

## Thermal Conductivity of Pure Indium\*

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The thermal conductivity of a pure indium specimen was measured in the normal and superconducting states in the range of temperatures 1.3–4.2°K. In the normal state the specimen showed sizable magneto-thermal resistivity effects which were not in agreement with Kohler's rule for thermal conductivity. Near the transition temperature the ratio of conductivities  $K_s/K_n$  exhibited the finite slope with temperature characteristic of electronic conduction limited by phonon scattering. The results were compared with a simple model proposed by Kadanoff and Martin and the agreement was found to be good.

### INTRODUCTION

IT is well known that the electronic thermal conductivity, which predominates in moderately pure metals, is diminished on passage from the normal to superconducting states. In recent years the ratio of the conductivities,  $K_s/K_n$ , has been the subject of several investigations.<sup>1</sup> In those specimens for which the electronic thermal resistance is predominantly caused by lattice imperfections,<sup>2</sup> the experimental results are in reasonably good agreement with the theoretical calculations of Bardeen, Rickayzen, and Tewordt<sup>3</sup> (hereafter referred to as BRT) based on the Bardeen, Cooper, and Schrieffer theory of superconductivity<sup>4</sup> (referred to as BCS). However, when the electrons are primarily scattered by phonons, the experimental measurements of  $K_s/K_n$  differ greatly from the behavior predicted by BRT. This study of a pure indium specimen was undertaken to give further information on the nature of the heat conduction process in this limiting case. The results are compared with the simple model proposed by Kadanoff and Martin,<sup>5</sup> and the agreement was found to be good.

### METHOD

The indium specimen was mounted in a vacuum can with one end in good thermal contact with the helium bath and the other end in contact with an electrical heater. A measurable heat flux, produced by the heater, was supplied to the specimen and the resultant temperature gradient down the length of the specimen measured by carbon resistance thermometers calibrated in the manner of Clement and Quinell.<sup>6</sup> A mechanical support made of linen delecto was used to support the weight of the

thermometer and heater assemblies since the pure indium was extremely soft and easily deformed. The parallel heat conductance provided by this support and the advance wire electrical leads was found to be negligible compared to the conductance of the pure indium specimen. During the course of the thermal conductivity measurements and the calibration of the carbon resistances, the helium was maintained at a constant temperature using a bellows-operated manostat and a thermal regulating device similar to that described by Sommers.<sup>7</sup> The relative accuracy of the experimental points was probably limited by the accuracy of the temperature determination and was worst at lower temperatures where the measured temperature gradients were smallest. This is consistent with the observed scatter of the data points which tended to be greatest at low temperatures.

The specimen was a polycrystalline spectroscopically pure indium wire which had been extruded to a diameter of about 0.5 mm. Since indium has a comparatively low recrystallization point, it is thought that many of the strains introduced by the extrusion process were removed in the several months between the time the wire was extruded and the time the thermal conductivity measurements were made. Although electrical measurements were not made on the specimen, it was estimated from the normal state thermal conductivity data that the residual resistance ratio ( $\rho_{\text{room temperature}}/\rho_{0^\circ\text{K}}$ ) was about 11 000 indicating that the specimen was extremely pure and that few strains or impurities were introduced in the experiment. Using the calculated residual resistance and the ratio of conductivity to mean free path measured by Pippard,<sup>8</sup> a mean free path of about 0.1 mm was computed. Since this figure was of the order of the grain size of the specimen, it is possible that grain boundaries represented the bulk of the static defects present.

### RESULTS AND DISCUSSION

The measured thermal conductivities in the normal and superconducting states corrected to zero field are shown in Fig. 1. By analyzing the temperature depend-

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<sup>1</sup> Two recent review articles are: P. G. Klemens, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 14, p. 266; K. Mendelssohn, *Progress in Low-Temperature Physics* (North Holland Publishing Company, Amsterdam, 1957), Vol. 1.

<sup>2</sup> C. B. Satterthwaite, Cambridge Superconductivity Conference, 1959 (unpublished).

<sup>3</sup> J. Bardeen, G. Rickayzen, and L. Tewordt, *Phys. Rev.* **113**, 982 (1959).

