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Thermal Conductivity in Solid He^3 and He^4 †

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The thermal conductivity of solid He^4 and He^3 has been measured over the temperature range 0.5 to 2.0°K at a number of molar volumes. In the hcp phase the thermal conductivities of both isotopes show the usual low-temperature maximum. Below the maximum the conductivity is characteristic of boundary scattering with a T^3 dependence and a phonon mean free path comparable to the sample diameter. The conductivity above the maximum varies exponentially with $1/T$ over about three orders of magnitude. This is characteristic of umklapp scattering. In the bcc phase of He^3 the anomalous behavior found by Walker and Fairbank is confirmed and the extension of the measurements to lower temperature permits observation of more of the anomalous region. The thermal conductivity in the bcc phase of He^4 has also been measured.

INTRODUCTION

THE atomic vibrations responsible for the propagation of thermal energy in dielectric crystals can be described in terms of quantized lattice waves called phonons.[1] Part of the understanding of the behavior of a solid depends on information about the behavior of the phonons which inhabit it. A portion of this information can be obtained by examining the scattering processes which impede the flow of phonon energy in a thermal-conductivity measurement.[2] We shall be concerned here with the scattering of phonons by crystal imperfections, boundaries, and by phonon-phonon interactions (the phonon-phonon interactions which contribute directly to thermal resistance are the umklapp processes, first described by Peierls).[3]

Solid helium is a particularly favorable substance for the study of heat conduction in a dielectric solid. It exists in two isotopic forms each having hexagonal close-packed and body-centered cubic crystal structures at easily attainable temperatures and pressures. The solid can be formed over a wide range of molar volumes allowing a study of variation in properties with Debye temperature. Both isotopes can be obtained chemically and isotopically very pure. Earlier thermal-conductivity measurements[4–8] have shown that both isotopes exhibit the exponential rise in thermal conductivity with decreasing temperature predicted by Peierls for umklapp processes. At the very lowest temperatures the phonon mean free path is expected to be limited by scattering at the crystal boundaries according to the theory of Casimir.[9] The earlier measurements of Webb, Wilkinson, and Wilks[5] on a single molar volume of He^4 are in agreement with this prediction.

The present work extends the temperature range of previous measurements to 0.5°K for both He^3 and He^4 for several molar volumes; the low-temperature measurements fall well within the range where boundary scattering predominates in the hcp phase and extend substantially the range of temperature for measurements in the bcc phase where earlier results showed anomalous behavior.[6] A preliminary report of part of this work was given at the Eighth International Conference on Low Temperature Physics.[7]

APPARATUS

Measurements were made on samples of solid helium 2.0 mm in diam and about 30 mm in length contained in a cylindrical stainless steel tube of 2.4 mm o.d. The ends of the tube were closed by pieces of high-conductivity copper as shown in Fig. 1. Power was fed through the sample from an electrical heater wound on the lower of these end pieces. The upper end piece was soldered to the He^3 refrigerator which served as the heat sink. The helium sample was condensed through the 0.006-in.-i.d. capillary filling line which was thermally anchored to the He^3 refrigerator and to the He^4 bath. Pressures up to 150 atm could be applied through the capillary from the high-pressure gas-handling system which has been described by Walker and Fairbank.[8] The cryogenics, except for the He^3 refrigerator, have also been described by these authors.

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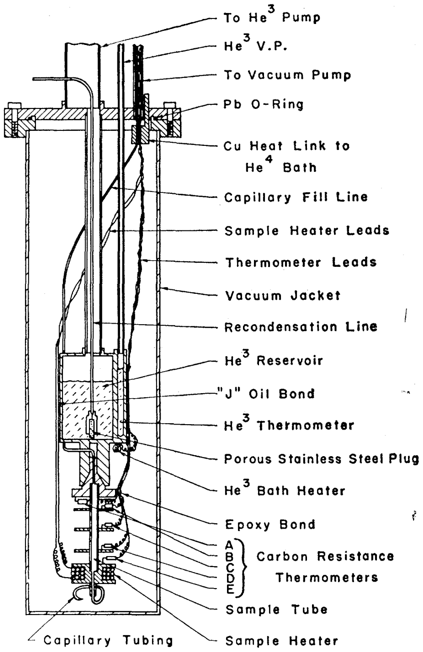

FIG. 1. Schematic drawing of conductivity cell and liquid He^3 bath.

