THE THERMAL CONDUCTIVITY OF SUPERCONDUCTORS IN THE INTERMEDIATE STATE

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The thermal conductivities of lead, tin and gallium in the intermediate state were measured between 0.15 and 3.7°K. It was found that the transition to the intermediate state is, for heat transport due to phonons, accompanied by an increase in thermal resistivity ΔW_{gi} which is proportional to T^{-3} over the whole temperature range. The magnitude of ΔW_{gi} is close to that calculated on the assumption that the phonons are scattered by conduction electrons in the normal state domains. For heat transport by electrons the increase in thermal resistivity ΔW_{ei} depends weakly on the impurity content of the specimen. Only the dependence of $\Delta W_{ei}/W_{es}$ on T/T_c is similar for all metals. This ratio decreases approximately 30 times between $0.4T_c$ and $0.9T_c$. At temperatures below ~ $0.4T_c$ the value of ΔW_{ei} is close to that calculated on the assumption that the electron mean free path is limited by the boundaries of the intermediate state domains.

N this work we study the additional thermal resistivity which arises when heat is conducted across alternate domains of normal and superconducting phases. An investigation of this effect makes possible an understanding of the phenomena occurring when particles cross the phase boundaries. As is known, a periodic structure arises in the intermediate state, for example, in a cylindrical specimen in a magnetic field $H > \frac{1}{2}H_c$, which is perpendicular to the axis of the cylinder. As a result of the effects studied, the thermal conductivity in the intermediate state may be several times smaller than the conductivity not only in the normal, but also in the superconducting phase. Mendelssohn and Olsen¹ were the first to find a reduction in the thermal conductivity of a specimen in the intermediate state, during measurements on leadbismuth alloys. This effect was also found in pure metals (see Abrikosov and Zavaritskii² for references), but there has been no systematic investigation over a wide temperature range.

Heat transport in a superconductor is effected by two mechanisms: by lattice phonons and by electrons. One or the other is dominant, depending on the temperature, the impurity content, etc. The problem posed for this work was the elucidation of the nature of the heat transport for each mechanism separately in the intermediate state.* We measured several metals for this purpose: lead, tin, and gallium. The results obtained for lead were used to determine the change in phonon conductivity, those for tin the change in phonon and electron conductivity and those for gallium the heat conduction by electrons.

The heat conductivity measurements were made on single crystal specimens ~50 mm long and ~1 mm in diameter. The tin and lead specimens were cast in vacuo in thin walled glass capillaries in which crystallization took place immediately after casting. The glass was then removed by etching in hydrofluoric acid. The gallium specimens were prepared as in our previous work.³ For the specimens, lead with ~ 10^{-3} % impurity content, tin with 2 × 10^{-3} %, and gallium with between 0.2 and ~5 × 10^{-4} % impurity were used. Tables I

TABLE I. Characteristics of the specimens used for studying phonon heat conductivity

Specimen	Diameter cm	Phonon mea			
		$\begin{array}{c c c c c c c c c c c c c c c c c c c $		10 ^a <i>l_{gn}</i> , cm	$10^{s} a_{s}$, cm (from (2) for $\eta = 0.5$)
Sn Pb-1 Pb-2	0.175 0.13 0.21	130 28 28	11.5 7 9	$1.2 \\ 1.1 \\ 1.1$	7 4.2 5.4
		at T	∼0,1°K		
Ga 3G-c	0.3	~ 200	~ 70	—	I —

In calculating a_s , results of the direct measurement of Δ were used for tin,⁹ and the values of Δ calculated from measurements of penetration depth¹⁰ according to the formulae given by Ginzburg¹¹ and Gor'kov¹² were used for lead.

^{*}The results obtained in this work show the inaccuracy of the earlier suggestions² that it is sufficient to consider only the change in lattice conduction in order to explain the decrease of thermal conductivity in the intermediate state.

