

## Thermal Conductivity of Isotopic Mixtures of Solid Helium\*

EDWARD J. WALKER AND HENRY A. FAIRBANK  
*Laboratory of Marine Physics, Yale University, New Haven, Connecticut*  
 (Received December 2, 1959)

The thermal conductivity of solid  $\text{He}^4$  containing zero, 0.56, 1.38, and 2.8  $\text{He}^3$  has been measured as a function of temperature from 1.1 to 2.1°K. For pure  $\text{He}^4$ , the results are in satisfactory agreement with those of Webb, Wilkinson, and Wilks. The addition of 1.38%  $\text{He}^3$ , at a sample density  $\rho=0.208 \text{ g cm}^{-3}$  caused a decrease in thermal conductivity by a factor of 5 at 1.1°K. The additional thermal resistance caused by adding  $\text{He}^3$  was substantially independent of temperature, rather than proportional to temperature, as would be expected from isotropically distributed point scatterers. Agreement with Klemens' theory in magnitude and temperature dependence can be obtained by assuming the  $\text{He}^3$  to be arranged on lines in the solid. Possible mechanisms to explain these results are discussed.

### I. INTRODUCTION

**I**n a dielectric solid, heat is transported entirely by quantized lattice waves, or phonons. Thermal resistance arises from any irregularities in the crystal which scatter the phonons. The thermal resistance of a perfectly regular, unbounded lattice would be zero, were it not for the "umklapp" processes described by Peierls,<sup>1</sup> in which phonons interacting through the anharmonicity of the interatomic forces in real crystals are scattered from the lattice in a process similar to Bragg reflection; this scattering introduces a thermal resistance which Peierls showed to be given, at low temperatures, by  $W_U=AT^\nu \exp(-\theta/bT)$ , where  $T$  is the absolute temperature,  $\theta$  the Debye temperature,  $b$  is approximately 2,  $\nu$  is a constant of order unity, and  $A$  is a constant characteristic of the material.

It was not until the measurements of Berman<sup>2</sup> and of Berman, Simon, and Wilks<sup>3</sup> in 1951 that the exponential dependence was observed. At present the exponential variation has been found in sapphire,<sup>2</sup> quartz,<sup>2</sup> lithium fluoride,<sup>4</sup> and solid helium,<sup>5</sup> but not in other dielectric crystals such as Ge,<sup>6,7</sup> Si,<sup>6</sup> KCl,<sup>8</sup> etc. It has been suggested recently by several authors<sup>4,9,10</sup> that in the crystals where this expected rapid change of thermal conductivity with temperature has not been found, the irregularities in the crystal lattice produced by the naturally-occurring distribution of isotopes may scatter phonons sufficiently to mask the effect of "umklapp" processes. Experimental support for this hypothesis has been reported by Geballe and Hull,<sup>11</sup> who have found that the thermal resistance of an

isotopically-enriched specimen of germanium (96%  $\text{Ge}^{74}$ ) is substantially lower than that of a specimen of natural germanium. This thermal resistance of the  $\text{Ge}^{74}$  specimen did not, however, show the exponential temperature dependence predicted for a pure crystal by Peierls' theory.

Solid helium was chosen as a highly suitable dielectric in which to investigate this isotope effect for several reasons: (a) specimens of any isotopic concentration including pure  $\text{He}^3$  and pure  $\text{He}^4$  can be prepared, (b) helium can easily be prepared in great chemical purity, (c) the temperature dependence of the thermal conductivity characteristic of "umklapp" processes had previously been found<sup>5</sup> in pure  $\text{He}^4$ .

### II. APPARATUS

The apparatus used in these measurements is shown schematically in Fig. 1. The cylindrical sample  $S$ , 0.57-cm diameter by 1.2 cm long, was contained in a stainless-steel tube  $T$ , with walls 0.77 mm thick, the ends of which were closed by copper blocks silver-soldered to  $T$ . Copper fins were silver-soldered to tube  $T$  about 0.3 cm apart. Carbon resistor thermometers  $A$ ,  $B$ ,  $C$ ,  $D$ , and  $E$  (Allen-Bradley 0.1 watt, 57 ohm)

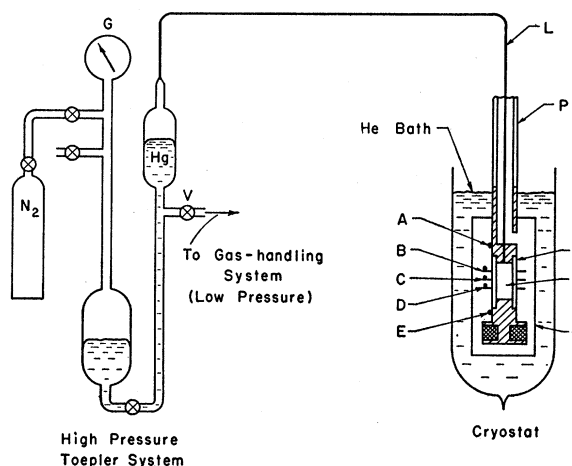


FIG. 1. Schematic drawing of the apparatus.

