

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

Isotope	Molar volume (cc)	Θ ($^{\circ}$ K)	A_1 ($\frac{\text{cm deg}}{\text{watt}}$)	T_1 deg K	T_2 deg K	A_2 ($\frac{\text{cm deg}}{\text{watt}}$)
He ³	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
	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 He³ 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 meas-

¹⁹ R. Berman and S. J. Rogers, Phys. Letters 9, 115 (1964).

ured 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}

ACKNOWLEDGMENTS

The authors wish to thank Miss Ann McNeal for her assistance in the reduction of some of the data, Dan Deis for his aid in taking the data, and Dr. P. G. Klemens for a number of illuminating discussions.

²⁰ J. H. Vignos and H. A. Fairbank, in *Proceedings of the Eighth International Conference on Low Temperature Physics*, edited by R. O. Davies (Butterworth's Scientific Publications Ltd., London, 1963), p. 31.

²¹ F. P. Lipschultz and D. M. Lee, Phys. Rev. Letters 14, 1017 (1965).

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 He³ in He⁴ 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 $x(1-x)$ 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 Callaway² and the subsequent interpretation by

Klemens and Maradudin³ 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 $1.2^{\circ}\text{K} \leq T \leq 2.0^{\circ}\text{K}$ 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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¹ E. J. Walker and H. A. Fairbank, Phys. Rev. 118, 913 (1960), hereafter WF.

² J. Callaway, Phys. Rev. 122, 5787 (1961).

³ P. G. Klemens and A. A. Maradudin, Phys. Rev. 123, 804 (1961).

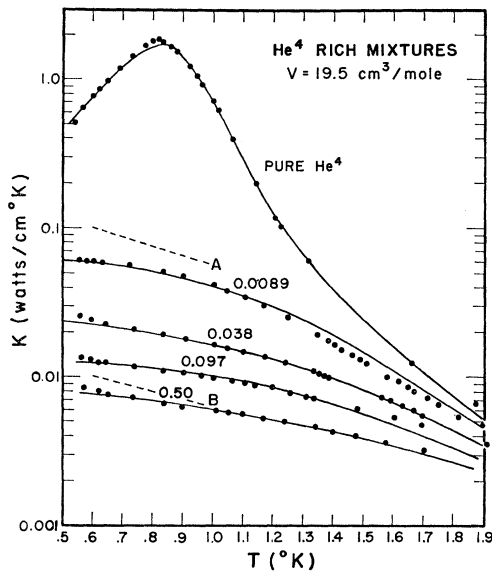


FIG. 1. The thermal conductivity of pure He^4 and He^4 with He^3 concentrations of 0.0089, 0.038, 0.097, 0.50 at molar volume $19.5 \text{ cm}^3/\text{mole}$. The data are shown by the black circles; the fits to the data using Eq. (2) are shown by the solid lines. The dotted lines A and B are the calculated thermal conductivity in the Ziman limit using the Klemens result for mass-fluctuation cross section.

concentrations, x , of 1, 4, 10, and 50% at two molar volumes, $V=19.5 \text{ cm}^3/\text{mole}$ and $V=20.2 \text{ cm}^3/\text{mole}$, over the temperature range $0.5^\circ\text{K} \leq T \leq 2.0^\circ\text{K}$. The additional low-temperature data ($T < 1^\circ\text{K}$) available to us permit a more extensive interpretation of the role

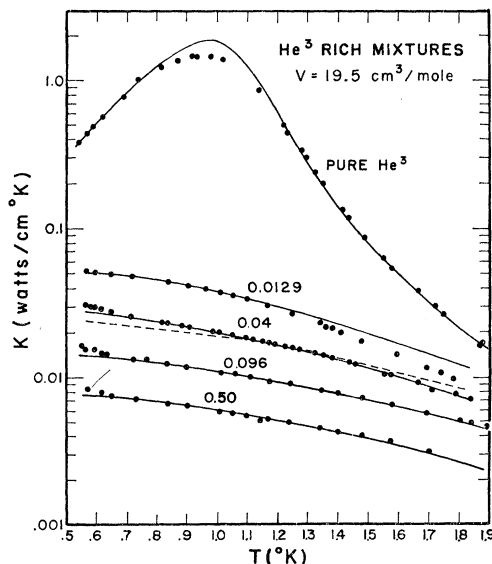


FIG. 2. The thermal conductivity of pure He^3 and He^3 with He^4 concentrations of 0.0129, 0.040, 0.096, 0.50 at molar volume $19.5 \text{ cm}^3/\text{mole}$. The data are shown by the black circles; the fits to the data using Eq. (2) are shown by the solid lines. The dotted line crossing the 0.040 data is the fit to that data using the values of the parameters B_N and A appropriate to the data at concentration 0.0129.

