## Anomalous Heat Flow in Superconductors

K. MENDELSSOHN AND J. L. OLSEN Clarendon Laboratory, Oxford University, England (Received July 24, 1950)

Measurements of heat flow in the superconductive and in the normal state have been carried out on pure lead and lead containing a small quantity of bismuth. The heat conduction has been measured in its dependence on magnetic field and temperature. The alloy shows a smaller heat conduction in the intermediate state than in either the superconductive or the normal state. This anomalous behavior is taken to indicate the existence in superconductors of a type of heat transport similar to that observed in liquid HeII.

## I. INTRODUCTION

R ECENT measurements on the heat transport in superconductors<sup>1</sup> have revealed the existence of two competing processes responsible for the variation of heat flow with temperature. One is the progressive passage of electrons into the superconductive state as the temperature is lowered. Since the "superconductive electrons" have zero entropy<sup>2</sup> they cannot take part in the ordinary heat conduction process and the heat flow through the metal is consequently decreased. This mechanism, in which at any given temperature below the transition point the heat conductivity of the superconductive metal is smaller than in the normal state (which can be re-established by a magnetic field), is dominant in pure metals.<sup>1,3</sup> In certain alloys on the other hand the superconductive state conducts heat better than does the normal one. Intermediate cases in which the curves for the normal and the superconductive heat flow intersect at some temperature below the transition point have been observed in columbium<sup>1</sup> and in pure lead to which controlled quantities of bismuth had been added.<sup>4</sup> Thus a second type of heat transport which opposes the reduction in heat flow mentioned above clearly exists, but its cause is not so certain. We have tentatively put forward the explanation<sup>1,5</sup> of a circulation current of electrons within the metal which somewhat resembles the heat transport in liquid HeII. One can think of a flow of superconductive electrons towards the warm end of the specimen which is compensated electrically by the return flow of normal electrons. Since in the pure superconductive metal the magnetic induction must be zero, this return current is prohibited. In a superconductive alloy, on the other hand, where as is well known the induction is different from zero "normal" return currents are possible. Their formation will even be facilitated because there are indications that the electrical conductivity of the normal enclosures in the metal is exceptionally high. An alternative explanation for the high heat conductivity in certain superconductors has been suggested by  $Hulm^6$  who considers that the lattice conduction may be increased when the metal becomes superconducting. Recent experiments on controlled impurities<sup>4</sup> however show that such a mechanism, though it may well exist, would be inadequate to account for the observed heat flows. In most researches the heat conductivity has been measured as it depends on the temperature but further information can be gained when these observations are supplemented by measurements of the heat conduction in its dependence on the magnetic field.7 Such measurements have now been carried out on pure lead and lead with a small quantity of bismuth.

## II. METHOD

The specimens used were cylindrical rods along which a temperature gradient of  $\sim 0.1^{\circ}$  per cm or less was established. At two places along the specimen, about 2 cm apart, gas thermometers were attached without the use of solder and the temperature measured differentially. The lead specimen was prepared from Johnson Matthey H.S. lead (laboratory number 1932) with an impurity content of less than 0.002 percent. The alloy specimen was prepared from the same metal to which a small quantity ( $\sim 0.02$  percent) of very pure bismuth from the same source had been added. A homogeneous magnetic field could be established in the direction of the specimen or normal to it. Heat was supplied electrically to one end of the specimen while the other end was firmly attached to the helium container of an expansion liquefier. By carefully regulating the escape of helium gas this apparatus could be used as a thermostat between the temperatures of liquid hydrogen and liquid helium.<sup>8</sup> Below the boiling point of helium the device was thermostated through the vapor pressure of the liquid.

