only when R_{1s} has no node), after applying this, we obtain

$$G(r)=r^2R_{1s}^2F=\int_\infty^r \frac{R_{1s}^2}{4\pi}\delta(r)dr-\langle\psi_{1s}|\delta(\mathbf{r})|\psi_{1s}\rangle \times\int_\infty^r r^2R_{1s}^2dr+G(\infty). \quad \text{(A8)}$$

Since we have boundary conditions that the wave function and its derivative go to zero at infinity, we then have $G(\infty)=0$. Also from Eq. (A8) we see that $G(0)=G(\infty)=0$. Equation (A8) therefore becomes

$$r^2R_{1s}^2F=-\langle\psi_{1s}|\delta(\mathbf{r})|\psi_{1s}\rangle\int_\infty^r r^2R_{1s}^2dr. \quad \text{(A9)}$$

Using Eq. (A2), we obtain

$$F(r)=-\frac{\langle\psi_{1s}|\delta(\mathbf{r})|\psi_{1s}\rangle}{P_{1s}^2(r)}\int_\infty^r P_{1s}^2(r)dr \quad \text{(A10)}$$

and

$$\delta\rho(r)=\int_\infty^r F(r)dr+\delta\rho(\infty), \quad \text{(A11)}$$

where $\delta\rho(\infty)$ is determined from the condition

$$\langle\delta\phi_{1s,N}|\psi_{1s}\rangle=0.$$

Doing so, we found

$$\delta\rho(\infty)=\int_\infty^0 P_{1s}^2(r)dr\int_\infty^r F(r)dr. \quad \text{(A12)}$$

Making use of Eqs. (A11) and (A12), Eq. (A4) becomes

$$\delta P_{1s,N}=P_{1s}\left\{\int_\infty^r F(r)dr +\int_\infty^0 P_{1s}^2(r)dr\int_\infty^r F(r)dr\right\}. \quad \text{(A13)}$$

From Eq. (A3), the expression for $\delta\phi_{1s,N}$ is then

$$\delta\phi_{1s,N}=\frac{1}{\sqrt{(4\pi)}}\frac{P_{1s}}{r}\left\{\int_\infty^r F(r)dr +\int_\infty^0 P_{1s}^2(r)dr\int_\infty^r F(r)dr\right\}, \quad \text{(A14)}$$

where $F(r)$ is given as in Eq. (A10).

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Thermal Resistivity at Pb-Cu and Sn-Cu Interfaces between 1.3 and 2.1°K*

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Measurements have been made of the thermal resistivity at lead-copper and tin-copper interfaces for $1.3\leq T\leq 2.1$°K. By the application of a magnetic field the lead or the tin could be transformed from the superconducting to the normal state. This permitted measurements of the changes in the thermal resistivity at the interface between two metals produced by allowing one of the metals to become superconducting. The thermal resistivity at an interface formed by vacuum casting lead onto copper was found to be $9.3\ T^{-3.7}$ °K cm²/W with the lead superconducting. An interface formed by growing a single crystal of lead onto a single crystal of copper had an interface resistivity of $9.3\ T^{-4.3}$ °K cm²/W with the lead superconducting. Upon application of a magnetic field to make the lead normal, the thermal resistivity at the interface dropped to a value too small to measure reliably, less than 0.04°K cm²/W. These measurements suggest that the heat transfer across Pb-Cu interfaces by electrons is increased considerably by changing the lead from the superconducting to the normal state. The thermal resistivity at the interface between tin and copper, with the tin superconducting, was about $\frac{1}{5}$ as great as that for lead and copper, with the lead superconducting. An alloy zone formed at tin-copper interfaces obscures to some extent the nature of the resistivity at these interfaces. However, heat transfer at Sn-Cu interfaces appears to be dominated by electrons with the tin either superconducting or normal. Heat transfer by electrons at all interfaces behaves qualitatively in a manner similar to the thermal conduction by electrons in the superconductor used. A rod made of alternate layers of Pb and Cu had a much larger thermal resistivity with the lead superconducting than with it normal. This suggests the use of such a sandwich structure as a thermal switch.

