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THE RICE INSTITUTE

THE PARAMAGNETIC EFFECT IN SUPERCONDUCTING
TIN, INDIUM, AND THALLIUM

by

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TABLE OF CONTENTS

I	INTRODUCTION	1
II	EXPERIMENTAL	6
III	DISCUSSION OF RESULTS	22
IV	CONCLUSIONS	38
V	APPENDIX	40
	REFERENCES	50
	ACKNOWLEDGEMENTS	52

I Introduction

Superconductivity has provided a wealth of interesting and intriguing problems since Kamerlingh Onnes¹⁾ first noticed that the resistance of mercury decreased to zero at 4.2 degrees Kelvin. New impetus was provided by the discovery of the Meissner²⁾ effect (perfect diamagnetism) in 1933. Recent efforts³⁾ have been aimed at the mechanism of the effect, with attention to the transition region between normal conduction and superconduction. Several theories⁴⁾ have been able to correlate the known experimental facts concerning the superconducting state and to predict others. However, only recently has there been adequate data to permit formulation of models of the intermediate state⁵⁾. Most such models are based on knowledge of the pure superconducting state and extrapolations are made into the mixed region using general electrodynamic and thermodynamic arguments. Experimental verification of such models and an enrichment of basic data on the intermediate state were taken as the broad aims of this thesis.

As has been stated, the superconducting state is characterized by zero magnetic induction and vanishing electrical resistance. The experiments reported here deal with the manner in which the induction and resistance vary from their finite normal state values to zero in the superconducting state. . In very pure metals and under "ideal" conditions the changes from the normal state to the superconduct-

2.

ing state are assumed to be discontinuous⁶⁾. Actually the transition is usually spread out over a short interval of the variable by which the transition is effected. In the experiments to be discussed, the quantities which determine the state of the specimen are the temperature, T , the external magnetic field, H , and the current which flows in the specimen, I . Figure 1 shows the phase separation of super and normal states in these variables for the metal indium.

The broadening of the transition may arise from two causes. First, chemical and physical impurities may enhance or impair the growth of the superconducting state in certain regions of the material. This leads to a large grained mixture of super and normal conducting material in the transition region and does not concern us here. The second source of broadening is present in cases of extreme purity, and may be termed geometrical broadening. In this latter case, thermodynamic and electrodynamic equilibrium considerations require⁷⁾ that in a magnetic field the metal break up into a fine grained mixture of normal and superconducting metal. The normal grains contain magnetic flux while the superconducting grains do not; this is called the intermediate state. A discussion of one case of geometrical broadening is contained in Appendix A.

Experimentally there are two general methods of observation. First, one may try to make measurements directly on the magnetic flux structure of the intermediate state.

Meshkovsky and Shalnikov⁸⁾ used a bismuth probe whose resistance depends on B to observe the magnetic flux structure in the gap between the two halves of a sphere. Schawlow and others⁹⁾ at the Bell Laboratories have recently photographed the flux pattern on cylinders and flat plates using a diamagnetic powder on the surface. In each of these experiments direct measure of the magnetic flux structure at the surface was obtained. The approach has the disadvantage that end effects and measuring procedures may tend to distort the magnetic field observed. Alternatively one can measure the average magnetization or resistance of the intermediate state over the bulk of a specimen and then try to determine which detailed mixture of normal and superconducting material will yield the observed behavior. The latter approach was employed in the experiments which form the basis of this thesis.

The work described herein is a continuation of investigations begun by Steiner and Schoeneck¹⁰⁾ and extended by Meissner, Schmeissner, and Meissner¹¹⁾. Steiner¹²⁾ found that under particular conditions of low longitudinal magnetic field and high longitudinal current a cylindrical superconductor in the intermediate state will exhibit extra longitudinal magnetic flux. Meissner et al. extended this work. They correlated Steiner's data and showed that the extra flux was due to an anisotropic resistance in the intermediate state causing the

current to flow in a solenoidal path. They proved that the apparent paramagnetism was not a volume magnetization. They also provided a name: the paramagnetic effect in superconductors.

Subsequent measurements made at Rice under equilibrium conditions showed^{13, 14)} that the extra magnetic flux existed for fixed values of H , I , and T . The transition was found to be reversible, and the effect was the same for current and field antiparallel as well as parallel. The flux increase occurred whatever variable (H , I , or T) was used to effect the transition.

The present investigation was initiated to observe the static and dynamic properties of the apparent paramagnetism. Later the scope of the study was broadened to include attempts to verify theories proposed by C. Kittel¹⁵⁾ and H. Meissner¹⁶⁾. Specifically: (1) the longitudinal magnetization of cylinders of tin, indium, and thallium was measured for constant values of H , I , and T ; (2) an upper limit was placed on the relaxation time for the decay or rise of the extra flux; and (3) the correlation between the resistance drop and the paramagnetic flux increase was determined for two indium specimens. Emphasis was placed on equilibrium measurements since the earlier determinations had been made by dynamic methods and these techniques led to doubts^{17, 18)} that the effect really existed under equilibrium conditions.

It was hoped that this work would lead to an unambiguous picture of the intermediate state and of the mechanism causing the flux increase. Qualitative verifications of the theories for the paramagnetism have been obtained. Quantitative conclusions about the structure of the intermediate state are still impossible.

BOUNDARY SURFACE BETWEEN NORMAL
AND SUPERCONDUCTING PHASES FOR IN

FIGURE 1

II Experimental

In the following paragraphs the samples, experimental apparatus, and measuring techniques will be described. These details are needed so that the results to be given in a later section will be more meaningful and the analysis in section III will be of more significance.

