

TABLE III. Values for effective mass, "Dingle temperature," and mobility.

	transv.	long.	
m^*/m	0.018	0.017	0.030, ^a 0.020, ^b 0.024 ^c
T_{AV}' (°K)	15.2	16.8	
μ (cm ² /v-sec)	1.57×10^4	$1.5^s \times 10^4$	2.13×10^4 ^d

^a Reference 12. ^b Reference 13. ^c Reference 14. ^d See Table I.

mechanism can be described by a classical collision time independent of the magnetic field. His result indicates a different power ($\frac{3}{2}$) of $(\hbar\omega/\zeta)$ in the coefficient of the oscillatory term.

Several years ago Dingle¹⁸ discussed the energy level broadening due to collisions. He showed that this broadening influences the damping of the oscillations; the form of the damping term remains the same except that the actual temperature T in the exponential is replaced by a somewhat higher temperature $(T+T')$, where $T' = \hbar/2\pi^2 k\tau$ (τ = collision time).

We have analyzed our data in terms of Eq. (2a) in which T is replaced by $T+T'$. It is evident that the coefficient of the cosine in the third term represents the amplitude A of the oscillations. Plotting $\ln(AH^{\frac{3}{2}})$ vs $1/H$, one expects a straight line with a slope of $-2\pi^2 k(T+T')/\beta^*$.¹⁹ From data at different temperatures T' and β^* can be calculated. Such a plot is shown in Fig. 5 and the results are listed in Table III. The

¹⁸ R. B. Dingle, Proc. Roy. Soc. (London) **A211**, 500 (1952).

¹⁹ It was mentioned before that there is some doubt with regard to the power of H in the proportionality factor of the amplitude. It was impossible, however, to make an unambiguous decision on the basis of our data.

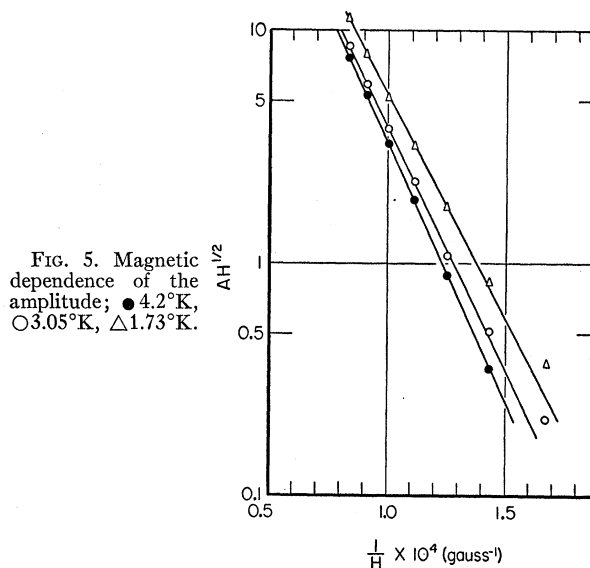


FIG. 5. Magnetic dependence of the amplitude; ● 4.2°K, ○ 3.05°K, △ 1.73°K.

accuracy of the values obtained is small due to the relatively large value of T' (causing very little difference in the slopes). The last column contains for comparison the effective masses reported in earlier experimental and theoretical work and the observed Hall mobility $R\sigma$. The mobility values agree much better than is commonly found in metals.⁴

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Lattice Conductivity of Tin*

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The thermal conductivities of several impure tin specimens have been measured at liquid helium temperatures. Impurities of antimony, bismuth, and indium between 0.1% and 6% were used to lower the electronic conductivity. The conductivities are found to be consistent with the equation $K = \alpha T + \beta T^2$, where the first term may be ascribed to the electronic conductivity and the second to the lattice conductivity. For impurities of 3% or less the values of α , combined with measurements of the residual electrical resistivity, agree to about one percent with the prediction of the Wiedemann-Franz law. For the same samples β is approximately constant with a mean value of $(3.5 \pm 0.4) \times 10^{-4}$ watt/cm deg³. The comparative constancy of this value of β indicates that it is characteristic of the intrinsic conductivity of the tin lattice.

INTRODUCTION

THIS paper will describe and discuss measurements of the thermal and electrical conductivities of several impure tin specimens between 1.7°K and 4.2°K.

