

Thermal Conductivity of Indium Antimonide at Low Temperatures

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Thermal conductivity measurements were made on a monocrystalline sample of indium antimonide from 10 to 50°K. Umklapp, isotope, and boundary scattering contributions to the thermal resistivity were calculated from theoretical expressions and then subtracted from the measured value of thermal resistivity. The subsequent deduction of impurity scattering gives a value for the number of point impurities which is compatible with that given by electrical measurements.

INTRODUCTION

INDIUM antimonide is a semiconductor crystallizing in a zinc blende structure. Its electrical and optical properties have been intensively investigated¹ and measurements of its thermoelectric power have been reported by Frederikse and Mielczarek,² Geballe,³ and Barrie and Edmond.⁴ This investigation was undertaken in order to determine the magnitudes of the various scattering mechanisms contributing to the low-temperature thermal conductivity. Previous measurements of its thermal conductivity have been made above room temperature by Stuckes,⁵ and Geballe has reported in an abstract³ on measurements below 300°K. Allowing for differences in impurity concentration, Geballe's measurements agree with ours.

EXPERIMENTAL DETAILS

Apparatus

Figure 1 shows the sample holder and sample. The sample was a single crystal of *p*-type InSb, which had been cut in the shape of an I by means of an ultrasonically driven die. The heat source, i.e., the heater which provided the thermal gradient, was wound directly onto the sample so as to insure the best possible thermal contact between the source and the sample. The heater was composed of No. 40 enamel-coated manganin wire held in place on the sample by G.E. 7031 Glyptal and wrapped with gold foil which served as a radiation shield. The other end of the sample was soldered with indium to a small tungsten bar which was screwed to the heat sink. In order to cut down on heat leakage to the sample all leads were first wound around the upper set of rods to bring them to bath temperature and then to the lower set of rods to bring them to the temperature of the heat sink.

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¹ L. Pincherle and J. M. Radcliffe, *Suppl. Phil. Mag.* **5**, 271 (1956).

² H. P. R. Frederikse and E. V. Mielczarek, *Phys. Rev.* **99**, 1889 (1955).

³ T. Geballe, *Bull. Am. Phys. Soc. Ser. II*, **2**, 56 (1957).

⁴ R. Barrie and J. T. Edmond, *J. Electronics* **1**, 161 (1955).

⁵ A. Stuckes, *Phys. Rev.* **107**, 427 (1957).

The sample holder was placed in a Dewar containing either liquid helium or liquid hydrogen. The sample and bath temperatures were equalized by introducing helium gas in the container; however, during the measurements a vacuum of 10⁻⁶ mm Hg was maintained. Temperatures higher than bath temperature were obtained by controlling the power into the heat sink.

Thermometry

In order to determine the thermal conductivity one must measure the thermal gradient developed when a known amount of power per unit area flows through a sample. In this experiment Allen Bradley resistors (150 ohms, ½ watt) were used as thermometers. The ceramic coating of the resistors was ground off. Then the resistors were cemented with Glyptal into small copper cylinders. In order to provide electrical insulation between the copper cylinder and the resistor, the resistor was wrapped with cigarette paper and a thin coat of Glyptal applied. A copper band 1 mm wide was wrapped and soft-soldered in place around the copper cylinder, the free ends of the band were tinned with indium using lactic acid as a flux and soldered to the sample.

The resistance of the carbon thermometer as a function of temperature can be expressed by the equation⁶

$$[(\log R)/T]^{\frac{1}{2}} = a + b \log R. \quad (1)$$

This relationship gives sufficient accuracy in the region from 70°K to 4°K. The thermometers were calibrated at 4.22°K and at 20.4°K; at the beginning of each run this calibration was checked at the former temperature. The sensitivity of the resistors was approximately 4000 ohms/deg at 4°K and 2.22 ohms/deg at 60°K. Typical resistance values were 6700 ohms at 4.2°K, 554 ohms at 20°K, and 280 ohms at 57°K.

Electrical Measurements

The resistance of the carbon thermometers was measured using a Leeds and Northrup Wheatstone Bridge. Care was taken to keep the current in the resistor small so that joule heating would be negligible.

