Thermal and Electrical Conductivities of Very-High-Purity Indium*

M. E. DE LA CRUZ, F. DE LA CRUZ,[†] J. M. COTIGNOLA,[‡] O. J. BRESSAN, AND C. A. LUENGO Centro Atómico Bariloche, Comisión Nacional de Energía Atómica San Carlos de Bariloche, Río Negro, República Argentina

(Received 12 June 1968)

Measurements of the thermal and electrical conductivities of a polycrystalline indium sample of very high purity have been performed between 1.5 and 4.2°K and in the presence of longitudinal magnetic fields up to 15 000 Oe. It was found that, within the experimental error, the electrical and thermal Kohler's rules were satisfied, the functional forms for the two being quite similar. The expression for the resistivity as a function of T for the normal state and in zero magnetic field are in good agreement with theory. The Lorenz number obtained by extrapolating to T=0 gives a very high value compared with the theoretical one. Measurements of the thermal conductivity for the superconducting state, in the same temperature range, have been made. The thermal-resistivity ratio W_n/W_s as a function of the reduced temperature does not agree with the results of Kadanoff and Martin.

I. INTRODUCTION

CCORDING to the Wiedemann-Franz law, the A ratio $L(T) = \rho/W_n T$, where ρ and W_n are the electrical and thermal resistivities at temperature T, should be a constant for all metals and equal to $L_0 = 2.44$ ×10⁻⁸ W Ω °K⁻².

The law will be satisfied if the electron scattering is elastic; this is the case when the temperature is low enough and the scattering of the electrons takes place mainly at the impurities.

Theory predicts that for low enough temperatures, ρ and $W_n T$ should be described by the following functions of T:

$$\rho = \rho_0 + A T^5, \tag{1}$$

$$W_n T = \alpha + \beta T^3. \tag{2}$$

The ratio between (1) and (2) gives the expression for L(T), and in the limit T=0 this expression should be equal to L_0 .

In formulas (1) and (2), ρ_0 and α are dependent on the purity, and the purer the materials under study, the smaller they become.

Thus, in samples of very high purity, the determination of the resistivity as a function of T will have a large influence upon the extrapolation to zero temperature, which should give the value L_0 .

Measurements carried out in indium by different authors¹⁻⁷ have shown that relations (1) and (2) are

* This work has been supported by the Consejo Nacional de Investigaciones Científicas y Técnicas (República Argentina) under Grant No. 2491/66.

† Present address: Department of Physics, Brown University, Providence, R. I.

‡ Present address: C. E. N. G., Boîte Postale 269, Grenoble, France.

¹ J. K. Hulm, Proc. Roy. Soc. (London) 204, 98 (1950)

² H. M. Rosenberg, Phil. Trans. Roy. Soc. London A247, 441 (1955)

⁽¹⁾ G. K. White and S. B. Woods, Rev. Sci. Instr. 28, 638 (1957).
⁴ A. M. Guenault, Proc. Roy. Soc. (London) A262, 420 (1961).
⁵ B. N. Aleksandrov and I. G. D'Yakov, Zh. Eksperim. i Teor. Fiz. 43, 852 (1962) [English transl.: Soviet Phys.—JETP 16, 603

(1963)]. ⁶ R. E. Jones and A. M. Toxen, Phys. Rev. **120**, 1167 (1960).

⁷ P. Wyder, Physik Kondensierten Materie 3, 263 (1965).

176

satisfied and that the Wiedemann-Franz law holds for T=0. However, the measurements reported here, in a sample of purity higher than those used by these authors, have shown an incompatibility between (2) and the extrapolation to the Lorenz number L_0 .

Indium becomes superconducting for temperatures below $T_c = 3.4^{\circ}$ K. Measurements in the normal state below this temperature were made by applying a longitudinal magnetic field H. To be able to obtain the resistivities at zero field, it was necessary to measure the electrical and thermal magnetoresistance. The electrical Kohler's rule was satisfied at several temperatures, and the values obtained were in good agreement with previous results.8

The thermal measurements satisfied Kohler's relation

$$[W_{n}(H) - W_{n}(0)]/W_{n}(0) = G[H/W_{n}(0)T], \quad (3)$$

where $W_n(H)$ and $W_n(0)$ are the resistivities for H and zero magnetic field, respectively, G is a characteristic function of the metal, and T is the absolute temperature.