played by each of the scattering processes and in particular allows a more accurate appraisal of the magnitude of lattice distortion due to isotopic impurities. A preliminary report of some of this work was presented at the Ninth International Conference on Low Temperature Physics.⁴

The data at $v=19.5 \text{ cm}^3/\text{mole}$ have been analyzed using the phenomenological equation of Callaway.⁵ The cross section for phonon scattering from a lattice distortion is calculated. From the cross sections we estimate the atomic misfit in solid-helium mixtures. These results are compared with the predictions of the theoretical work of Klemens *et al.*⁶

RESULTS

The thermal-conductivity apparatus used and the data-taking procedures for this experiment were iden-

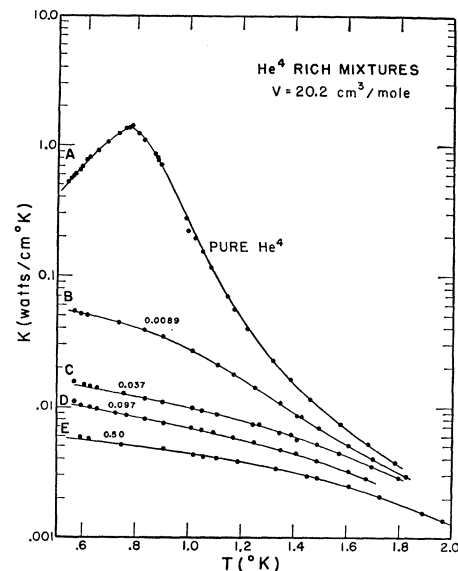


FIG. 3. The thermal conductivity of pure He^4 and He^4 with He^3 concentrations of 0.0089, 0.038, 0.097, 0.50 at molar volume $20.2 \text{ cm}^3/\text{mole}$.

tical to those reported in the previous paper.⁷ The experimental results are given in Figs. 1-4.

In the remainder of this paper we discuss in detail the data at $V=19.5 \text{ cm}^3/\text{mole}$. For all concentrations the $19.5 \text{ cm}^3/\text{mole}$ mixtures have a hexagonal close-packed (hcp) structure. Comparison of Figs. 1 and 2 with Figs. 3 and 4 shows that the gross features of the data in mixtures are the same at both molar volumes.

⁴ B. Bertman, Clark W. White, and Henry A. Fairbank, in *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt, D. V. Edwards, F. J. Milford, and M. Yagub (Plenum Press, Inc., New York, 1965).

⁵ J. Callaway, *Phys. Rev.* **113**, 1046 (1959).

⁶ P. G. Klemens, R. De Bruyn Ouboter, and C. Le Pair, *Physica* **30**, 1863 (1964).

⁷ B. Bertman, C. W. White, H. A. Fairbank, and Michael J. Crooks, *Phys. Rev.* **140**, 74 (1966), preceding paper, hereafter I.

When isotopic impurities are introduced into the host lattice the thermal conductivity exhibits the following qualitative behavior: (1) The addition of as little as 1% He³ into a He⁴ lattice depresses the conductivity by as much as a factor of 50. (2) At all concentrations in the temperature range $0.5^\circ\text{K} \leq T \leq 1.2^\circ\text{K}$ scattering of phonons by isotopic impurities dominates the other resistive processes (umklapp scattering and boundary scattering). Even at the lowest isotopic concentrations used (1%) and the lowest temperatures reached (0.5°K), boundary scattering does not make a major contribution to the thermal resistance. (3) For high temperatures, $T \approx 2^\circ\text{K}$, the thermal conductivity approaches the value expected from the scattering of phonons by umklapp processes. As we have discussed in I, the high-temperature region in both pure He³ and pure He⁴ shows behavior in good agreement with that expected by phonon scattering dominated by umklapp processes.

