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**Autor:** Olsen, J.L. / Waldvogel, A. / Wyder, P.  
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## Size Effect in the Intermediate State Thermal Resistivity Maxima in Indium and Lead

by J. L. Olsen, A. Waldvogel, and P. Wyder<sup>1)</sup>

Institut für kalorische Apparate, Kältetechnik und Verfahrenstechnik,  
Swiss Federal Institute of Technology, Zürich, Switzerland

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*Abstract.* We have measured the size dependence of the intermediate state thermal resistivity maxima in superconducting lead and indium. At temperatures near 2°K phonon conduction is dominant in the intermediate state in lead while electron conduction dominates in indium. For the phonon case the maximum resistivity is inversely proportional to the square root of the specimen diameter. In the electron conduction case the ratio of maximum resistivity to the resistivity in the superconducting state is independent of specimen size at temperatures above ca.  $T_c/2$ .

### Introduction

Cylinders of type I superconducting material in a transverse magnetic field change their magnetic moment and electrical resistivity approximately linearly from the superconducting to the normal value as the field is increased from  $1/2 H_c$  to the critical field  $H_c$ . A similar linear change is found in some cases in the thermal resistivity. MENDELSSOHN and OLSEN [1, 2]<sup>2)</sup> found, however, that the thermal resistivity in the intermediate state can be much greater than that corresponding to such a linear change, and that there may be a pronounced maximum in the thermal resistivity greatly exceeding its value in either the normal or superconducting state. This resistivity increase has since been studied experimentally by several authors. It exists both in the low temperature range where phonons make the major contribution to the thermal conductivity in the superconducting state [3–9], and in situations where electron transport dominates in both normal and superconducting states [10–15]. Theoretical treatments of the first case where phonon transport is important have been given by CORNISH and OLSEN [16], ABRIKOSOV and ZAVARITSKIĬ [17], and by LAREDO and PIPPARD [9]. The second case has been discussed by HULM [12], STRÄSSLER and WYDER [18], WYDER [13], and by ANDREEV [19] who all conclude that the maximum is caused by scattering of electrons at the normal-superconducting phase boundaries.

The temperature dependences of the effects have been investigated by various authors [7, 12, 14], but no detailed measurements of the effect of specimen size appear to exist. For the phonon transport case all three treatments lead in first approximation to an increase in the thermal resistivity proportional to the number of boundaries in

<sup>1)</sup> Now at the Department of Physics, University of California, Berkeley.

<sup>2)</sup> Numbers in brackets refer to References, page 368.

the intermediate state structure for given phonon and electron bulk mean free paths. In the pure electron transport case ANDREEVS [19] calculations which are valid at very low temperatures lead to a similar dependence on the scale of the intermediate state structure. WYDERS [13] calculations may be extended to give the same result at low temperatures. They indicate, however, that at temperatures close to the critical the anomalous resistivity increase should be proportional to the thermal resistance in the superconducting state.

We have thought it important to test these predictions. The results on lead at low temperatures where phonon conduction dominates the superconducting state heat conduction are found to be in agreement with the theory. In indium in a range where electron conduction is dominant we find agreement with the results of WYDERS modified treatment.

### Phonon Case – Lead

Electron transport dominates in the normal state in all but the most impure specimens. In the superconducting state phonon transport will dominate at sufficiently low reduced temperatures  $t = T/T_c$ . The reduced temperature at which this occurs depends upon the purity and increases with increasing  $T_c/\theta_D$  where  $\theta_D$  is the Debye characteristic temperature. We chose lead where  $T_c$  is large and  $\theta_D$  small to allow us to study this situation over as wide a range of temperature as possible.