Because of the high purity of the sample, the main contribution to the thermal resistivity in the normal state for $T = T_c$ arises from the scattering of electrons by phonons. The thermal resistivity in the superconducting state W_s was measured in order to obtain W_n/W_s as a function of T/T_c . The results show considerable departures from the curves of Kadanoff and Martin.9

II. EXPERIMENTAL DETAILS

One slice was cut out of an ingot of 99.9999% pure indium from Consolidated Mining and Smelting Co., Canada, Ltd.; the material was rolled between Teflon plates to a thickness of 1.5 mm. Much care was taken with the cleanliness during the rolling process so as to avoid contamination of the material.

A sample in the form of a rectangular prism with the ratio of cross section to length $A/L=3.96\times10^{-3}$ cm was cut from the rolled material. The geometry factor

⁸ F. de la Cruz, M. E. de la Cruz, and J. M. Cotignola, Phys. Rev. 163, 575 (1967).

⁹L. P. Kadanoff and P. C. Martin, Phys. Rev. 124, 270 (1961). 871



872

FIG. 1. Experimental arrangement. A—He⁴ evaporator, B thermal shield, C—vacuum jacket, D—coupling between evaporator and Mylar frame, E—Mylar frame, F—indium sample, G—heater, H—superconducting magnet, I—cotton threads used to center the sample, J—nylon_cap, K—He⁴ bath, R₁, R₂—carbon resistor.

was determined by measuring the resistance of the sample at room temperature and assuming that ρ_{293} °_K = 9.1×10⁻⁶ Ω cm.¹⁰

The ratio between resistivities at room temperature and at 4.2° K was found to be 2×10^4 .

The experimental arrangement is shown in Fig. 1. The He⁴ evaporator (A) was thermally isolated from the helium bath by a vacuum jacket (C). Pumping on the liquid helium of the evaporator made it possible to lower the temperature to 1° K.

The sample (F) and the thermometers (R_1) and (R_2) were fixed on a frame (E). This was made of a 0.005in.-thick Mylar sheet which was bent longitudinally into a U shape in order to obtain more rigidity. The frame was glued on to an Epoxy 100 A piece (D) which was screwed to the evaporator. In order to avoid deformations due to the different expansion coefficients, the sample was glued (with GE 7031 varnish) into the frame at only one end, above the upper thermometer R_2 . At a distance of 1 cm above the R_2 thermometer, the sample was widened and fixed to the evaporator with Apiezon N grease; this part of the sample was covered with a 0.1-mm-thick copper sheet, which was wrapped around the evaporator in order to get a larger contact surface. Thermal contact among the sample, the copper sheet, and the evaporator was made by means of Apiezon N grease. The contact area was about 4 cm².

The thermometers R_1 and R_2 consisted of $10 \cdot \Omega$, $\frac{1}{4}$ -W Allen-Bradley carbon resistors. Contact between the thermometers and the sample was made by soldering a 1-mm-diam copper wire to the sample; the other end of each wire was soldered to a piece of coil-foil¹¹ wrapped tightly upon the resistor and in contact with it by means of Apiezon N grease. In all cases indium was used as soldering material in order to avoid the presence of other superconductors near the sample. The electrical wiring for the thermometers was made from 0.05mm-diam manganin wire.

The heater (G) was made with manganin wire coiled on a piece of Mylar sheet and glued to it with GE 7031 varnish. The thermal contact between the heater and the sample was made with Apiezon N grease. The distance from the heater to R_1 was of the order of 1 cm. The electrical wiring to measure the voltage on the heater was made with 0.05-mm-diam manganin wire. To avoid heat dissipation outside the heater, the wires supplying current to it were made of copper¹² and were coiled in the shape of springs to avoid mechanical work on the sample. The length of the copper wires between heater and evaporator was of the order of 30 cm.

After assembly, the arrangement was put into a liquid-air bath for testing, and no deformation could be detected.



FIG. 2. Thermal resistivity $W_n(H)$ versus longitudinal magnetic field H at T=3.6 and 2.2° K.

¹⁰ Wright Air Development Division Technical Report No. 60-56, Part IV (unpublished).

¹¹ A. C. Anderson, G. L. Salinger, and J. C. Wheatley, Rev. Sci. Instr. **32**, 1110 (1961). ¹² Anaconda copper wire B&S No. 30.



FIG. 3. The unbroken curve gives the thermal Kohler's rule for indium in a longitudinal magnetic field. H as a function of $H/W_n(0)T$. Temperatures corresponding to symbols are:

$T = 3.81^{\circ}$ K,	$\diamond T = 3.60^{\circ} \mathrm{K},$	$\times T = 3.17^{\circ} \text{K},$
$\nabla T = 2.69^{\circ} \mathrm{K},$	$\circ T = 2.22^{\circ} K$,	$\Box T = 1.2$ °K,
$+T = 1.53^{\circ} K$		

The dashed curve gives the electrical Kohler's rule for indium in a longitudinal magnetic field H as a function of $\gamma H/\rho(0)$. $\gamma=2.06 \times 10^{-8} \text{ W } \Omega \text{ }^{\circ}\text{K}^{-2}$.

