Thermal Conductivity of Normal and Superconducting Aluminum

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The thermal conductivity of three specimens of aluminum differing widely in residual resistivity was measured in the normal and superconducting states. The experimental method yielded quite accurate results of the ratio of the superconducting state thermal conductivity to that in the normal state, κ_s/κ_n. The ratios κ_s/κ_n, for specimens differing in electronic mean free path by as much as a factor of 140, were essentially the same over the range of temperature covered by the experiments, 0.32 to 1.20°K. Agreement of the experimental results with the theoretical predictions of Bardeen, Rickayzen, and Tewordt was quite good.

INTRODUCTION

In the range of temperature in which superconductivity occurs, the transport of heat in reasonably pure metals is predominantly by the conduction electrons. The predominant scattering mechanism, however, may be either elastic scattering by static imperfections such as impurity atoms, lattice defects, or specimen or grain boundaries, or it may be scattering from the thermally excited lattice vibrations. In many instances both scattering processes are important in limiting the flow of heat.

In order to study in detail this important transport phenomenon and its relation to superconductivity, one would choose systems in which only one of these scattering processes is important. Elastic scattering of the electrons by static imperfections is the major mechanism for limiting the conduction of heat by the electrons in superconductors for which the Debye temperature (Θ) is very high relative to the transition temperature. Furthermore, increasing the concentration of impurities or imperfections tends to increase this elastic scattering and cause it further to predominate.

For a study of the electronic thermal conductivity limited by elastic scattering, aluminum is perhaps the best suited superconductor. In the normal state, if we are to extrapolate from the measurements of Andrews *et al.*,[1] we find that for specimens of normal laboratory dimensions the electronic mean free path at the transition temperature calculated on the basis of thermal scattering alone would be many times the dimensions of the specimens. So that for even the purest aluminum the thermal scattering may be completely ignored over the whole superconducting range of temperature.

We therefore have a system in which to study the detailed temperature dependence of the thermal conductivity in the normal and superconducting states for comparison with theory and to study the ratio of superconducting to normal thermal conductivities at various purity levels (i.e., at various electronic mean free paths) maintaining the same mechanism over the range of purity.

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[1] F. A. Andrews, R. T. Webber, and D. A. Spohr, Phys. Rev. **84**, 994 (1951).

EXPERIMENTAL

Cryostat

In order to cover an appreciable fraction of the superconducting temperature range of aluminum, it was necessary to work at temperatures below 1°K. A liquid He^3 cryostat described briefly below served this purpose well. This cryostat was similar to that used by Biondi and Garfunkel[2]; in fact, some of the apparatus served both experiments.

The liquid He^3 reservoir and the thermal conductivity specimen with its heater and thermometers, as shown in Fig. 1, were suspended in a vacuum can which was immersed in liquid He^4. The vacuum can was supported by stainless steel tubes which communicated to the outside world. In addition to the cold part of the system which is shown, there were (a) a high-vacuum system for maintaining thermal isolation between the He^3 system and the He^4 bath; (b) a closed He^3 system consisting of a He^3 gas reservoir for storing the 4 liter-atmospheres of gas, a mechanical pump and a diffusion pump with appropriate valving; and (c) McLeod gauge and mercury monometer for measuring the vapor pressure of He^3.

For operation, the He^4 bath was maintained at approximately 1.2°K by lowering the pressure of the evaporating He^4 with a large vacuum pump. The He^3,

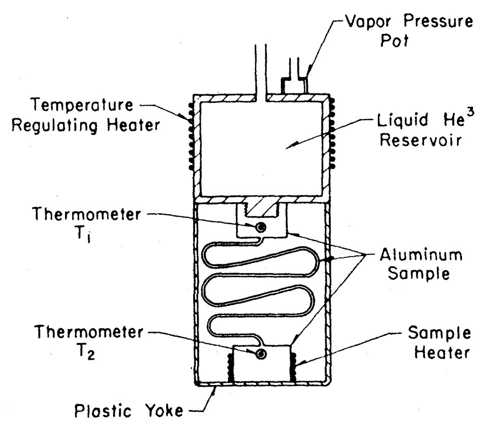


FIG. 1. Thermal conductivity apparatus.

[2] M. A. Biondi and M. P. Garfunkel, Phys. Rev. **116**, 853 (1959).

when admitted to the cold part of the system, liquified by contact with the wall of the tube passing through the liquid He^4 and ran down into the liquid He^3 reservoir. By evaporating and recondensing the He^3, the reservoir was cooled to the temperature of the He^4 bath without using exchange gas, thus avoiding extraneous heat transfer through residual gas. He^3 was similarly condensed into the vapor pressure bulb. Desired temperatures between 0.32° and 1.2°K were reached by adjusting the rate at which He^3 was pumped off by means of a throttling valve. The temperature was further regulated by varying the power supplied to a heater wound on the reservoir.

