

## THERMAL AND ELECTRICAL CONDUCTIVITY OF Ag AND Pt BELOW 1°K\*

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Results obtained in the temperature range 0.3–1.2°K give direct evidence that electron-electron scattering in the transition metal Pt is at least an order of magnitude greater than in Ag. Our results do not confirm the anomalous behavior in thermal conductivity which has been reported for numerous metals below 1°K.

Theoretically, the thermal conductivity of normal metals is expected to vary linearly with temperature in the limit of very low temperatures. In recent years, however, several experimental papers have reported oscillatory deviations from this behavior for a variety of metals in the temperature range 0.3–1.0°K.<sup>1-4</sup> No attendant anomalies were observed in the electrical conductivity, nor in the thermal conductivity of the superconducting state.<sup>4</sup> One might therefore infer that the phenomenon is not found in the thermal transport contributed by phonons, but rather by electrons. In addition, the effect would be associated with small-angle electron scattering which is of importance in thermal conductivity as opposed to electrical conductivity. The fact that the magnitude of the anomalies was found to scale roughly with the magnitude of the thermal conductivity, i.e., with the purity of the metal,<sup>1,5</sup> is suggestive of an error in thermometry, but thermal-conductivity data for the superconductors and for sapphire which varied as  $T^3$  appeared to rule this out.<sup>4</sup> Nevertheless, in view of the unusual nature of the above-mentioned implications and since all previous measurements were evidently made in the same cryostat, we decided to make similar measurements on Ag and Pt. This also allowed us, in a temperature range where electron scattering by phonons should contribute less than 1:10<sup>3</sup> to the total scattering probability, to compare electron scattering in a noble metal with that in a transition metal. The latter has a greater density and variety of states at the Fermi surface because of a more significant overlap of *s* and *d* bands.

The technique was to establish a known thermal flux  $\dot{Q}$  in a sample of cross sectional area *A* and length *l*, measure the (time-independent) temperatures  $T_C$  and  $T_H$  at points *C* and *H* a distance *l* apart on the sample, and compute the thermal conductivity *K* from  $K = l\dot{Q}/A(T_H - T_C)$ , where  $T_H - T_C \lesssim 0.1T_H$ . An uncalibrated resistance thermometer clamped to *C* main-

tained that point at a fixed temperature ( $T_C$ ) by means of an electrical heater and electronic (or manual) regulation. With  $\dot{Q} = 0$ ,  $T_C$  was measured with a cerium magnesium nitrate (CMN) magnetic thermometer attached to *H*. Turning on  $\dot{Q}$  then permitted the same magnetic thermometer to measure  $T_H$  while  $T_C$  remained constant. Since the thermometric parameter of CMN—namely the inverse susceptibility—varies linearly with *T*, errors in calibration could at most produce a slight curvature in a plot of *K* vs *T*. The CMN was calibrated against the vapor pressure of liquid He<sup>3</sup> in the range 1–2°K. No corrections were required. The calibration reproduced to 1:10<sup>3</sup> from run to run, and was frequently checked during a run against a calibrated resistance thermometer. In measuring the electrical conductivities, the thermometer contacts were utilized as the potential contacts to the sample.

The results on thermal conductivity obtained for Ag<sup>6</sup> and Pt<sup>7</sup> are shown in Fig. 1. The scatter is ±0.2% but the total systematic error may be as large as ±0.5%.<sup>8</sup> Both samples had been vacuum annealed, but were strained somewhat during mounting. Sharma's results on Ag<sup>4</sup> are also shown in Fig. 1 for comparison. The anomalous, oscillatory behavior cannot be found in the present data. Both our samples were remeasured, and the results (not shown in Fig. 1) agreed to within 1% with the first runs. The slight change may have been caused by a limited handling of the samples at room temperature. A second, unannealed sample of Ag<sup>9</sup> was also measured and showed no anomalies.

The electrical resistivity  $\rho$  for Ag was  $0.203 \times 10^{-8} \Omega \text{ cm}$ , and was independent of *T* below 4°K to within ±1%.<sup>10</sup> The Pt data could be fitted by the expression  $\rho = (1.687 + 0.0020T^2) \times 10^{-8} \Omega \text{ cm}$ , which, for the  $T^2$  term, is in reasonable agreement with the data White and Woods obtained above 2°K.<sup>11</sup> This temperature dependence in  $\rho$  is typical of the transition elements.<sup>11</sup>