\* Supported by the Office of Naval Research.

<sup>1</sup> R. Peierls, *Ann. Physik* **3**, 1055 (1929).

<sup>2</sup> R. Berman, *Proc. Roy. Soc. (London)* **A208**, 90 (1951).

<sup>3</sup> R. Berman, F. E. Simon, and J. Wilks, *Nature* **168**, 277 (1951).

<sup>4</sup> R. Berman, E. L. Foster, and J. M. Ziman, *Proc. Roy. Soc. (London)* **A237**, 344 (1956).

<sup>5</sup> F. J. Webb, K. R. Wilkinson, and J. Wilks, *Proc. Roy. Soc. (London)* **A214**, 546 (1952).

<sup>6</sup> H. M. Rosenberg, *Proc. Phys. Soc. (London)* **A67**, 837 (1954).

<sup>7</sup> G. K. White and S. B. Woods, *Can. J. Phys.* **33**, 58 (1955).

<sup>8</sup> G. A. Slack, *Phys. Rev.* **105**, 832 (1957).

<sup>9</sup> I. Pomeranchuk, *J. Phys., U.S.S.R.* **6**, 237 (1942).

<sup>10</sup> G. A. Slack, *Phys. Rev.* **105**, 829 (1957).

<sup>11</sup> T. H. Geballe and G. W. Hull, *Phys. Rev.* **110**, 773 (1958).

were soldered to the upper copper block, to each fin along the tube, and to the lower copper block respectively, permitting the measurement of the temperature at each of these five points. Resistances were measured on a dc Wheatstone bridge; the thermometers were calibrated against the vapor pressure of the helium bath. Power was supplied electrically to a heater coil wound upon the copper block closing the lower end of the sample tube. This power flowed through the sample and sample tube and out into the liquid helium bath, the temperature of which could be maintained at any desired temperature between 1.1 and 4.2°K by a simple bath pressure controller described elsewhere.<sup>12</sup> The sample tube assembly was enclosed in a vacuum can *C*, which was evacuated through the pumping line *P*. All electrical leads and the fill line were brought up to room temperature through the pumping line *P*. The electrical leads were thermally locked to a copper block extending into the helium bath, using epoxy resin. The sample was introduced through a 0.005-in. diameter cupro-nickel fill line *L*. The pressure necessary to solidify the helium (from 25 to 150 atmospheres) was obtained from a cylinder of nitrogen gas; the nitrogen was separated from the helium sample by a stainless-steel high-pressure Toepler pump containing mercury, as shown schematically in Fig. 1. Pressures were measured on a precision Bourdon gauge<sup>13</sup> *G*, and corrected for the difference in pressure due to the mercury in the high-pressure Toepler. The isotopic mixtures were prepared in a glass gas-handling system (not shown) by mixing appropriate amounts of pure He<sup>3</sup> and He<sup>4</sup>, and the concentrations determined from the measured values of the volume, pressure and tem-

perature of each component. The concentrations so determined were in satisfactory agreement with values determined mass-spectrometrically.

### III. EXPERIMENTAL PROCEDURE

The sample of solid helium was prepared in a manner similar to that described by Webb, Wilkinson, and Wilks.<sup>5</sup> With the mercury lowered in the right-hand column of the high-pressure Toepler (Fig. 1) the requisite amount of helium gas was admitted through valve *V* to the upper Toepler volume. The sample volume *S* was filled with liquid by condensation through fill line *L*. Valve *V* was closed and by admitting high pressure gas to the left hand column of the Toepler the pressure of the sample was adjusted to the desired value. The bath temperature was then adjusted to be about 0.05°K above the melting point<sup>14</sup> for the sample pressure. After waiting ten minutes to insure reaching thermal and pressure equilibrium, the bath temperature was rapidly lowered, causing solidification to take place in the fill line, thus blocking it so that the sample solidified at constant volume. Several investigators<sup>15,16</sup> have reported that this method produces one or a very few large crystals of solid helium. Measurements were taken at the starting pressures, densities, and concentrations shown in Table I. The data at zero He<sup>3</sup> concentration were taken to permit comparison with the results of Webb, Wilkinson, and Wilks on pure He<sup>4</sup>.

The thermal conductivity  $\kappa$  of the sample was calculated from the equation  $Q - Q_t = Q_s = \kappa A \Delta T / \Delta x$ , where  $Q$  is the total heat flow,  $Q_t$  is the heat flow through the sample tube, calculated from measurements made in a preliminary run on the empty tube,  $Q_s$  is the heat flow through the sample,  $A$  is the sample cross-sectional area, and  $\Delta T$  is the temperature difference between adjacent thermometers  $\Delta x$  apart. In no case did  $Q_t$  exceed 10% of  $Q$ , and it was usually less than 2% of  $Q$ .  $\Delta T$  varied between 10 and 100 millidegrees.