High-conductivity copper fins were silver soldered to the sample tube at 6-mm intervals. Temperature measurements were made at these fins and the copper end pieces using 33-Ω 0.1 W Allen Bradley carbon resistors as thermometers. A 33-cps Wheatstone Bridge of the type described by Briscoe[10] was used to measure the resistances; the measuring power was about 10^{-11} W. Measurement at constant. temperature using higher powers yielded the same resistances indicating that the measured resistance was truly characteristic of the fin temperatures.

The reservoir serving as a heat sink was filled with He^3 for measurements below 1.2°K, and with He^4 for measurements at higher temperatures. The siphon used for filling the He^4 bath from an external storage container was fitted with a needle valve at the bath end so that the bath could be refilled below atmospheric pressure, and solid samples could be retained as long as necessary.

EXPERIMENTAL PROCEDURE

The resistance thermometers were calibrated against the vapor pressure of He^3[11] and He^4.[12] These calibrations were found to remain valid for periods of over three weeks if the apparatus was maintained at or below nitrogen temperature.

The sample was solidified using the blocked-capillary method. The gas was admitted to the high-pressure system, and a pressure appropriate to the desired molar volume was applied. The He^4 bath was pumped to a temperature about 5 mdeg above the freezing temperature at this volume. He^3 gas was then admitted to the He^3 reservoir pumping line to aid in transferring heat out of the sample into the bath, and the resistors were monitored. Thirty minutes after equilibrium was reached the He^4 bath was pumped rapidly to about 1.1°K. The portion of the capillary leading to the sample tube was thermally anchored to the He^4 bath and hence blocked with solid as the bath temperature was reduced below the freezing point. The sample, therefore, remained subsequently at constant volume.

The sample was then annealed by holding the He^4 bath at 1.1°K and raising the sample to a temperature well within the liquid-solid mixed-phase region by applying power to the He^3 bath heater. This temperature was maintained for about 30 min, and then smoothly reduced to bath temperature over a period of 90 min. The He^3 bath was then slowly pumped to about 0.5°K.

The thermal conductivity measurements were made by applying an appropriate power to the lower end of the sample chamber and measuring the resistance of resistor B, and the differences in resistance between B and C, and between B and D. Two pairs of resistances were measured in order to provide a check on the consistency of the data. These resistances were then converted to temperatures and temperature differences. By taking data on the empty stainless-steel sample chamber the power flowing through its walls could be measured. Because of the relatively low conductivity of the stainless steel this correction was less than 1% for all measurements except those near 2°K where it was as large as

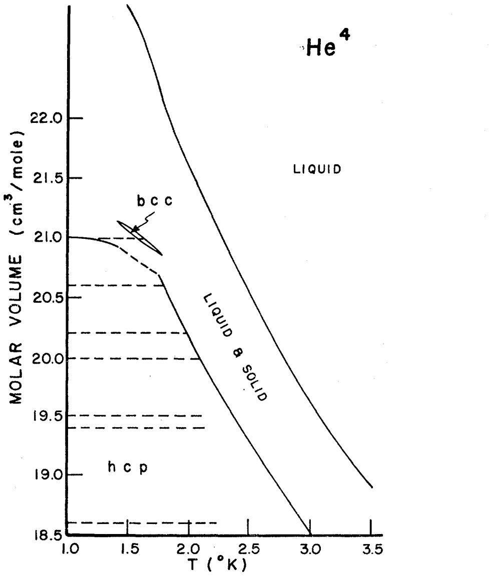

FIG. 2. Equilibrium molar volume versus temperature for He^4. The dashed lines correspond to volumes at which data were taken.

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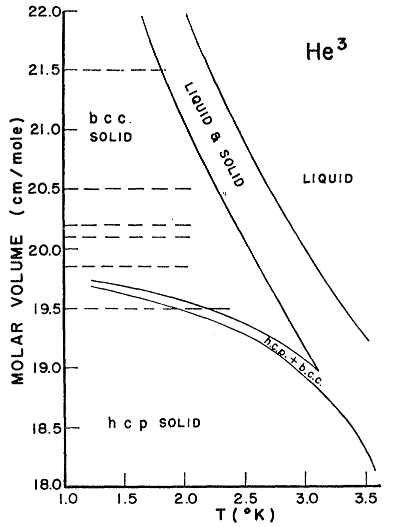

FIG. 3. Equilibrium molar volume versus temperature for He^3. The dashed lines correspond to volumes at which data were taken.