For measurements in the absence of a magnetic field the earth's (laboratory) field was nulled to within ± 0.02 oe in the vicinity of the sample by three mutually perpendicular Helmholz coils. For normal state measurements, fields up to 120 oe were generated by a battery-operated solenoid immersed in the liquid N_2 bath surrounding the He^4 bath.

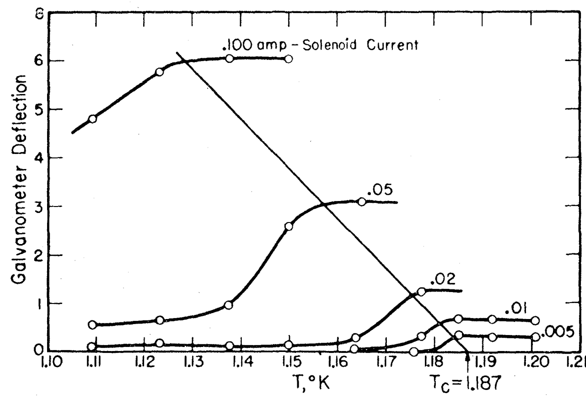


FIG. 2. Plot of ballistic galvanometer deflection as a function of temperature for various solenoid currents for extrapolating to the transition temperature at zero measuring field.

Specimens

Three specimens were used in the measurements of widely different impurity content. They are designated by their approximate residual resistivity ratio $R_{273}/R_{1.2}$. Thus, the most impure specimen, Al-26, with 0.3% copper added impurity had a residual resistivity ratio $R_{273}/R_{1.2}$ of 26. Al-430 was nominally pure aluminum from Alcoa and Al-3660 was zone refined aluminum obtained from Aluminum Industrie A.G., Switzerland, through their U. S. distributor, United Mineral and Chemical Corporation.

Al-430 and Al-3660 were prepared from $\frac{5}{8}$-in. bars 7 in. long which had been work-hardened for easy machining. The end sections, which carried thermometers and heater and made contact with the heat sink, were machined to cylinders $\frac{1}{2}$ in. in diameter and $\frac{3}{8}$ in. long. The center portion 15 cm long was milled to a ribbon $\frac{1}{2}$ mm thick and 2 mm wide. They were then folded into the shape shown in Fig. 1 on a template of pure aluminum and annealed in the final shape at 525°C for two hours. Specimen Al-26 was similar except that the center section $\frac{1}{2}$ mm$\times$2 mm in cross section was straight and 3.2 cm long.

Measurements

The working thermometers were $\frac{1}{4}$-watt carbon resistors. Speer 1200-ohm resistors were used over most of the temperature range, however, the sensitivity became so low near the transition temperature that measurements in the region from 0.75°K to 1.2°K were repeated using Allen Bradley 12-ohm resistors for Al-430 and Al-3660.

Electrical leads from the thermometers and the sample heater were of number 40 B and S gauge Manganin wire which was tin-coated so that it would be superconducting at the temperature of the experiments. The leads were thermally bonded to the He^3 reservoir and again to the vacuum can in contact with the He^4 bath where they were connected to copper leads which extended out of the cryostat.

The resistance of the thermometers and the power to the heater were measured potentiometrically using two Rubicon potentiometers and Leeds & Northrup dc breaker amplifiers. The off-balance signal from the potentiometer measuring thermometer T_1 was fed to an electronic temperature controller which maintained the temperature of T_1 constant to within 10^{-5} degree or better by regulating the power to the heater on the liquid He^3 reservoir.

By the method used for making the measurements, it was possible to determine quite accurately the ratio of the thermal conductivity in the superconducting state to that in the normal state κ_s/κ_n, in fact, much more accurately than either of the quantities individually. With T_1 controlled at a constant temperature, the resistance of T_2 was measured in the equilibrium state with no power supplied to the sample heater and again in a steady state with measured power P supplied. In absence of a magnetic field the superconducting thermal conductivity was determined by

$$\kappa_s=\alpha(P_s/\Delta R)(dR/dT),$$

where α depended upon the geometry of the sample, ΔR was the change in resistance of T_2 resulting from power P in the heater, and dR/dT was obtained from a calibration of the thermometer against the vapor pressure of He^3. The normal-state thermal conductivity κ_n was determined similarly in a magnetic field of approximately 100 oe duplicating the resistance of T_2 both without and with power to the heater. Thus the same ΔR was obtained as in the superconducting state but the power was adjusted to a new value, P_n, so that

$$\kappa_n=\alpha(P_n/\Delta R)(dR/dT).$$

Whereas the individual values of κ_s and κ_n depended upon the slope of the $R-T$ curve and the dimensions of the sample, the ratio κ_s/κ_n was just dependent upon the

ratio of powers P_s/P_n and thus errors inherent in the individual measurements of κ_s and κ_n were eliminated from the ratio κ_s/κ_n.