### IV. EXPERIMENTAL RESULTS

Figure 2 gives the results of our measurements on pure He<sup>4</sup> (solid circles) together with smoothed data

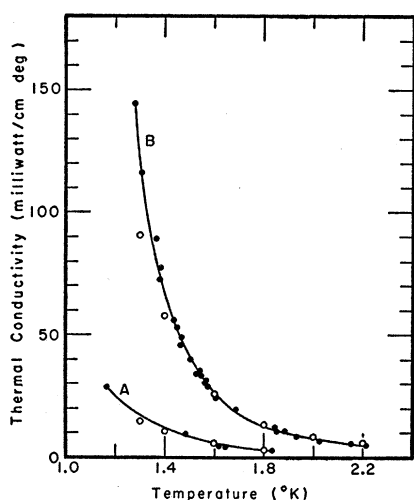


FIG. 2. The thermal conductivity of pure solid He<sup>4</sup> as a function of temperature. The present results (solid circles) are compared with the measurements of Webb, Wilkinson, and Wilks (reference 5) (open circles). Curve *A* is for a helium density of 0.194 g/cm<sup>3</sup>; curve *B*, a density of 0.208 g/cm<sup>3</sup>.

<sup>12</sup> E. J. Walker, Rev. Sci. Instr. **30**, 834 (1959).

<sup>13</sup> Heise Bourdon Tube Company, Inc., Brook Road, Newtown, Connecticut.

TABLE I. Conditions under which measurements were made.

Pressure, atm	Density solid He <sup>4</sup> , g cm <sup>-3</sup>	Atomic percent He <sup>3</sup>
56.2	0.194	0.00
96.0	0.208	0.00 0.56 1.38 2.80
131.0	0.218	0.56

<sup>14</sup> C. A. Swenson, Phys. Rev. **89**, 538 (1953).

<sup>15</sup> W. H. Keesom, *Helium* (Elsevier Publishing Company, Amsterdam, 1942).

<sup>16</sup> D. G. Henshaw, Phys. Rev. **109**, 328 (1958).

of Webb, Wilkinson, and Wilks<sup>5</sup> (open circles) at comparable pressures. It is seen that the two sets of measurements are in substantial agreement. The rapid variation of thermal conductivity with temperature is evident; these data, when replotted as  $\log(\kappa)$  vs  $1/T$  yield good straight lines in the temperature range covered (approximately  $\theta/13$  to  $\theta/24$ ) in agreement with the prediction of Peierls for umklapp resistivity. On this type of plot a slight systematic deviation between our results and those of Webb, Wilkinson, and Wilks is evident as a small difference in slope.

Figure 3 shows the dependence of thermal resistivity upon temperature for four values of He<sup>3</sup> concentration, at a fixed starting pressure of 96 atmospheres ( $\rho=0.208$  g/cm<sup>3</sup>). Smoothed values of these data are given in Table II.

Figure 4 compares the thermal resistivity of an 0.56% He<sup>3</sup>-He<sup>4</sup> solid mixture at a starting pressure of 96 atmospheres ( $\rho=0.208$  g/cm<sup>3</sup>) with that of the same mixture at a starting pressure of 131 atmospheres ( $\rho=0.218$  g/cm<sup>3</sup>).

## V. DISCUSSION

### (a) Theory Using the Additive Resistance Approximation

To a first approximation, the resistivities due to individual scattering processes are additive, i.e.,

$$W = W_u + W_1 + W_2 + \dots, \quad (1)$$

where  $W$  is the total resistivity,  $W_u$  is the intrinsic resistivity due to umklapp scattering, and  $W_i$  is the resistivity due to the  $i$ th scattering process. In the temperature range studied, boundary scattering is inappreciable. As a first step in interpreting the data, we assume that Eq. (1) holds, with only one scattering process,  $W_1$ , in addition to  $W_u$ , and that  $W_u$  is the same as the resistivity  $W_0$  of pure solid He<sup>4</sup> at the same starting pressure (i.e., it is assumed that the small amount of He<sup>3</sup> added does not change the Debye  $\theta$  of the lattice). Curves 1, 2, and 3 in Fig. 5 show values of  $(W - W_u)/n$  for the three concentrations  $n$  used, calculated from the data in Table II, as a function of tem-

TABLE II. Thermal resistivity of mixtures of solid He<sup>3</sup> and He<sup>4</sup> at density  $\rho=0.208$  g cm<sup>-3</sup>. Values in watts<sup>-1</sup> cm °K. Subscripts on  $W$  refer to He<sup>3</sup> concentrations in atomic percent.

$T$ (°K)	$W_0$	$W_{0.56}$	$W_{1.38}$	$W_{2.80}$
1.1	1.8	18.6	36.4	55.2
1.2	4.4	21.4	39.4	58.8
1.3	8.2	25.7	44.1	63.8
1.4	14.6	32.2	51.3	71.4
1.5	24.7	41.4	61.3	82.2
1.6	38.4	53.6	74.0	96.6
1.7	55.1	69.7	90.5	114.4
1.8	76.4	90.4	111.7	135.7
1.9	103.5	116.3	137.7	161.3
2.0	135.3	145.3	167.5	189.8
2.1	169.0	175.8	200.2	219.7