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The investigation was carried out in order to obtain a measure of the lattice contribution to the thermal conductivity of tin.

The theory of thermal conductivity has been extensively treated in the literature¹ and we merely state

¹ A summary of the theory and an exhaustive bibliography are given by P. G. Klemens, *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1956), Vol. 14, p. 198.

some relevant results. In a metal both electrons and lattice waves carry energy. The total heat current may be considered as flowing along two parallel paths, so that the thermal conductivity K may be written as the sum of an electronic conductivity K_e and a lattice conductivity K_g .

For temperatures below one tenth of the Debye temperature the electronic thermal conductivity may be written in the form

$$1/K_e = (\alpha T)^{-1} + AT^2. \quad (1)$$

The first term represents the thermal resistivity arising from collisions of electrons with impurities; the second, for which the symbol $1/K_i$ is usually used, represents the thermal resistivity caused by collisions of electrons with phonons. The parameter α is related to the residual electrical resistivity ρ_0 by the Wiedemann-Franz law $\alpha\rho_0 = L_0$, where L_0 is the Lorenz number 2.445×10^{-8} volt² deg⁻².

In a well-annealed metal the most important resistive process for lattice waves is the scattering by electrons. For temperatures far below the Debye temperature, the conductivity K_E determined by this process varies with the square of the temperature. The lattice waves may also be scattered by dislocations. For the conductivity resulting from this process a variation with T^2 is also expected.

In pure metals the lattice conductivity is much smaller than the electronic conductivity. By adding impurities to the metal the electronic conductivity is reduced so that the lattice conductivity will be a greater fraction of the total thermal conductivity. Furthermore, the term AT^2 in Eq. (1), which at liquid helium tem-

peratures is smaller than $(\alpha T)^{-1}$ even for pure samples, becomes less important with the addition of impurities. For sufficiently impure specimens, therefore, the expected expression for the thermal conductivity becomes

$$K = \alpha T + \beta T^2. \quad (2)$$

APPARATUS

Most of the samples used (2.9% Sb, 3.1% In, 6% Sb) were obtained from Budnick, Lynton, and Serin,² who describe their preparation. The other two specimens were made according to the same procedure. Their diameter was about 4 mm and their length between 8 and 13 cm. Annealing times ranged from two to sixteen months.

The calorimeter, with a specimen in place, is illustrated in Fig. 1. The samples were melted into collars (G) of tellurium copper (99.5% Cu, 0.5% Te). This material may be machined easily and has a thermal conductivity of the same order of magnitude as commercial copper.³ Two thermometers (I , J) and a heater (H) were attached to the sample. The heater consists of about 200 turns of 0.0025-inch diameter, Formvar coated Advance wire, wound directly on the sample and secured in place with Glyptal lacquer. The thermometers are $\frac{1}{2}$ -watt, 56-ohm, Allen-Bradley carbon composition resistors. They were cemented into copper holders with Araldite resin after most of the insulation was ground off. The holders were soldered to the sample with Rose's metal.

The sample collar is bolted to the cap of the calorimeter with a gold gasket (K). To complete the adiabatic enclosure a brass can (A) is bolted to the cap with a second gold gasket (B). The calorimeter is evacuated through a $\frac{3}{8}$ -inch diameter 0.010-inch thick stainless steel tube (L). This tube does not connect directly into the calorimeter. It is soldered to a tellurium-copper tube (C), $\frac{3}{32}$ inch thick, which passes through the calorimeter cap and extends down for about 4 inches.

Electrical connections to the heater and thermometers are brought out of the calorimeter in the following manner: Two Advance wires, 0.0025 inch diameter, 10 inches long, are carried from each thermometer to a terminal strip (E), molded of a mica-filled epoxy resin.⁴ Two niobium wires 0.004 inch diameter, 40 inches long, connect the heater to the terminal strip. Niobium is used for these leads because of its superconductivity over the entire liquid helium range. As a result, even though the wires carry appreciable current, no heat is developed in them and there is no potential drop across them. Advance wires, 0.004 inch in diameter, are used for the connections from the terminal strip to the outside of the apparatus. About 2 $\frac{1}{2}$ feet of each wire are wrapped around the tellurium-copper pumping tube to