⁶ P. H. Keesom and N. Pearlman, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 14, No. 1, p. 297.

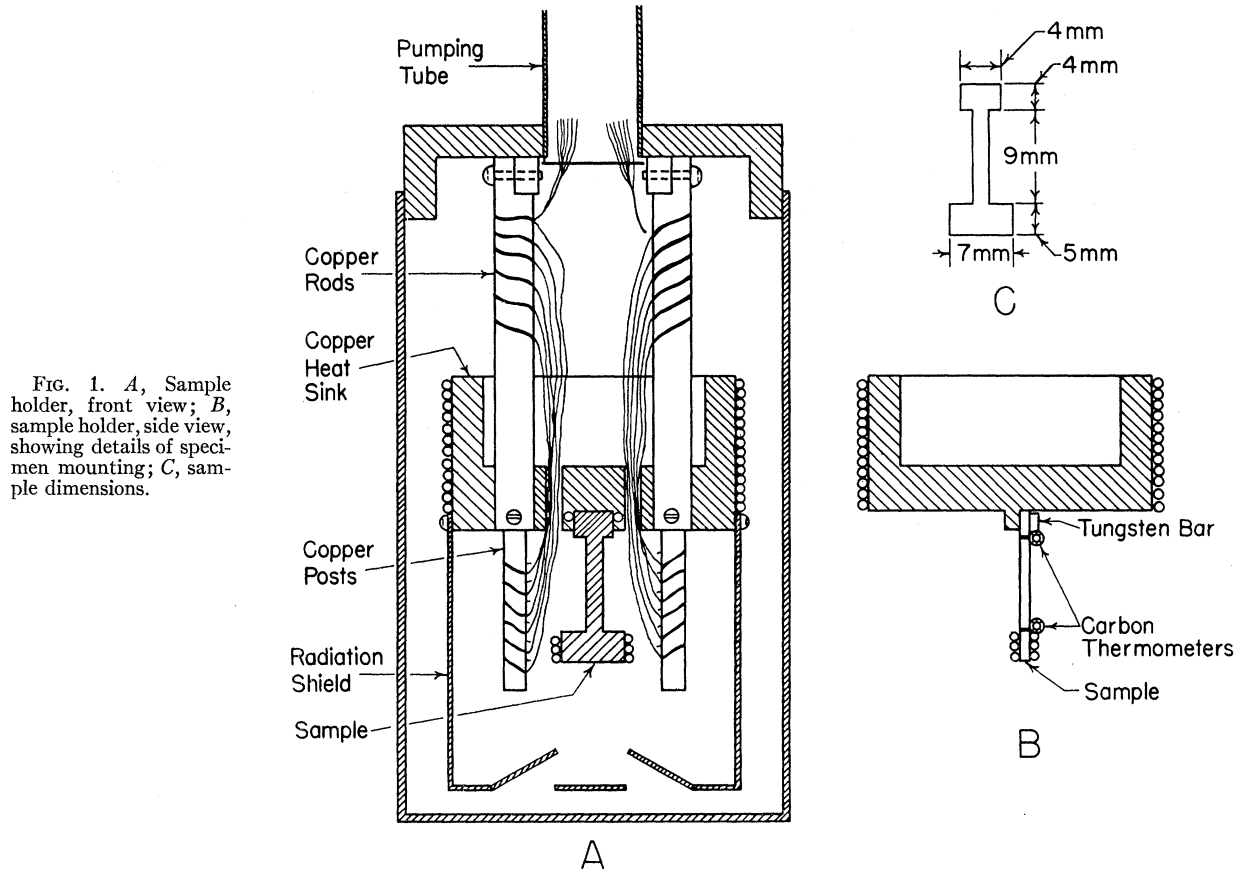


FIG. 1. A, Sample holder, front view; B, sample holder, side view, showing details of specimen mounting; C, sample dimensions.

With this precaution one can measure to an accuracy of 0.001° at 4°K and 0.2° at 60°K .