## III. RESULTS

Figure 1 gives the change of the heat conduction of the pure lead specimen with a longitudinal magnetic field. The mean temperatures for the two experiments were 4.6°K and 2.7°K. The values of magnetic field at

<sup>&</sup>lt;sup>1</sup> K. Mendelssohn and J. L. Olsen, Proc. Phys. Soc. London A

<sup>63, 2 (1950).</sup> <sup>2</sup> J. G. Daunt and K. Mendelssohn, Proc. Roy. Soc. A 185, 225

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<sup>3</sup> W. J. de Haas and A. Rademakers, Physica 7, 992 (1940).
<sup>4</sup> K. Mendelssohn and J. L. Olsen, Proc. Phys. Soc. A63, 1182 (1950)

<sup>&</sup>lt;sup>5</sup> K. Mendelssohn, Royal Coll. Sci. J. 16, 105 (1946).

<sup>&</sup>lt;sup>6</sup> J. K. Hulm, M.I.T. Conf. Very Low Temp., (1949), p. 90. <sup>7</sup> K. Mendelssohn and R. B. Pontius, Phil. Mag. 24, 777 (1937). <sup>8</sup> D. K. C. MacDonald and K. Mendelssohn, Proc. Roy. Soc. A 202, 103 (1950).

which the change occurs are in fair agreement with the threshold values Hc for the electrical resistance.<sup>9</sup> As this value is reached and the specimen changes from the superconductive into the normal state the heat conductivity rises suddenly; a similar change in the reverse direction is observed on reducing the field. There is a very small amount of hysteresis between the two curves; this is slightly more pronounced at the lower temperature. However, as on the return curve the field is again reduced to zero the value of the heat conductivity is sensibly the same as before superconductivity was destroyed. Observations in transverse magnetic fields yield similar results except that the transition was, of course, gradual between  $\frac{1}{2}Hc$  and Hc.

The addition of a very small quantity of bismuth ( $\sim 0.02$  percent) affects the absolute value of the heat conduction but the character of the transition at a relatively high temperature is hardly influenced. Figure 2 gives the longitudinal and the transverse transitions of this specimen a little above 5°K. The change in heat conduction again agrees well with that of electrical resistance and the hysteresis is still small. After superconductivity was destroyed and the field reduced to zero the conductivity is now actually slightly higher than originally.

The situation changes completely, however, as the temperature is lowered. Figure 3 shows the longitudinal and transverse transition curves for the same specimen at  $\sim 2.9^{\circ}$ K. As the field is increased and superconductivity is destroyed there is again a sharp increase in the longitudinal case; however, on decreasing the field the



FIG. 1. Heat conductivity of pure lead; variation with longitudinal magnetic field. Full line: field increasing; broken line: field decreasing.





FIG. 2. Heat conductivity of Pb Bi 0.02 percent; variation with magnetic field. Full line: field increasing; broken line: field decreasing.

heat conductivity now falls well below the original value and at zero field the heat flow through the specimen in the superconductive state is 22 percent lower than at first. An even more striking behavior is exhibited on the destruction of superconductivity by a transverse field. The heat conduction remains constant until a field of about  $\frac{1}{2}Hc$  is reached and then begins to fall to a minimum value well below the heat flow in either the superconductive or the normal state. On further increasing the field the heat conductivity rises again until the normal value is reached at Hc. On returning the specimen into the superconductive state by reducing the field a monotonic fall of the heat conduction similar to that of the longitudinal case takes place.

Further information on this effect is provided by the observations of the heat conduction of this specimen in dependence on temperature given in Fig. 4. It is clear that the curve for the normal state is the continuous one. The curve for the superconductive specimen departs at a large angle and is in this respect rather similar to that of pure lead. Two values are given for the heat conduction in the superconductive state, one before and one after superconductivity had been temporarily destroyed. In the latter case some of the magnetic flux was evidently "frozen in." As can be seen, the values with and without the frozen in field coincide at the higher temperatures but below  $\sim 4^{\circ}$ K the heat flow with frozen in field is consistently smaller, decreasing to about two-thirds of the value of that without frozen in field at the lowest temperature.