INTRODUCTION

The thermal resistivity ρ at the interface between two different media is defined as the temperature discontinuity ΔT assumed to exist at the interface, divided by the rate at which heat is transferred across the interface per unit area. This paper presents results of an experimental investigation for $1.3\leq T\leq 2.1$°K of the thermal resistivities at interfaces between lead and copper, and between tin and copper. Since application of a magnetic field could transform the lead or tin from the superconducting to the normal state, it was possible to obtain information concerning the con-

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tribution of electrons to the transfer of heat across these interfaces. A preliminary account of some of this work has appeared elsewhere.[1]

The thermal resistivity at the interface between liquid He^4 and a solid[2–4] and liquid He^3 and a solid[5] is generally referred to as the Kapitza resistivity. This is usually assumed to be due to the imperfect transmission of lattice waves or phonons across the interface as a result of the acoustic mismatch. The magnitude of the Kapitza resistivity is measured to be as small as 1/50 of that calculated on the basis of the acoustic mismatch.[6,7] Surface conditions such as oxide layers, adsorbed gases, and surface irregularities affect the thermal resistivity at the interface by an amount which is difficult to determine theoretically or experimentally.[2,4]

The acoustic-mismatch theory[7] has been more successful in accounting for measurements of the thermal resistivity at the interface between two solids. Imperfections of the interface such as irregularities, impurities, alloy zones, and defects due to differential thermal contraction exist but affect the thermal resistivity less for solid-solid interfaces than for solid–liquid He interfaces. Much of the information available on solid-solid interface resistivity has come from studies of practical problems. Berman[8] studied the thermal resistivity at the interface formed by pressing two metals together. Miedema *et al.*[9] and Anderson *et al.*[10] measured the thermal resistivity at the interface formed by pressing a powdered paramagnetic salt around pieces of copper and found it to vary as T^{-3}, as predicted by Little,[7] but the magnitude of this resistivity was about 6 times as large as expected. Recently, Neeper and Dillinger[11] measured the thermal resistivity at the interface between indium and sapphire and found both its temperature dependence and magnitude to be within the predictions of Little's theory.

In the case of the Kapitza resistivity and the resistivity at the interface between a metal and a dielectric such as indium and sapphire, lattice conduction is the only direct means of heat transfer across the interface. At a metal-metal interface free of imperfections, one would expect the conduction of heat by electrons to exceed that by phonons, as in the case of ordinary thermal conduction in metals at low temperatures. Conduction of heat by electrons will essentially "short out" the lattice resistivity.

A situation intermediate to the metal-dielectric and metal-metal cases is represented by a superconducting metal–normal metal interface. When a metal is in the superconducting state, the number of electrons or quasiparticles available for participation in thermal conduction is reduced from that in the normal state.[12] At temperatures considerably lower than the superconducting transition temperature, the density of excited quasiparticles is reduced to a point such that thermal conduction by electrons is negligible compared with lattice conduction.[13] Since only the excited quasiparticles participate in the process of thermal diffusion in a superconductor, it is reasonable to assume that only these particles can transfer heat across the superconducting-normal interface, in addition to heat transfer by the lattice. The bound Cooper pairs at the Fermi surface cannot participate in the thermal diffusion process. Thus, the transfer of heat by electrons across the interface will behave in a manner similar to the transfer of heat through the body of the superconductor. Therefore, at a sufficiently low temperature the superconducting-normal metal interface will behave as a dielectric-metal interface with heat being transferred across the interface only by the lattice. The observed thermal resistivity at the interface would be that due to the acoustic mismatch between the two metals. Upon applying a magnetic field to transform the superconductor to a normal conductor, the resistivity at the interface should decrease as the transfer of heat by electrons increases, as was pointed out by Little.[14] The acoustic mismatch at the metal-metal interface is affected by a negligible amount by the superconducting-to-normal transition of one of the metals.

Measurements of the thermal resistivity at superconducting-normal metal interfaces to be reported here are in accord with the behavior as described above. Lead and tin were used because of their different thermal conduction properties in the temperature range to be used. This difference is mainly due to the difference in their energy gaps 2Δ. For lead, $2\Delta_0 \approx 4k_BT_C$, in which $T_C = 7.2°K$. For tin, $2\Delta_0 \approx 3.5k_BT_C$, in which $T_C = 3.7°K$.

In the range of temperatures used, thermal conduction by the lattice is the dominant process in superconducting lead,[15] whereas in tin thermal conduction by electrons still dominates.[16] Data to be presented show that for a Pb-Cu interface, heat transfer by electrons is reduced by a large amount when the lead becomes superconducting. However, there is a much

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smaller change in the thermal resistivity at the interface between Sn and Cu when the Sn is transformed from the superconducting to the normal state, indicating much less change in the contribution to the transfer of heat by the electrons. Studies of a composite rod made of alternate layers of Pb and Cu showed that a large decrease in thermal resistivity was produced by applying a magnetic field to transform the lead from superconducting to normal. This decrease could be accounted for by changes in the resistivity at the Pb-Cu interfaces in series rather than by changes in the bulk lead present.