<sup>4</sup> J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Phys. Rev.* **108**, 1175 (1957).

<sup>5</sup> L. P. Kadanoff and P. C. Martin (to be published).

<sup>6</sup> J. R. Clement and E. H. Quinell, *Rev. Sci. Instr.* **23**, 213 (1952).

<sup>7</sup> H. S. Sommers, Jr., *Rev. Sci. Instr.* **25**, 793 (1954).

<sup>8</sup> A. B. Pippard (private communication).

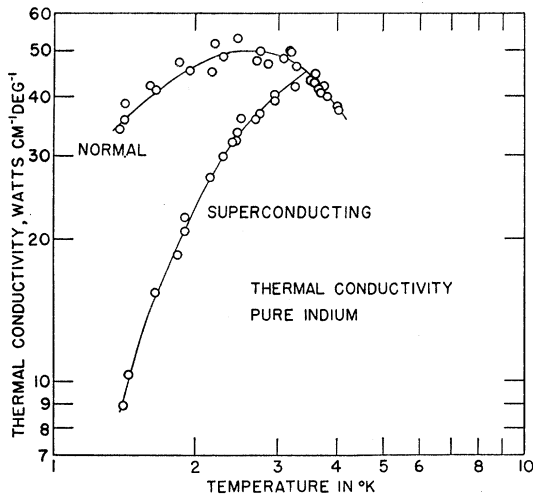


FIG. 1. Thermal conductivity of indium in the superconducting and normal states corrected to zero magnetic field.

ence and magnitude of the normal state conductivity, it is possible to deduce quantitatively the part of the resistance due to scattering by static defects and the part due to scattering by phonons at each temperature in the normal state. For, assuming the additivity of imperfection and phonon resistances (Matthiessen's rule), the normal state thermal resistance is given by the following expression:

$$W = W_i + W_p = A/T + BT^2. \quad (1)$$

Consequently,  $WT$  may be expressed as a sum of a constant representing imperfection scattering and a phonon term varying as  $T^3$ . Figure 2 shows the plot of  $WT$  versus  $T^3$  for the normal state data. The coefficient  $A$  found from this plot was  $0.034 \text{ cm}^2\text{K/watt}$ , which is lower by an order of magnitude than previously measured values for indium,<sup>9,10</sup> as would be expected considering

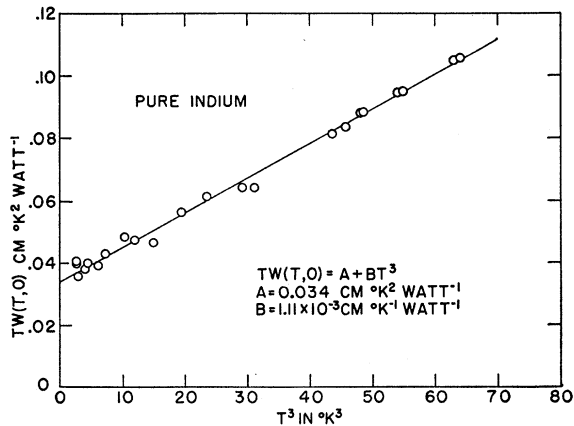


FIG. 2.  $W(T,0)$  vs  $T^3$  for the normal state corrected to zero magnetic field.

<sup>9</sup> J. K. Hulm, Proc. Roy. Soc. (London) A204, 98 (1950).

<sup>10</sup> H. M. Rosenberg, Phil. Trans. Roy. Soc. (London) A247, 441 (1955).

the high purity of the specimen. However, the coefficient  $B$ , which to a first approximation should be independent of purity, was  $1.11 \times 10^{-3} \text{ cm}^2/\text{K}^2\text{-watt}$ , compared to the values of  $1.89 \times 10^{-3}$ <sup>9,10</sup> and  $1.85 \times 10^{-3} \text{ cm}^2/\text{K}^2\text{-watt}$  reported previously. The exact reason for this discrepancy is not known, although one possible explanation may lie in the fact that the previously reported values were mainly determined from data in a higher temperature range and that in order to have the data agree with Eq. (1) at every temperature the parameter  $B$  must vary with temperature.