If the scattering of phonons in isotopic mixtures is the analogue of electron scattering in an alloy,⁸ the additional resistance to heat flow due to the introduction of an isotopic impurity concentration x should be proportional to $x(1-x)$. A plot of $W_i/x(1-x)$ versus T , where $W_i = K_i^{-1}$ is the thermal resistivity and $W_i = W - W_{\text{pure}}$, should yield a common curve for all mixtures. This quantity is plotted in Fig. 5. The expected concentration dependence is not observed, a conclusion reached earlier from the data of WF on He³-He⁴ mixtures. The data of WF have been analyzed by Callaway using the phenomenological equation developed by him to include the effect of normal scattering processes.² (These nonresistive scattering processes enhance the effectiveness of the resistive scattering mechanisms by redistributing momentum among the phonon modes; they do not themselves contribute directly to the resistance.) Callaway was able to obtain the suggested concentration dependence. We find that our data can be analyzed satisfactorily using Callaway's approach. However, we find: (1) The dominant scattering agent operative in these crystals is lattice distortion around the site of an isotopic impurity. (2) Phonon scattering due to lattice distortion depends more strongly on the concentration than $x(1-x)$.

If, (a) the scattering mechanism associated with the addition of an isotopic impurity is Rayleigh-like scattering from mass fluctuations,^{9,10} (b) the normal processes are infinitely rapid, and (c) other scattering mechanisms can be ignored, the thermal conductivity is given by the Ziman limit¹¹

$$K = [\hbar / (120\pi^2 v T A)] [1/x(1-x)], \quad (1)$$

where N is Avogadro's number, v is the velocity of

⁸ N. F. Mott and H. Jones, *Theory of Metals and Alloys* (Dover Publications, New York, 1958), p. 296.

⁹ P. G. Klemens, Proc. Roy. Soc. A208, 108 (1951).

¹⁰ P. Carruthers, Rev. Mod. Phys. 33, 92 (1961).

¹¹ J. M. Ziman, Can. J. Phys. 34, 1256 (1956).

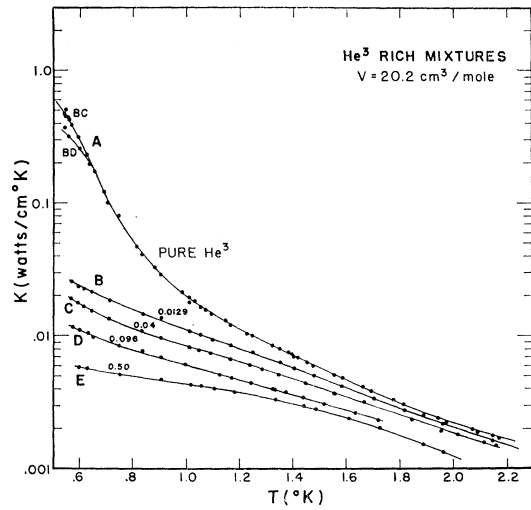


Fig. 4. The thermal conductivity of pure He³ and He³ with He⁴ concentrations of 0.0129, 0.040, 0.096, 0.50 at molar volume $20.2 \text{ cm}^3/\text{mole}$.

sound and $A = V/4\pi N v^3 (\Delta m/m)^2$ for mass fluctuations after Klemens.⁹ This is the smallest thermal conductivity that is possible for a given Rayleigh-like scattering mechanism, i.e., for a given value of A . When Eq. (1) is evaluated using the parameters appropriate to He⁴ at $x=0.01$ and $x=0.10$ the results are as shown by the dashed lines in Fig. 1. Two conclusions follow: (1) Since, at $x=0.0089$ the experimental conductivity is less than the Ziman limit, there must be a scattering mechanism operative in the crystal that is stronger than mass fluctuation scattering. It is this result which leads us to consider lattice distortion.³ (2) The experi-

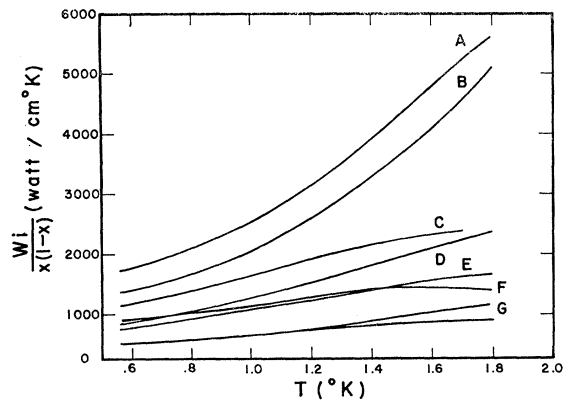


Fig. 5. The isotope resistance of helium mixtures, $W_i/x(1-x)$ is plotted versus temperature for the isotopic mixtures at $19.5 \text{ cm}^3/\text{mole}$.

- A 0.0089 He³ in He⁴
- B 0.0129 He⁴ in He³
- C 0.037 He³ in He⁴
- D 0.040 He⁴ in He³
- E 0.096 He⁴ in He³
- F 0.097 He³ in He⁴
- G 50% mixture.