A single crystal of lead supplied by Semi Elements Inc. and stated to contain less than 5 ppm of metallic impurity was investigated. Thermal conductivity measurements were made at a range of diameters. Between each set of measurements the diameter was reduced by electrolytic etching and the surface was subsequently smoothed by electropolishing. The sizes investigated are shown in Table I. The temperature dependence of the normal state and superconducting conductivities are

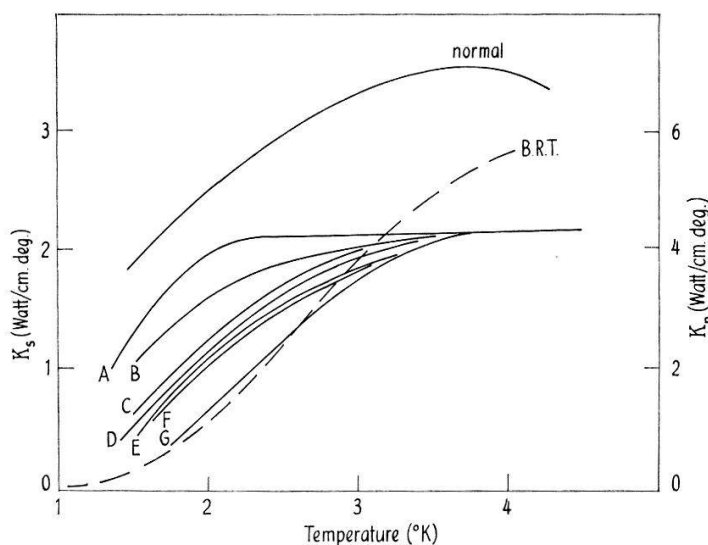


Figure 1

Heat conduction of lead specimens in normal and superconducting states. Top curve gives normal state conductivity for all specimens. (For this curve right hand scale applies.) Curve A: specimen No. 1 a; Curve B: No. 1 b; Curve C: Nos. 3, 6, and 7; Curve D: No. 4; Curve E: No. 5; Curve F: No. 2; Curve G: No. 8. The dashed curve shows result of Bardeen, Rickayzen, Tewordt theory.

shown in Figure 1. The normal state conductivity was independent of diameter. This is natural since the normal state electronic free path was of the order of  $10^{-3}$  cm and thus much smaller than the diameter of the specimens.

Table I  
Lead Specimens

Specimen No.		Diameter mm
Pb 1	as supplied	13.04
Pb 1 a	slightly strained	13.04
Pb 2	rough surface	12.26
Pb 3	smooth surface	9.94
Pb 4	smooth surface	9.25
Pb 5	rough surface	9.05
Pb 6	smooth surface	7.37
Pb 7	smooth surface	4.15
Pb 8	smooth surface	1.35

There is a noticeable difference between the superconducting thermal conductivities of the different specimens. This cannot be attributed to a size effect since the phonon free path in specimen 1 is approximately 0.5 mm at 1.5°K. We conclude that the difference is due to a gradual increase in internal strain due to the unavoidable handling of the specimens connected with the etching procedure. Our situation is thus different from that considered by LINDENFELD and ROHRER [20] who showed that the electronic contribution to the lattice thermal resistivity depended upon the residual electrical resistivity. This is the same for all our specimens.

The field dependence of the thermal resistivity  $W = \kappa^{-1}$  in one specimen for a range of temperatures is shown in Figure 2. The behaviour to be expected for the present case was discussed by CORNISH and OLSEN [16] who pointed out that imperfect thermal relaxation between electrons and lattice would lead to a resistivity maximum. They also discussed the influence of the degree of thermal coupling between lattice and electrons upon the height of the maximum. LAREDO and PIPPARD [9] assumed parallel but entirely uncoupled heat conduction processes in lattice and electrons. This is an extreme case of the CORNISH OLSEN model. LAREDO and PIPPARD's treatment, however, takes into account that the two contributions due to lattice and electrons may be modified further by the intermediate state structure. In particular they consider the possibility of electron scattering at the phase boundaries. This effect is treated in more detail by STRÄSSLER and WYDER [18] and most recently by ANDREEV [19].

From ANDREEV's theoretical work and from WYDER's experiments we conclude that the electronic conductivity in the intermediate state will be negligible for these specimens at the lowest temperature investigated. If so we may try to use LAREDO and PIPPARD's [9] estimate of the intermediate state lattice conductivity  $\kappa_{gi}$  to determine the height of the maximum  $W_{max}$ . For thick normal laminae they find

$$\kappa_{gi} = \frac{1}{4} v C_g a$$

where  $v$  is the phonon velocity,  $C_g$  the lattice specific heat, and  $a$  is the scale of the laminar structure. It is clear that this leads to a  $T^{-3}$  dependence for  $W_{gi} = \kappa_{gi}^{-1}$  if  $a$  is independent of temperature. Figure 3 shows the temperature dependence observed for  $W_{max}$ . It will be seen to be in good agreement with this prediction.