TABLE II. Characteristics of the specimens used for studying electronic heat conductivity

Ga specimen	Diameter cm	K _{Tc} *, W•cm ⁻¹ deg ⁻¹	₽₀/₽ ₂₉₃ °K	% ** impurity	$\frac{\Delta W_{ei}}{W_{es}}$ at T=3,5T _c	$\frac{\Delta W_{ei}}{\left(\Delta W_{ei}\right)_{\text{spec. c-10}}}$
20 0	1 0 2	1 2 5 40-2	4 4 40-2	1 9 40-1		
3G-C	0.5	3.3.10 -	1,4.10	2.10-1		
C-0	0.12	0,9	5.4·10 ⁻⁴	7.7.10-3	0,5	2
c-7	0,083	2,8	1.6.10-4	$2.3 \cdot 10^{-3}$	0,9	1.5
c-8	0.16	4	$1.2 \cdot 10^{-4}$	1.7.10-3	1.25	1.4
c-9	0,115	8	6.10-5	8.6.10-4	2.0	1.2
c-1 0	0.11	~13	$\sim 3.6 \cdot 10^{-5}$	5.10-4	2.7	1
a -5	0,16	16	8•10-5	$2 \cdot 10^{-3}$	1.25	_
b-4	0.16	34	8.10-5	2.10-3	0.5	
b-5	0.083	~120	$2.4 \cdot 10^{-5}$	5.5.10-4	1.1	_
Sn specimen	0,175	48	1,6.10-4	2.10-3	1.0	_

*For a number of specimens $K_{T_{\rm C}}$ is the result of extrapolating the measured $K_{\rm S}(T)$ (according to Zavaritskii').

**The impurity content is calculated from analysis of specimen 3G-c on the assumption that ρ_0/ρ_{293} is proportional to the impurity content of the metal.



and II show the characteristics of the specimens used.

A method similar to that used previously⁴ was employed to measure thermal conductivity. The dependence of thermal resistivity on the strength of the magnetic field perpendicular to the specimen axis was measured at constant temperature. During each measuring cycle the temperature of one of the thermometers on the specimen was held constant. A correction was applied to take into account the change in mean temperature of the specimen as the temperature gradient varied during the experiment; this reached some tenths of one per cent in different cases. In the work on FIG. 1. The variation of thermal resistance for: A - lead (Pb-1) and B - tin in the intermediate state for various temperatures (degrees Kelvin). The dash-dot curve corresponds to $a\Delta W_{ei}$ = const.

gallium, the specimen was heated above the critical temperature after each measuring cycle and brought back into the superconducting state in a magnetic field compensated to within ~ 0.2 oe.

RESULTS AND DISCUSSION

The results of measurements of the thermal resistivity of specimens of lead, tin, and gallium in the intermediate state are shown in Figs. 1 and 2. The conductivity of a number of specimens in the superconducting state is plotted in Fig. 3.

As can be seen from Figs. 1 and 2, the transition from the superconducting to the intermediate state (at a field $\frac{1}{2}H_{C}$) is generally accompanied









FIG. 2. The variation of thermal resistance for gallium specimens in the direction of the c axis in the intermediate state for various temperatures: A - specimen 3G-c, B - specimen c-6, C - specimen c-8, D - specimen c-9. The data of Zavaritskii³ were used to determine the value of H_c for each curve.

by an increase in thermal resistivity W.* This increase was reproduced in all the control measurements (see, for example, the data for the two series of measurements in Fig. 2C for 0.62°K). The magnitude of the increase is independent of the heat flow along the specimen, as shown by extra experiments in which the heat flow was changed by a factor of up to ten. Hysteresis effects were often found on the reverse transition from the normal to the superconducting state (by reducing the magnetic field). Some of these (see, for example, Fig. 2D) are apparently related to a supercooling of the normal phase, as a result of which the transition from the normal to the intermediate state only takes place in a field less than

*We should note that the increase in resistivity in the intermediate state makes it possible to reduce by several-fold the conductivity of a superconducting "heat switch" in the "open" position. H_c . Others (see, for example, Fig. 2A) are connected with "frozen-in" normal phase domains, which cause the thermal resistivity to remain larger than in the superconducting phase after the field has been switched off. These effects, which are due to the existence of "frozen-in" domains, are found only in gallium specimens with 2×10^{-1} % impurity. In what follows we will discuss only the results obtained with the magnetic field increasing.