#### **IV. CONCLUSIONS**

It is known from our other experiments<sup>1,4</sup> that as the bismuth content is increased the heat conduction in the



FIG. 3. Heat conductivity of Pb Bi 0.02 percent; variation with magnetic field at  $\sim 2.9^{\circ}$ K. Full line: field increasing; broken line: field decreasing.

superconductive state grows steadily in comparison with the normal heat conductivity until eventually their relation is completely reversed. It is therefore significant that the superconductive curve without frozen in field shown in Fig. 4 indicates higher heat flows than the curve with frozen in flux. Moreover, the shape of the curve definitely gives the impression that the frozen in values represent a true continuation of the original curve from which the values without frozen in flux branch off. We are clearly encountering here the first sign of the new heat transmission process in the superconductive state and the conclusion is reached that this transmission of heat is inhibited by the magnetic flux frozen into the specimen. The interesting point is that when the alloy specimen at 2.9°K is partly in the superconductive and partly in the normal state, the heat conduction is *smaller* than in either of these states. This shows that the observed heat flow cannot be simply due to a mixture of superconductive and normal materials each having its specific heat conductivity. At the higher temperature and in pure lead on the other hand this is evidently the case.

While thus any explanation simply based on different heat conductions in the two states must fail to account for our results, they seem to offer an unexpected confirmation of our model. We assume that as with falling temperature the alloy character of the specimen increases,<sup>9</sup> and with it the deviation from zero induction grows, a circulation of electrons along the specimen will be established. The superconductive electrons take up their *total* entropy which is returned by the flow in the opposite of normal electrons. For this reason the circu-

lation flow is a powerful means of heat transport and as it develops can outstrip the negative effect of electrons being lost into the superconducting state. The present results seem to show that at higher temperatures or in a pure metal no anomalous effects occur because no circulation flow exists. Circulation flow can take place in the alloy specimen at low temperatures but it will be broken up by the formation of normal enclosures in the specimen. In this respect the transverse transition in Fig. 3 is of particular significance. When at  $\frac{1}{2}Hc$  the threshold value on the surface of the cylindrical specimen is exceeded the rod passes into the intermediate state. At the establishment of this state normal laminae perpendicular to the axis of the specimen are formed<sup>10</sup> which gradually increase at the expense of the superconductive material as the field is increased. Although therefore at the beginning of the transition the actual amount of normal material within the specimen is small the circulation process will be inhibited very soon and the total heat flow will fall instead of rising. Only when enough normal material is present will the original value of the conductivity be reached again and the actual value for the normal state will ultimately be attained. No such intermediate decrease is observed in the longitudinal destruction where no transverse laminae are formed. On the other hand, as the field is decreased to zero and the frozen in lines of force curl up like those in a permanent bar magnet the circulation flow is evidently again inhibited.

It would thus appear that the heat flow given by the experiments with the frozen in field is nearer to the true value of the heat conduction in the superconductive material than that before magnetic penetration. This fact must be borne in mind when analyzing the heat conduction of a superconductor. Apart from its theoretical interest the anomalous heat conduction has some bearing on the practice of cryogenic technique. It has



FIG. 4. Heat conductivity of Pb Bi 0.02 percent; variation with temperature. Small circles: zero field; crosses: field >Hc; large circles: field frozen in.

<sup>10</sup> It can easily be shown that laminae in any other direction to the cylinder axis are unstable.

been suggested by us<sup>1</sup> (and independently by Daunt and Heer<sup>11</sup>) to make use of the change of heat conduction in superconductors for make and break thermal contacts at very low temperatures. The present results

<sup>11</sup> C. V. Heer and J. G. Daunt, Phys. Rev. 76, 854 (1949).

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# The Disintegration of Praseodymium 142

Erling N. Jensen, L. Jackson Laslett, and D. J. Zaffarano Institute for Atomic Research and Department of Physics, Iowa State College, Ames, Iowa\* (Received July 26, 1950)

The radiations from Pr<sup>142</sup> have been examined with a thin lens spectrometer. One gamma-ray was found having an energy of 1.576 Mev. The beta-spectrum is complex, the two groups observed having maximum kinetic energies of  $2.15_4$  Mev and  $0.63_6$  Mev. It is suggested that the high energy beta-group gives a transition to the ground state of Nd<sup>142</sup> and that the gamma-ray follows the low energy beta-transition. The half-life was found to be 19.1 hr. The electron distribution produced by bremsstrahlung, due to the absorption of the beta-rays, was observed.