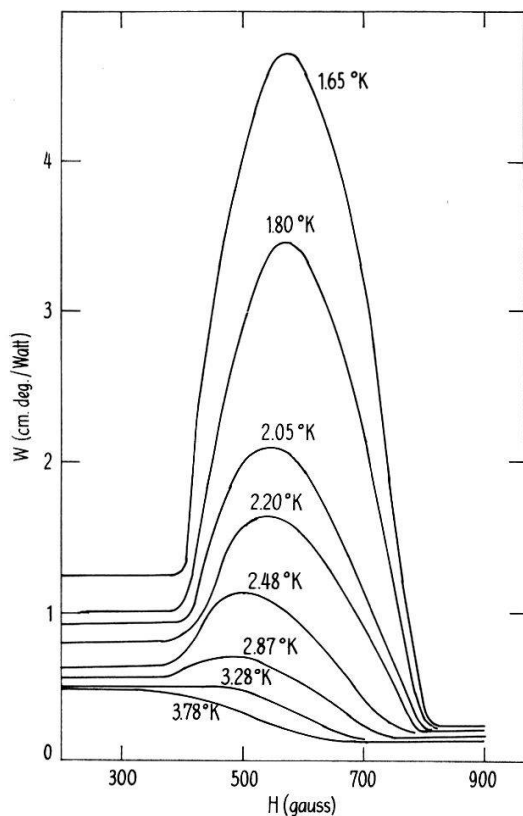


Figure 2

Field dependence of thermal resistivity of specimen Pb 7 as a function of magnetic field at the temperatures indicated.

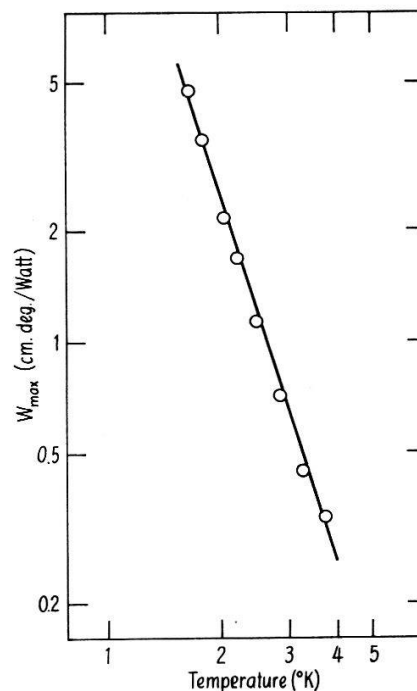


Figure 3

Temperature dependence of the maximum thermal resistivity,  $W_{max}$ , in Pb 7. The slope of the line corresponds to  $W_{max} \propto T^{-3}$ .

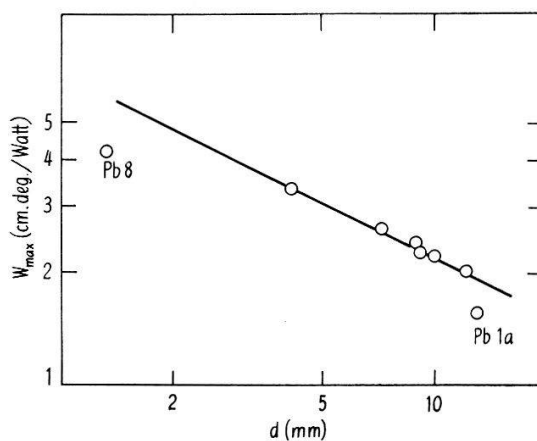


Figure 4

Dependence of  $W_{max}$  on specimen diameter at 1.78°K. The slope of the line corresponds to  $W_{max} \propto d^{-1/2}$ .