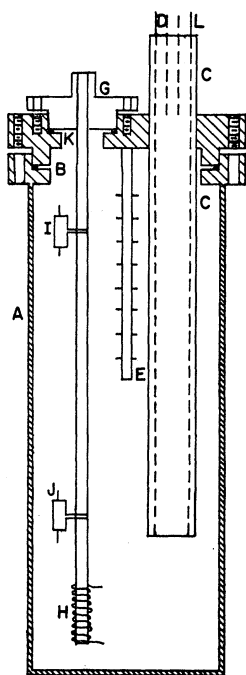


FIG. 1. View of calorimeter with sample in place.

² Budnick, Lynton, and Serin, Phys. Rev. **103**, 286 (1956).

³ Powell, Roder, and Rogers, J. Appl. Phys. **28**, 1282 (1957).

⁴ Epocast PRH 103, Permacel Tape Corporation, New Brunswick, New Jersey.

TABLE I. Resistivity and resistivity ratio.

Sample	Composition, atomic percent	$\rho_{4.2}$ ohm cm	ρ_{300} ohm cm	$\rho_{4.2}$
				$\rho_{300} - \rho_{4.2}$
1	0.11% Bi	0.0721×10^{-6}	1.169×10^{-5}	0.00621
2	1.1% Bi	0.758×10^{-6}	1.261×10^{-5}	0.0639
3	3.1% In	1.591×10^{-6}	1.226×10^{-5}	0.1492
4	2.9% Sb	2.142×10^{-6}	1.605×10^{-5}	0.1538
5	6.0% Sb	3.483×10^{-6}	1.491×10^{-5}	0.3047

establish thermal contact with the helium bath. The wires are brought out of the calorimeter through a thin stainless steel tube (D), and through a wax seal at the top.

A water-cooled Helmholtz coil surrounded the Dewar assembly. This coil provided a field of about 560 gauss at the sample during all measurements in order to keep the sample in the normal state at all temperatures. No evidence was found that this field had a measurable effect on any of the conductivities.

THERMAL CONDUCTIVITY MEASUREMENTS

Power was supplied to the heater from a storage battery. It was measured by determining the current and voltage with two potentiometers.

Temperatures were determined by measuring the resistances of the thermometers with a Wheatstone bridge. Current through the resistors was kept at 4 microamperes. For the more impure samples, however, in order to keep the power dissipated in the thermometers much smaller than that in the heater, the current was, at low temperatures, reduced to 0.8 microampere. Since the resistance of the thermometers depends on the current, a separate calibration had to be made for each current.

The resistance values were converted to temperatures by using the equation⁵

$$1/T^* = A^2/\log R + B^2 \log R + 2AB.$$

The constants A and B were calculated from two calibration points, one at 4.2°K and one just below the lambda point, with the absolute temperature T given by the T_{55E} temperature scale.⁶ Measurements of the vapor pressure and of the resistances at other temperatures were used to construct correction curves of $T - T^*$ vs T^* . Such a curve is shown in Fig. 2. From these curves, each value of T^* was converted to absolute temperature.

ELECTRICAL CONDUCTIVITY MEASUREMENTS

Subsequent to each thermal conductivity run a measurement was made of the electrical conductivity at 4.2°K. The brass can (A) was removed and copper wires were attached to the sample with Rose's metal.

⁵ J. R. Clement, in *Temperature, its Measurement and Control in Science and Industry* (Reinhold Publishing Corporation, New York, 1955), p. 382.

⁶ Clement, Logan, and Gaffney, *Phys. Rev.* **100**, 743 (1955).

A current lead was soldered to the free end of the sample and potential leads were soldered to the copper thermometer holders. In this way the geometry for the electrical measurement was the same as that for the thermal one. The current and voltage were measured with potentiometers. The electrical resistivities are listed in Table I.

ERRORS

It is important to guard against heat flow into the sample along the leads going to the top. A carbon resistance thermometer was therefore taped over the wires wound on the tellurium-copper pumping tube, and thermal conductivity measurements were not started until this thermometer indicated that the leads in the calorimeter had cooled to 4.2°K. Heat conduction from the sample to the bath along the leads was less than 0.1% of the conduction along the sample and could therefore be neglected.