The vacuum jacket (C) was removed to make the electrical measurements. The electrical connections to measure the voltage drop were made on the thermal contacts between the sample and thermometers without modifying the arrangement. The current leads were soldered on the widening of the sample and below the heater.

The magnetic field was applied with a Magnion superconducting coil, model CF 20-200-800.

When the measurements were finished, the portion of the sample between R_1 and R_2 was cut out. The thermal conductance of the rest was measured and was found to be negligible compared with that of the part taken out.

III. EXPERIMENTAL METHOD

In order to measure the thermal conductivity, thermometers R_1 and R_2 were calibrated over the whole range of temperature against the vapor pressure of the He⁴ in the evaporator. This was done at the beginning of each experiment. A pressure regulator¹³ was used to maintain constant temperature.

It was checked that no magnetoresistive effects were present on the thermometers due to the applied magnetic fields. Resistances were measured by means of an ac bridge with a frequency of 33 cps. By regulating the He⁴ pressure it was possible to obtain the desired working temperature. Once the system attained equilibrium, power was applied to the heater and the helium pressure was changed so as to maintain R_2 constant. The temperature difference between the ends of the sample was obtained from the values of R_1 and R_2 . Power was calculated from voltage measurements made with a K-3 potentiometer.

The electrical measurements were made by putting a constant current through the sample and measuring the voltage drop. The current was measured with a Weston model 931 ammeter (3 A full scale) and the voltage was measured with a Keithley model 149 millimicrovoltmeter ($0.1 \,\mu V$ full scale maximum sensitivity). Several current values were used in order to ensure that no heating of the sample took place; the value of the current finally used was 2 A.

IV. EXPERIMENTAL RESULTS

A. Thermal and Electrical Magnetoresistance

Measurements of the electrical and thermal magnetoresistance were carried out alternately, with intervals of some days, during which the sample was allowed to attain room temperature.

The isothermic variation of W_n as a function of the magnetic field was measured at several temperatures. Figure 2 shows two of the curves obtained. It is easy to appreciate that W_n depends very strongly on the magnetic field; consequently, it is very important to find a good extrapolation procedure which enables one to find the values of $W_n(0,T)$ for $T < T_c$.

Wyder' has developed an extrapolation method which allows the determination of $W_n(0,T)$ from the values obtained for $T > T_c$. The method is based on the existence of a certain characteristic function whenever the representations on log-log paper of $W_n(H,T)$ as a function of H for different values of T can be made to coincide one with another by means of a simple displacement parallel to the axis. This was possible in our case, and the corresponding characteristic function was that given for Kohler's relation (3). Figure 3 shows the results.



FIG. 4. Thermal resistivity of the normal and superconducting states versus temperature.

¹³ P. G. Strelkov and E. J. Walker, Rev. Sci. Instr. 30, 834 (1959).



Measurements of the electrical magnetoresistance were made on the same sample and in the same range of temperature. The values of $\rho(0,T)$ below the critical temperature were obtained with the same method used in the thermal case. These values were used in the determination of the electrical Kohler's function, which was found to be in good agreement with previous results.⁸

B. Dependence of Thermal and Electrical Resistivities on the Tempersture

In Fig. 4 the results obtained for the thermal resistivity of the normal state with zero field are shown. These results satisfied relation (2) within the experimental error.

where

and

$$W_n(I)I = \alpha + \beta I^\circ,$$

 $\beta = 9.6 \times 10^{-4} \text{ cm} \,^{\circ}\text{K}^{-1} \,^{\circ}\text{W}^{-1}$.

$$\alpha = 1.8 \times 10^{-3} \text{ cm }^{\circ} \text{K}^2 \text{ W}^{-1}$$

Figure 5 shows $W_n(T)T$ as a function of T^3 .

The results for the electrical resistivity $\rho(T)$ can be expressed, within the experimental error, by expression (1), i.e.,

$$\rho(T) = \rho_0 + A T^5,$$

where $\rho_0 = 1.0 \times 10^{-10} \ \Omega$ cm and $A = 3.33 \times 10^{-13} \ \Omega$ cm





 $^{\circ}$ K⁻⁵. The corresponding representation is shown in Fig. 6.