APPARATUS AND PROCEDURE

The metal-metal interfaces studied were formed by vacuum casting cylinders of Sn or Pb onto the ends of Cu cylinders. One sample was prepared by growing a single crystal of lead onto a single crystal of copper. The rods formed in this manner were approximately 3 in. long and $\frac{1}{4}$ in. in diameter, with the two metal segments being of about equal lengths.

A heat current was initiated at one end of the sample rod by an electrical heater, and the other end was placed in contact with a liquid He^4 bath which served as a heat sink. Four carbon resistor thermometers (47Ω, $\frac{1}{10}$-W Allen-Bradley carbon resistors) were located on the rod with two on each metal segment. Two of the thermometers were placed as close as possible (within 1 mm) to the interface. Thus, the temperature profile along each segment of the rod could be determined during the passage of a heat current. Extrapolation of these profiles to the interface determined the temperature discontinuity (if any) at the interface from which the thermal resistivity ρ of the interface could be determined.

The sample rod was mounted inside an evacuated capsule which was submerged in a liquid He bath. The top end of the rod was soldered into a stainless steel tube which was open to the bath. This permitted measurements of the Kapitza resistivity at the interface between one of the metals and liquid He^4 in addition to the metal-metal interface resistance.

Measurements from 1.3 to 2.1°K were taken with the Pb or Sn superconducting. A longitudinal magnetic field, sufficient to transform the superconducting metal to the normal state, was then applied and measurements over this temperature range were repeated.

The pertinent physical details of the experimental apparatus have been given previously.[1] Details concerning the mounting of thermometers on the specimens, thermometer calibration, and techniques of measurement are also presented elsewhere.[11]

Absolute temperatures measured with the heat current present were not used to determine the temperature profile along the rod. Instead, the changes in temperatures of the thermometers observed when the heat current was initiated were used. This method of determining the temperature profile eliminated some of the errors involved in determining absolute temperatures.

The total temperature drop along the sample during passage of a heat current was kept below 0.1°K. In some samples the determination of the temperature discontinuity ΔT at the interface had to take into account the nonlinearity of the temperature profiles resulting from a slight change in thermal conductivity with temperature gradient. This correction was significant only when ΔT was small (≈ 0.005°K) and the thermal conductivity of the superconducting segment was small. This correction was usually only 1 to 2% and amounted to 10% in the worst case.

The combined effect of random experimental errors on the values of interface resistivity determined is as follows. For the Pb-Cu samples it is about ±5% corresponding to $\Delta T \approx 0.015$ to 0.020°K. For the Sn-Cu samples it is about ±20% with $\Delta T \approx 0.005$°K. Estimated errors are shown on the curves by error bars. The systematic error introduced by inexact knowledge of the locations of the thermometers and the area of the interface could be as large as ±6%.

RESULTS

Lead-Copper Interfaces

Two different Pb-Cu samples were investigated and will be referred to as Pb-Cu 1 and Pb-Cu 2. Pb-Cu 1 was prepared by vacuum casting a lead rod onto the end of a copper rod in a reactor-grade graphite mold at a pressure of 10^{-6} mm Hg. Both metals were American Smelting and Refining Company 99.999% pure material. Before casting, the end of the copper rod was repeatedly polished and etched in order to obtain as smooth and unstrained a surface as possible. This sample was prepared by Allis-Chalmers Research Laboratories at Milwaukee, Wisconsin. Pb-Cu 2 consisted of a single-crystal rod of lead grown onto the end of a single-crystal rod of copper and was prepared by Semi-Elements Inc. at Saxonburg, Pennsylvania. Neither sample was annealed, since this would have damaged the interface by diffusion.