Uncertainties in the distance between thermometers arise by virtue of the finite width of the thermometer holders. The holders made contact with the sample over a distance of about 0.5 mm. For a typical distance of 5 cm this leads to an error in K at most equal to two percent. This error and the error in the measurement of the area (less than one percent) are constant for each sample and therefore do not contribute to the scatter of the data and have no effect on the Wiedemann-Franz ratio.

The power is known to about 0.1%. The sensitivity of the thermometers was such that at 4°K, dT/dR was about 2×10^{-3} deg/ohm, rising to 7×10^{-5} deg/ohm near 2°K. The uncertainty in R at 4° was about 0.3 ohm and at 2° about 10 ohms so that the error in T^* was about 7×10^{-4} degree. We estimate that the temperature difference ΔT was known to about 10^{-3} degree. The values of ΔT were greater than 0.1 degree so that the error in ΔT was always less than one percent. More-

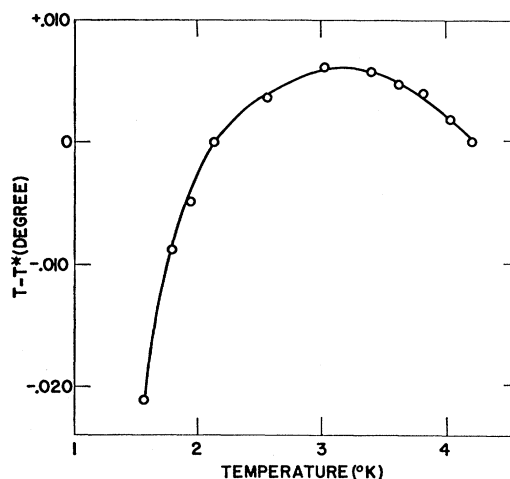


FIG. 2. Correction curve for 56-ohm $\frac{1}{2}$ -watt Allen-Bradley resistor.

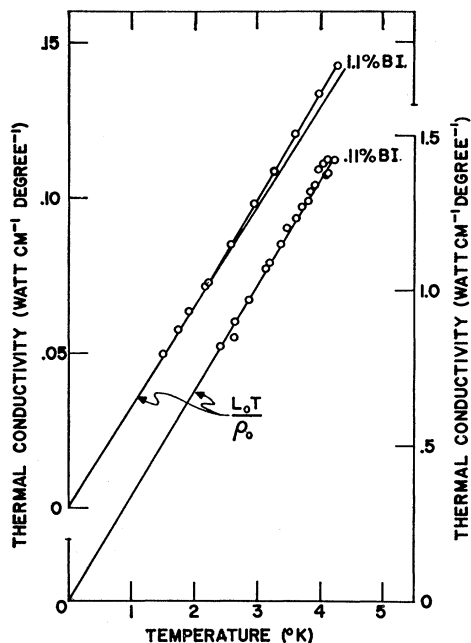


FIG. 3. Thermal conductivity of specimens 1 and 2.

over, in error in K introduced by the finite size of ΔT is negligible for our specimens because of their nearly linear variation of thermal conductivity with temperature.

RESULTS AND DISCUSSION

In general, the data are consistent with the theory outlined in the introduction. Sample 1, containing 0.11% bismuth, has a thermal conductivity (Fig. 3) varying linearly with the temperature. Evidently the

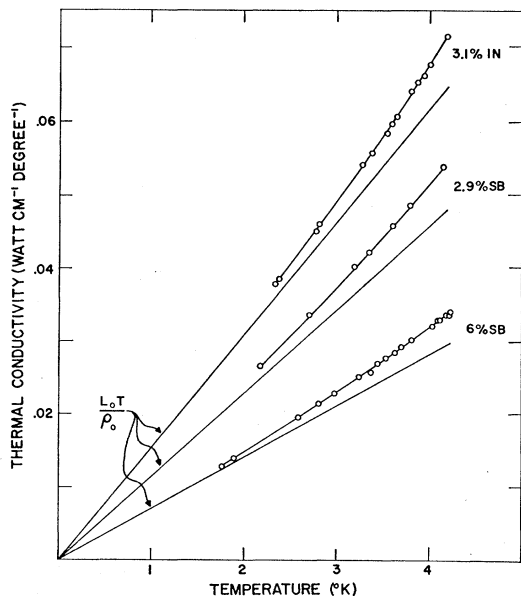


FIG. 4. Thermal conductivity of specimens 3, 4, and 5.