Knowing the constants  $A$  and  $B$ , one can calculate the ratio of phonon to defect resistances at any temperature. Over the measured range of temperatures,  $W_p/W_i$  goes from about 0.1 at  $1.5^\circ\text{K}$  to over 3 at  $4.2^\circ\text{K}$ . The value of this ratio at the transition temperature ( $T_c = 3.407^\circ\text{K}$ ), 1.3, will be used in the later discussion of the superconducting state.

To measure the normal state conductivity below the superconducting transition temperature, a longitudinal

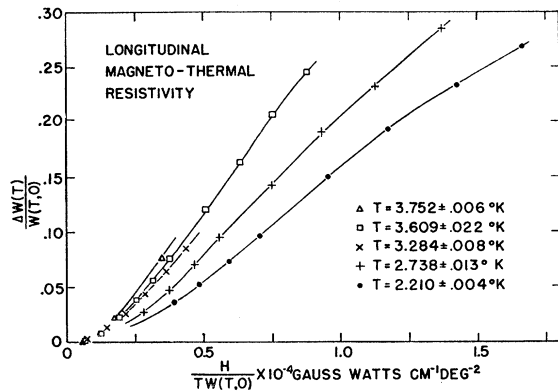


FIG. 3. Relative change of thermal resistance plotted against the coordinate  $H/W(T,0)$ . This plot would give a reduced curve if Kohler's rule was obeyed.

magnetic field was used to switch the sample into the normal state. It was found, however, that even at fields of a few hundred gauss, the indium specimen showed a considerable increase in the thermal resistance with increasing fields. Hence, in order to obtain the normal conductivity at zero field, it was necessary to establish the variation of the thermal resistivity with changing magnetic field and temperature. Previous experimenters have found that the relative variation of thermal resistance with magnetic field was in approximate agreement with Kohler's rule for thermal conductivity,<sup>11</sup> i.e.,

$$\Delta W(T)/W(T,0) = G(H/TW(T,0)), \quad (2)$$

where  $W(T,0)$  is the thermal resistance in zero magnetic field and  $G$  is a function characteristic of each metal. However, for our data a plot of the relative change of resistance versus the coordinate  $[H/TW(T,0)]$  failed

<sup>11</sup> M. Kohler, Ann. Physik 6, 18 (1949); Naturwiss. 36, 186 (1949).

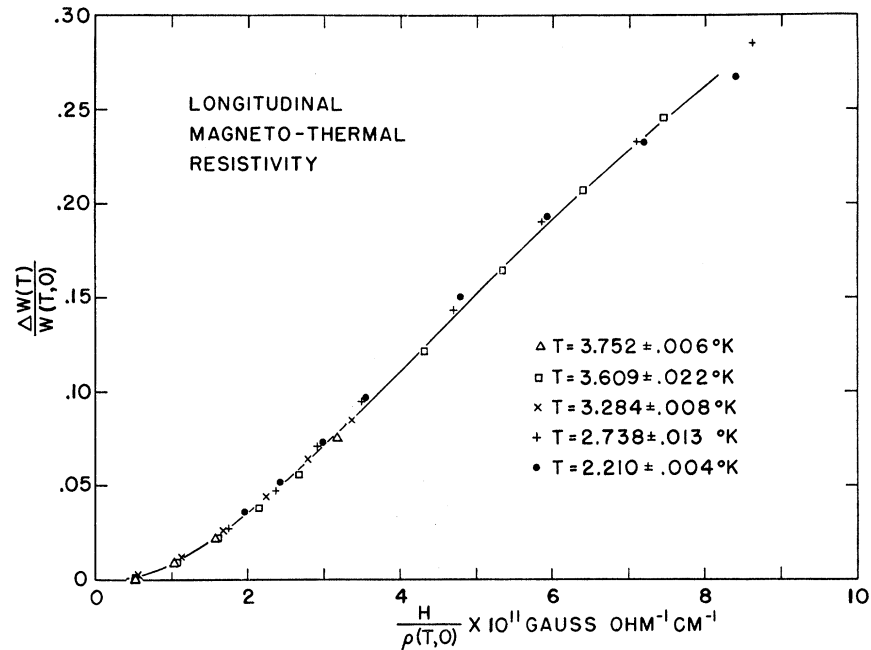


FIG. 4. Relative change of thermal resistance plotted against the coordinate  $H/\rho(T,0)$ .