A. Apparatus and Measuring Techniques

The paramagnetic effect was investigated in three metals: tin, indium, and thallium. Indium received the lion's share of attention only because it was the least troublesome to handle. The samples were all prepared in approximately the same manner, the only differences in treatment were due to the differing properties of the metals. Each solid cylindrical specimen was grown from pure metal (better than 99.9%) in a clean glass tube in vacuo. The metal was first heated considerably (100 C degrees) above its melting point and outgassed. The furnace was then slowly lifted (6 cm/hr) from about the sample. This caused solidification to progress slowly from bottom to top in a manner designed to produce single crystals. Two of the samples, where lead was the reference conductor (see below), were grown directly on the lead. The lead cylinder was first grown in the usual manner, then was slipped from the glass envelope and one end tinned with 50-50 Sn-Pb solder. It was returned to the envelope with a precast indium cylinder touching the tinned end. The

furnace was turned on, the indium, but not the lead, melted and recrystallized in the usual manner. A good clean uniform joint was thereby obtained. After cooling, the glass was removed by dissolving it in HF in the case of indium and thallium and by merely slipping the sample out of the tube in the case of tin. Subsequent etching (with HF for Sn and In; HNO_3 for Tl) showed the samples to be composed of a few large crystals with several smaller crystals near the ends. Thus the measurements were made on essentially monocrystalline specimens. In addition, samples In III and In V were vacuum annealed for several days after mounting. Table I gives the various physical dimensions of the specimens.

Table I

Sample	Diameter (mm)	Length (mm)
Sn	6.15	72
In I	6.25	70
In III	5.83	70
In IV	6.05	70
In V	5.69	80
Tl	6.91	70

The flux measuring system was based upon a technique described by Mendelsohn, Squire, and Teasdale¹⁸⁾. The method involved a comparison of the flux content of the sample with the known flux content of some material under the same conditions as the sample. A schematic diagram is given in Figure 2. The longitudinal flux content of the cylindrical

specimen (A) was measured by quickly moving a concentric detection coil (B) from the sample to a conductor (C) of known flux content and observing the deflection of a ballistic galvanometer (G) connected to the coil. Two types of reference conductors were used. In most cases the reference was a normal conductor (copper) whose small diamagnetism was negligible relative to the large paramagnetic effect (or superconducting diamagnetism). For samples In IV and In V the reference was a cylinder of lead. The lead remained superconducting throughout the experiment and provided a reference of zero flux content and zero resistance.

The procedure for observing a transition from the super to normal state will now be described. The sample was initially established in the superconducting state (with zero frozen flux) and in a specified external magnetic field, H_{20} , less than the critical field appropriate to the temperature. Then a small current was turned on and the flux content of the sample (i.e., galvanometer deflection) measured for various steady currents in the superconducting, intermediate, and normal states (see either curve of Figure 4). Potentiometer measurement of the current permitted high accuracy; it was found that a precision of 0.1 amp was adequate to obtain a smooth function as in Figure 4. Transitions were made for various combinations of H , I , and T for each of the six speci-

mens.

A second aspect of the equilibrium measurements was a determination of the resistance of the specimen concurrent with the flux determination. Preliminary measurements on an extremely pure crystal of indium indicated that the normal state resistance was too low to be easily observable by usual techniques. Especially so since the quantity of interest was the variation of resistance from the zero superconducting state value to the normal state value. Therefore rather sophisticated techniques were devised. Later measurements on a slightly less perfect specimen (In V) showed that direct measurement was possible. Both techniques were used.

The indirect resistance measuring system, also shown schematically in Figure 2, was based on a similar scheme used by Meissner and Doll¹⁹⁾ to measure the resistance change at the superconducting transition. The device consisted essentially of a two-branch parallel network; one branch (A-C) was the sample whose resistance was of interest (the reference conductor was Pb with zero resistance), the other (D) was a fixed resistor. The branch with fixed non-zero resistance contained in it a coil which produced a magnetic field when a current flowed. If the total current and the current in branch D are known then the current in branch A is easily computed. Since the voltage drop across the two branches is the same, one can obtain a quantity proportional to the sample

resistance, R_A :

$$R_A = R_D I_D / I_A \ll I_D / I_A$$

If either R_A or R_D is known then the value of the other is known.

The problem is twofold. First, the normal state resistance of the sample, R_{A0} , is small, of the order of 10^{-6} ohms; thus if an appreciable fraction of the current is to flow through R_D , R_D must also be of the same order of magnitude. In the system used R_D was composed of a (superconducting) tantalum wire, of diameter 0.005 inches, in series with a copper strip 5 mm wide, 15 mm long, and 0.1 mm thick. The copper strip was the fixed resistance R_D . The tantalum wire was wound in a coil of about 15 cm length and 1 mm diameter, the pitch was 50 turns/cm, and the coil produced about 60 gauss/amp.

Second, since the whole apparatus must be placed in the helium bath (to keep resistances low) conventional current measuring devices were not applicable. This problem was solved by observing the magnetic field set up by the current in the tantalum coil. The magnetic field set up by the coil was detected and measured by using a 0.010 inch diameter bismuth wire down the center of the coil (following a suggestion by H. E. Rorschach). The resistance of bismuth at helium temperatures is strongly dependent on the magnetic field applied to the bismuth²⁰⁾. Thus measurement of the resistance of the bismuth and its vari-

ation with current in the tantalum coil afforded a measure of the resistance of the indium sample. By using a large number of turns in the tantalum coil a considerable amplification was obtained, in that the bismuth resistance changes were large (about 0.15 ohm). Thus resistances greater than 5% R_{A0} could be measured as the transition between normal and superconduction progressed. The variation of bismuth resistance with current in the tantalum coil is shown in Figure 3. The sensitivity at low currents is low, since the variation of bismuth resistance with field is quadratic in that region.

The resistance of the bismuth was measured with a type K-2 Leeds and Northrup potentiometer. With 1 ma measuring current the voltage drop was of the order of 1 mv, the precision of the galvanometer null indicator and the type K (used always on its lowest scale) was such that potentials were known to ± 0.0000003 volt; this gives the bismuth resistance to one part in 4000. However, 0.1 amp in the tantalum produced an initial change of only about one part in 2000, hence the initial precision in R_A quoted above. For larger currents in the tantalum, a change of 0.1 amp produced a change of one part in 200 in the bismuth and a concomitant increase in the precision of determination of R_A .

The type K potentiometer was also used for the direct resistance determination. Potential leads were at-

tached to the sample at the same points as the tantalum leads had been attached. In the direct measurement, potential drops of from 1.0 to 40.0 microvolts were observed, the precision was again ± 0.3 microvolt. Thermal and contact potentials remained constant throughout a series of transitions, and thus introduced no error.

The measuring cycle was the same as for the flux measurements, in fact the two were carried out concurrently. Sufficient time was allowed at each step for the heat flush, caused by the dropping of the coil to die away before any resistance measurements were made.