The power in the source heater, i.e., the heat transported through the sample, was measured using a four probe potentiometric method. The voltage was measured with an accuracy of 0.1% with a *K-2* potentiometer and the current with an accuracy of 2% with a precision milliammeter.

THEORY

In insulators and semiconductors the thermal conductivity at low temperatures is determined by the heat transported by the lattice. This lattice conductivity involves three main scattering mechanisms: (1) Umklapp processes, (2) scattering by crystalline boundaries, and (3) scattering by point imperfections, i.e., isotopes and impurity atoms. The total heat resistance can then be written as follows:

$$W_{\text{total}} = W_{\text{Umklapp}} + W_{\text{iso}} + W_{\text{imp}} + W_{\text{bound}} \quad (2)$$

The contributions of the different scattering mechanisms to the heat resistivity have recently been discussed by Klemens⁷ and by Slack.^{8,9} The thermal

conductivity as limited by Umklapp processes is given by the following expressions⁷:

(a) for $T > \theta$,

$$K_{\text{Umklapp}} = \frac{3 k^3 M a \theta^3}{10 \pi^3 \hbar^3 \gamma^2 T} \quad (3)$$

where k is Boltzmann's constant, M is the mass of a unit cell, a is the lattice constant, θ is the Debye temperature, \hbar is Planck's constant divided by 2π , T is the temperature, and γ is the Grüneisen constant. The Grüneisen constant can be determined from the bulk modulus λ , the volumetric coefficient of expansion $d\Delta/dT$, and the specific heat C_v :

$$\gamma = (1/C_v \lambda) d\Delta/dT \quad (4)$$

(b) for $T < \theta$,

$$K_{\text{Umklapp}} \propto (\theta/T)^2 e^{\theta/\alpha T} \quad (5)$$

where α is a parameter approximately equal to 2. No expression for the absolute value of K_{Umklapp} has been calculated for $T \ll \theta$. Leibfried and Schlömann¹⁰ have shown that a relation exists between the Umklapp conductivity at low temperature (K_{Umklapp}) and that at

⁷ P. G. Klemens, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 14, No. 1, p. 198.

⁸ G. Slack, *Phys. Rev.* **105**, 832 (1957).

⁹ G. Slack, *Phys. Rev.* **105**, 829 (1957).

¹⁰ G. Leibfried and E. Schlömann, *Nachr. Akad. Wiss. Göttingen, Math.-physik. Kl.* **4**, 71 (1954).

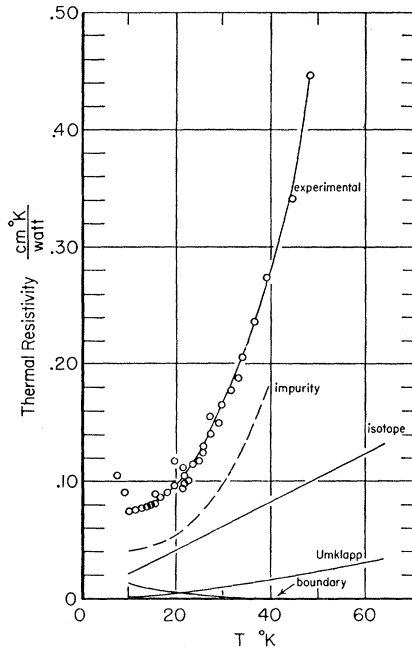


FIG. 2. Thermal resistivity of InSb at low temperatures. Isotope, Umklapp, and boundary resistivity were calculated from theoretical expressions. Impurity conductivity was deduced by subtracting the isotope, Umklapp and boundary resistivity from the total resistivity as measured experimentally.

$$T = \theta(K_\theta)$$

$$K_{\text{Umklapp}}/K_\theta = f(\theta/T). \quad (6)$$

$f(\theta/T)$ is a universal function which may be found from experimental results on other materials.