## I. INTRODUCTION

HE characteristic radiations of Pr<sup>142</sup> were investigated by Amaldi<sup>1</sup> and others, but in more detail recently by absorption and coincidence methods by Bothe,<sup>2</sup> Mandeville,<sup>3</sup> and Jurney.<sup>4</sup> Data obtained with spectrometers have been reported by DeWire et al.,5 Peacock et al.,6 Cork et al.,7 and Rae.8

The radiations found by these authors are shown in Table I.

The coincidence work of Mandeville and Jurney has established that the 1.6-Mev gamma-ray probably follows the soft beta-transition, while the higher energy beta results from a disintegration to the ground state of Nd142. Delayed coincidence work by DeBenedetti and and McGowan<sup>9</sup> showed that there are no metastable states in  $Nd^{142}$  with half-lives in the range  $10^{-6}$  to  $10^{-3}$ sec.

DeWire et al.<sup>5</sup> remark that the higher energy beta-ray arises from a first-forbidden transition, according to Konopinski's notation,<sup>10</sup> and a characteristic "forbidden shape" is suggested by their Kurie plot. The fact that Nd<sup>142</sup> is a "magic number" nucleus in neutrons, and the

<sup>1</sup> Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, and Segrè, Proc. Roy. Soc. **149A**, 522 (1935). <sup>2</sup> W. Bothe, Zeits, f. Naturforsch. **1a**, 173 (1946).

somewhat conflicting evidence for the decay scheme of Pr<sup>142</sup> led the authors to the present study.

show that in order to get a great difference of heat flow

in the two states very pure metals should be used. On

the other hand, it now seems to be possible to design

thermal switches which break a contact on magnetiza-

tion by employing an alloy.

Two 100-mg samples of spectrographically pure (contaminants less than 0.1 percent) Pr<sub>6</sub>O<sub>11</sub> (empirical formula) made available through the courtesy of Dr. F. H. Spedding and Mr. T. A. Butler of this laboratory were bombarded in the Oak Ridge pile and then examined with a thin-lens spectrometer<sup>11</sup> modified to incorporate ring focusing.<sup>12</sup>

## II. HALF-LIFE

The half-life of Pr<sup>142</sup> was determined by following the activity of a sample of PrCl<sub>3</sub> with a Lauritsen electroscope for more than four half-lives. A value of 19.1 hr. was obtained. This is in good agreement with the values reported by Bothe<sup>2</sup> (19.2 hr.) and DeWire, Pool, and Kurbatov<sup>5</sup> (19.3 hr.).

## **III. SECONDARY ELECTRON SPECTRUM**

The spectrometer was calibrated by means of the Fconversion line of ThB (1385  $H\rho$ ). All the data reported in this paper were obtained with a spectrometer resolution of 2.1 percent (half-width) and a Geiger counter window of Formvar with a cut-off at 15 kev. The data have not been corrected for window absorption.

Since praseodymium has a single isotope,<sup>13</sup> namely Pr<sup>141</sup>, it is assumed that only the radiations of Pr<sup>142</sup> were observed in this investigation.

To obtain the secondary electron spectrum, the Pr<sup>142</sup> was placed in a Lucite holder and covered with a copper cap, of surface density  $1.4 \text{ g/cm}^2$ , in order to absorb the

<sup>\*</sup> Contribution No. 112 from the Institute for Atomic Research and Department of Physics, Iowa State College, Ames, Iowa. Work was performed in the Ames Laboratory of the AEC

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<sup>8</sup> E. R. Rae, Proc. Phys. Soc. (London), 63A, 292 (1950).
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<sup>&</sup>lt;sup>13</sup> Inghram, Hess, and Hayden, Phys. Rev. 74, 98 (1948).