Typical measurements of the thermal resistivity ρ at the interface between lead and copper are plotted in Fig. 1. It is seen that ρ is much greater when the Pb is superconducting than when it is normal. ρ was less than 0.04 (°K cm^2 W^{-1}) for both samples with the Pb normal. For Pb-Cu 1 with the Pb superconducting $\rho = 9.3T^{-3.7}$ (°K cm^2 W^{-1}) and $\rho = 9.3T^{-4.3}$ for Pb-Cu 2. Data for Pb-Cu 1 show that values of ρ were the same for heat flowing across the interface from Pb to Cu as for flow from Cu to Pb. To reverse this flow the heater was attached to the opposite end of the specimen, a different end was soldered into the stainless steel tube, and a different set of thermometers was mounted on the specimen. The reversed heat flow (Cu to Pb) data were taken about two months after the other data. Agreement among these data shows that ρ is the same

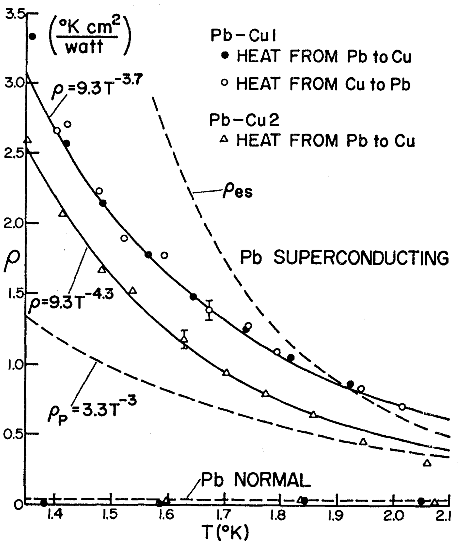

FIG. 1. Thermal resistivity at the interface between lead and copper.

in either direction and gives added confidence in the reliability of the measurements.

The contribution of the lattice ρ_P to the thermal resistivity at the interface is plotted in Fig. 1 as calculated from the following theoretical expression[7,17] which is valid for small values of ΔT.

$$\frac{1}{\rho_P}=2\times10^{10}\left[\frac{\Gamma_L}{C_L^2}+\frac{2\Gamma_T}{C_T^2}\right]T^3\frac{\mathrm{W}}{{}^\circ\mathrm{K\ cm}^2}. \tag{1}$$

In this expression Γ_L and Γ_T are related to the transmission of longitudinal and transverse phonons, respectively, and C_L and C_T are the velocities of sound of the two polarizations. Values for C_L and C_T for lead[18] and copper[19] have been reported previously as determined by ultrasonic techniques at liquid He temperatures. Values for Γ_L and Γ_T were obtained from calculations by Little,[7] who gave them as functions of the ratios of the densities and longitudinal sound velocities of the two materials. The curve labeled ρ_{es} in Fig. 1 represents an estimate, to be discussed later, of the electronic thermal resistivity at the interface with the Pb superconducting.

If the transfer of heat at the interface by electrons is independent of the transfer by the lattice, $1/\rho=1/\rho_e+1/\rho_P$, in which ρ_e and ρ_P are the electronic and lattice resistivities, respectively. The data of Fig. 1 indicate that $\rho_{en}\ll\rho_P$, making $\rho\approx\rho_{en}$ when the Pb is in the normal state. ρ_{en} is the thermal resistivity due to heat transfer by electrons with the lead normal. Although the curves of Fig. 1 suggest that $\rho_{es}>\rho_P$, with values of ρ in between them when the Pb is superconducting, the values of ρ_P calculated from Eq. (1) are not exact and may be somewhat larger than shown. In any case it is evident that $\rho_{es}\gg\rho_{en}$.

The samples were sectioned perpendicular to the interfaces and carefully polished and etched in order to determine the structure by microscopic examination. The lead in Pb-Cu 1 within a region extending about 0.1 mm out from the interface was highly polycrystalline. The remainder of the lead was composed of crystals 1 to 2 mm in diameter The lead in Pb-Cu 2 was a single crystal with no polycrystalline layer next to the interface. There was no evidence in either sample of a lead-copper diffusion zone at the interface. This is in accord with the very small values of ρ measured with the Pb normal. Such a diffusion zone would reduce the conduction of heat by electrons and cause an increase in the measured thermal resistivity at the interface. Each sample was then sectioned six more times to look for voids at the interface. None was found in Pb-Cu 1 and only one was found in Pb-Cu 2. It was approximately 0.05 mm in diameter too small to affect the data significantly.

Several factors could account for the measured values of ρ with the Pb superconducting having greater magnitudes and temperature dependences than the values ρ_P calculated from Eq. (1). This equation was derived by Little for a semi-infinite, perfectly plane interface, whereas the interfaces of actual samples are microscopically rough. It has been suggested[7] that such roughness would increase the temperature dependence of ρ. If the temperature is lowered to a value at which the dominant phonon wavelength becomes larger than the scale of the roughness, ρ should revert back to a T^{-3} dependence.