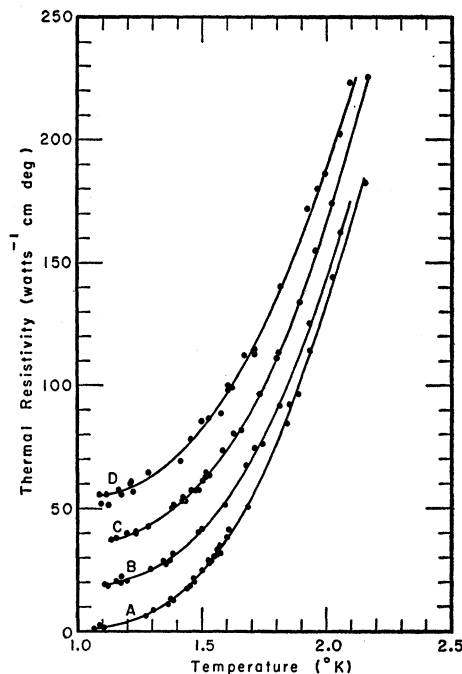


FIG. 3. The thermal resistivity of solid helium as a function of temperature for several He<sup>3</sup> concentrations at a density of 0.208 g/cm<sup>3</sup>. Curve A, pure He<sup>4</sup>; curve B, 0.56% He<sup>3</sup>; curve C, 1.38% He<sup>3</sup>; curve D, 2.80% He<sup>3</sup>.

perature. It is seen that the increase in resistivity caused by the addition of He<sup>3</sup> is roughly independent of temperature. If the He<sup>3</sup> atoms were behaving as isotropically distributed point scatterers, according to the theory of Klemens<sup>17,18</sup> they should produce an increase

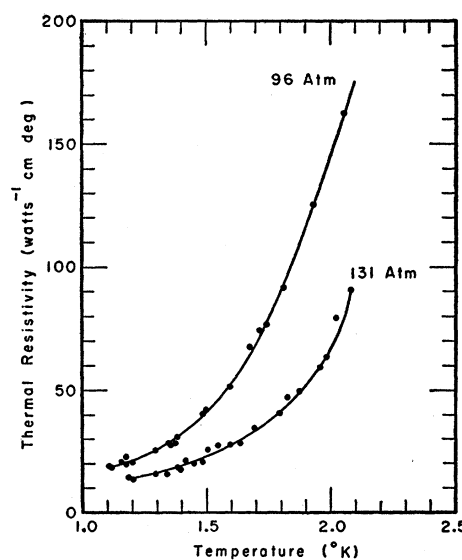


FIG. 4. The thermal resistivity of a He<sup>3</sup>-He<sup>4</sup> solid mixture containing 0.56% He<sup>3</sup> as a function of temperature for two different starting pressures.

<sup>17</sup> P. G. Klemens, Proc. Roy. Soc. (London) A208, 108 (1951).

<sup>18</sup> P. G. Klemens, Proc. Roy. Soc. (London) A68, 1113 (1955).

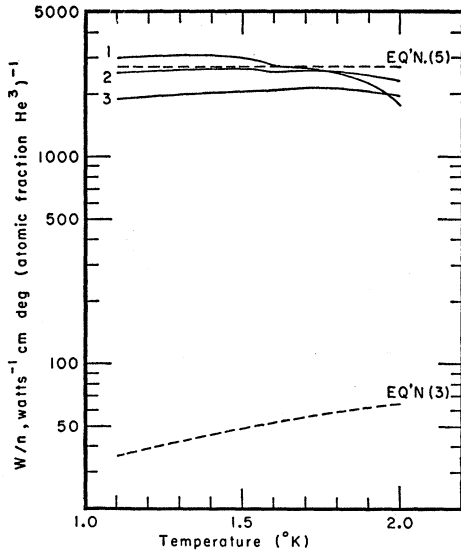


FIG. 5. The isotope contribution to the thermal resistance of solid helium divided by the atomic fraction of  $\text{He}^3$  present as a function of temperature. In curves 1, 2, and 3,  $W - W_u/n$  calculated from the experimental results is plotted for  $n = 0.56, 1.38,$  and  $2.80\%$   $\text{He}^3$ , respectively. The upper and lower dashed curves are obtained from Klemens' theory for line and point scattering, respectively.

in resistivity given, for small  $n$ , by

$$\frac{W_P}{T} = \frac{10^7 \pi^2 a^3 b n}{0.90 h v^2} \left( \frac{\Delta M}{M} \right)^2, \quad (2)$$

where  $W_P$  is the point scattering resistance ( $\text{watts}^{-1} \text{ } ^\circ\text{K cm}$ ),  $a^3$  is the volume per unit cell,  $n$  the point defect concentration (atomic fraction),  $\Delta M$  is the difference between the mass  $M$  of a normal unit cell and that of a unit cell containing an impurity atom,  $v$  is the velocity of sound, and  $b$  is the number of atoms per unit cell (2 for  $\text{He}^4$ ). Evaluating this for the case of small concentrations of  $\text{He}^3$  in solid  $\text{He}^4$ , and using  $v = 3.2 \times 10^4 \text{ cm sec}^{-1}$ ,  $a^3 = 6.38 \times 10^{-28} \text{ cm}^3$ ,  $(\Delta M/M)^2 = \frac{1}{4}$ , we find

$$W_P/n = 32.5T. \quad (3)$$

A plot of Eq. (3) is shown on Fig. 5; it is seen that the increase in resistivity caused by adding  $\text{He}^3$  disagrees both in magnitude and in temperature dependence with that predicted by Eq. (3) for isotropically distributed point scatterers.