13% in the worst case. The thermal conductivity was then calculated using the equation

$$Q_s = Q - Q_t = KA\Delta T/L,$$

where Q_s is the heat flow through the sample, Q is the total heat input, Q_t is the heat flow through the stainless steel, A is the cross-sectional area of the sample, and L is the distance between resistors.

RESULTS

The thermal-conductivity measurements were carried out at constant volume. Figures 2 and 3 are plots of molar volume versus temperature along the liquidus and solidus curves for He^4 and He^3, respectively. Measurements were made at volumes corresponding to the dashed lines on these graphs. The data were taken over the temperature range 0.5°K to the melting temperatures in both the hcp and bcc phases for both isotopes. The volume range corresponded to values of the Debye temperature, Θ, of 23.5 to 34.6°K for He^3 and 17.0 to 33.9°K for He^4.[13,14] These measurements are consistent with earlier results, the present data having somewhat less scatter.

TABLE I. Constants for Eq. (1) in hcp helium.

Isotope	Molar volume (cm^3)	Θ (°K)	$A\left(\frac{\text{watts}}{\text{cm deg}}\right)$	$\frac{\Theta}{b}$	b
	18.6	33.9	3.7×10^{-5}	11.7	2.9
	19.4	30.4	3.0×10^{-5}	10.3	3.0
	20.0	28.0	1.8×10^{-5}	10.2	2.7
He^4	20.2	27.2	2.0×10^{-5}	9.4	2.9
	20.6	25.7	1.8×10^{-5}	8.9	2.9
	21.0	24.4	1.6×10^{-5}	8.4	2.9
He^3	19.5	34.6	5.0×10^{-5}	11.2	3.1

hcp Phase

Figure 4 is a plot of log$K$ versus T for two samples of He^4 and one of He^3 in the hcp phase. The thermal conductivity shows the usual peak just below 1°K. At temperatures above the peak the thermal conductivity shows the characteristic behavior of phonon scattering by umklapp processes and is in excellent agreement for both He^3 and He^4 over three orders of magnitude with the expression derived by Peierls for this case

$$K = AT^n \exp(\Theta/bT), \tag{1}$$

where A is a constant related to the scattering strength, n is a small number, Θ is the Debye temperature, and b is a number related to the phonon dispersion. Curve fitting of our data to this expression, both graphically and by computer, shows agreement for $n \cong 0$ (best fit by computer: $n = 0.4 \pm 0.5$). The constants A and b in Eq. (1) have been evaluated from the data for $n = 0$ using the Debye temperatures calculated by Heltemes and Swenson[13] and are given in Table I; the fit is shown graphically in Fig. 5 where log$K$ versus $1/T$ is plotted for several molar volumes of He^4 over the temperature range where Eq. (1) applies.

At temperatures below the peak, the thermal conductivity is dominated by scattering of phonons at the crystal boundaries. Casimir[9] has shown that the conductivity should be proportional to T^3 in this case. The splitting of the data for He^3 at 19.5 cm^3/mole into two

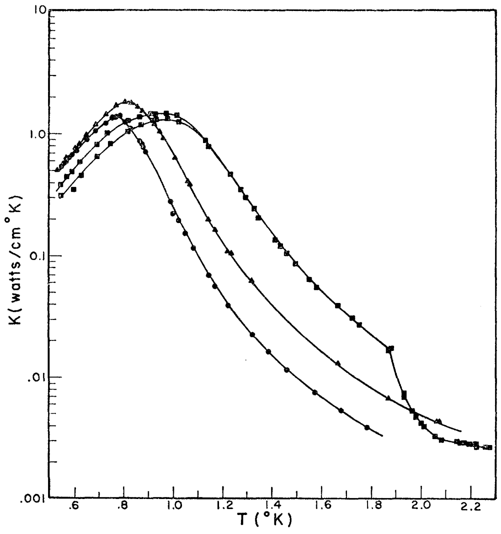

FIG. 4. Thermal conductivity versus temperature for three samples of solid helium. The triangles are for He^4 at 19.5 cc/mole. The squares are for He^3 at 19.5 cc/mole. The closed circles are for He^4 at 20.2 cc/mole.