Also, the presence of strain and grain boundaries very near the interface, although properties of the bulk material, can contribute to the measured interface resistivity if they alter the resistivity of the bulk material. Strains can exist because the thermal contraction of lead, when it is cooled from room temperature to 4°K, is twice as great as that of copper. At temperatures shown in Fig. 1, strain is known[20] to increase the thermal resistivity of superconducting lead by an amount which varies as T^{-3}. The additional resistivity due to grain boundaries[21] is thought to vary as T^{-2} to T^{-3}.

Taking into consideration the size of the samples and placement of thermometers used here, together with experimentally determined magnitudes[20] of the combined effects of strain and grain boundaries near the interface, it is estimated that these effects could account at most for about 20% of the measured values

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plotted in Fig. 1. The larger magnitudes and small temperature dependence of the Pb-Cu 1 values relative to the Pb-Cu 2 values could be due to the layer of polycrystalline lead adjacent to the interface. After allowing for effects on the results due to the above considerations concerning the lattice, there remains a large decrease in the thermal resistivity at the interface when the lead is changed from superconducting to normal. This must be due primarily to an increase in the transfer of heat across the interface by the electrons when the lead is changed from superconducting to normal.

Tin-Copper Interfaces

Three Sn-Cu samples were prepared in the same manner as Pb-Cu 1 and studied. Since Cu dissolves slowly in molten Sn, despite precautions a thin alloy zone was present at the interface of each sample. Microscopic examination revealed an alloy zone in the Sn adjacent to the interface which was 0.1 to 0.2 mm thick. It had the appearance of a very dilute alloy. There were numerous small inclusions in this alloy zone of a Sn-Cu eutectic.

The data obtained for two of these samples are given in Fig. 2. Data for the third have been reported elsewhere.[1] It is seen that ρ for Sn-Cu with the Sn superconducting is very much smaller than ρ for Pb-Cu with the Pb superconducting. ρ for Sn-Cu with the Sn normal was significantly greater than ρ for Pb-Cu with the Pb normal. The contribution of the lattice ρ_P to the thermal resistivity at the interface between Sn and Cu with the Sn superconducting as calculated from Eq. (1) is also plotted in Fig. 2.

ρ_P is significantly larger than ρ for Sn-Cu with the Sn either superconducting or normal. In view of the alloy zones described above and measurements of the effect of alloying on the thermal resistivity of tin,[22] it is reasonable to conclude that the measured values of ρ for Sn-Cu with the tin either superconducting or normal are characteristic of this alloy zone and not the interface. Since values of ρ characteristic of the interface alone would be smaller than those plotted, it is seen that the transfer of heat across a Sn-Cu interface is dominated by electrons with the Sn either superconducting or normal.

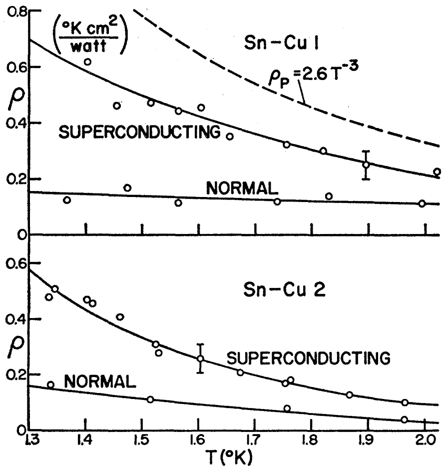

FIG. 2. Thermal resistivity at the interface between tin and copper.

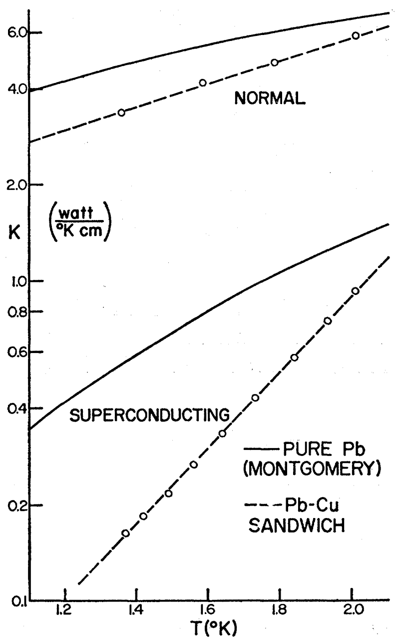

FIG. 3. Thermal conductivity of a lead-copper sandwich and of pure lead.