Figure 7 shows values of the Lorenz number as a function of T:

$$L(T) = \rho(T) / W_n(T) T, \qquad (4)$$

as it was obtained from relations (1) and (2). If (1) and (2) are valid in the limit where T=0, then

$$L(0) = 5.6 \times 10^{-8} \text{ W } \Omega \,^{\circ} \text{K}^{-2}$$

The values of L(H,T)/L(0,T), as a function of H for the range of temperature where the present measurements were carried out, can be seen in Fig. 8.

The thermal resistivity of the superconducting state $W_s(T)$ was always measured before switching on the magnetic field. The results for $W_s(T)$ can be seen in Fig. 4.

V. DISCUSSION

The estimated error in the measured values, which is of the order of 3%, arises mainly from the error in ΔT .

Measurements made by Mendelssohn and Rosenberg¹⁴ in indium, in regions where the scattering is mainly due to impurities, with perpendicular magnetic fields up to 4 kOe and in a temperature range 2–4.5°K, showed that relation (3) is satisfied. Guenault⁴ verified the same relation for indium in longitudinal magnetic fields, but because of the lack of graphical representation of his results it was not possible to compare them with ours. Other authors^{6,7} did not find an agreement between relation (3) and their experimental results.

Figure 3 also shows the electrical Kohler's rule as a function of $\gamma H/\rho(0)$. The constant $\gamma = 2.06 \times 10^{-8}$ W Ω °K⁻² was introduced to make a convenient change in the scale to show that, in this representation, both of Kohler's functions can be made to coincide by means of a translation parallel to the ordinate axis. Assuming



FIG. 7. Lorenz number for zero field versus temperature. The unbroken line corresponds to the measured range.

¹⁴ K. Mendelssohn and H. M. Rosenberg, Proc. Roy. Soc. (London) **218**, 190 (1953).

874



FIG. 8. Lorenz number versus magnetic field.

that both functions continue to be parallel up to saturation values of the field, we can define a more general function

$$\frac{(\Delta W/W_n(0))}{(\Delta W/W_n(0))_{\infty}} = \frac{(\Delta \rho/\rho(0))}{(\Delta \rho/\rho(0))_{\infty}} = f(H/Z),$$

where the value of the denominator for each case is the corresponding saturation value of the Kohler's function.

The parameter Z takes different values depending on whether one wishes to express the electrical or the thermal magnetoresistance.

$$Z_{\rm el} = \rho(0), \quad Z_{\rm th} = W_n(0)T.$$

The coefficient β in expression (2) is in good agreement with values obtained by Jones and Toxen⁶ and Wyder.⁷ On the other hand, the coefficient of T^5 in expression (1) does not agree with the value obtained by Aleksandrov and D'Yakov,⁵ but it agrees with the values obtained by other authors.^{3,7}

The values of ρ_0 and α in (1) and (2), which are dependent on purity, are much smaller than the ones obtained in previous work.

A least-squares adjustment was made to find the best value of the exponent in the exponential dependence of W_nT on T. A similar method was used to find the dependence of ρ on T in expression (1). Both functions are in good agreement with experimental results of other authors and with the predictions of existing theories. However, the value of L(0) which results is much larger than the theoretical one. On the other hand, in our range of measurements, this value is very sensitive to the T dependence of $W_n(T)T$. The dependence



FIG. 9. W_n/W_s versus T/T_c . The dashed line corresponds to the values obtained from curves of Fig. 4, and the unbroken line gives the theoretical curve of Kadanoff and Martin for $a = \infty$.

of ρ on T is not so important, since we are working near the residual zone.

It is important to note that if we take the value 3.4 for the power to which T is raised in Eq. (2), the Lorenz number obtained by extrapolating to zero temperature equals the theoretical one. In that case the dispersion of the experimental values is only 20% larger than the dispersion which results with the T^3 law.

It is thus possible to conclude from the present measurements that the theoretical value of the Lorenz number is incompatible with the fulfillment of the T^3 law and vice versa.

Figure 9 shows the ratio W_n/W_s as a function of T/T_c . By means of expression (2) it is possible to determine that the contribution of phonon scattering to the resistivity is of 95%. In this case the curve of Kadanoff and Martin⁹ for $a = \infty$ should be applicable. It can be seen from Fig. 9 that the present results do not agree with the curve corresponding to $a = \infty$, and that they are in the region between the Kadanoff and Martin curve and the values corresponding to mercury¹ and lead.^{2,15}

¹⁵ W. J. de Haas and A. Rademakers, Physica 7, 992 (1940).