The behavior of the heat transfer through the successive domains of normal and superconducting phases is given, to first order, by the quantity

$$\Delta W_i = W_i - \{(1-\eta)W_s + \eta W_n\}, \qquad (1)$$

where W_i , W_S and W_n are the thermal resistivities in the intermediate, superconducting, and normal states and η is the concentration of normal phase. For a cylindrical specimen in a magnetic field perpendicular to its axis, η changes from 0 to 100% as the field changes from $1/_2 H_C$ to H_C . At the same time, the period of the intermediate state structure, a, also changes; according to E. M. Lifshitz and Sharvin, 5

$$a = \sqrt{\Delta d / \varphi(\eta)} \tag{2}$$

 $(\Delta$ is the surface tension between the normal and superconducting phases, d is the specimen diameter, and $\varphi(\eta)$ is a dimensionless parameter tabulated by Lifshitz and Sharvin⁵*, and must also be taken into account.

^{*}Although relation (2) was obtained⁵ by considering the intermediate state of an infinite sheet, it can also be used for determining the period of the structure in a cylindrical specimen, according to a calculation by Sharvin.

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1. THE LATTICE HEAT CONDUCTIVITY

In order to understand the mechanism of heat transfer by phonons, the most useful experimental results are those obtained in the temperature region where in the superconducting state the electronic heat conductivity is negligibly small compared with the phonon conductivity. According to Montgomery,⁶ this occurs at temperatures lower than $\sim 1^{\circ}$ K for lead and lower than $\sim 0.3^{\circ}$ K for tin.⁴

As can be seen in Fig. 1, the transition to the intermediate state in this temperature region is accompanied by a sharp increase in thermal resistivity (6 times for tin and 2.5 times for lead). The relative increase in resistivity $\Delta W_{gi}/W_{gs}$ changes only negligibly with temperature. At constant temperature ΔW_{gi} varies in inverse proportion to the period of the intermediate state structure.* In all specimens investigated, the temperature variation of the absolute magnitude

of ΔW_{gi} was close to T^{-3} . As the temperature was reduced from 1 to 0.15°K, the change in the value of $T^{3}\Delta W_{gi}$ was not more than 20 or 30% (see Fig. 4).

At temperatures so much lower than T_c , the thermal resistivity of the normal domains is negligibly small compared with that of the superconducting domains.* The extra resistance ΔW_{gi} can therefore only relate to a change in conductivity of the superconducting phase, which is determined by phonon heat transport. The phonon heat conductivity K_g can be expressed in terms of the phonon mean free path I_g , the velocity u, and the lattice heat capacity per unit volume C, by the relation

$$K_g \sim \frac{1}{3} l_g \, uC \tag{3}$$

or, on substituting numerical values,

$$K_g \sim Gl_g T^3 W / cm-deg,$$
 (3a)

^{*}In all cases the change in phonon resistivity in the intermediate state departed from the relation $a\Delta W_{gi} = const$ by less than 10% with the proportion of normal phase varying from 20 to 80%.

^{*}For tin at 0.3°K, W_n/W_s is -10⁻³; for lead at 1° and 0.3°K this ratio is -5 × 10⁻² and 5 × 10⁻³.



where $G_{\rm Pb} = 4$, $^7 G_{\rm Sn} = 1$, 4 and $G_{\rm Ga} = 0.5$. From expression (3a), $\Delta W_{\rm gi}$ can be related to the change in the inverse of the phonon mean free path, produced by extra scattering in the intermediate state. We obtain

$$1/l'_{gi} = 1/l_{gi} - 1/l_{gs} \sim GT^{3} \Delta W_{gi}/(1-\eta)$$
 (4)

 $(l_{gi} \text{ and } l_{gs} \text{ are the phonon mean free paths in the intermediate and superconducting states). The magnitude of <math>l'_{gi}$ changes little in the temperature range studied, as can be seen from Fig. 4, but possibly increases slightly with decreasing T. Since the dimensions of the superconducting domains in the intermediate state $a_s = a(1 - \eta)$, it follows from the proportionality of ΔW_{gi} and i/a and relation (4) that l'_{gi} varies in the entire intermediate range in the same way as the dimensions of the superconducting layers. A calculation of the absolute value of l'_{gi} is close to the dimensions of the superconducting layers.