If boundary scattering were the only resistance to the phonon flow, one would find

$$K_{\text{bound}} = \frac{4\pi k^4 T^3}{h^3 v^2} L \int_0^\infty \frac{x^4 e^x}{(e^x - 1)^2} dx, \quad (7)$$

where L is the smallest linear dimension of the crystal, x is $h\nu/kT$, and the value of the integral is 25.658.¹¹

In the case of point imperfections, the thermal conductivity is given by

$$K_{\text{pt. imp.}} = \frac{1}{T} \frac{h v^2}{(2\pi)^2 3} \frac{G}{V_0 S^2} \left[\int_0^1 \frac{x^4 e^x dx}{(e^x - 1)^2} + \int_1^\infty \frac{e^x dx}{(e^x - 1)^2} \right], \quad (8)$$

where V_0 is the volume of a unit cell, G^{-1} is the concentration of impurities per unit cell, S is a scattering parameter which can usually be taken as equal to 1. The value of the integral is 0.897.¹²

¹¹ D. K. C. MacDonald and L. T. Towle, *Can. J. Phys.* **34**, 418 (1956).

¹² P. G. Klemens, *Proc. Phys. Soc. (London)* **A68**, 1113 (1955).

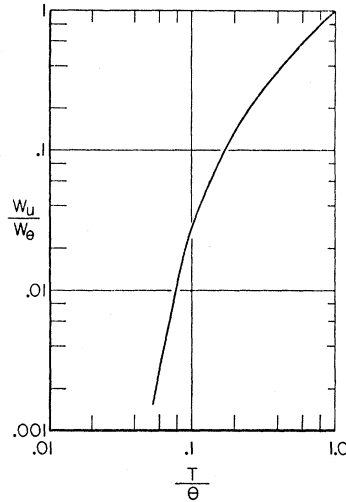


FIG. 3. The universal function $W_u/W_\theta = f(T/\theta)$.

For isotope scattering one finds

$$K_{\text{iso}} = \frac{0.897 h v^2}{12 \pi^2 T V_0 B \Gamma}. \quad (9)$$

This formula is identical with the one for point imperfections, Eq. (8), only now G/S^2 has been replaced by $1/(B\Gamma)$, where B is a numerical parameter evaluated by Slack⁸ to be 0.33, and $\Gamma = \sum_i f_i (1 - M_i/M)^2$ where M_i is the mass of a unit cell, M is the average mass for all unit cells, and f_i is the fraction of unit cells with mass M_i . A perusal of Klemens' paper¹² (especially Sec. 7) shows how Eq. (8) can be adapted to Eq. (9), a form more convenient for handling isotope scattering, i.e., use of the natural abundances.

RESULTS

Figure 2 shows the experimentally measured thermal resistance of a sample of InSb as a function of temperature. Also shown in Fig. 2 are the Umklapp, isotope, and boundary contributions as calculated from the theory outlined in the previous section.

The Umklapp contribution at $T = \theta (= 205 \pm 5^\circ \text{K})$ was evaluated using values for the bulk modulus and the thermal expansion coefficient as given by Potter.¹³ Since InSb is diatomic, the specific heat at $T = \theta$ was assumed to be $6R$, where R is the gas constant. For $T < \theta$ the contribution of Umklapp scattering to the thermal resistance was calculated on the basis of the relation by Leibfried and Schlömann¹⁰ [Eq. (6)]. The universal function of (θ/T) was deduced from Slack's paper⁹; this function is plotted in Fig. 3.

The thermal resistance due to the presence of different isotopes was calculated using the natural abundances as given in a tabulation of nuclear spins, moments, natural abundances, etc. (article by Pake¹⁴), and

¹³ R. F. Potter, *Phys. Rev.* **103**, 42 (1956).

¹⁴ G. E. Pake, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2.

TABLE I. Comparison of calculations with electrical data.

Temp. °K	Impurity concentration from thermal conductivity	Impurity concentration from Hall data	Electrical conduc- tivity (ohm ⁻¹ cm ⁻¹) at 78°K	Hall coefficient cm ³ / coulomb at 78°K	Mobility cm ² /volt sec at 78°K
10	2.26 × 10 ¹⁷ cm ⁻³	5.1 × 10 ¹⁸ cm ⁻³	14.1	145	2450
20	1.44 × 10 ¹⁷ cm ⁻³				
30	1.80 × 10 ¹⁷ cm ⁻³				
40	2.52 × 10 ¹⁷ cm ⁻³				
48	3.76 × 10 ¹⁷ cm ⁻³				
Av 2.89 × 10 ¹⁷ cm ⁻³					

assuming that at $T < \theta$ the indium and antimony vibrate as a single mass point.