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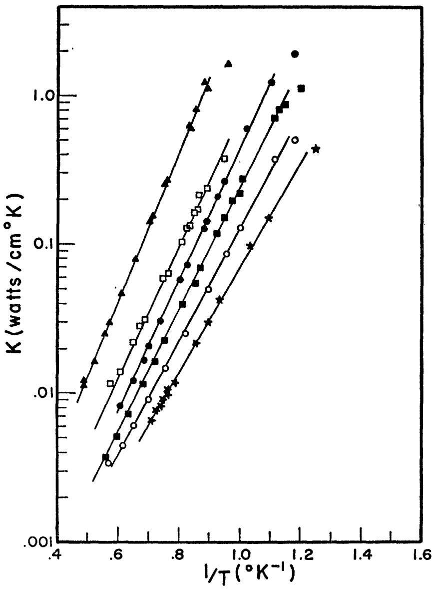

FIG. 5. Umklapp behavior in solid He⁴. Plot of log$K$ versus $1/T$ showing agreement with Eq. (1). The volumes are as follows:

Triangles	18.6 cc/mole
Open squares	19.4 cc/mole
Closed circles	20.0 cc/mole
Closed squares	20.2 cc/mole
Open circles	20.6 cc/mole
Stars	about 21 cc/mole.

branches in this region corresponds to measurements taken over two different paths along the sample. A qualitative discussion can be given by writing an expression for the thermal conductivity of a phonon gas by analogy with a well-known result of kinetic theory:

$$K=\tfrac{1}{3}Cvl. \qquad (2)$$

This expression is used to define a phonon mean free path, l. C is the specific heat per unit volume; the velocity of sound v is calculated using a Debye model, $v=(k_B\Theta/\hbar)(6\pi^2N/V)^{-1/3}$ where N is Avogadro's number, V is the molar volume, and k_B is the Boltzmann constant. Casimir's result shows $R\leq l\leq 1.5\,R$, where R is the diameter of a cylindrical sample, v is constant, and C (and thus K) varies as T^3.

Figure 6 is a plot of thermal conductivity versus T^3 at temperatures below about 0.8°K for several samples. The lower three curves have mean free paths somewhat smaller than the sample diameter, probably due to imperfect annealing.

Preliminary measurements showed that at temperatures in the boundary scattering-region the effect of annealing was considerable. Figure 7 shows three sets of thermal conductivity results on a sample of hcp He³; the differences among the three curves is due to different states of annealing. The total effect of annealing was to increase the thermal conductivity, and hence the maximum phonon mean free path, by as much as a factor of 20. The maximum mean free path calculated using Eq. (2) was found to be about 1 mm, or half the sample diameter in most cases, (although it was 3 mm in one instance). This is somewhat less than predicted by Casmir's theory. As found by previous investigators[15,16] samples of solid helium formed by the blocked-capillary technique usually consist of a number of crystals and even after careful annealing the sample probably consisted of a few large crystals rather than a single crystal. A thermal conductivity result in He⁴ at about 18.5 cc/mole recently reported by Mezhov-Deglin[17] corresponds to a maximum phonon mean free path of about 1.5 sample diameters. This measurement was made in a glass chamber; the helium sample could be visually observed during crystallization and measurement.

bcc Phase

Results in the bcc phase of solid helium are of interest because of the anomalous behavior observed in this phase by Walker and Fairbank.[8] Four of the molar volumes of He³ measured are identical with those measured by these authors above 1°K. In the region of overlap the agreement between the two sets of data is excellent, the scatter being somewhat smaller in the present work.

The thermal conductivity in this phase is lower than in the hcp phase and a plot of log$K$ versus $1/T$ shows a distinct change in slope in the neighborhood of 0.7(°K)⁻¹. Such plots are made in Fig. 8 for two molar volumes. The change in slope can be described by as-

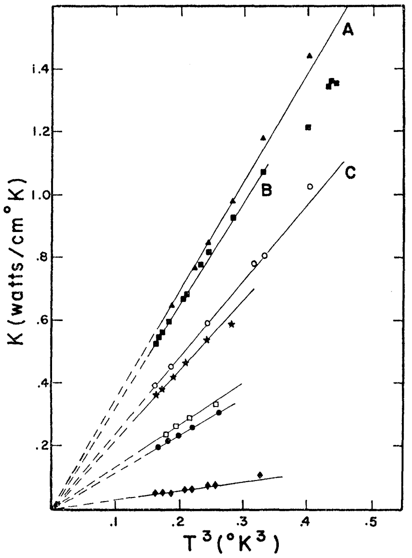

FIG. 6. Thermal conductivity versus T^3 in the boundary scattering region. A is He⁴ at 19.5 cc/mole. B is He³ at 19.5 cc/mole. C is He⁴ at 20.2 cc/mole. The three lower curves are less well annealed samples of He⁴. Open squares and closed circles are 19.5 cc/mole data; diamonds are 18.6 cc/mole data.