The final set of measurements was essentially different from the foregoing in that a dynamic quantity was under scrutiny. Consider Figure 4. The curves labeled H' and H'' are the transitions one obtains at two slightly different external fields, H' and H'' . It is apparent that the flux content at a given current is quite different, for example, there is more flux in the sample at the low field H' at 11 amp than at the higher field at the same current. One might inquire: if the field were quickly increased from H' to H'' , would the galvanometer deflection obtained when the change occurs ever indicate a flux decrease in the sample? Experiments were carried out to ascertain the behavior of the flux changes with such field changes. First, two transition curves (such as Figure 4) were taken at some fixed temperature. Then the current

was set at some arbitrary value (less than 20 amps), the external field switched from H' to H'' , and the galvanometer deflection recorded (called initial deflection below). About 7 seconds after the field was changed the coil was lifted from the sample to the copper and the flux measured. If the flux content did not correspond to curve H'' , this flux measurement was repeated until the proper value was reached. Next the process was reversed, the field quickly changed from H'' to H' and flux measured at field H' until equilibrium was again reached. This procedure was repeated at various currents, yielding the flux change in the coil when an external field change occurs. The net change in the sample alone may be determined by correcting for: the leakage flux in the superconducting state due to the gap between coil and sample; and the frozen flux which may occur if a field decrease puts the sample in the superconducting state.

An over-all view of the apparatus is given in Figure 5. The sample is at SN and the copper (or Pb) rod is at CU. Silk threads (W) linked the flip coil (C) to an iron slug (I) which could be moved with a strong solenoid (not shown) placed outside the gas-tight envelope housing the iron slug. The exact position of the coil was controlled by the stops (L).

The direct current leads to the specimen are shown in Figure 5 entering through gas-tight seals and then spir-

aling around a vessel which contained liquid nitrogen. Below that point and extending on down into the liquid helium dewar the current carrying wires were of smaller diameter, so that neither heat leaks nor Joule heating effects were excessive. The current could be sent through the specimen in either direction by a reversing switch in the power supply (large Edison cells). It will be noted in the figure that the return lead is by the concentric copper shell (R) which was slotted so that liquid helium was always in direct contact with the specimen. Not shown in the figure are the external nitrogen dewar and the external solenoid for producing small homogeneous fields (the magnet calibration was 19.87×10^2 amp-turn/meter per ampere) whose direction was along the specimen cylinder. The external magnetic field could also be reversed in direction. Also omitted from the diagram is the resistance measuring system (see Figure 2).

Calibration of the meters used gave current accuracy of ± 0.02 amp, and magnetic field accuracy of ± 4 amp/meter. The horizontal component of the earth's magnetic field was cancelled by a pair of Helmholtz coils.

The temperature was determined from the vapor pressure of the helium bath using the 1949 agreed temperature scale, and was regulated to ± 0.001 K deg near the tin and indium critical temperatures, and within ± 0.002 K deg for the thallium data.

FIGURE 2

FIGURE 3

TYPICAL TRANSITIONS AT CONSTANT TEMPERATURE AND FIELD

$$\tilde{K}_m = 1 - (\delta_m / \delta_0) = 1 + (4.9 / 1.9)$$

$$= 3.6$$

FIGURE 4

FIGURE 5 APPARATUS

The results obtained with the procedure outlined above are given in the next paragraphs, and an interpretation and discussion is given in section III.

B. Results

The determinations of flux content can be correlated most succinctly by defining an "apparent relative permeability", first introduced by Meissner et al.¹¹). If $B = \mu_0 H$ in free space, and $B = \mu H$ in a magnetic medium, then one can define a dimensionless relative permeability by $K = \mu / \mu_0$. Then $B = K \mu_0 H$, with $K = 0$ in a perfectly diamagnetic medium (superconductor) and $K = 1$ in a non-magnetic medium. In this work the symbol $\tilde{K}$ will be used to emphasize that the permeability discussed is due to a circular current component and not to some basically atomic magnetism. Let $\mathcal{J}$ be the galvanometer deflection when the coil is moved from the sample to the copper, and let $\mathcal{J}_0$ be the deflection when the sample is in the pure superconducting state; then $\tilde{K}$ is given by:

$$\tilde{K} = 1 - (\mathcal{J} / \mathcal{J}_0) \quad (1)$$

and it is a function of the current and field at which $\mathcal{J}$ is obtained. Thus $\tilde{K}$ is zero in the superconducting state ($\mathcal{J} = \mathcal{J}_0$), unity in the normal state, and greater than unity in the paramagnetic region. See Figure 4 for a sample computation. Special notice will be given to the maximum value, $\tilde{K}_m$.

The magnitude of $\tilde{K}$ for each metal was determined

at various fields, temperatures, and currents. The magnitude of the maximum paramagnetic permeability, $\tilde{K}_m$, was found to increase linearly with current for a given external field, once some threshold current dependent on the field had been exceeded. The curves of $\tilde{K}_m$ versus current using different constant external fields are shown in Figures 6, 7, and 8 for two different indium specimens and thallium, respectively. The lines drawn through the points were fitted by the method of least squares. We have found similar curves for tin¹³⁾. Each point corresponds to a different temperature and external field combination where the total field equals the critical field.

The intercept of the line of $\tilde{K}_m$ versus current for a given field yields the threshold current characteristic of that field. These current intercepts plotted against their associated fields form the threshold curves of Figures 9, 10, and 11. The considerable scatter is due to the derived nature of the quantities involved. Following Meissner¹¹⁾, one may represent the threshold curve by:

$$I_0 = I_g + \pi \gamma d H_{z0} \quad (2)$$

where d is the sample diameter and H_{z0} the external field (all in mks units). The factor π is included in the second term to facilitate later discussion. I_g is the threshold in zero field, which is obtained by extrapolation since some field is necessary for observation, and is a constant of the material. Presumably²¹⁾ γ also depends

upon the metal used. In a field H_{z0} , paramagnetism is observed if the transition occurs for currents greater than I_0 . Our experimental values of I_g and γ are given in Table II, and also values reported by other workers. In the case of thallium there exists a sharp discrepancy be-

Table II

Specimen	I_g (amp)	γ (dimensionless)	Reference
tin	1.6 ± 0.7	0.82 ± 0.2	present work
	1.2	0.68	(11)
	1.2	0.92	(22)
indium I	0.33 ± 0.15	0.83 ± 0.04	present work
	III 0.37 ± 0.15	0.83 ± 0.04	present work
	0.6	0.68	(11)
thallium	0.9 ± 0.3	0.84 ± 0.04	present work
	0.6	0.36	(12), (11)

tween the present results and those quoted by Meissner et al. from Steiner's data (Meissner did not experiment with Tl rather re-interpreted Steiner's data to calculate γ). Our work indicates that thallium is more like the other superconductors.