1. Upper curve $W_i = W - W_{\text{He}^3}$, 2. Lower curve $W_i = W - W_{\text{He}^4}$.

TABLE I. Lattice-distortion scattering parameters.

	(a)	(b)	(c)	(d)	(e)	(f)	(g)	(h)	
	Concentration	$B_N[\text{sec}(\text{°K})^{-3}]$	$A_{\text{expt}}(\text{sec}^3)$	$A_{\text{expt}}/x(1-x)$	$A_{\text{expt}}(1-x)/x$	Relative scattering strength from (d)	Cluster factor	Relative scattering strength from (d) using (f)	Relative volume change
He ³ in He ⁴	$x \rightarrow 0$	9.6×10^{-17}	...	9.2×10^{-38}
	0.0089	1.14×10^{-16}	8.5×10^{-40}	9.7×10^{-38}	9.5×10^{-38}	1.00	1.20	1.00	1.00
	0.038	1.70×10^{-16}	4.5×10^{-39}	1.23×10^{-37}	1.14×10^{-37}	1.24	1.75	0.88	0.94
	0.097	2.25×10^{-16}	1.6×10^{-38}	1.83×10^{-37}	1.66×10^{-37}	1.85	2.55	0.86	0.93
	0.50	2.0×10^{-16}	3.3×10^{-38}	1.32×10^{-37}	3.3×10^{-37}	0.442	...	0.52	0.72
He ⁴ in He ³	0.096	1.7×10^{-16}	1.0×10^{-38}	1.15×10^{-37}	9.4×10^{-38}	1.55	2.55	0.80	0.90
	0.040	1.5×10^{-16}	2.5×10^{-39}	6.5×10^{-38}	6.0×10^{-38}	1.02	1.80	0.70	0.84
	0.0129	1.2×10^{-16}	6.2×10^{-40}	4.88×10^{-38}	4.7×10^{-38}	0.817	1.30	0.78	0.88
	$(1-x) \rightarrow 0$	9.6×10^{-17}	...	4.2×10^{-38}

mental conductivity for $x=0.097$ is greater than the Ziman limit given by (1). Hence, at high concentration the normal processes are quite probably not very much more rapid than the resistive scattering. In fact we will find that the normal processes occur at about the same rate as the resistive processes.

DATA ANALYSIS

Following Callaway we use the phenomenological equation

$$K = \frac{k_B}{2\pi^2 v} \left(\frac{k_B T}{\hbar} \right)^3 (I_1 + \beta I_2), \quad (2)$$

where I_1 , I_2 , and β as defined by Callaway are

$$I_1 = \int_0^{\Theta/T} \tau_C(x) x^4 \frac{e^x}{(e^x - 1)^2} dx$$

$$I_2 = \int_0^{\Theta/T} \frac{\tau_C(x)}{\tau_N(x)} x^4 \frac{e^x}{(e^x - 1)^2} dx$$

$$\beta = I_2 / \int_0^{\Theta/T} \frac{\tau_C(x)}{\tau_N(x)} \frac{1}{\tau_R(x)} x^4 \frac{e^x}{(e^x - 1)^2} dx$$

where Θ is the Debye temperature, $x = \hbar\omega/k_B T$, and the relaxation time we have used for the various phonon scattering mechanisms are as follows:

$$\tau_N^{-1} = B_N T^3 \omega^2, \quad \text{normal processes}$$

$$\tau_u^{-1} = B_u T^3 \omega^2 \exp(-a/T), \quad \text{umklapp processes}$$

$$\tau_D^{-1} = A \omega^4, \quad \text{point-defect scattering processes}$$

τ_B , the boundary scattering relaxation time, is found from analysis of the low-temperature T^3 region.⁷ τ_C and τ_R are defined by

$$\tau_C^{-1} = \tau_N^{-1} + \tau_R^{-1}, \quad \tau_R^{-1} = \tau_u^{-1} + \tau_D^{-1} + \tau_B^{-1}.$$

Equation (2) was derived by Callaway from a relaxation-time approximation to the collision operators in a

linearized Boltzmann equation.⁵ It has recently been obtained from a linearized Peierls equation in such a way that the identification of collision operators with relaxation times is made only as a final step,¹² thus providing justification for Callaway's method of including the effect of normal processes. (Further the Callaway equation, Eq. (2), and the conductivity derived via the Ziman variational procedure have been shown to be equivalent.¹³)

The data at 19.5 cm³/mole were fit using Eq. (2). The Debye temperatures were taken from the data of Heltemes and Swenson;¹⁴ the velocity of sound was calculated using these temperatures and a Debye model for the phonon spectrum, $v = (k_B \Theta / \hbar) (6\pi^2 N / V)^{-1/3}$.