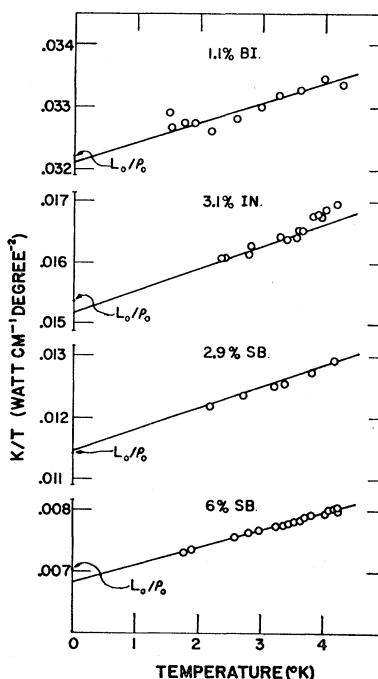
electronic conductivity is determined by scattering from impurities, and is sufficiently large to mask the lattice conductivity.

The curves of K versus T for samples 2 to 5 (Figs. 3 and 4) deviate from straight lines. In each case equation (2) may be used to describe the data. Figure 5 shows graphs of K/T versus T for these specimens. Straight lines were fitted to the points by a least-squares analysis. The values obtained for α , β , and L_0/ρ_0 are shown in Table II.

For specimens 1, 2, 3, and 4, α and L_0/ρ_0 agree to about one percent. For specimens 2, 3, and 4, the results are consistent with a value for β equal to $(3.5 \pm 0.4) \times 10^{-4}$ watt/cm deg³, constant within the accuracy of this experiment.

In the introduction it was pointed out that a lattice conductivity proportional to T^2 may arise from the scattering of phonons by electrons or from the scattering of phonons by dislocations. The relative constancy of β in samples 2, 3, and 4 with respect to the nature of the impurity and with respect to the concentration indicates that for these samples the conductivity βT^2 arises from the first mechanism, which is inherent in the metal, rather than from the second, which depends on circumstances which are difficult to control.

The most impure specimen (No. 5) has a lattice conductivity significantly lower than the others. The increase in the thermal resistance is of the same order of magnitude as that observed for silver-palladium and silver-cadmium alloys by Kemp, Klemens, Sreedhar,

FIG. 5. K/T for samples 2, 3, 4, and 5.

and White,⁷ and for indium-thallium alloys by Sladek.⁸ This extra resistance was ascribed by Kemp *et al.* to dislocations associated with the impurity. The density of single dislocations necessary to explain the extra resistance⁹ of sample 5 is of the order of 6×10^{11} per cm^2 . This seems improbably high for an annealed specimen. It is possible that, as suggested by Kemp *et al.* for their strained samples, the scattering is due to clusters of dislocations. Further experiments would be necessary to clarify this point.

For sample 5, the values of α and L_0/ρ_0 differ by about three percent. It is conceivable that the 6% impurity raised the transition temperature sufficiently¹⁰ for superconducting threads to be present in spite of the applied magnetic field.² Such threads might have resulted in an erroneously low measurement of the electrical resistivity.

Other measurements of the thermal conductivity of dilute tin alloys at liquid helium temperatures have been made by Hulm and by Shiffman. Hulm¹¹ measured a sample of tin with four percent mercury. The lattice conductivity varied as T^3 , and at 4°K was an order of magnitude higher than the one which we measured. His specimen consisted of two phases so that the result probably does not represent a property of the pure metal. Shiffman¹² measured a number of specimens with indium impurities over a much wider temperature range than that of this experiment. His data in the liquid helium range are not, however, sufficiently accurate to allow a quantitative comparison with the results of this experiment.