We carefully ascertained that the maximum flux content, i.e., $\tilde{K}_m$, exists when the total field due to the current and to the external solenoid equals the critical field. Thus at the maximum one can write:

$$H_c^2 = H_{z0}^2 + (I/2\pi a)^2 \quad (3)$$

where a is the specimen radius and H_{z0} is the external field. Equation (3) states that the maximum flux occurs

just at the critical field as defined by the Silsbee hypothesis²³⁾. Since:

$$H_c^2 = H_0^2 [1 - (T/T_c)^2]^2, \quad (4)$$

at a given temperature and external field there is only one I satisfying equation (2). Hence the paramagnetic effect is observed only on that portion of the surface defined by (3) and (4) (see Figure 1) bounded by the planes $H = 0$, $T = 0$, and $I_0 = I_g + \pi \chi_d H_{z0}$. Equation (3) forms the experimental basis for the theories subsequently developed.

As mentioned above, resistance measurements were made on two indium specimens. For the work with bismuth, a typical transition is shown in Figure 12. By means of the calibration (Figure 3) this could be reduced to the usual form of flux and resistance as a function of the current in the specimen (instead of total current as in Figure 12). The result of the conversion for Figure 12 is shown in Figure 13. The discontinuous rise at the same current as the paramagnetic peak was characteristic of all the data taken with the parallel tantalum coil. Direct measurements of the resistance had greater sensitivity at the low end of the resistance curve, and failed to show the discontinuous rise. Figure 14 shows a typical transition, again correlated with the flux change. The residual resistance in the normal state, R_{A0} , was found to be 2.48×10^{-6} ohm at 3.4

deg K. Such a resistance yields a conductivity of 1.2×10^9 mho/meter for this sample. A low residual conductivity implies that the sample (In V) was not as good a crystal as was hoped. Other workers report values as high as 2×10^{10} mho/meter and specimen In III was known to have a residual resistance less than 10^{-7} ohm. It has been known for some time²⁴⁾ that the imperfections which cause a high residual electrical resistance in the normal state have no effect on the superconducting state. However, they imply the existence of imperfections in the crystal structure which might influence the behavior in the intermediate state.

The solid lines in Figures 13 and 14 are theoretical and the circles are the experimental points. The theory is derived in Appendices S and D, and discussed in section III.

Let us now examine the results of the relaxation time studies which we indicated on page 12. The result of the flux change measurements with thallium is given in Figures 15 and 16. Figure 15 shows the total flux change in the coil when the external field was changed from H' to H'' (see Figure 4). In the superconducting state the flux change (and flux) is zero, and in the normal state the flux change is simply due to the external field change. Hence to correct for leakage flux one uses the dotted line shown in Figure 15 as a zero and measures deflections from it. The corrected data are shown by the crosses in Figure 16. By referring to the original transition curves in Figure 4

one can predict what the flux change should be for any current. Simply subtract the galvanometer deflections obtained by moving the coil in the constant fields H' and H'' . Calibration reveals that a deflection of 1 cm corresponds to a flux change of 5.4×10^{-9} weber. Here one must remember the fact that the deflection obtained in the superconducting state depends on the external field even though the induction in the sample is zero. For this reason one adds a constant amount to each difference obtained by subtraction. The solid line of Figure 16 shows the predicted flux change (i.e., deflection). A positive sign indicates that the flux change in the sample went in the same direction as the field change. A flux increase in the sample was found when the external field was decreased in the neighborhood of 11 amps.

Similar results were obtained on the tin specimen and In III sample. On the basis that the initial galvanometer deflection followed the prediction, the relaxation time was estimated to be much less than the period of the galvanometer (5 sec), perhaps as low as 0.1 sec.

For one imperfect indium specimen (In I), the time variation of the flux content after the field was changed indicated an exponential decay (or increase) of the form $\exp(t/40)$, t in seconds. The points were taken over several transition sequences and indicate that the long delay was reproducible. Another sample (In III) was grown and

particular pains taken to insure its chemical and physical purity; it was also annealed after mounting. For that crystal the relaxation time was short as in the cases of the original tin and thallium crystals. Both indium samples gave the same results for the magnitude of the paramagnetism at a given current and field. The long relaxation time for the imperfect crystal was probably due to flux trapping by grain boundaries similar to that which occurs in frozen flux experiments²⁵⁾.

MAGNITUDE OF PEAK PARAMAGNETIC PERMEABILITY AS A FUNCTION OF CURRENT AND FIELD

FIGURE 6

MAGNITUDE OF PEAK PARAMAGNETIC PERMEABILITY AS A FUNCTION OF CURRENT AND FIELD

FIGURE 7

MAGNITUDE OF PEAK PARAMAGNETIC PERMEABILITY AS A FUNCTION OF CURRENT AND FIELD

FIGURE 8

THRESHOLD FOR THE PARAMAGNETIC EFFECT

FIGURE 9

FIGURE 10

THRESHOLD FOR THE PARAMAGNETIC EFFECT

FIGURE 11

RAW DATA FOR FLUX AND RESISTANCE
VARIATION AT THE
SUPERCONDUCTING TRANSITION

FIGURE 12

FLUX AND RESISTANCE AT THE SUPERCONDUCTING TRANSITION

FIGURE 13

FLUX AND RESISTANCE AT THE SUPERCONDUCTING TRANSITION

FIGURE 14

III Discussion

The results given in the preceding section enable one to construct a reasonable picture of the mechanism responsible for the flux increase. The purpose of this section is to assemble the arguments for and against various interpretations and to provide the background for the model to be finally presented.