The procedure for fitting the data was as follows:

(a) The pure data were fitted using the value of the boundary relaxation time found from the data at the lowest temperatures (where $K \propto T^3$) with the parameters B_N (characterizing normal scattering) and B_u and a (characterizing umklapp scattering) adjusted to obtain the best fit. No unique set of parameters (B_N, B_u, a) could be determined from the pure data although B_N was varied over a wide range. However, the pure data could be used to find the best B_u and a appropriate to a given B_N . The relationships $B_u = B_u(B_N)$ and $a = a(B_N)$ established from the pure data are not expected to be altered greatly by introducing isotopic impurities.

(b) For each set of parameters (B_N, B_u, a) the mixture data were fitted by varying the parameter A . The mixture data in the temperature range 0.5°K < T < 1.4°K were particularly sensitive to the choice of B_N and A . Hence, the data in this range provide a very good test of the proper choice of (B_N, A). In this temperature range B_u and a are relatively unimportant.

¹² R. A. Guyer and J. A. Krumhansl (to be published). See also J. A. Krumhansl, Proc. Phys. Soc. (to be published).

¹³ J. M. Ziman, *Electrons and Phonons* (Oxford University Press, London, 1960).

¹⁴ E. C. Heltemes and C. A. Swenson, Phys. Rev. **128**, 1512 (1962).

For each concentration a different choice of B_N gave the best fit. The values of these quantities are given in columns (a) and (b) of Table I. It was possible using this method to determine unambiguously B_N and A to better than $\pm 10\%$. The solid lines in Figs. 1 and 2 are the fits to the mixture data obtained with the values of B_N and A in Table I. The fits to the pure data shown in Figs. 1 and 2 are made using the set (B_N, B_u, a) corresponding to the value of B_N obtained by extrapolating $B_N(x)$ to zero concentration. These data are fitted with the parameters

$$\begin{aligned} \text{pure He}^4: B_N &= 9.6 \times 10^{-17} \text{ sec}(\text{°K})^{-3}, \\ B_u &= 1.20 \times 10^{-13} \text{ sec}(\text{°K})^{-3}, \\ a &= 10.4 \text{°K}; \\ \text{pure He}^3: B_N &= 9.6 \times 10^{-17} \text{ sec}(\text{°K})^{-3}, \\ B_u &= 4.0 \times 10^{-14} \text{ sec}(\text{°K})^{-3}, \\ a &= 11.7 \text{°K}. \end{aligned}$$

It is interesting that the values of a are very nearly those that are obtained by taking the slope of the $\log K$ -versus- $1/T$ plot of the pure data in the umklapp region.

The Callaway expression for the thermal conductivity is the sum of two terms, I_1 and βI_2 . When the normal process scattering is relatively slow compared with momentum loss scattering, I_1 dominates βI_2 in the expression for K . In the opposite limit, when normal processes become very rapid, βI_2 makes the major contribution to K . We found that in fitting the pure data, βI_2 played a major role in determining K ; however, in the case of the mixture data, βI_2 was at most a small correction. This is expected since the large lattice-distortion cross section leads to relatively rapid momentum-loss scattering. We found the following empirical law for the relationship of τ_N to τ_D ; $\tau_N/\tau_D = 0.076T/x$. Thus it is seen that for our data τ_D/τ_N was sufficiently small for all values of T and x to make βI_2 a small correction to I_1 in the calculation of K from Eq. (2).¹⁵

The procedure of fitting the mixture data with sets of (B_N, B_u, a) determined from the pure data alone does not permit B_u and a to vary as a function of x independent of B_N . Since B_N depends on concentration it is not unreasonable that B_u and a do also. The fact that the 1% fits for both He³- and He⁴-rich mixtures are relatively bad in the umklapp region bears this out. For the high concentration mixtures the fits are better in the umklapp region; for these concentrations the isotope scattering dominates sufficiently so that an error in B_u or a is obscured.

Finally as a demonstration of the sensitivity of the choice of B_N and A we show in Fig. 2 the fit to the 4%

data in He³ obtained using a proper A and the set of parameters, (B_N, B_u, a) , appropriate to the 1% data.