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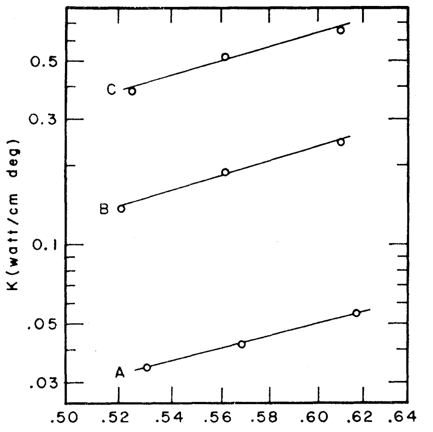

FIG. 7. The thermal conductivity of a sample of solid He^3 for different annealed states. Curve A is for an unannealed sample. Curve B is for a sample annealed slightly below the mixed-phase region. Curve C is for a sample annealed well in the mixed-phase region.

suming two competing phonon scattering mechanisms, each of which contributes a thermal conductivity of the form of Eq. (1), and adding their thermal resistances:

$$K^{-1}=W=A_1\exp(-T_1/T)+A_2\exp(-T_2/T)\,, \qquad (3)$$

where k_BT_1 and k_BT_2 are the excitation energies of the individual scattering processes. Table II lists the pertinent parameters for this fit. If the high-temperature behavior is taken to be due to umklapp scattering the values of T_2 yields b's between two and three; the A_2's are also roughly consistent with the hcp results.

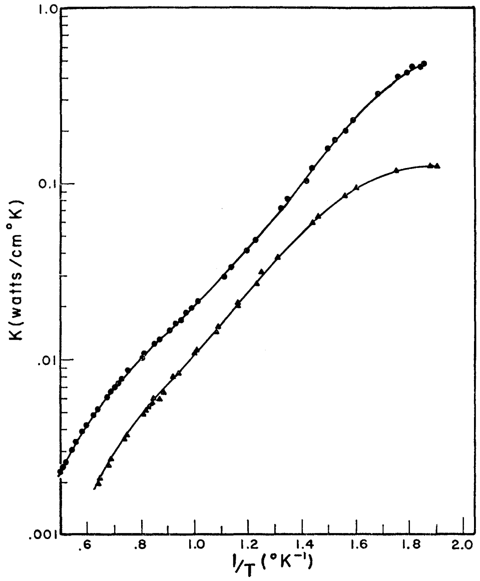

FIG. 8. Thermal conductivity versus $1/T$ for two volumes of He^3 in the bcc phase. The circles are for 20.2 cc/mole. The triangles are for 21.5 cc/mole.

In addition to the above behavior, there exist slight dips in the thermal conductivity curves, centered around 1°K. This behavior suggests the possibility of a resonance phonon scattering mechanism.[18] These thermal conductivity results indicate that the bcc phase of He^3 presents a very complicated problem.

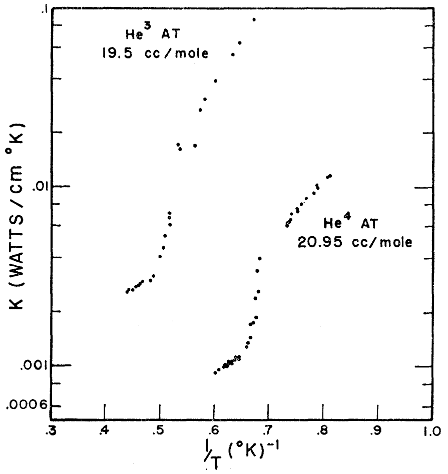

FIG. 9. Thermal conductivity versus $1/T$ for He^4 and He^3 showing transition from hcp to bcc phase. The upper curve is He^3 at 19.5 cc/mole. The lower curve is He^4 at 21 cc/mole. The crystals are in the bcc phase in the left portion of each curve, in the hcp phase in the right portion and in a two-solid mixed phase in the middle portion.