to give a reduced curve as can be seen in Fig. 3. This implies a failure of the above form of Kohler's rule in the region of transition from impurity to phonon scattering. On the other hand, on a plot of the relative change of thermal resistance versus  $H/\rho(T,0)$ , where  $\rho$  is the electrical resistivity, the experimental points lay on a single smooth curve (see Fig. 4). [In fact both  $\Delta W/W(T,0)$  and  $\rho$  are nearly independent of temperature over the range 2.21–3.75°K.] The values of  $\rho(T,0)$  for the specimen were calculated by using Matthiessen's rule for electrical resistivity:

$$\rho(T,0) = \rho_0 + \rho_i(T), \quad (3)$$

where the residual resistivity,  $\rho_0$ , was calculated from the coefficient  $A [= 3/\pi^2(e/k)^2\rho_0]$  and the temperature dependent resistivity,  $\rho_i(T)$ , was calculated from the data of White and Woods.<sup>12</sup>

Thus, it appears that the relative magnetothermal resistivity in the region of transition from impurity to phonon scattering is of the form:

$$\Delta W(T)/W(T,0) = F(H/\rho(T,0)). \quad (4)$$

In the regions of purity and temperature in which it is possible to define a universal relaxation time, this formula would reduce to Eq. (2), since in these regions  $\rho$  is given by the expression:

$$\rho = TWL_0,$$

where  $L_0$  is the Sommerfeld value of the Wiedemann-Franz-Lorentz number. For the further discussion of this expression and Kohler's rules, see the Appendix.

Among the recent theoretical papers on supercon-

ductivity, there are two in which an attempt is made to calculate the ratio  $K_s/K_n$  in the region where phonon scattering is important. In the first of these, by BRT,<sup>3</sup> the results indicated that the ratio should rise as the temperature fell below the transition temperature in the limit that all the resistance was due to phonons. This rise is in contradiction with all the experimental results. In the second of these papers, Kadanoff and Martin<sup>5</sup> have proposed an expression of the following form:

$$\frac{K_s}{K_n} = \frac{3}{2\pi^2} \int_0^\infty d(\beta\epsilon) (\beta\epsilon)^2 \operatorname{sech}^2 \frac{\beta E}{2} \times \left[ 1 + a \left( \frac{T}{T_c} \right)^3 \right] \left[ \frac{(\beta\epsilon)}{(\beta E)} + a \left( \frac{T}{T_c} \right)^3 \right]^{-1}, \quad (5)$$

where we have rewritten the expression in terms of the symbols used by BCS; the additional parameter,  $a$ , is defined as the ratio of phonon to impurity resistance in the normal state at the transition temperature. In the limiting case of all impurity scattering in the normal state ( $a=0$ ) this expression is identical to that calculated by BRT for this case. In the other limit,  $a \rightarrow \infty$  (all phonon scattering), the equation predicts a monotonic decrease of  $K_s/K_n$  with decreasing reduced temperature. For intermediate values of  $a$ ,  $K_s/K_n$  at a given reduced temperature is between the values of  $K_s/K_n$  for the two limiting cases; however for these intermediate cases it is not possible to write the general expression for  $K_s/K_n$  in terms of the two limiting expressions after the manner of Hulm.<sup>9</sup> A direct evaluation of the integral [Eq. (5)] must be made.

<sup>12</sup> G. K. White and S. B. Woods, Rev. Sci. Instr. 28, 638 (1957).

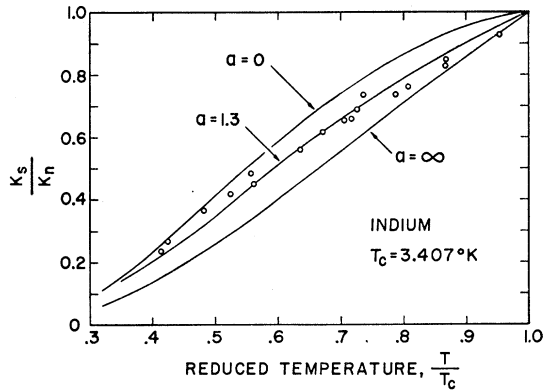


FIG. 5.  $K_s/K_n$  versus  $T/T_c$  for the experimental points and the theoretical curves of Kadanoff and Martin for the cases  $a=0$ ,  $\infty$ , and 1.3.