DISCUSSION

The parameter A measures the strength of the scattering due to the introduction of isotopes. For mass-fluctuation scattering the relaxation time is given by^{9,10} $\tau_D^{-1} = A\omega^4$, where $A = A_0x(1-x)$ and

$$A_0 = \frac{V}{4\pi N} \frac{1}{v^3} \left(\frac{\Delta m}{\bar{m}} \right)^2 \quad (3)$$

and $\bar{m} = xm_3 + (1-x)m_4$. For small concentrations of He⁴ in He³, $A_0 = 4.83 \times 10^{-39} \text{ sec}^3$, and for small concentration of He³ in He⁴, $A_0 = 4.48 \times 10^{-39} \text{ sec}^3$. The values of $A/x(1-x)$ calculated from the experimental data are given in column (c) of Table I. There are two notable features of our result: (1) $A/x(1-x)$ is greater than A_0 as predicted by Eq. (3) by a factor 10–20, (2) $A/x(1-x)$ contains a residual concentration dependence that is not removed by the factor $x(1-x)$.

Following the suggestion of Klemens and Maradudin we assume that there is Rayleigh-like scattering from each isotope impurity due to lattice distortion in addition to that of mass fluctuations. Scattering from lattice distortion leads to a relaxation time of the form¹⁶

$$\tau_{LD}^{-1} = (V/4\pi N)(1/v^3)564\epsilon^2\omega^4 = A_{LD}\omega^4. \quad (4)$$

The matrix element which describes the strength of mass fluctuation scattering goes as $(\Delta m/\bar{m})^2 \sim \frac{1}{10}$, that for lattice distortion goes as $564\epsilon^2$ and must be of order 1. The parameter ϵ is a measure of the atomic misfit of an atom in the host lattice, $\epsilon = \delta R/R$. From the analysis of the data, the numbers in column (c) of Table I, it is possible to evaluate the atomic-misfit parameter for a He³ atom in a He⁴ host lattice and vice versa; we find ϵ_3 in $4 \simeq 4\%$ and ϵ_4 in $3 \simeq 5\%$. However, the numerical factors in Eq. (4) are not without some uncertainty; hence, the comparisons which we will make will primarily involve ratios. Before doing this we examine the concentration dependence that is expected for lattice distortion scattering.

The concentration factor $x(1-x)$ for mass fluctuation scattering follows from arguing that in an isotopic mixture the low-frequency phonons, those which carry the heat current, are characteristic of the average mass of the crystal, $\bar{m} = xm_3 + (1-x)m_4$. These \bar{m} phonons are different from the phonons of pure He³ and pure He⁴; hence, they scatter from both He³ atoms and He⁴ atoms.¹⁰ For this reason the square of the matrix element, $V' = \sum_n \Delta m_n e^{i(q-q') \cdot r_n}$, which leads to mass fluctuation scattering, has a diagonal contribution only. (The isotopic impurities are assumed to be randomly distributed throughout the host lattice.)

¹⁵ See Ref. 2, in particular the discussion relevant to Fig. 1. Recent work by Berman and Brock (to be published) makes a careful analysis of the contribution of I_1 and βI_2 to the thermal conductivity in LiF.

¹⁶ The numerical factor in this formula is obtained by combining Eqs. (11) and (12) of Ref. 3 and using the value 2 for the Gruneisen constant.

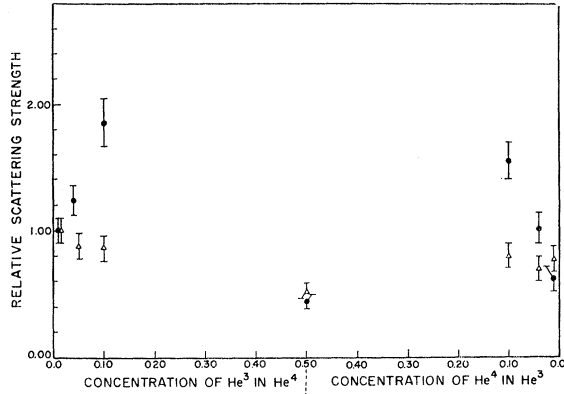


FIG. 6. The relative scattering strength of a lattice distortion site as a function of concentration. The black circles are the results from column (d) of Table I, the triangles are from column (g).

$$|V'|^2 = x(m_3 - \bar{m})^2 + (1-x)(m_4 - \bar{m})^2 + [x(m_3 - \bar{m}) + (1-x)(m_4 - \bar{m})] \sum_{n \neq 0} e^{i(\mathbf{q} - \mathbf{q}') \cdot \mathbf{n}}, \quad (5)$$

$$|V'|^2 = (m_3 - m_4)^2 x(1-x).$$

The sum in V' is over all equilibrium lattice sites, $\mathbf{x}_n = \mathbf{n}$; \mathbf{q} and \mathbf{q}' are the wave vectors of the incoming and outgoing phonons. No enhancement of scattering occurs because of the possibility that two impurities may be neighbors and scatter coherently (this would require off-diagonal terms).