Both the electronically limited phonon conduction K_E and the phonon-limited electronic conduction K_i depend on the strength of the electron-phonon interaction. Klemens^{13,14} has developed an equation connecting these two conductivities, in which the electron-phonon interaction parameter does not appear:

$$K_E = 313(T/\Theta)^4 N_a^{-\frac{1}{3}} K_i. \quad (3)$$

TABLE II. Summary of thermal conductivity and resistivity data.

Sample		$\beta \times 10^4$ watt/cm deg ³	$\alpha \times 10^4$ watt/cm deg ²	$(L_0/\rho_0) \times 10^4$ watt/cm deg ²
1	0.11% Bi	...	3410	3380
2	1.1% Bi	3.2 ± 0.3	321.0 ± 0.9	322
3	3.1% In	3.65 ± 0.28	151.8 ± 0.8	153.7
4	2.9% Sb	3.47 ± 0.18	114.7 ± 0.6	114.2
5	6% Sb	2.83 ± 0.02	68.24 ± 0.08	70.23

⁷ Kemp, Klemens, Sreedhar, and White, Proc. Roy. Soc. (London) **A233**, 480 (1956).

⁸ R. J. Sladek, Phys. Rev. **97**, 902 (1955).

⁹ P. G. Klemens, Proc. Phys. Soc. (London) **A68**, 1113 (1955).

¹⁰ W. F. Love, Phys. Rev. **92**, 238 (1953).

¹¹ J. K. Hulm, Proc. Roy. Soc. (London) **A204**, 98 (1950).

¹² C. A. Shiffman (to be published).

¹³ P. G. Klemens, Australian J. Phys. **7**, 57 (1954).

TABLE III. Parameters entering into Eqs. (3) and (4) for Cu, Ag, In, and Sn.

	N_a	$1/K_i T^2$ cm/watt deg	K_E/T^2 watt/cm deg ³	Θ deg ^a	E	D
Cu	1	2.55×10^{-5} b	1.8×10^{-3} c	339	2	6.2
Ag	1	6.4×10^{-5} b	2×10^{-3} d	225	1	5.2
In	3	18.5×10^{-4} e	6×10^{-4} f	109	2.2	7.7
Sn	4	3.9×10^{-4} b	3.5×10^{-4}	189	3.5	18

^a P. H. Keesom and N. Pearlman, *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1956), Vol. 14, p. 382.

^b See reference 1.

^c G. K. White and S. B. Woods, Can. J. Phys. **33**, 58 (1955).

^d See reference 7.

^e H. M. Rosenberg, Trans. Roy. Soc. (London) **247**, 441 (1955).

^f See reference 8.

Here N_a is the number of conduction electrons per atom and Θ is the Debye temperature.

We may describe the success of Eq. (3) by calculating a number E defined by the relation $E = (1/313)N_a^{\frac{1}{3}} \times (\Theta/T)^4 K_E/K_i$. This number is equal to one when the experimental values of K_E , K_i , and Θ are consistent with Eq. (3). In Table III we list the pertinent data and the resulting value of E for tin and for three other metals whose lattice conductivities have been measured.

Similar comparisons of experiment and theory are often made by combining the measurements of K_i and K_∞ , the high temperature limit of the thermal conductivity, using the equation¹

$$K_\infty = 64N_a^{\frac{1}{3}} K_i (T/\Theta)^2. \quad (4)$$

Klemens¹ tabulates the number $D = 64N_a^{\frac{1}{3}} (K_i/K_\infty) \times (T/\Theta)^2$ for the monovalent metals to show the consistency of the data with this equation. The relevant values for D are also listed in Table III. It is seen that the agreement with Eq. (3), in the few cases where sufficient data exist, is significantly better than the agreement with Eq. (4). This is expected since Eq. (3), in contrast to Eq. (4), compares data taken in the same temperature range. As has been pointed out by Klemens,¹³ differences in the phonon spectrum at different temperatures could lead to differences in the sound velocity and in the electron-phonon interaction parameter which are neglected in the derivation of Eqs. (3) and (4). Furthermore, umklapp processes, which are also neglected, should have a greater effect at high temperatures.

The two equations are derived on the assumption of spherical energy surfaces and close agreement can certainly not be expected for the polyvalent metals. The data listed in Table III suggest that Eq. (3) may nevertheless be used with some confidence.

ACKNOWLEDGMENTS

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