The above results lead one to look for an imperfection which produces a resistance independent of temperature. Klemens<sup>19</sup> points out that long cylinders whose diameters are small compared with the phonon wavelengths involved will produce a thermal resistance independent of temperature. (This can be seen from the kinetic-theory formula  $\kappa = \frac{1}{3} C v l$ ; at low temperatures,  $C \propto T^3$ ,  $v = \text{constant}$ , and a classical calculation of

scattering from a thin cylinder shows  $l \propto \omega^{-3}$ . But the dominant phonon wavelength is given approximately by  $\hbar\omega = kT$ , so  $l \propto T^{-3}$ , and  $\kappa$  is thus independent of temperature.)

If the assumption is made that the  $\text{He}^3$ , instead of being distributed isotropically, is arranged in the  $\text{He}^4$  matrix in lines oriented at random, it can be shown, using the formula given by Klemens for the relaxation time due to a linear mass inhomogeneity of this type [Eq. (65), reference 18], and his general formula for thermal conductivity in a crystal [Eq. (67), reference 17], that

$$\frac{W_L}{n} = \frac{0.55 \times 3\pi^2 a^2 b \times 10^7}{2k v B} \left( \frac{\Delta M}{M} \right)^2, \quad (4)$$

where

$$B = \int_0^1 \frac{e^x x^4 dx}{(e^x - 1)^2} + \int_1^\infty \frac{x e^x dx}{(e^x - 1)^2} = 1.3578,$$

and  $\Delta M$  is the change in mass of a unit cell due to the presence of a line of impurity atoms.

It is seen that Eq. (4) gives a temperature-independent isotope resistivity, in agreement with our experimental results. Furthermore, the order of magnitude of the resistance predicted agrees with experiment. Close agreement with the experimental results is obtained if one assumes all the  $\text{He}^3$  to be arranged in lines oriented at random and  $(\Delta M/M) = \frac{1}{4}$ . (This assumes two  $\text{He}^3$  atoms per unit cell in the line.) At a density of  $0.208 \text{ g cm}^{-3}$ , Eq. (4) then reduces to

$$W_L/n = 2.70 \times 10^3 \text{ watts}^{-1} \text{ cm } ^\circ\text{K}. \quad (5)$$

A comparison of the experimental results with the prediction of Eq. (5) is given in Fig. 5. It should be emphasized that this close agreement in magnitude is the result of making favorable assumptions regarding the orientation of the lines and the number of atoms included in a line per unit cell. If the lines were oriented normal to the heat flow, instead of at random, the predicted thermal resistivity would be larger by 1/0.55; if the lines included only one scattering atom per unit cell, the predicted thermal resistivity would be smaller by a factor of 4.

TABLE III. Values of  $I_1(R_1)$ .

$R_1$	$I_1(R_1)$
0.01	0.010348
0.05	0.050623
0.10	0.098600
0.50	0.40993
1.00	0.68229
2.00	1.03199
3.00	1.25180
5.00	1.51786
7.50	1.70511
10.00	1.82024
50.00	2.19159
$\infty$	2.3169

<sup>19</sup> P. G. Klemens, *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1958), Vol. 7, p. 1.

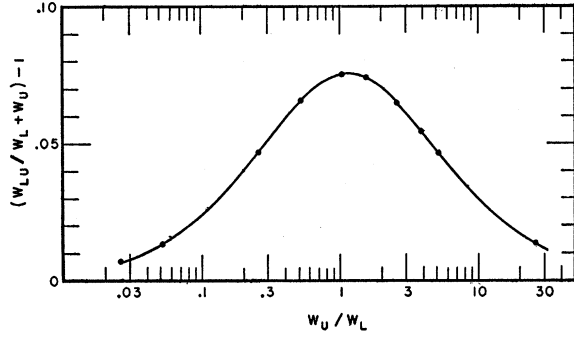


FIG. 6. A plot of Eq. (8). See text.

Comparing Eqs. (3) and (5), it is seen that  $W_L/W_P = 83/T$ ; thus the agglomeration of  $\text{He}^3$  into lines increases the isotope resistance by almost two orders of magnitude at 1°K.