When the phonon scattering is due principally to lattice distortion around impurity sites, the proper phonons with which to analyse the thermal conductivity continue to be those characterized by the average mass \bar{m} . The unperturbed Hamiltonian is

$$H_0 = \sum_i \frac{1}{2} \bar{m} v_i^2 + V^{(2)},$$

where $V^{(2)}$, the harmonic part of the potential energy, is essentially the same for He^3 and He^4 . This follows from the experimental observation $\Theta(\text{He}^3)/\Theta(\text{He}^4) \simeq (m_4/m_3)^{1/2}$,¹³ the principle change in the character of the phonon spectrum as the solid goes from pure He^4 to pure He^3 at constant molar volume is accounted for by the change in mass alone.

When there is a concentration x of He^3 atoms in a He^4 crystal of N atoms, the \bar{m} phonons scatter from xN He^3 distortion sites. The matrix element for this process is proportional to $|\delta V_{3 \text{ in } \bar{m}}|^2$, where $\delta V_{3 \text{ in } \bar{m}}$ is the change in volume around a He^3 atom in a host lattice of average mass \bar{m} . The xN He^3 atoms occupy an additional volume $xN\delta V_{3 \text{ in } \bar{m}}$. If the He^3 atoms are regarded as uniformly squeezing all of the He^4 atoms, then the solid contains $(1-x)$ distortion sites that scatter phonons with strength proportional to $|\delta V_{4 \text{ in } \bar{m}}|^2$, where

$$x\delta V_{3 \text{ in } \bar{m}} + (1-x)\delta V_{4 \text{ in } \bar{m}} = 0 \quad (6)$$

by conservation of volume. (The suggestion that a few He^3 atoms squeeze all of the He^4 is open to criticism at low concentration although as $x \simeq 0.50$ this idea becomes

more tenable.) By analogy with (5) the probability for lattice distortion scattering depends on concentration thus

$$|V_{\text{LD}}'|^2 = x|\delta V_{3 \text{ in } \bar{m}}|^2 + (1-x)|\delta V_{4 \text{ in } \bar{m}}|^2, \quad (7)$$

where again the nondiagonal term is zero because of (6). However, using (6) in (7) yields the concentration dependence

$$|V_{\text{LD}}'|^2 = [x/(1-x)]|\delta V_{3 \text{ in } \bar{m}}|^2, \quad (8a)$$

$$|V_{\text{LD}}'|^2 = x(1-x)|\delta V_{3 \text{ in } 4}|^2, \quad (8b)$$

where $\delta V_{3 \text{ in } \bar{m}}$ depends on concentration. The step from (8a) to (8b) follows from the approximation $\delta V_{3 \text{ in } \bar{m}} = (1-x)\delta V_{3 \text{ in } 4}$. When the concentration dependence given in (8a) is divided out of A the numbers recorded in column (d) are obtained. We take for the matrix element for lattice distortion the expression (4); we can estimate the relative scattering strength of a distortion site by dividing out the velocity dependence of A using a velocity linearly interpolated from pure He^3 to pure He^4 . The numbers recorded in column (e) are the relative scattering strength for each concentration; they are proportional to the square of $\delta V_{3 \text{ in } \bar{m}}$. We make the following observations:

(1) The ratio of the volume change for He^3 in He^4 at $x \simeq 0.01$ to that for He^4 in He^3 at $x \simeq 0.01$ is $\simeq 1.1$. Actually this result is not terribly sensitive to the argument regarding concentration dependence; since A is proportional to x at low concentration, the best comparison is given by using the values of A extrapolated to zero concentration. Then, we still find $\delta V_{3 \text{ in } 4}/\delta V_{4 \text{ in } 3} \simeq 1.1$ as above.⁶

(2) The volume change at $x=0.50$ is less than the volume change for either pure material. We would expect it to be on the order of $\frac{1}{2}$.

(3) For the low-concentration data the relative volume change increases by a factor 1.4 and the scattering strength by a factor of 2 (see Fig. 6) as x goes from 0.01 to 0.10. This change is far outside the uncertainty in our measurement. We advance two possible explanations of this effect:

(i) Suppose that at low concentrations a He^3 impurity does not uniformly squeeze all of the He^4 atoms but in fact its effect is very localized. Equation (6) is no longer valid. A phonon sees a medium containing essentially only He^3 distortion sites in a He^4 host. In Eq. (5) the nondiagonal terms are no longer identically zero. If $\delta V_{4 \text{ in } \bar{m}}$ is effectively zero for $x < x_c$, some critical concentration, then, for concentrations less than x_c the nondiagonal terms contribute to the scattering.