The size dependence is determined by the proportionality of  $\kappa_{gi}$  to  $a$ . In its turn  $a$  is proportional to  $(d/\Delta)^{1/2}$  where  $d$  is the specimen diameter and  $\Delta$  the surface energy parameter of the normal superconducting phase boundary. The field dependence of  $a$  has been discussed in detail by LIFSCHITZ and SHARVIN [22], but as both SHAWLOVS [23] powder work and the beautiful recent work of WALTON [15] show, special steps must be taken to obtain clearly defined normal and superconducting laminae extending right across the specimen. For this reason we consider only the maximum values  $W_{max}$  of  $W_{gi}$  near  $3/4 H_c$  where it is likely that the laminar structure is most complete for our comparison with theory.

Figure 4 shows the maximum resistivities observed at 2.06°K as a function of specimen diameter. All but specimen No. 8 show maximum resistivities proportional to  $d^{-1/2}$ . This is in agreement with our remarks above. The deviation of specimen No. 8 is not at present understood.

### Electron Transport Case – Indium

Here the resistivity maximum is caused by electron scattering at the boundaries between normal and superconducting material. This effect has been discussed by HULM [12], by STRÄSSLER and WYDER [18], by WYDER [13], and by ANDREEV [19]. While HULM's discussion is essentially qualitative the treatments due to STRÄSSLER and WYDER and ANDREEV successfully suggested the temperature dependence to be expected. There is, however, an apparent discrepancy between the effect of the scale of the structure derived from the two treatments.

STRÄSSLER and WYDER's treatment makes use of a relation due to BARDEEN, RICKAYZEN, and TEWORDT [20] stating that the thermal conductivity  $\kappa$  is given by

$$\kappa = \int - [2 N(0) v_0 / 3 T] l(E) E^2 (\partial f / \partial E) dE \quad (1)$$

where the energy  $E$  is measured relative to the Fermi energy  $E_F$ ,  $N(0)$  is the density of states at the Fermi surface,  $v_0$  the velocity of the electrons,  $T$  is the temperature,  $l(E)$  the free path as a function of the energy, and  $f(E)$  is the Fermi distribution function. The expression is independent of the density of states function and the real physical problem lies in the choice of a suitable function for  $l(E)$ . For this STRÄSSLER and WYDER chose a step function with

$$\begin{aligned} l(E) &= 0 & E &\leq (1 + \beta) \varepsilon_0 \\ l(E) &= l & E &> (1 + \beta) \varepsilon_0 \end{aligned} \quad (2)$$

where  $\beta$  is a temperature independent parameter and  $2 \varepsilon_0$  is the energy gap.  $l$  is the electronic free path in the absence of normal-superconducting interfaces, and is given by

$$l = l_s = l_n \quad (3)$$

$$1/l = 1/l_b + 1/d \quad (4)$$

where  $l_b$  is the bulk mean free path and  $d$  is the specimen diameter. This leads to the following expression for the thermal resistivity at the maximum

$$W_{max}/W_s = f[y]/f[(1 + \beta) y] \quad (5)$$

where  $W_s$  is the thermal resistivity in the superconducting state,  $y = \varepsilon_0/k T$ ,

$$f[y] = 2 F_1(-y) + 2 y \ln(1 + e^{-y}) + y^2/(1 + e^y) \quad (6)$$

and

$$F_1(-y) = \int_0^\infty z (1 + e^{(z+y)})^{-1} dz. \quad (7)$$

In this form  $W_{max}/W_s$  is independent of the specimen diameter.  $W_s$  will be diameter dependent due to the ordinary size effect in the free path, so that

$$W_{max} = W_{s,b} (1 + l_s/d) \quad (8)$$

where  $W_{s,b}$  is the bulk value of  $W_s$ .

This result is different from that obtained by ANDREEVS calculation which is valid only at low temperatures. It leads to a resistivity increase in the intermediate state proportional to  $d^{-1/2}$ .

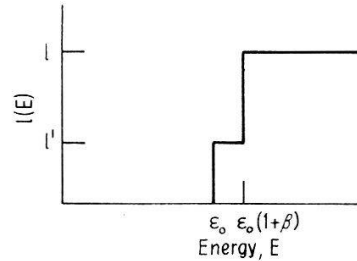


Figure 5

Electronic free path as function of energy.