When the specimen is in the superconducting state its properties are well known⁷⁾. If a small magnetic field is applied, the flux will not penetrate since a superconductor is diamagnetic. One says that non-dissipative supercurrents flow on the surface in such a manner as to create a magnetic field which just cancels the external field and maintains zero induction in the specimen. If a small electric current is passed through the specimen no potential drop can be observed since a superconductor has no resistance. Also the externally sustained current flows on the surface, for otherwise there would be a non-zero magnetic field inside the specimen. In the intermediate state we have quite a different situation. We find that the external field penetrates and that the circular currents have reversed themselves so that the longitudinal flux exceeds that flux due to the applied magnetic field; however, the current flow is no longer free from dissipation. We also know what happens if the fields and currents far exceed the critical values, for the specimen then has only its normal,

non-superconductive, properties. This discussion will concentrate on the intermediate state, where the behavior is the one bearing on our experiments.

We have several broad experimental bases to build the theory upon. First, there is certainly extra flux in the specimen at the transition, and the flux exists under equilibrium conditions. Second, the extra flux extends over a considerable range of currents when a transition is made at constant field and temperature. Third, there is some resistance at the point at which the maximum flux occurs, though the normal state resistance does not appear until the current far exceeds the critical current. Fourth, we know that the maximum flux occurs just at the critical field. That is, when $\bar{K} = \bar{K}_m$, one can write:

$$H_s^2 + H_{z0}^2 = H_c^2 \quad (1)$$

where H_s is the surface value of the azimuthal field due to the longitudinal current, H_{z0} the applied field, and H_c the critical field. And finally we know that extra flux is observed only if the current at the transition exceeds some threshold current. Let us begin by understanding the mechanism for the extra flux.

Theoretical description of the paramagnetic effect has been attempted by C. Kittel¹⁵⁾, H. Meissner¹⁶⁾, and C. J. Gorter²⁶⁾. None of the theories is complete since only a portion of the experimental data can be explained.

C. J. Gorter's proposed theory for the flux increase is given first. He assumes a domain structure much like that of Kittel (see below), with the major difference that Gorter's domains are in motion instead of static. He envisions alternate concentric tubes of superconducting and normal material appearing at the surface and moving inward. Each superconducting tube carries a certain amount of trapped flux in the adjacent normal region. The longitudinal trapped flux builds up at the center until it reaches the magnitude of the critical field. Thus at the peak of paramagnetism one has two general regions: a core of normal metal with a longitudinal field equal to the critical field in it, and a sheath of alternate tubes of normal and superconductive material in a constant state of motion toward the center. A given tube moves toward the center until it reaches the core, then disappears (leaving its load of flux behind) and simultaneously another tube appears at the surface. No calculations of observed quantities based on this model have been published.

Kittel's theory (see details in Appendix B) deals only with the peak of the paramagnetism, and attempts to describe the slow drop off on the normal state side of the transition have failed. The general idea is that a circular system of currents must be set up in order to satisfy boundary conditions in the intermediate state, and the magnetic field of these currents simulates

a paramagnetic moment. Kittel supposes that the domain structure of the intermediate state consists of a large number of thin coaxial tubes, alternately of normal and of superconductive metal. It is argued that a reasonable assumption is that on each boundary surface the total magnetic field must be equal to the critical field, H_c , appropriate to the temperature. Then if H_ϕ denotes the azimuthal field due to the longitudinal current, and H_z is the total longitudinal magnetic field, at each boundary he requires:

$$H_\phi^2 + H_z^2 = H_c^2 \quad (2)$$

If one assumes next that the circular field has the same form in the intermediate state as in the normal, then H_ϕ falls off inside the cylinder, and to match H_ϕ to H_c at every point an extra longitudinal field, H_z , is built up. H_z is produced by a circular concentric current system which is set up spontaneously to satisfy the boundary conditions. The model accounts in a natural way for the observation by Meissner¹¹⁾ that the paramagnetic effect occurs in a hollow cylinder, but disappears if a slit is made through one side of the tube, thereby interrupting the path of the circular currents. However, no explanation of the threshold is indicated.

A most ambitious theoretical study has been made by Hans Meissner at The Johns Hopkins University. He pos-

tulates that the intermediate state consists of needle shaped superconducting domains embedded in a matrix of normal metal. The thermodynamic arguments quoted earlier imply that the grains will be oriented parallel to the magnetic field²⁷⁾. Meissner is able to derive a pair of differential equations describing the longitudinal and azimuthal fields. The solution of these equations permits straightforward computation of the effective permeability as a function of the applied field and the current. Meissner imposes the condition above, equation (2), on the average field throughout the specimen. The theory of the effect contains as a parameter the ratio of the effective length, l , to the effective diameter, a , of the superconductive particles less one, i. e., $C = (l/a) - 1$. In the limit of long thin particles, $C \rightarrow \infty$, the result is the same as Kittel's, for smaller values of the parameter, the magnitude of the paramagnetism is reduced and a threshold appears. The description is extended²⁸⁾ into the region on the normal state side of the maximum by invoking the London²⁹⁾ picture of a normal sheath surrounding a core of metal in the intermediate state. A more complete exposition of this theory is to be found in Appendix C, but we quote here the main features.

In the limit of long thin domains of superconductor, Meissner's result is:

$$\chi_m = (2/3) \mu_0^{-2} \left[(1 + \mu_0^2)^{3/2} - 1 \right] \quad (3)$$

where ϕ_0 is the ratio of the azimuthal field at the surface to the longitudinal applied field. For currents such that the net field at the surface exceeds the critical field, the observed permeability is given by:

$$\bar{K} = 1 + (\bar{K}_m - 1) \left[2(I/I_c)^2 - 1 - 2(I/I_c) \sqrt{(I/I_c)^2 - 1} \right] \quad (4)$$

when the transition is effected with fixed longitudinal field and temperature. I is the current, I_c being the value derived from equation (1) of this section.

Some immediate consequences of the expression for maximum effective permeability (equation 3) are: (i) since dependence is on ϕ_0^2 , the effect is the same for current and field parallel or antiparallel; (ii) for ϕ_0 greater than about two, and at fixed external field, the magnitude of the peak is linearly dependent on the current; (iii) there is no threshold; (iv) the same curve holds for all superconducting metals, unless there is some variation in the length-diameter parameter C , between different materials. Predictions i, ii, and iv are borne out rather well by experiment. However, as has been mentioned, a threshold is characteristic of all the data.