The empirical Lorenz number  $L (= \rho K / T)$  for

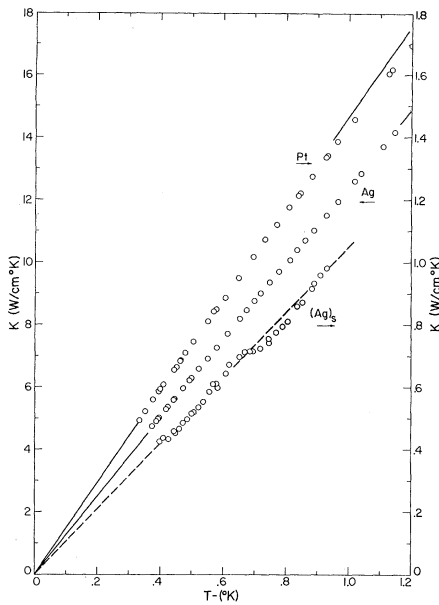


FIG. 1. The thermal conductivity  $K$  of Pt and Ag versus temperature. Data labeled Ag and Pt are from the present work. That labeled  $(Ag)_0$ , taken from Ref. 4 for comparison, has been multiplied by a constant factor of 0.78 for purposes of clarity. Ag is referred to the left ordinate, Pt and  $(Ag)_0$  to the right.

our sample of Ag is  $2.53 \times 10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$  as compared to the theoretical value of  $L_0 = 2.45 \times 10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$ . Willott found a similar effect in Al with data obtained above  $1.5^\circ\text{K}$ .<sup>12</sup> He suggested this was due to an additional thermal conduction contributed by transverse phonons, and that the empirical Lorenz number would tend to  $L_0$  at lower temperatures. If such a conduction mechanism is present in Ag, it has not started to decrease with decreasing temperature at  $0.35^\circ\text{K}$ .

As is obvious from Fig. 1, the data for Pt definitely deviate from a linear temperature dependence. Using only data below  $0.6^\circ\text{K}$ , which extrapolate rather well to the origin, gives an empirical Lorenz number of  $2.50 \times 10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$ , again greater than  $L_0$ . A logarithmic plot of the deviation from this straight line indicates a  $T^2$  dependence. Therefore, justifiably assuming the validity of Matthiessen's rule, we plot  $T/K$  vs  $T^2$  and obtain the relation  $T/K = 0.676 + 0.022T^2 \text{ cm } ^\circ\text{K}^2/\text{W}$ . The  $T^2$  temperature dependence in both  $\rho$  and  $T/K$  is indicative of electron-electron scattering, for which Herring has suggested that an effective Lorenz ratio of  $L' \approx 1.6 \times 10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$  should be appropriate.<sup>13</sup> Using the  $T^2$  terms in  $\rho$  and  $T/K$  from our data on Pt, however, gives  $L'$

$\approx 0.1 \times 10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$ . Other recent measurements on the transition metals at higher temperatures also indicate low values of  $L'$ , with 1.0 for Ni,<sup>14</sup> 0.7 for Pd,<sup>15</sup> and 0.5 for Re,<sup>15</sup> each in units of  $10^{-8} \text{ W } \Omega \text{ } ^\circ\text{K}^{-2}$ .<sup>16</sup>

In summary, the present experiment clearly shows that cusp- or oscillatory-shaped anomalies are not inherently present in the thermal conductivity of pure metals. It has given further evidence that the details of electron-electron scattering in the transition metals vary greatly from metal to metal, i.e., a simple Lorenz ratio is not applicable. It has demonstrated explicitly that, in a temperature range uncomplicated by phonon scattering, evidence for electron-electron scattering is present in both the thermal conductivity and the electrical conductivity for the transition metal Pt, while for the noble metal Ag any such effects are at least an order of magnitude smaller. It has not resolved the question concerning the high empirical Lorenz number in the limit of low temperatures. Here it should be noted, however, that conclusions based on this and other low-temperature experiments involving absolute magnitudes must be considered tentative pending further refinement of the international temperature scales which utilized the vapor pressure of liquid He<sup>3</sup> or He<sup>4</sup>.<sup>8</sup>

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<sup>1</sup>G. Davey and K. Mendelssohn, Phys. Letters **7**, 183 (1963).

<sup>2</sup>K. Mendelssohn, J. K. N. Sharma, and I. Yoshida, Bull. Inst. Intern. Froid, Annexe **45**, 49 (1965).

<sup>3</sup>G. Davey, K. Mendelssohn, and J. K. N. Sharma, in Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965), p. 1196.

<sup>4</sup>J. K. N. Sharma, Cryogen, **7**, 141 (1967). The results on Ag in this paper appear to be in error by a factor of 10.

<sup>5</sup>M. H. Jericho, Phil. Trans. Roy. Soc. London **A257**, 385 (1965). The lack of similar anomalies in Jericho's measurements on Cu would then be explained by the impure state of his commercial grade Cu, with  $R_{300}/R_0 \approx 85$ .

<sup>6</sup>The Ag, obtained from Cominco American, was 99.999% pure. It was of rectangular cross section with minimum dimension  $1.27 \times 10^{-2} \text{ cm}$ . The resistivity ratio when mounted in the cryostat was  $R_{300}/R_0$

=740, its  $A/l = 1.36 \times 10^{-4}$  cm.