Figure 9 is a plot of log$K$ versus $1/T$ in the bcc phase of He^4. The anomalously low thermal conductivity present in He^3 is not observed because of the small range of temperature and pressure over which this solid exists. The lower curve of Fig. 9 is bcc, mixed phase, and hcp in He^4 at 21 cc/mole. A similar transi-

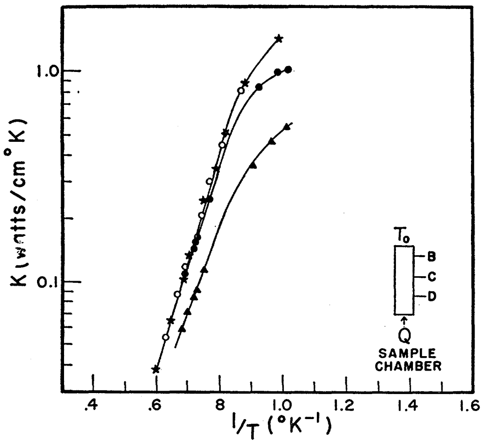

FIG. 10. Illustration of the effect of anisotropy on thermal conductivity. The symbols represent the following conditions: solid circles are for path BC after initial annealing. Triangles are for path BD after initial annealing. Open circles and stars are for both paths after reannealing.

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TABLE II. Parameters for Eq. (3) in the bcc phase of solid helium.

Isotope	Molar volume (cc)	Θ (°K)	$A_1\left(\frac{\text{cm deg}}{\text{watt}}\right)$	T_1 deg K	T_2 deg K	$A_2\left(\frac{\text{cm deg}}{\text{watt}}\right)$
	19.8	27.9	1.2×10^3	3.35	15	1.2×10^5
	20.1	27.2	1.3×10^3	3.35	14	1.8×10^5
He^3	20.2	27	1.8×10^3	3.77	13.5	6.2×10^5
	20.5	26.1	1.7×10^3	3.53	16	5.2×10^5
	21.5	23.5	4.4×10^3	3.85	11	1.3×10^5
He^4	21	17			5.2	2.5×10^4

tion in He^3 at 19.5 cc/mole is shown for comparison in the upper curve. These results are in agreement with the recent work of Berman and Rogers.[19]

Anisotropy

Except for a few isolated cases, the results were reproducible from sample to sample. The exceptional cases typically yielded results exemplified by Fig. 10. The two lower curves are thermal conductivities measured between different points on the sample, and were taken after an initial annealing. After reannealing the same sample, the upper curve was measured over both path lengths, it is consistent with the usual data. We interpret the anomalous behavior as due to anisotropy; two or more crystals were oriented in some arbitrary direction with respect to each other. The usual reproducibility is a reflection of a preferred orientation during crystal growth. The samples were always grown the same way, and always from one end of a long cylindrical chamber. There is also evidence for anisotropy in solid helium from sound-velocity measurements.[20,21]

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Thermal Conductivity in Isotopic Mixtures of Solid Helium*

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This paper reports thermal-conductivity measurements in isotopic mixtures of solid helium at concentrations of 1%, 4%, 10%, and 50% of He^3 in He^4 and vice versa. The data were taken at $V=19.5$ cm³/mole and $V=20.2$ cm³/mole; the data at 19.5 cm³/mole are analyzed using the phenomenological equation of Callaway. Resistance to heat flow is assumed to be due to boundary scattering, normal-process scattering, umklapp-process scattering, and a Rayleigh-like scattering mechanism which may be a combination of mass-fluctuation scattering and lattice-distortion scattering. The strength of these scattering mechanisms is determined by the analysis. The major contribution to the thermal resistance at low temperature is found to be the lattice distortion around the site of an isotopic impurity. The intrinsic strength of lattice-distortion scattering is evaluated and found to depend on concentration more strongly than the $x(1\text{-}x)$ expected by analogy to mass-fluctuation scattering. Two arguments are given in explanation of this increased concentration dependence.

INTRODUCTION

THERMAL-conductivity measurements in isotopic mixtures of solid helium have been reported by Walker and Fairbank.[1] The analysis of their data by Callaway[2] and the subsequent interpretation by Klemens and Maradudin[3] suggest that the scattering of phonons by lattice distortion may make a major contribution to thermal resistance in addition to the contribution of mass fluctuation scattering. The data of Walker and Fairbank (WF) are in the temperature range 1.2°K $\leq T \leq 2.0$°K and the concentration range, $x<3\%$, where the umklapp processes are still competitive with the scattering due to isotopic impurities. In this paper we report thermal conductivity measurements in mixtures of each helium isotope in the other at

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