U + c/L)}{Ex^2 + c/L} H(x) dx \right\},$$

where

$$E = E_N + E_U = (k/\hbar)^2 T^5 (B_N + B_U e^{-\theta/\alpha T}),$$

$$G(x) = x^4 e^x/(e^x - 1)^2, \quad H(x) = x^6 e^x/(e^x - 1)^2.$$

Values of the parameters B_N, $B_U e^{-\theta/\alpha T}$, c/L, and α which give the best fit to experimental results are given in Table II. Values of B_N and c/L are accurate up to 5% in all cases. B_U and α are accurate up to 10% for $\rho = 0.262$ and 0.282 g/cm^3, while for $\rho = 0.194$ g/cm^3 they are accurate up to 20%.

It may be seen from Table II that the values of the constants B_N and B_U are affected by the density, but at low temperatures it is not possible to give reliable

TABLE V. Contribution of the correction term $(k/2\pi^2 c)\beta I_2$ to total thermal conductivity at different temperatures for density $\rho = 0.282$ g/cm^3.

T (°K)	$(k/2\pi^2 c)I_1$ (W cm^{-1} deg^{-1})	$(k/2\pi^2 c)\beta I_2$ (W cm^{-1} deg^{-1})	$K = (k/2\pi^2 c)(I_1 + \beta I_2)$ (W cm^{-1} deg^{-1})
1.33	0.23	0.07	0.30
1.75	0.36	0.32	0.68
2.0	0.39	0.61	1.00
2.5	0.37	0.64	1.01
3.0	0.26	0.14	0.40
3.5	0.134	0.023	0.16
4.0	0.059	0.004	0.063

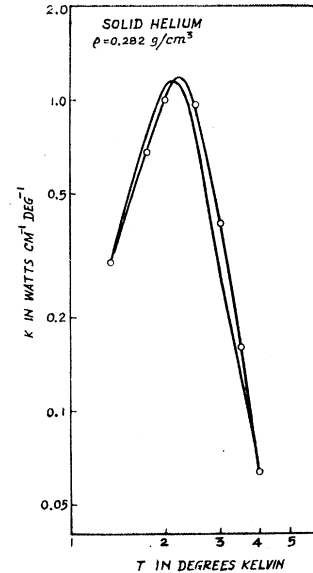[11] Bal Krishna Agrawal and G. S. Verma, Phys. Rev. **126**, 24 (1962).

FIG. 3. Thermal conductivity of solid He^4 for density $\rho = 0.282$ g/cm^3. The simple curve is the experimental one given by Webb and Wilks. Circles mark the theoretical curve calculated on the basis of Callaway's model, taking $\alpha = 2.4$. $\theta_D = 57$°K.

numerical estimates of the density variation of these constants.

The values of $(k/2\pi^2 c)I_1$ and $(k/2\pi^2 c)\beta I_2$ for different temperatures at three densities are given in Tables III to V. It may be noticed from these tables that the correction term containing β in the final expression for thermal conductivity makes an important contribution and can not be neglected. As a matter of fact, near the conductivity maximum this becomes the dominant term in two cases. Hence, the additivity of the reciprocal relaxation times is not valid in the present case, although this assumption was valid for our earlier calculations of lattice thermal conductivity, where either the point-defect scattering or the umklapp scattering was large and the β term was negligible.

It may be seen from the figures that the agreement between the theory and the experiments is good. The interesting part of the present calculations is that the value of α turns out to be the same in all the three cases. Since, upon varying the density, there is no change in the structure of the substance, it is expected that α should remain the same. The value of α as obtained for He^4, which is hexagonal in structure, is 2.4.

In view of the fact that α is constant, the θ dependence of umklapp processes is verified in the framework of Callaway's theory. Further, the present calculations also indicate the importance of normal processes near the conductivity maximum, where contributions due to boundary and umklapp scatterings are negligible and the correction term due to normal processes in Callaway's expression for thermal conductivity dominates, or makes a significant contribution.

Comparison of the boundary relaxation time, as obtained experimentally with the theoretical value, indicates the presence of internal boundaries in the crystal.