<sup>7</sup>The nominal  $5.1 \times 10^{-2}$ -cm-diam Pt wire, obtained from Matthey Bishop, Inc., was 99.999% pure. The resistivity ratio when mounted in the cryostat was  $R_{300}/R_0 = 633$ , its  $A/l = 2.02 \times 10^{-4}$  cm.

<sup>8</sup>This does not include any deviation which may exist between the  $T_{62}$  (or  $T_{58}$ ) temperature scale and the thermodynamic scale. A deviation that ranged as abruptly as that reported by D. T. Grimsrud and J. H. Werntz, Jr., Phys. Rev. **157**, 181 (1967), would completely change the quantitative, though not the qualitative, conclusions of the present paper. In private conversations with Dr. H. Plumb and Dr. C. A. Swenson we learned that recent work above 2°K indicates a smoother deviation as a function of temperature. See H. Plumb and G. Cataland, Metrologia **1**, 127 (1966), and J. S. Rogers, R. J. Tainsh, M. S. Anderson, and C. A. Swenson, to be published.

<sup>9</sup>This Ag sample was taken from the same spool as that of Ref. 6 and used in the "as received" condition. The electrical properties were not measured; the thermal conductivity was  $5.4T$  W/cm °K<sup>2</sup>. We are indebted to Professor J. S. Koehler for providing the Ag samples.

<sup>10</sup>This large uncertainty resulted from the small resistance of the Ag sample plus the small measuring

current used in order to keep the resulting rise in temperature of the sample (situated in a vacuum) small. These measurements were made possible through the kind loan of a nanovolt potentiometer by Professor R. O. Simmons.

<sup>11</sup>G. K. White and S. B. Woods, Phil. Trans. Roy. Soc. London, Ser. A **251**, 273 (1959).

<sup>12</sup>W. B. Willott, Phil. Mag. **16**, 691 (1967).

<sup>13</sup>C. Herring, Phys. Rev. Letters **19**, 167, 684(E) (1967).

<sup>14</sup>G. K. White and R. J. Tainsh, Phys. Rev. Letters **19**, 165 (1967). The value of  $L'$  for Ni is in question because the metal is ferromagnetic; see F. C. Schwerer and J. Silcox, Phys. Rev. Letters **20**, 101 (1968).

<sup>15</sup>J. F. Schriempf, Phys. Rev. Letters **19**, 1131 (1967).

<sup>16</sup>A  $T^2$  term having a magnitude similar to that found in the electrical resistivity of the transition metals has been reported for single-crystal Ga above 1°K [M. Yaqub and J. F. Cochran, Phys. Rev. **137**, 1182 (1965)], but recent thermal-conductivity data [R. I. Boughton and M. Yaqub, Phys. Rev. Letters **20**, 108 (1968)] reflect only electron-phonon scattering with a magnitude in reasonable agreement with the earlier work of H. M. Rosenberg, Phil. Trans. Roy. Soc. London, Ser. A **247**, 411 (1955).

## INTRINSIC QUANTUM FLUCTUATIONS IN UNIFORM FILAMENTARY SUPERCONDUCTORS\*

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Small depressions of supercurrent and critical temperature that indicate intrinsic thermodynamic fluctuations have been revealed in thin superconducting tin whisker crystals.

Quantum fluctuations are supposed to manifest their existence in thin superconductors by depression of the critical supercurrent densities and the critical temperature, according to several recent investigations.<sup>1-6</sup> However, disparities among the various accounts of this effect illustrate the present uncertainty about the fundamental nature and even the existence of fluctuations in systems characterized by long-range coherence. This Letter reports some new experiments on the critical supercurrent depression due to intrinsic fluctuations that may help to clarify this general problem.

Langer and Ambegaokar<sup>1</sup> have developed an elegant theory of the effect of intrinsic fluctuations on the resistive transition in thin superconductive filaments. They found that a fluctuation of order parameter in a volume  $\sigma\xi(T)$ , where  $\sigma$  is the cross-sectional area and  $\xi(T)$  is the coherence length, has the minimum activation energy that permits phase slippage,

and they calculated the steady-state temperature and current-density dependence of the consequent resistance near the critical temperature. However, all of the experiments—in thin microbridges formed by vapor deposition of thin films of tin—have shown a substantially larger effect. Typically, depressions of the effective critical temperature  $\Delta T_C \gtrsim 10^{-2}$  °K have been observed instead of the predicted  $\Delta T_C \sim 10^{-3}$  °K in comparable cross sections  $\sim 10^{-9}$  cm<sup>2</sup>. Groff, Marčelja, Masker, and Parks<sup>6</sup> introduced a model based on phase fluctuations in a volume  $\sigma L$  extending the full length  $L$  of their microbridges to explain their experiments. Hunt and Mercereau<sup>5</sup> assumed a fluctuation volume  $\sigma l$ , where  $l$  is the electron mean free path, and included a noise temperature seven times the critical temperature to explain theirs.

Because the principal experimental difficulty in measurements of intrinsic fluctuations is elimination of overwhelming extraneous ef-