In Fig. 5 our experimental points are compared with the theoretical curves for  $a=0$  and  $a=\infty$  and with the curve which should be applicable in our case, namely  $a=1.3$ . In each case the energy gap at 0°K was taken to be  $2\epsilon_0(0) = 3.5kT_c$ . The overall agreement is reasonably good, although there appear to be systematic deviations below the theoretical curve near the transition temperature and deviations above the theoretical curve at low temperatures.

#### APPENDIX

To understand the disagreement between the present thermal magnetoresistance measurements and the Kohler rule for thermal conductivity, it is helpful to consider the derivation of Kohler's rules for electrical and thermal conductivity. It can be shown directly from the Boltzmann equation (see, for example, Klemens<sup>1</sup> or Ziman<sup>13</sup>) that, if one assumes a relaxation time solution, then the relative change in the distribution function for a state  $K$  in the presence of a magnetic field  $H$  is a function of  $H\tau$ . From this the similarity relations proposed by Kohler follow:

$$\Delta\rho(T)/\rho(T,0) = F(H/\rho(T,0)), \quad (\text{A1})$$

$$\Delta W(T)/W(T,0) = G(H/TW(T,0)), \quad (\text{A2})$$

where  $F$  and  $G$  are functions which depend on the details of the band structure. Similar results were obtained by

<sup>13</sup> J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, 1960), p. 491.

Sondheimer and Wilson<sup>14</sup> on the basis of a special model, the two-band model.

At temperatures below the Debye temperature, the scattering of electrons by phonons cannot be characterized by a relaxation time. For this reason, when the phonon part of the electrical or thermal resistivity becomes appreciable, one cannot really define a relaxation time. Over the temperature range 2.2–3.7°K, the range covered by the magnetoresistance measurements,  $\rho_i(T)$  as defined by Eq. (3) comprises 1–14% of the total resistivity, whereas  $BT^2$  as defined by Eq. (1) comprises 26–62% of the total thermal resistivity over the same temperature range. Hence, it seems reasonable that a relaxation time solution of the Boltzmann equation would be a better approximation to the electrical magnetoresistivity than to the thermal. Hence, Equation (A1) would be more valid than Eq. (A2).

If we define a quantity  $L$  by the relation  $\rho = LWT$ , then  $L$  will in general be a function of  $H$  and  $T$ . If upon the application of a magnetic field, the relative change in  $L$  is small compared to the relative change in  $W$ , then  $\Delta W/W(T,0)$  is approximately equal to  $\Delta\rho(T)/\rho(T,0)$  and the thermal magnetoresistivity is given by Eq. (A1). If one makes the assumption that  $L$  is a function of  $H/\rho(T,0)$  rather than approximately independent of field, then  $\Delta W(T)/W(T,0)$ , although not equal to  $\Delta\rho(T)/\rho(T,0)$ , is still a function of  $H/\rho(T,0)$ . Therefore, under either of these assumptions, the result follows that

$$\Delta W(T)/W(T,0) = g(H/\rho(T,0)), \quad (\text{A3})$$

where  $g$  is a function characteristic of a given metal. Indeed, this is the relation which the data satisfy, as can be seen from Fig. 4.

At temperatures low enough (or defect concentrations high enough) that a universal time of relaxation can be defined, then Eq. (A3) reduces to Kohler's rule for thermal conductivity, Eq. (A2), since now  $L$  is equal to  $L_0 = \pi^2/3(k/e)^2$ , and  $\rho$  is equal to  $L_0WT$ .

#### ACKNOWLEDGMENTS

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<sup>14</sup> E. H. Sondheimer and A. H. Wilson, Proc. Roy. Soc. (London) **A190**, 435 (1947).