Figure 17 shows a comparison of the data obtained with the theoretical prediction from equation (3). The agreement of theory and experiment is satisfactory for ϕ_0 greater than 1.5. However, for lower values the data drop below the curve and indicate a "threshold" at about 0.8.

This is not quite the threshold of equation (2) of section II. For, if paramagnetism is observed for κ greater than 0.8, then the threshold current, I_0' , for a given field is:

$$I_0' = 0.8(2\pi a) H_{z0} = \pi (0.8)d H_{z0} \quad (5)$$

Thus no " I_g " is implied. This value of the parameter κ compares rather well with the estimate from the empirical threshold equation (see Table II).

One might try to adjust the Meissner theory by using a finite value of C . Since the differential equations for the fields are non-linear and have no closed solutions (except for infinite C), the equations for finite C analogous to the above are based on approximate solutions. Numerical integration of the differential equations reveals that the approximations are reasonable in one of two limits: either small κ and any C , or large C and any κ . (Comparison of the calculations is given in the appendix.) Considerable numerical computation would be required for finite values of C and has not been completely done. A value of $C = 10$ would give a threshold of approximately 0.8; however, the upper portion of the curve would be displaced downward and remove the agreement found there (e.g., for $C = 10$ and $\kappa = 2.5$, $\bar{K}_m = 1.32$, instead of 2.0 given by equation (3)). Thus adjustment of C does not appear to be the solution to the threshold problem.

For comparison of the data with theory, Meissner¹⁶⁾

replaces μ with a variable β which he defines by:

$$\beta = \mu \left[1 - (I_g/I) \right] \quad (6)$$

This amounts to a varying constriction of the abscissa and provides reasonable agreement and a threshold. But there is no theoretical justification for the substitution.

We believe the threshold which one observes (equation 2 of section II) to be due to the presence of superconducting grains in the intermediate state. The diamagnetism of these grains reduces the extra flux observed. Thus when the extra longitudinal field is small, it is overshadowed by the diamagnetism of the superconducting domains. Extra flux is observed only when the paramagnetic term in the permeability exceeds the diamagnetic; that is when the superconducting volume becomes small. The average longitudinal field in the specimen at the critical field, may be expressed by:

$$B = \mu_0 [(1-\alpha)K_m' - \alpha] H_{z0}$$

where α is the volume fraction of superconducting material, and K_m' is the paramagnetic permeability due to the circular component of the externally sustained current. The paramagnetic effect is observed when the expression in the square brackets exceeds unity. Using the experimentally observed threshold, and the theoretical K_m from equation (3) for K_m' , one arrives at a value of 0.07 for α . In the form of Kittel's and Meissner's theories leading to equation (3) the superconducting volume is neglected as being

essentially zero. Our experimental value of α is at least reasonable. This provides a source for the coefficient of the H_{20} term in the threshold equation, but says nothing about I_g .

Meissner and his students find values of I_g for mercury¹¹⁾ and NbN³⁰⁾ to be 1.7 and 2.35 respectively. Comparison of these values with Meissner's values for In, Tl and Sn (Table II) reveals that they stand in the ratio of 1:2:3:4 to each other. The values reported here show no such simple relation, but precision is such that agreement with Meissner is within experimental error. No explanation of I_g , much less the apparent relationship, has been proposed. Any complete theory for the paramagnetic effect must include a derivation of the threshold equation, especially I_g .

The theoretical description of the flux behavior for currents greater than I_c (Appendix D) has had mixed success. Figure 13 shows a typical case where the flux matches at the peak but drops off much faster than predicted. For some transitions, the theory and experiment agree rather well as shown in Figure 18. Still a third comparison is shown in Figure 19. The open circles are the actual data; the solid circles are obtained by matching the data to theory at the maximum and adjusting all other points by the same fraction. In this latter case the value of $\tilde{K}_m$ was apparently wrong but the form of the curve

was correct.

So far as has been ascertained, there are no consistent differences in procedure or environment between the three types of behavior exemplified by Figures 13, 18, and 19. One might suspect that heating would cause the flux to decrease more rapidly since I/I_c would be effectively increased. Were this the case, the transitions at lower currents should agree better than those at higher currents. No such trend was observed. Another possible explanation is that strains or other impurities might distort the circular current path and thereby reduce the extra field. One would think though, that this would also decrease K_m , yet Figure 13 shows agreement at I_c and not at higher currents. Also no differences were discernible between the better and poorer specimens as to the rate of drop off. The most plausible inference is that the same mechanism that produces the threshold causes the flux to drop off more rapidly than one would anticipate on the basis of existing theory. Measurements reported by Meissner²⁸), who found excellent agreement when the experimental curve of K_m versus β (see equation 6) was used, support this view.

Further verification of the London model of the behavior for I greater than I_c is provided by the resistance measurements. Figure 14 shows excellent agreement once the steeply rising region at $I \approx I_c$ is passed. For the theoretical resistance curve in Figure 14, I_c was taken as the

current at which $\tilde{K}_m$ occurred. Since the normal state resistance was known, the computation of the experimental points was straightforward, and no other empirical fitting was used. For sample In IV the normal state resistance was undetermined since data was not taken far enough above the transition. Hence the point at which the rise deviated from the vertical was arbitrarily defined as $\frac{1}{2}R_0$. The agreement with theory is naturally sensitive to this choice.

It should also be stated that the choice of R_0 varied from transition to transition for In IV. In fact, in several cases of small ϕ_0 the data showed that a choice like the above was impossible since values of R/R_0 in excess of unity would thereby result. One concludes that for large fields and small currents the model may not be quite correct. However, for In V the agreement was independent of ϕ_0 over the range observed (1.0 to 9.0). This discrepancy remains unresolved.