Let us compare our results with the mechanism for extra scattering of phonons, proposed independently by Abrikosov and Zavaritskii² and by Laredo and Pippard.¹³ The essentials of this mechanism are as follows: because of the presence of conduction electrons, the phonon free path in the normal domains, l_{gn} , is small* (see Table I) and therefore the transition of a phonon from a superconducting to a normal region is accompanied by its immediate scattering. If the phonon mean free path in the superconducting state is limited only by scattering at lattice defects and at the specimen boundaries, then in the intermediate state the dimensions of the superconducting domains will also limit it, leading to a considerable reduction of $l_{\rm g}$ (see Table I). The direct proportionality between $l'_{\rm gi}$ and $a_{\rm s}$ and their close agreement are explained in this way. Some difference between the absolute values of $l'_{\rm gi}$ and $a_{\rm s}$ in our specimens may be due to the approximation in Eq. (4), since the real structure of the intermediate state in a cylindrical specimen¹⁵ must differ somewhat from the assumed structure consisting of alternating layers crossing the whole specimen. An increase in $l'_{\rm gi}$ can also be produced by phonons "jumping" through the normal domains without being scattered; this becomes especially probable if $l_{\rm gn}$ is comparable with the width of the normal domains. The slight increase in $l'_{\rm gi}$ with decreasing temperature may be connected with this effect.

The mechanism described above can, in this way, explain at least qualitatively the whole range of phenomena observed for phonon heat transport in the intermediate state.

2. THE ELECTRONIC HEAT CONDUCTIVITY

We used the data obtained for gallium and tin to elucidate the behavior of the electron heat transport in the intermediate state. We discuss here only the properties of gallium specimens prepared from metal with less than ~0.01% impurity, for which the lattice conductivity is negligibly small compared with the electronic heat conductivity (Fig. 3) over the whole range of measurements (0.15 to 1°K). For tin we use the results obtained between 0.7 and ~3°K, with a correction applied below 1°K for the contribution Kg of the lattice heat conductivity.*

Let us first consider the results of measurements on the thermal resistivity of gallium specimens with their length along the crystallographic c axis. The large number of specimens studied enabled us in this case to find the main features of the phenomenon of interest. The transition to the intermediate state for all specimens is accompanied by the appearance of an increase in thermal resistance. There is apparently no direct proportionality between the change in ΔW_{ei} and the inverse of the period of the structure $1/a^{\dagger}$, as a result of which a maximum of ΔW_{ei} is usually found at $\eta \sim 0.5 - 0.7$ (see Fig. 2). In what follows we shall consider just $(\Delta W_{ei})_{max}$.

^{*}The variation of lattice thermal conductivity in the normal state K_{gn} is at sufficiently low temperatures, K_{gn} = BT² where B_{Pb} = 1.3×10^{-3} W cm⁻¹ deg⁻³, B_{Sn} = 3.5×10^{-4} W cm⁻¹ deg⁻³.^{6,14} It follows from this relation and Eq. (3) that the phonon mean free path in the normal state $l_{gn} = l/T$, where $l_{Pb} = 3.25 \times 10^{-4}$ cm·deg and $l_{Sn} = 3.5 \times 10^{-4}$ cm·deg.

^{*}In calculating the lattice conductivity it is assumed that for tin between 0.7 and 1°K in the superconducting state $K_{gs} = 0.135T^3$, and in the intermediate state $K_{gi} = 0.025T^3$. The correction for the lattice conductivity is ~10% at 1°K and ~50% at 0.7°K.

[†]In all the curves of the change of electronic thermal resistance in the intermediate state, both for gallium and for tin specimens, a systematic increase in the value of $a\Delta W_{ei}$ is observed, reaching 300% when the fraction of normal phase varies from 20 to 80%.