The temperature was determined at each point by measuring the vapor pressure of He^3 when the sample had reached equilibrium without power. These measurements provided not only the temperature to assign to each thermal conductivity point (corrected very slightly to the average temperature of the sample) but also a set of R vs T values from which a calibration curve could be drawn. Absolute temperatures were calculated from measured vapor pressures, after first applying a correction for thermomolecular pressure,[3] from the He^3 vapor pressure-temperature scale.[4]

TRANSITION TEMPERATURES

Since it was desired to make comparisons in terms of reduced temperatures T/T_c, the transition temperature of each specimen was determined. A pickup coil of fine copper, wound on the end of the specimen over the sample heater, was connected to a ballistic galvanometer. The deflection of the galvanometer was observed on reversing the solenoid field for a series of currents in the solenoid at each of a series of temperatures. (The field in the vicinity of the specimen was made null except for the solenoid field.) For several solenoid currents the deflection was plotted against temperature as illustrated in Fig. 2. The first departure from normal-state deflection was considered the temperature of transition for the field corresponding to the solenoid current. On this plot itself an extrapolation was made to zero current since deflection was proportional to current and the point so obtained on the zero-current axis was considered T_c.

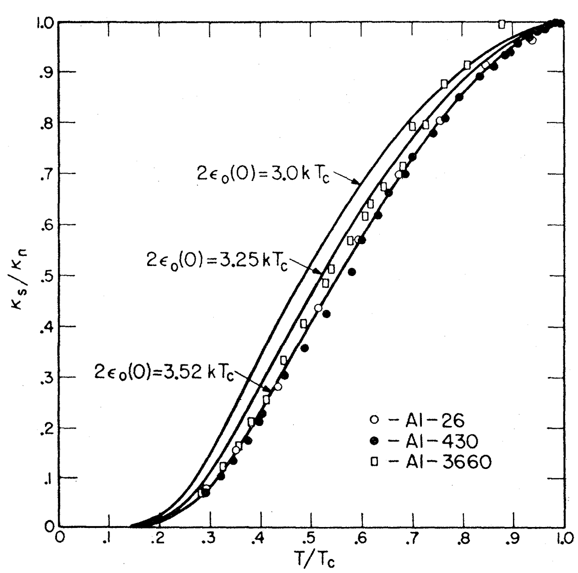
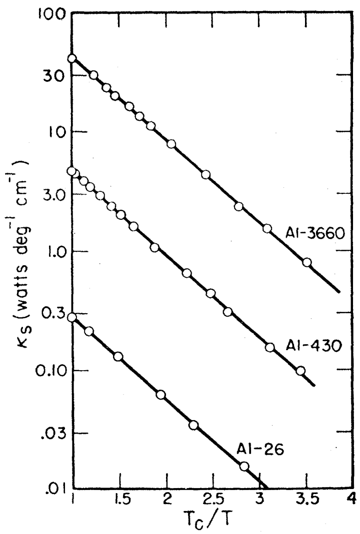


FIG. 3. Ratio of superconducting to normal thermal conductivity for aluminum.

[3] T. R. Roberts and S. G. Sydoriak, Phys. Rev. **102**, 304 (1956).
[4] S. G. Sydoriak and T. R. Roberts, Phys. Rev. **106**, 175 (1957).

FIG. 4. Thermal conductivity in the superconducting state for various aluminum specimens.



The values of T_c relative to mean free path found for the specimens of this study are in good agreement with those reported by Chanin *et al.*[5] Below are listed the mean free path and transition temperatures for the three specimens measured. The mean free path l for each of the specimens was determined from the normal-state thermal conductivity, assuming the Wiedemann-Franz law and a value for l/σ of 4×10^{-12} ohm cm^2.[6]

	l (cm)	$1/l$ (cm^{-1})	T_c (°K)
Al-26	4.0×10^{-5}	2.5×10^{4}	1.149
Al-430	6.7×10^{-4}	1.5×10^{3}	1.178
Al-3660	5.6×10^{-3}	1.8×10^{2}	1.187