An explanation of the differences in the initial resistance increase is in order. When the bismuth detection was used (see Figure 13) a parallel path was available to the current. Thus when the total current was slightly increased the increment would divide between the two branches according to the ratio of their resistances. However, at I_c the resistance of the indium is undefined (according to existing theory), it may assume any value between zero and half its normal state value. Thus the total

current can increase while the current in the sample remains constant. The IR drop is the same in each branch; in one branch the current increases and in the other the resistance increases. Thus one observes the intermediate steps in the discontinuous rise of the resistance. However, for sample In V there was no alternative path for the current. Thus one cannot observe intermediate values of R less than $\frac{1}{2}R_0$. The expected result would be to find a sudden jump from quite small resistances to about $\frac{1}{2}R_0$. Instead a steep rise over a narrow range of currents was found. The breadth of the transition is interpreted as an indication of the imperfection of the specimen, and not as the true intermediate state.

Finally we look again at the measurements of flux change. A comparison of the deflection obtained when the field was changed from one value to another and the expected deflection indicates that the flux change occurs just as predicted. This is true for all the samples tested, if the crystals were pure and homogeneous. The relaxation time of the currents responsible for the extra flux is estimated to be less than 0.1 second as a result of the experiments on good single crystals. The fast response time coupled with the reversible nature of the transition again emphasizes the dependence on current and field and not on history of the specimen. The response time in imperfect specimens is longer, one presumes that this is

because the grains of superconducting material are not as free to re-orient themselves to the new field when impurities are present.

In this dependence on crystal imperfection can be found one possible source of the difference between our χ for thallium and the value derived from Steiner's data. Steiner's data were taken by placing a coil about the specimen and observing the deflection of a ballistic galvanometer as the field was commuted. If the crystal was poor (as our In I) then the deflection in the paramagnetic region would be smaller than it should be since the process would not have time to set up or decay during the period of the galvanometer. One might suspect that the decay time would be proportional to $\frac{1}{K_m}$ since the amount of reorientation would increase with K_m . This would lead to a lower threshold and the low value of χ given by Steiner's data. Thallium is prone to form poor crystals since it has a phase transition about 100° below its melting point. Even though the metal crystallized from the melt as a single crystal, it would undergo a distortion at the phase change.

We will now attempt to draw a complete and consistent picture of the behavior of a solid cylinder of superconductive metal below its critical temperature. If no magnetic fields are extant and no currents flow, then the metal is superconducting. If a small magnetic field is turned on, it does not penetrate the specimen and we say

that the rod exhibits perfect diamagnetism. If a small current flows through the specimen we can measure no potential drop, and therefore say the rod has zero resistance. We also know that all of the current flows on the surface, else the magnetic field due to the current would penetrate in violation of the perfect diamagnetism. Thus $B = R = 0$ is characteristic of the pure superconducting state.

As we increase the current the conditions described above persist until we reach the critical current characteristic of the temperature and field. At that point the sample enters the intermediate state. We are unable to say just how the transition from the superconducting state to the intermediate region takes place except that it must be abrupt in the absence of impurities. However, at the critical current, we will assume the domain structure postulated by Meissner to be set up. That is, long thin threads of superconductive material are embedded in a matrix of normal conductive material. In order to minimize the free energy of the specimen, the superconducting domains must be aligned parallel to the magnetic field. Since the rod is in the intermediate state, the magnetic field and current are allowed to penetrate, and we assume an almost uniform current density. The current distribution produces an azimuthal magnetic field whose magnitude at a given radius is directly proportional to that radius except near the surface. Thus the total magnetic field at

a given distance from the center of the specimen is helical. The pitch of the helix decreases as the distance from the axis of the rod decreases. This helical system of superconducting domains offers a low resistance path for the current since there is a maximum amount of superconducting path along the helix. The current therefore spirals as it traverses the rod and the circular component produces a longitudinal magnetic field.

Formally one postulates that the average total magnetic field throughout the specimen is the critical field. From this requirement, and the helical current path, we find that the longitudinal field must increase as one approaches the axis of the rod. Thus the longitudinal magnetic moment of the specimen exceeds that moment due to the external longitudinal magnetic field. The specimen thus appears to be paramagnetic. From the foregoing model one can calculate the magnitude of this paramagnetic moment as a function of current, field, and temperature. The experimental data corroborate this calculation.

We have thus far considered only the contribution of the zero resistance property of the superconducting domains. We must also include their diamagnetic susceptibility. The diamagnetic grains act to reduce the observed paramagnetic moment. The longitudinal paramagnetic moment must exceed the diamagnetic moment before a flux increase is observed. This implies a field-dependent threshold cur-

rent. Empirically one finds that about 5% of the specimen is superconducting at $\tilde{K}_m$.

The preceding description of the flux increase mechanism applies just at the critical current. Suppose we now increase the current slightly. For currents in excess of the critical current the field at the surface exceeds the critical field. Following London, we say that the outer layer of the cylinder enters the normal state, while the interior remains in the mixed state. The boundary is defined by the radius at which the total field is the critical field. Now only the inner core contributes to the paramagnetism since the sheath of normal metal is non-magnetic. However, some of the current flows in the normal layer so that a portion of the resistance is restored. The effective average permeability and the resistance of the specimen can be calculated. The calculations agree reasonably well with the experiments.

Eventually, the fields so far exceed the critical field that the specimen contains no superconducting domains and its behavior is completely normal. It has its normal (temperature dependent) resistance and the external magnetic field penetrates completely.

BEHAVIOR OF FLUX ABOVE PARAMAGNETIC PEAK

FIGURE 18

FIGURE 19

IV Conclusions

We have found that the so-called paramagnetic effect is an equilibrium property of the mixed region between the normal and superconducting states. Its magnitude is dependent on the current and magnetic field that exist at the transition. The maximum paramagnetic permeability occurs when the vector sum of the azimuthal and longitudinal fields (at the surface) is the critical field associated with the temperature of the specimen. The maximum occurs when the resistance ratio R/R_0 is equal to one half. We also found that there exists a field-dependent threshold current which must be exceeded ere the effect is observed.

As we have seen, the theory developed by H. Meissner is able to account for nearly all of the experimental observations. The use of the London model when the current slightly exceeds the critical current, enables one to include the resistance behavior and the long "tail" of the paramagnetism in the theoretical framework. Though the Meissner theory compares satisfactorily with the experimental facts taken as a whole, the approximations made for most calculations are too crude to permit adequate comparison of detailed predictions. Numerical computation of fields and the effective permeability for many values of the parameters is needed.

The term in the threshold equation dependent upon the external magnetic field is tentatively understood.