An estimate of the magnitude of the contribution of the nondiagonal terms is obtained by regarding the solid as a random distribution of scattering sites occurring with probability x and nonscattering sites occurring with probability $(1-x)$. Helium at $V=19.5$ cm³/mole is in the (hcp) phase with each atom having 12 nearest

neighbors. At $x=0.01$, the probability that a given He^3 atom have no, one, and two He^3 neighbors is 0.90, 0.10, and 0.00, respectively. Since, for

$$\lambda = \hbar\omega/k_B T \ll d = (V/N)^{1/3},$$

a cluster of n scattering sites scatters with n^2 times the strength of an isolated site, at $x=0.01$ the scattering due to xN distortion sites is increased over that of xN isolated sites by the cluster factor, $f(x)$,

$$f(x=0.01) = \frac{0.90 \times 1 + 0.10 \times 4}{0.90 \times 1 + 0.10 \times 2} \sim 1.20,$$

where the denominator normalizes the total number of scatterers. The cluster factor for the experimental concentrations are recorded in Table I-(f). When the low-concentration data are corrected by this factor the intrinsic single site scattering is as recorded in Table I-(g); the corresponding relative volume changes are in I-(h). For the 50% data we have continued to use the argument based on Eq. (6).

In review: at low concentration, $x < x_c$, Eq. (6) is not valid and random clustering leads to an enhancement of the scattering due to distortion sites. For high concentrations Eq. (6) obtains and both He^3 and He^4 sites scatter phonons; the concentration dependence of (8a), (8b) is recovered. This argument has led to a much more satisfactory concentration dependence of the scattering strength, (see Fig. 6).

(ii) Suppose that Eq. (6) remains valid at all experimental concentrations, then, an enhanced scattering at low concentrations could be brought about by nonrandom clustering due to a tendency toward short-range order. This nonrandom clustering would lead to a contribution from the nondiagonal terms in (5). Such an explanation would require that at $x=0.10$ the probability that a He^3 atom have a He^3 neighbor must be on the order of $0.20 = 2x$, i.e., twice the probability expected due to concentration alone.

If short-range ordering is the proper explanation of our results, a nonrandom cluster factor similar to the cluster factor produced by (i) would give satisfactory results. Further, short-range ordering would correct the result at 50% in a direction more in line with the expectation announced above.

CONCLUSION

We have reported thermal-conductivity measurements in isotopic mixtures of He^3 and He^4 over the complete concentration range. The results of the analysis of these data are interpreted in terms of phonon scattering from lattice distortion around the site of an isotopic impurity, boundary scattering, normal process scattering and umklapp-process scattering. The strengths of these various scattering mechanisms are determined from the analysis. We are able to relate the magnitude of lattice-distortion scattering to the volume changes $\delta V_{4 \text{ in } 3}$ and $\delta V_{3 \text{ in } 4}$. We find $\delta V_{4 \text{ in } 3} \simeq \delta V_{3 \text{ in } 4}$.

However, we find that the scattering strength due to lattice distortion depends on concentration more strongly than is expected. We have advanced two arguments in explanation of this result. In the first of these we suggest that phonon scattering from lattice distortion sites is not the exact analogue of phonon scattering from mass fluctuations. At low concentration an additional cluster factor is necessary. This argument when made quantitative removes the anomalous concentration dependence. Our second argument takes the form of a conjecture: if nonrandom clustering occurs with the right probability as a function of concentration our results can be explained. We have not made this argument quantitative.

The present experiment is not able to discriminate between the above two alternatives. At this time the state of our knowledge about scattering from distortion sites and the features of He^3 - He^4 mixtures requires that our conclusions be regarded as tentative.

Note: Since the preparation of this manuscript, it has come to our attention that thermal-conductivity measurements on solid-helium mixtures have been made by Berman, Bounds, and Rogers.¹⁷ They find values of A_{expt} much closer to the theoretically expected ones than those given in the present work. Their samples were all at the same pressure rather than the same molar volume, and they assumed that normal-process relaxation time varied as T^2 rather than T^3 . A discussion of this question will be given by one of us (RAG) in a forthcoming paper.

¹⁷ R. Berman, C. L. Bounds, and S. J. Rogers (to be published).