The relative value of the change in thermal resistance $\Delta W_{ei}/W_{es}$ depends on the impurity concentration. As the amount of impurity is decreased from 10^{-2} % (specimen c-6) to 5×10^{-4} % (specimen c-10) the magnitude of $\Delta W_{ei}/W_{es}$ increases (at T ~0.1°K) from 0.8 to 6. In the temperature range studied $\Delta W_{ei}/W_{es}$ is not constant, but continously decreases with increasing temperatures, most sharply in the temperature region above ~ 0.4 °K. This temperature dependence of the ratio $\Delta W_{ei}/W_{es}$ appears to be universal for all specimens (see Fig. 5). The temperature dependence of the absolute value of ΔW_{ei} – the change of thermal resistance in the intermediate state (Fig. 6) - likewise agrees for all specimens. The impurity content has only little influence on the value of ΔW_{ei} . As the impurity concentration is reduced almost 15-fold, ΔW_{ei} is only reduced to one half (see Fig. 2).



FIG. 6. The variation in the absolute value of the resistance ΔW_{ei} with temperature in gallium specimens.



FIG. 7. The dependence of thermal resistance change in gallium and tin specimens in the intermediate state on reduced temperature. $\Delta W_{ei}/W_{es}$ was taken as 1 at $T/T_c = 0.35$. The broken curves correspond to the dependence for gallium specimens along the c axis (according to Figs. 6 and 7).

Gallium specimens of different orientation differ both in the absolute value of ΔW_{ei} and in the law of its temperature variation (see Fig. 7A). The anisotropy of ΔW_{ei} to some extent reflects the anisotropy in the thermal conductivity of gallium in the superconducting state.³ The largest value of ΔW_{ei} is found along the c axis, corresponding to the minimum thermal conductivity Kes and in this direction the temperature variation of both K_{es} and ΔW_{ei} is more gradual than in the other directions. There is not, actually, a complete correspondence between the anisotropies in K_{es} and $\Delta W_{ei}.$ This is seen most clearly in the character of the temperature dependence of the relative change of thermal resistance ΔW_{ei} Wes. This dependence for gallium agrees over the whole temperature range studied only along the a and b axes (see Fig. 7). Along the c axis the dependence of $\Delta W_{ei}/W_{es}$ on T is considerably different at temperatures below $\sim 0.35 T_{C}$. We should remark that the c direction is also differentiated from the other two in the anisotropy of the energy spectrum of the superconductor excitations.³

The results for tin, in which there is a considerably more rapid variation of ΔW_{ei} than for gallium (Fig. 7), confirm the absence of a general relation between ΔW_{ei} and T/T_c for all superconductors. It is therefore all the more interesting that these metals display a similar dependence of the relative change in thermal resistance $\Delta W_{ei}/W_{es}$ on the reduced temperature. At temperatures $T \ge 0.35T_c$ this dependence is the same, within the experimental accuracy, for all the specimens



FIG. 8. The field dependence of resistance of gallium specimen c-9, with the magnetic field perpendicular to its axis, for two values of the current through the specimen.

studied, and below ~ $0.35 T_{C}$ only gallium along the c axis shows a departure from the general relation. It appears that the variation of the relative change in thermal resistance with reduced temperature is one of the main features of heat transport by electrons across alternate regions of normal and superconducting phases.

We suggest, unlike, for example, Hulm,¹⁶ that the increase in resistance in the intermediate state is determined mainly by the change in electronic heat transport in the superconducting domains (we are concerned here with the so-called "normal" electrons of the superconductor). Both the occurrence of an appreciable effect at temperatures much below the critical, where the resistivity of the normal phase is negligibly small compared with that of the superconducting, and the results of the following additional experiments bear out this suggestion.

a) The electrical resistivity of the specimens R was measured in the intermediate state. We found that $R = \eta R_n$, where R_n is the resistivity of the specimen and η is the concentration of normal phase. No additional increase in resistivity of the normal phase was found in the intermediate state (Fig. 8) which might be expected if there were an increase in the thermal resistivity of the normal domains.

b) The thermal resistivity W of a tin single crystal of high purity with the length along the four-fold axis was measured. Measurements were made with the directions of a magnetic field along the directions of maximum and minimum variation of thermal conductivity in the normal state (the angle between these directions is 22.5°). Although the thermal resistivity in these two directions differs by a factor of 1.5 in the normal domains, the difference in the absolute value of ΔW_{ei} did not exceed 5 - 10% (see Fig. 9). FIG. 9. The variation of thermal resistance of a high purity tin specimen (impurity ~ 5×10^{-5} %, $W_s \approx 9$ × 10⁻³ cm·deg·W⁻¹) in the intermediate state (at 2.2°K), for the field directed: •along the minimum of conductivity change, o - along the maximum.