### (b) More Exact Theory of Klemens

As pointed out by Klemens,<sup>19</sup> Slack,<sup>8,10</sup> and others, the additive resistance approximation, Eq. (1), is rather inaccurate when more than one term on the right is large. The total resistivity  $W_{Lu}$  due to line scattering and umklapp scattering is obtained by adding reciprocal mean free paths,

$$1/l_{Lu} = 1/l_L + 1/l_u, \quad (6)$$

and then substituting in the formula

$$1/W_{Lu} = \kappa_{Lu} = \frac{1}{3} \int [(\partial/\partial T)E(k, T)] l_{Lu}(k) v(k) dk. \quad (7)$$

The result is, for the case of line and umklapp scattering, using Klemens' cutoff approximation,<sup>19</sup>

$$\kappa_{Lu} = Al_{u0} \left[ 0.3172 \frac{R_1}{1+R_1} + I_1(R_1) \right],$$

where

$$A = 4\pi K^4 T^3 / v^2 h^3,$$

$K$  = Boltzmann constant;  $h$  = Planck constant,

$$l_{L0} = \frac{h^3 v^3}{0.55 \times 6\pi^3 K^3 T^3 a^2 b n} \left( \frac{M}{\Delta M} \right)^2,$$

$$l_{u0} = \frac{U}{a} \left( \frac{v h}{2\pi K T} \right)^2 e^{\theta/2T},$$

$$R_1 \equiv \frac{l_{L0}}{l_{u0}} = \frac{W_u C}{W_L B},$$

$$B = \int_0^1 \frac{x^4 e^x dx}{(e^x - 1)^2} + \int_1^\infty \frac{x e^x dx}{(e^x - 1)^2} = 0.3172 + 1.0407 = 1.3578,$$

$$C = \int_0^1 \frac{x^4 e^x dx}{(e^x - 1)^2} + \int_1^\infty \frac{x^2 e^x dx}{(e^x - 1)^2} = 0.3172 + 2.3169 = 2.6341,$$

$$I_1(R_1) = \int_1^\infty \frac{x^2 e^x dx}{(1+x/R_1)(e^x - 1)^2},$$

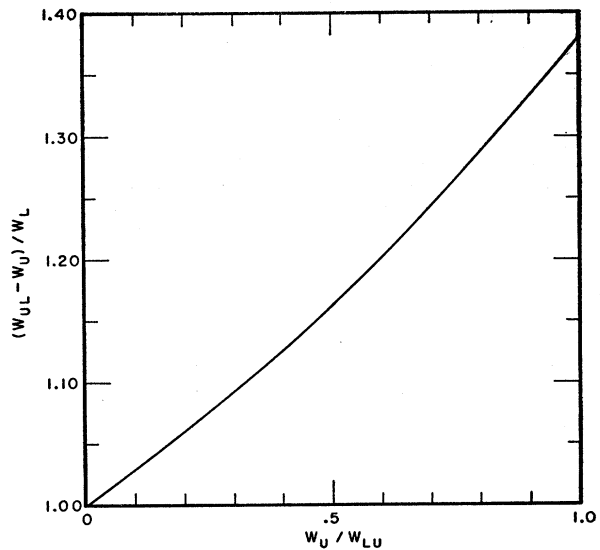
$U$  = constant.

Following Slack,<sup>8</sup> we may calculate  $W_{Lu}/(W_L + W_u)$ ; the result is

$$\frac{W_{Lu}}{W_L + W_u} = \frac{B(1+R_1)}{[1 + (B/C)R_1][0.3172 + (1+1/R_1)I_1(R_1)]}. \quad (8)$$

The integral  $I_1(R_1)$  was evaluated on an IBM-650 computer; the results are given in Table III. A plot of  $W_{Lu}/(W_L + W_u) - 1$  as a function of  $W_u/W_L$  is given in Fig. 6. The maximum correction occurs around  $W_u/W_L = 1.02$  and amounts to 7.6%, as against 28% for the case of point plus umklapp scattering calculated by Slack. This is reasonable, as the frequency dependence of  $l_L$ ,  $l_L \propto \omega^{-3}$ , is closer to that of umklapp scattering,  $l_u \propto \omega^{-2}$ , than is that of point scattering,  $l_P \propto \omega^{-4}$ .

For purposes of comparing experimental data with theory, one requires the quantity  $(W_{Lu} - W_u)/W_L$  as a function of the experimentally observable quantity  $W_u/W_{Lu}$ ; the calculation is similar to that leading to Eq. (8), and the result is given graphically in Fig. 7. This graph can be looked upon as giving the relative enhancement of line scattering due to the presence of


 FIG. 7. Auxiliary graph for calculating  $W_L$  from the experimentally-determinable quantities  $W_{Lu}$  and  $W_u$ .

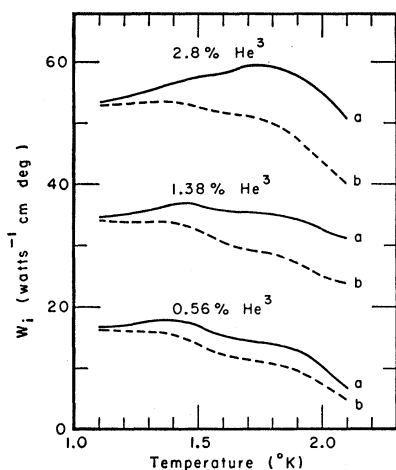


FIG. 8. The isotope contribution to the thermal resistance for three different  $\text{He}^3$  concentrations as a function of temperature, at a density of  $0.208 \text{ g/cm}^3$ . Curves *a* are obtained from the experimental data by using the additive resistance approximation ( $W_L = W - W_u$ ). Curves *b* give  $W_L$  as corrected using Fig. 7.