Thermal resistance caused by boundary scattering was computed and found to be negligible above 30°K.

All the contributions to the thermal resistivity have

now been evaluated except W_{imp} . Using our experimentally measured value for W_{total} in Eq. (2), we can then solve for W_{imp} . The number of impurities can be calculated from Eq. (8). This value can then be compared with that derived from Hall effect and electrical conductivity measurements¹⁵ made on the same sample. (The electrical measurements give only a lower limit because the sample might be compensated.) This comparison is given in Table I.

The thermal conductivity data lead to an impurity concentration which is 4 or 5 times larger than is indicated by Hall data. The hole mobility (2450 cm²/volt sec at 78°K) shows, however, that the acceptors are considerably compensated by donor impurities.

It is our feeling that the agreement is quite satisfactory.

¹⁵ Electrical measurements were made by Mr. W. R. Hosler.

Influence of Collective Effects on the Magnetoresistance of Metals

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We have investigated the influence of the correlations between carriers on the magnetoresistance β of metals. Using the expressions derived by Fletcher and Larson, the change in β is evaluated for a one-band model. We have also used Landau's theory of a Fermi-liquid as modified by Silin to apply to the conduction electrons in a metal. The change in β both in a one-band and in a two-band model is considered. It is found that for a semiconductor like InSb the change would be negligible, but that for a metal such as Al there would be a change of about ten percent.

IT is by now a well-established fact that the correlation between the conduction electrons in a metal, which are due to their long-range Coulomb interactions, have an important bearing on the various properties of a metal. Some of the changes to be introduced in the theory of transport properties and of specific heats and x-ray band widths have been discussed by a number of people¹⁻⁵ on the basis of the Bohm-Pines theory (for an account of this theory see, for instance, reference 1). In the present paper we shall only be concerned with the magnetoresistance and we shall attack the problem using two different methods. The first one is the one used by Blatt, Barrie, or Fletcher and Larson, while the second one uses Landau's theory⁶ of a Fermi liquid as

extended by Silin⁷ to a Fermi-system with long-range interactions.

In applying the Bohm-Pines theory to magnetoresistance, we shall follow the procedure adopted by Blatt,² Barrie,³ and Fletcher and Larson,⁵ and use the standard expressions for spherical energy surfaces, but in evaluating these expressions we shall use the energy-wave number relation obtained by splitting off the long-range plasmon modes. If there is only one band which gives a contribution to the transport properties, we have for the magnetoresistance β the expression^{8,9}

$$\beta = [\rho(H) - \rho(0)] / \rho(0) = BH^2 / [1 + CH^2], \quad (1)$$

where $\rho(0)$ and $\rho(H)$ are the resistivities in zero magnetic field and magnetic field H , respectively, while B

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¹ D. Pines, *Solid State Physics* edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1955), Vol. 1, p. 367.

² F. J. Blatt, *Phys. Rev.* **99**, 1735 (1955).

³ R. Barrie, *Phys. Rev.* **103**, 1581 (1956).

⁴ M. Tsuji, *J. Phys. Soc. Japan* **13**, 133 (1958).

⁵ J. G. Fletcher and D. C. Larson, *Phys. Rev.* **111**, 455 (1958).

⁶ L. D. Landau, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **30**, 1058 (1956) [translation: *Soviet Phys. JETP* **3**, 920 (1956)].

⁷ V. P. Silin, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **33**, 495, 1282 (1957) [translation: *Soviet Phys. JETP* **6**, 387, 985 (1958)].

⁸ F. Seitz, *The Modern Theory of Solids* (McGraw-Hill Book Company, New York, 1940), p. 184.

⁹ R. A. Coldwell-Horsfall and D. ter Haar, *Phil. Mag.* **46**, 1149 (1955); the factor $\frac{1}{2}$ in front of $d \ln \lambda / d \ln e$ should in this reference be replaced by 2.