By analogy with the case of phonon heat transport, it seems natural to connect the reduction in electronic thermal conductivity in a structure consisting of alternate layers of normal and superconducting phases, with the existence of an additional "scattering" of electrons. Since the change in the absolute value of ΔW_{ei} with impurity content is insignificant, we can assume that this mechanism and the scattering of electrons by impurities are additive to a first approximation. Assuming, in line with present theory, ¹⁷⁻¹⁹ that the electron mean free path in a superconductor, due to scattering by impurities, is independent of temperature,* then the reciprocal free path for the extra scattering mechanism is

$$\frac{1}{l_{ei}'} = \frac{1}{l_{ei}} - \frac{1}{l_{es}} \approx \frac{\Delta W_{ei}}{W_{es}} \frac{LT_{\mathbf{c}}\sigma/l}{K_{T_{\mathbf{c}}}(1-\eta)},$$

where K_{T_c} is the thermal conductivity at the criti-

cal temperature, L is the Lorenz number and σ/l is the ratio of the conductivity of the specimen to the mean free path, measured in the normal state. Using the value $\sigma/l \approx 5 \times 10^{10}$ derived from experiments on the size effect on electrical resistance,^{20,21} we find that for $T \leq 0.4T_{C}$ the free path $l'_{\rm ei}$ is close to the dimensions of the superconducting domains.† Thus, at least qualitatively, the reduction in the electronic heat conductivity in the intermediate state is explained by the assumption that the electron mean free path is limited by the phase boundaries. We should note that this reduction in mean free path can, in fact, be produced either by an extra scattering of the electrons or by their elastic reflection at the phase boundaries. In the present case it only makes sense to talk of reflection according to classical laws since the width

^{*}Except for the tin specimen used in the additional experiments, all the specimens were studied in the temperature region where the electron mean free path is mainly determined by impurity scattering.

[†]For a change in concentration of normal phase from 0.2 to 0.8 the ratio l'_{ei}/a_s in tin decreases from 1.2 to 0.5.

of the transition layer between the phases ($\sim 10^{-5}$ cm) is many times greater than the electron wavelength ($\sim 10^{-7}$ cm). A qualitative calculation of the resistance produced by reflection shows that it is sufficient to assume that $\sim 50\%$ of the electrons are elastically reflected at the phase boundaries in order to explain the results obtained on tin for $T \lesssim 0.4T_c$.

The mean free path l'_{ei} is only close to the dimensions of the superconducting domains a_s at the lowest temperatures. It follows from relation (5) and from the dependence $\Delta W_{ei}/W_{es}$ on T that the ratio l'_{ei}/a_s increases with increasing temperature. This may be taken as an indication that the additional scattering (or reflection) of electrons at a phase boundary disappears as the critical temperature is approached.* From the data of Fig. 7 it follows that the decrease in scattering (or reflection) of electrons is almost the same for all the superconductors.

While we could relate the reduction in mean free path for phonon heat transport to scattering of phonons by conduction electrons in the normal domains, in the case of electronic thermal conductivity the effect of scattering of electrons by electrons is too small to explain the observed effect. It is not impossible that the appearance of an extra thermal resistivity in this case is connected with the different energy spectra of electrons in the superconducting and normal phases of the metal. If, in fact, the energy-momentum relations are different for electrons in the two phases, then there will be additional scattering when the phase boundary is crossed. We should mention that we are concerned here with the so-called "normal" electrons of a superconductor, which do not show superconducting properties. This difference in the energy spectra, together with the decrease in this difference as the temperature approaches T_c , as follows from the form of the $\Delta W_{ei}/W_{es}$ - T/T_c relation, agrees qualitatively with the present theory. We can ascertain whether all the results obtained can be explained quantitatively only after making the appropriate calculations.

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^{*}The change in the period of the intermediate state structure is small, over the temperature range studied, compared with the the change in $\Delta W_{ei}/W_{es}$. Thus, while $\Delta W_{ei}/W_{es}$ decreases nearly 30-fold between 0.5 and 0.9 T_c, the increase in the period of the structure, a, amounts to only ~40%.⁹