umklapp scattering. If one assumes that the umklapp resistance  $W_u$  is unaffected by the (small) quantity of  $\text{He}^3$  added, then  $W_u$  is equal to  $W_0$ , the resistance of pure solid  $\text{He}^4$ , and  $W_{Lu} = W$  is the resistance of the mixture of solid  $\text{He}^4$  and  $\text{He}^3$ . Curves (a) in Fig. 8 are the same data as curves 1-3, Fig. 5, replotted on a linear scale, i.e., curves (a) give  $W_{Lu} - W_0$  as a function of  $T$  for the three  $\text{He}^3$  concentrations measured. Curves (b), Fig. 8, gives  $W_L$  corrected using Fig. 7. Although curves (b) show some decrease at higher temperatures, the general trend of the data is not changed substantially by applying Klemens' more exact theory; thus the conclusion that the added isotope resistance is approximately independent of temperature still holds. It should be mentioned that near  $1^\circ\text{K}$ ,  $W_L$  makes the major contribution to the thermal resistance, while at  $2^\circ\text{K}$ ,  $W_u$  contributes the major part. Thus the precision of  $W_L$  is lower at the higher temperatures. In this connection it is worth noting that the differences between individual curves in Fig. 8 are more nearly independent of temperature than are the curves themselves. This type of behavior would be expected if  $W_L$  were indeed independent of temperature and  $W_u$  were slightly in error at the higher temperatures, since an error in  $W_u$  affects all curves in Fig. 8 equally.

The dependence of  $W_L$  on  $\text{He}^3$  concentration  $n$  at  $1.1^\circ\text{K}$  where the precision of the data is highest is shown in Fig. 9. The experimental results are compared with the predictions of Eq. (5).  $W_L$  is nearly proportional to the  $\text{He}^3$  concentration at low concentrations but deviates from proportionality at the highest point. On the assumption that the isotope resistance arises from agglomeration of  $\text{He}^3$  atoms into lines, this suggests a saturation effect at higher concentrations and would be consistent with Klemens' theory if an in-

creasing number of  $\text{He}^3$  atoms were remaining in solid solution and acting as point scatterers. As pointed out previously, the scattering resistance due to isolated atoms is about 80 times smaller at  $1.1^\circ\text{K}$  than that for the same atoms arranged on lines.

### (c) Results at Higher Density

From the measurements made on a  $0.56\%$   $\text{He}^3$  mixture at a density of  $0.218 \text{ g/cm}^3$ , values of  $W_L$  were calculated using both the additive resistance approximation and the more exact treatment given in the previous section. The results are given in Fig. 10 in curves *a* and *b*, respectively. In these calculations the experimental values of Webb, Wilkinson, and Wilks<sup>5</sup> for thermal resistance of pure  $\text{He}^4$  were used. In accordance with expectations, the magnitude of the isotope resistance is decreased at higher density and the temperature dependence is substantially the same.

## VI. POSSIBLE MECHANISMS PRODUCING LINEAR INHOMOGENEITIES

### (a) Phase Separation

In view of the fact that the crystal structure of solid  $\text{He}^4$  is hexagonal close packed, while that of  $\text{He}^3$ , in the pressure range investigated, is body-centered cubic, a two-phase, mixed crystal region would be expected at some intermediate concentrations. In the mixed-crystal region, it is possible to have linear (needle-shaped) crystals of one component freezing out in a matrix of the other component. Our data indicate that linear arrays are formed at  $\text{He}^3$  concentrations as small as  $0.56\%$ ; because of the chemical similarity of  $\text{He}^3$  and  $\text{He}^4$ , it is unlikely that the solid-solution range is as small as this would indicate. For this reason, phase separation as an explanation of our results is unlikely. Parenthetically, it might be added that a separation

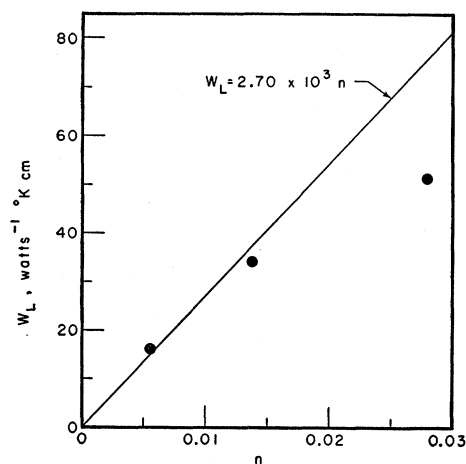


FIG. 9. Isotope resistance at  $1.1^\circ\text{K}$  as a function of concentration  $n$ . The straight line is a plot of Eq. (5).

into two phases occurs in liquid He<sup>3</sup>-He<sup>4</sup> mixtures, but only below about 0.9°K.<sup>20</sup>