The difference between the two results is understood if we notice that in WYDERS calculation  $l$  is taken to be zero up to energies  $(1 + \beta) \varepsilon_0$ . At sufficiently low temperatures only a negligible number of electrons having energies greater than this are excited, and we are thus ignoring most of the excitations in this calculation. This situation may be improved by also allowing these electrons to take part in the heat transport mechanism and assuming a non-zero free path  $l'$  for electrons having energies between  $\varepsilon_0$  and  $(1 + \beta) \varepsilon_0$ . The resultant free path distribution becomes that shown in Figure 5. It is plausible to suppose that these electrons are scattered mainly at the phase boundaries, and  $l'$  must therefore be of order of the thickness of the laminae. As a rough approximation we assume

$$l' = 2\sqrt{\Delta d} \quad (9)$$

where  $\Delta$  is the surface energy.

The expression (5) now has to be modified to read

$$\frac{W_{max}}{W_s} = \frac{l f[y]}{l f[(1 + \beta) y] + l' \{f[y] - f[(1 + \beta) y]\}} \quad (10)$$

and using (4) we obtain

$$\frac{W_{max}}{W_s} = \frac{f[y]}{f[(1 + \beta) y] + l' (1/l_n + 1/d) \{f[y] - f[(1 + \beta) y]\}}. \quad (11)$$



For temperatures near the transition temperature (say  $t > 0.5$ ) and for free paths longer than the scale of the laminae this expression tends to (5).  $W_{max}/W_s$  then becomes independent of  $d$ . For low temperatures on the other hand where

$$f[y] \xrightarrow{y \rightarrow 0} e^{-y}$$

so that

$$\frac{W_{max}}{W_s} \rightarrow \frac{l}{l'}$$

or

$$W_{max} \rightarrow \frac{l}{l'} W_s = \frac{l_b}{l'} W_{s,b}$$

so that from (9) we find  $W_{max} \propto (\Delta d)^{-1/2}$ .

Thus WYDERS modified formula leads to the same diameter dependence as that of ANDREEV at low temperatures, while at high temperatures we expect  $W_{max}/W_s$  to be independent of diameter.

An examination of Figure 12 in WYDERS [13] paper show this to be the case for a range of specimens. Of course, this implies that

$$W_{max} \propto (1 + l/d) \quad (13)$$

and also

$$\Delta W \propto (1 + l/d) \quad (14)$$

where  $\Delta W$  is the increase in resistivity due to the scattering at phase boundaries.

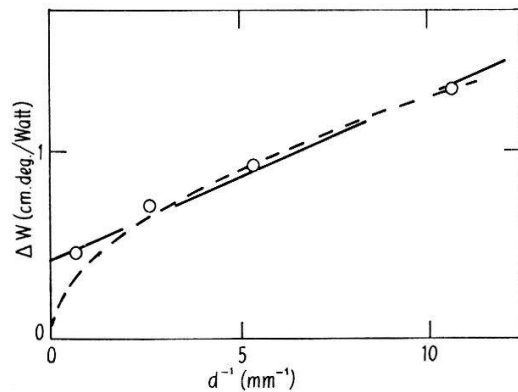


Figure 6

Dependence on thickness of  $\Delta W$  of the additional intermediate state heat resistivity in pure indium specimens at 2.1 °K.

To test this relation we have plotted  $\Delta W$  at 2.1 °K for a range of specimens in Figure 6. The slope of the line corresponds to a free path of 0.18 mm. This is in fair agreement with the value of 0.13 mm found by WYDER [24] from thermal conductivity size effect measurements in the normal state. For comparison we have shown dashed a curve  $\Delta W \propto d^{-1/2}$  scaled to fit the points at  $d = 0.1$  mm. It appears that the results may be described by either of the formulae although expression (14) gives a slightly better fit than that due to ANDREEV.



### Conclusion

The additional resistivity in the phonon case is found to be proportional to the inverse square root of the specimen diameter. This is in agreement with current theory. In the electron case either ANDREEVS formula or STRÄSSLER and WYDERS modified formula may be used to describe the present experiments relatively close to the transition temperature. For lower temperature no experimental data are available.

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