THERMAL CONDUCTIVITY RESULTS

The thermal conductivity data are shown in Fig. 3 plotted as κ_s/κ_n vs T/T_c for all three specimens. For comparison, curves calculated from Bardeen, Rickayzen, and Tewordt (BRT)[7] are also shown. Curves for values of the energy gap at the absolute zero of $3.0kT_c$, $3.25kT_c$, and $3.52kT_c$ were calculated from Eq. (3.6) of BRT by inserting values for $2\epsilon_0(0)$ of $3.0kT_c$, $3.25kT_c$, and $3.52kT_c$, retaining the temperature variation of the energy gap $2\epsilon_0$ given by Bardeen, Cooper, and Schrieffer (BCS).[8]

The uncertainties in κ_s/κ_n for specimens Al-26 and Al-430 did not exceed $\pm1\%$; however, for Al-3660 precision was not as high because of the very high thermal conductivity of the specimen necessitating smaller temperature differences. Furthermore, at the time the experiments were carried out on Al-3660, some electrical interference had appeared that could not be completely

[5] G. Chanin, E. A. Lynton, and B. Serin, Phys. Rev. **114**, 719 (1959).
[6] T. E. Faber and A. B. Pippard, Proc. Roy. Soc. (London) **A231**, 336 (1955).
[7] J. Bardeen, G. Rickayzen, and L. Tewordt, Phys. Rev. **113**, 982 (1959).
[8] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

eliminated by shielding so that the accuracy of the measurements in this case was somewhat less than for other specimens.

The normal-state data for each of the specimens approximated a linear temperature dependence; however, there was considerable scatter presumably due to uncertainties in the slope of the R vs T curve, dR/dT, for the carbon thermometers. The best linear expression fitted to the data for each specimen gave the following formulas for κ_n:

Specimen	
Al-3660	$\kappa_n=34.08T$ watt cm^{-1} deg^{-1}
Al-430	$\kappa_n=4.00T$ watt cm^{-1} deg^{-1}
Al-26	$\kappa_n=0.242T$ watt cm^{-1} deg^{-1}.

Since we would expect κ_n to follow a linear temperature dependence quite accurately in this range of temperature, the above expressions should represent the normal-state conductivity with the errors due to uncertainties in dR/dT pretty well averaged out. The superconducting-state thermal conductivity may now be determined from $\kappa_s=\kappa_n P_s/P_n$ and again errors due to uncertainties in dR/dT of the thermometer are greatly reduced.

In Fig. 4 values for κ_s are plotted as $\log\kappa_s$ against $1/T$ and, as noted by Zavaritskii[9] for aluminum and some other superconductors, an expression of the form $\kappa_s \propto e^{-bT_c/T}$ is very closely approximated for all specimens.

CONCLUSIONS

Regarding the data shown in Fig. 3, one can make the following observations:

(a) The experimental data and the theoretical prediction of BRT for the elastic scattering case are in essentially complete agreement.

(b) The BCS value for the energy gap, $2\epsilon_0(0)=3.52kT_c$, appears to fit the data as well as any other choice. The thermal conductivity is not a sensitive enough function of the energy gap nor are the present data of high enough accuracy, particularly considering the slight difference from specimen to specimen, to claim disagreement with other measurements of the energy gap which range from $3.25kT_c$ to $3.37kT_c$.[2,10–12]

(c) Variation of the mean free path by as much as a factor of 140 does not appreciably change the κ_s/κ_n ratio as a function of reduced temperature. It should be remembered that the transition temperature T_c varies slightly with mean free path so that the same κ_s/κ_n ratio for specimens of different mean free path would indicate slightly different energy gaps. The difference is consistent with Anderson's theory of dirty superconductors.[13] The slight variation of κ_s/κ_n from specimen to specimen is not systematic with mean free path and probably should not be taken very seriously.

The experimental data of the present study are in agreement with measurements of Zavaritskii[9] although the precision of the latter was considerably less. The present results are in qualitative agreement with the type of expression he proposes, $\kappa_{es}=Ae^{-bT_c/T}$, as shown in Fig. 4; however, failure to agree in detail is masked by the logarithmic scale. In particular, an expression of the above type would predict a finite slope to the κ_s/κ_n vs T curve near T_c instead of the zero slope observed and predicted by BRT. Furthermore, it is highly doubtful that such an expression could be justified on the basis of present theory.

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[9] N. V. Zavaritskii, J. Exptl. Theoret. Phys. USSR 34, 1116 (1958).

[10] Piotr B. Miller, Phys. Rev. **118**, 928 (1960).

[11] A. G. Redfield, Phys. Rev. Letters **3**, 85 (1959).

[12] Norman E. Phillips, Phys. Rev. **114**, 676 (1959). One can deduce from the fit to the expression $C_{es}=Ae^{-bT_c/T}$, with $b=1.34$ in the range $2.0<T_c/T<4.5$, that the energy gap at the absolute zero should be about $3.3kT_c$.

[13] P. W. Anderson, J. Phys. Chem. Solids **11**, 26 (1959).