### (b) Decoration of Dislocation Lines

A more probable explanation of the production of these linear arrays is the adsorption of He<sup>3</sup> atoms on dislocation lines. One might reasonably ask why an attractive force should exist between a dislocation in He<sup>4</sup> and a solute He<sup>3</sup> atom, since the radii of the atoms concerned are, in the gaseous state, virtually identical. A possible answer lies in the differing zero-point energies of the two atoms; in a given potential well, He<sup>3</sup> will, because of its smaller mass, have a greater zero-point energy than He<sup>4</sup>. In a mixture, therefore, motion of He<sup>3</sup> atoms toward the regions of expansive strain around dislocations will lower the total lattice zero-point energy. Donohue,<sup>21</sup> using data of Henshaw<sup>22</sup> and Schuch,<sup>23</sup> has recently compared nearest-neighbor spacings in He<sup>3</sup> and He<sup>4</sup> at the same pressure, and found the spacings in He<sup>3</sup> larger than those in He<sup>4</sup> by 2.3%, due to the larger zero-point energy in He<sup>3</sup>. This is equivalent to having a larger effective radius for He<sup>3</sup>, and implies the existence of a potential energy between solute He<sup>3</sup> atoms and regions of strain in the He<sup>4</sup> matrix. Because of the constant-volume method in which our crystals were grown, a reasonably large deformation takes place during crystal growth; the deformation occurring during density equalization is as high as 7%. Thus it is likely that a large density of dislocations will be produced in the crystal. If all the He<sup>3</sup> present at approximately 1% concentration is adsorbed on dislocation lines with one He<sup>3</sup> atom per atomic plane of dislocation, one would expect a dislocation density of about  $3 \times 10^{12}$  lines per cm<sup>2</sup>, a high, but possible, density. If the lines contained more than one He<sup>3</sup> atom in a cross section of the line, the density of dislocations necessary would be proportionately smaller.

### VII. OTHER INTERPRETATIONS OF THE DATA

A word of caution is in order; the calculation of thermal conductivity is a difficult problem, and present solutions are approximate at best.<sup>24</sup> Callaway,<sup>25</sup> Ziman,<sup>26</sup>

<sup>20</sup> G. K. Walters and W. M. Fairbank, *Phys. Rev.* **103**, 262 (1956).

<sup>21</sup> J. Donohue, *Phys. Rev.* **114**, 1009 (1959).

<sup>22</sup> D. G. Henshaw, *Phys. Rev.* **109**, 328 (1958).

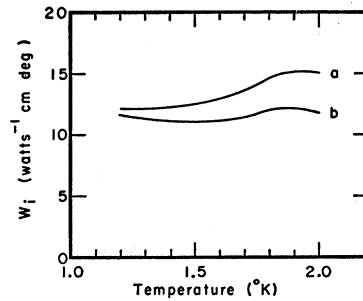
<sup>23</sup> A. F. Schuch, *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 79.

<sup>24</sup> J. M. Ziman, *Nuovo cimento Suppl.* **7**, 353 (1958).

<sup>25</sup> J. Callaway, *Phys. Rev.* **113**, 1046 (1959).

<sup>26</sup> J. M. Ziman, *Can. J. Phys.* **34**, 1256 (1956).

FIG. 10. The isotope contribution to the thermal resistance for a He<sup>3</sup> concentration of 0.56%, at a density of 0.218 g/cm<sup>3</sup>, as a function of temperature. Curve *a* is obtained from the experimental data by using the additive resistance approximation ( $W_i = W - W_u$ ). Curve *b* gives  $W_L$  as corrected using Fig. 7.



and Carruthers<sup>27</sup> have presented alternative approaches to that of Klemens. We have used the theory of Klemens since it is in a form permitting direct quantitative comparison with our results. Our tentative conclusion that He<sup>3</sup> is not homogeneously distributed in these dilute mixtures is predicated on the validity of Klemens' theory. Objections to Klemens' cutoff approximation, in particular, have been made, and it is possible that a more refined theory may be able to explain the present experimental results without the necessity of assuming nonhomogeneous distribution of the He<sup>3</sup> atoms. Further measurements extending to lower temperatures would be useful and are in progress. Independent experimental evidence as to the distribution of the isotopes in these solid mixtures would be helpful.

### VIII. CONCLUSIONS

The addition of small amounts of He<sup>3</sup> to solid He<sup>4</sup> produces a substantial increase in the thermal resistance, approximately independent of temperature, in the range of the measurements, 1–2°K. This result is in good agreement with the theory of Klemens, on the assumption that the He<sup>3</sup> is aggregated along lines in the crystal. Of the possible mechanisms considered which might produce this effect, the decoration of dislocation lines by He<sup>3</sup> atoms seems the more likely. It is also possible that other interpretations of the data not involving aggregation of the He<sup>3</sup> atoms may be possible. Further experimental and theoretical work is required.

### IX. ACKNOWLEDGMENTS

The authors are indebted to Professor David Mintzer, Professor C. T. Lane, and Professor Lars Onsager for stimulating discussions and advice during the course of this work. Thanks are also due to Mrs. Lois Frampton for carrying out the numerical integration of  $I_1(R_1)$ .

<sup>27</sup> P. Carruthers (to be published).