Lead-Copper Sandwich

In work at very low temperatures one frequently needs to have two reservoirs in good contact and then to thermally insulate them from each other. A lead wire connecting the two reservoirs serves as an adequate "thermal switch" for some applications. With the lead superconducting, its thermal conductivity K is much smaller than when it is normal. It can be switched from the superconducting to the normal state by applying a magnetic field. The two solid curves of Fig. 3 show the thermal conductivity of a single crystal of lead when it is superconducting and normal as measured by Montgomery.[15]

The large change in thermal resistivity at the interface between lead and copper when the lead is changed from superconducting to normal suggests the possibility of using this interface resistivity to enhance the operation of a lead switch.[14] The two dashed curves of Fig. 3 show measurements of thermal conductivity K obtained for a multiple-layered sample of lead and copper. This sample was prepared by vacuum casting seventeen oxygen-free high-conductivity (OFHC) copper cylinders $\frac{1}{8}$ in. long and $\frac{1}{4}$ in. in diameter together

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with a thickness of about 0.005 cm of lead between adjacent cylinders.

Measurements on the sandwich with the lead normal agree with what one would expect from data for the thermal conductivity of OFHC copper and pure lead. This is in accord with measurements presented above showing that the thermal resistivity at an interface between Pb and Cu is very small with the Pb normal. With the Pb superconducting, K of the sandwich was decreased considerably below that of pure lead. These results together with those of Challis and Cheeke[23] suggest that such sandwich structures should be useful as thermal switches for some applications.

Kapitza Resistivity

During the course of the above measurements, the Kapitza resistivities of tin, lead, and copper were also measured with the tin and lead superconducting and normal. The data obtained agree quantitatively in both magnitude and temperature dependence with those reported by other observers. The Kapitza resistivity of superconducting lead was observed to be 2 to 3 times larger than its value for normal lead,[2,3] whereas it was only about 10% greater for superconducting tin than for normal tin.[3]

Heat Transfer by Electrons at an Interface

The curve of Fig. 1 representing estimates of the electronic thermal resistivity ρ_{es} at the interface between Pb and Cu with the Pb superconducting was determined as follows. Strässler and Wyder[24] have considered the transfer of heat by electrons across interfaces between superconducting and normal material which characterize the intermediate state of a type-I superconductor. Electrons in the normal material with energies lying within the energy gap of the superconductor are assumed to be totally reflected at the super-normal interface. Additional scattering by electrons with energies above the gap was taken into consideration by using an effective gap $\Delta' = \Delta + \beta\Delta$, in which Δ is the normal energy gap and $\beta\Delta$ is a term which takes into account additional scattering at the interfaces by electrons with energies above the gap. With this assumption and the Bardeen, Rickayzen, and Tewordt[12] theory of thermal conduction by electrons in superconductors, they were able to account for the anomalous thermal resistivity of indium, with $\beta = 0.172$. Challis and Cheeke[23] have used the arguments of Strässler and Wyder in interpreting some of their measurements of interface resistivities. These arguments lead to the following

$$(K_{es}/K_{en})(\Delta') = \rho_{en}/\rho_{es}. \qquad (2)$$

K_{es} and K_{en} are the thermal conductivities of the superconducting metal due to the electrons when the metal is in the superconducting and normal states, respectively. As used previously, ρ_{en} and ρ_{es} are the thermal resistivities due to the electrons at an interface when the superconducting metal is in the normal and superconducting states, respectively.

As seen from Fig. 1, ρ_{en} was <0.04 for all Pb-Cu interfaces studied. For substitution into Eq. (2) it is taken as 0.02 and assumed to be independent of temperature in the range of the data covered by Fig. 1. Assuming that $\beta = 0$ and using values of the ratio K_{es}/K_{en} calculated according to Bardeen, Rickayzen, and Tewordt,[12] Eq. (2) gave values of ρ_{es} plotted in Fig. 1 for a Pb-Cu interface. Equation (2) predicts that the behavior of electrons, in contributing to the thermal resistivity at an interface between a superconducting and normal metal, is similar to the behavior of electrons in contributing to the thermal resistivity of the superconducting metal. The measurements presented in previous sections here are in accord with this.

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