One would hope, however, that the derivation could be made less ad hoc. The quantity I_g is at present understood neither on a theoretical nor on an intuitive basis.

Both experiment and theory lack sufficient precision to permit quantitative conclusions about the effective length or diameter of the superconducting inclusions in the intermediate state.

V Appendix

A. Origin of the Intermediate State

The intermediate state was said to arise from geometrical effects. As an example of geometrical broadening of the transition, consider a superconducting wire carrying a current. When the current attains a value such that the critical field is reached at the surface of the wire, the surface might be expected to become normal with the interior of the wire remaining superconducting. If this were to occur, all of the current would flow in the superconducting interior; which would lead to an increase of the field at the normal-superconducting interface and more of the metal would become normal. Finally the current would be so constricted that the entire wire would be normal. A uniform current would result. This puts the field below the critical field in most of the metal and restores the structure first hypothesized. The process would begin again. Thus if an equilibrium situation is to exist (when the critical current $I_c = 2 \pi R H_c$ is reached) a fine grained mixture of normal and superconducting regions is required. (Large domains would lead to the same instability due to the demagnetizing effects of their shapes.) And the total field will be just the critical field at every interface and in the normal regions. For currents slightly above the critical current a normal sheath is created, but the current flows parti-

ally in the normal region. Thus some resistance is restored. London has worked out this model in some detail and the variation of resistance with current is shown in Figure 13 for one sample. A similar development is given in part C of this appendix for a wire carrying a current in the presence of an external longitudinal magnetic field. The result is the same except that I_c must be redefined to include the applied field. The intermediate state also occurs when the demagnetization factor of the sample is such that the magnetic field at an interior point differs from the applied field³¹⁾.

B. Kittel Theory

Kittel, in his theory for the paramagnetic effect, requires that the total field at every interface be the critical field. He assumes that the domains are concentric hollow tubes, alternately normal and superconducting.

Since:

$$H_c^2 = H_z^2 + H_\phi^2 \quad (1)$$

at each interface, and presumably in the normal tubes also, an effective longitudinal permeability, $\bar{K}$, can be defined by:

$$\bar{K} = \frac{2\pi}{\pi a^2 H_{z0}} \int_N [H_c^2 - H_\phi^2]^{1/2} r dr \quad (2)$$

where H_{z0} is the applied longitudinal magnetic field, a is the radius of the rod, and the integral extends over the normal portion, N , of the cross section. For the maximum,

$\tilde{K}_m$, he assumes: (1) the superconducting volume is negligible, the tubes increasing in number and decreasing in thickness, and (2) the current density is uniform. Then:

$$\tilde{K}_m = \frac{2}{\pi^2 H_{z0}} \int_0^2 \left[H_c^2 - (I_c r / 2\pi a^2)^2 \right]^{1/2} r dr \quad (2a)$$

where I_c is the current flowing at the maximum. For H_{φ} defined by:

$$H_{\varphi} = I_c / 2\pi a$$

and $\rho = r/a$ the integral becomes:

$$\begin{aligned} \tilde{K}_m &= \frac{2}{H_{z0}} \int_0^1 \left[H_c^2 - H_{\varphi}^2 \rho^2 \right]^{1/2} \rho d\rho \\ &= \frac{2}{3} \left(\frac{H_{z0}}{H_{\varphi}} \right)^2 \left[\left(\frac{H_{z0}^2 + H_{\varphi}^2}{H_{z0}^2} \right)^{3/2} - 1 \right] \end{aligned} \quad (3)$$

Now define $\varphi_0 = H_{\varphi} / H_{z0}$ and (3) becomes:

$$\tilde{K}_m = (2/3) \varphi_0^{-2} \left[(1 + \varphi_0^2)^{3/2} - 1 \right] \quad (3a)$$

This is the equation quoted in section III, and predicts the maximum apparent permeability observed in a transition as a function of current, field, and sample diameter; the temperature also enters through equation (1).

C. Meissner Theory

Meissner's theory for the paramagnetic flux increase (at the maximum only) is considerably more complex than Kittel's since Meissner makes an attempt to base it on a more accurate model of the intermediate state. Meissner assumes that the intermediate state is composed of ellipsoidal domains of superconducting material embedded in a

matrix of normal metal. Free energy arguments imply that the long axis of the superconducting domains will be parallel to the field. See, for instance, Progress in Low Temperature Physics I, Chapter IX. The total field at any point is composed of longitudinal components from the external field and circular components from the field due to the current. The net result is a spiral field. If the particles of superconductor orient themselves parallel to this field, the current will have a preferred path along the helical lines of force. A spiraling current acts like a solenoid to produce extra longitudinal flux.

In order to put these ideas into a mathematical form, consider a layer of the cylinder between r and $r + dr$. We will spread this layer out into a plane so that equations can be initially formulated in familiar rectangular coordinates. Choose the z -axis parallel to the original axis of the cylinder, and the y -axis parallel to the original φ direction. The local value of the magnetic field in the normal regions will always be H_c . We define the macroscopic magnetic induction, B , as:

$$\vec{B} = \mu_0 \xi_2 \vec{H} \quad (4)$$

where:

$$\xi_2 = d/(2+d) \quad (5)$$

We have chosen a/d as the ratio of the thickness of the superconducting particles to the spacing between the particles. We are led to this choice by consideration of the

fraction, ξ_2 , of the material which is in the normal state. H has the magnitude H_c , and the direction of the average of the local field; it will therefore make an angle α with the y -axis.

By a similar process, we find:

$$\begin{aligned} E_z &= E_y \sin \alpha + E_x \cos \alpha \\ E_y &= E_x \cos \alpha - E_z \sin \alpha \end{aligned} \quad (6)$$

where E_x and E_z are the electric fields parallel and perpendicular to B , respectively. In terms of the local fields e_x and e_z :

$$E_x = \xi_1 e_x ; \quad E_z = \xi_2 e_z \quad (7)$$

$$\xi_1 = d/(\ell + d) \quad (8)$$

where ℓ/d is the ratio of the length of the particles to the spacing between particles. In the normal regions, the conductivity is the usual normal state value, σ_n , hence:

$$J_y = \sigma_n e_y ; \quad J_z = \sigma_n e_z \quad (9)$$

The applied electric field, however, has only a z component. Hence, $E_y = 0$, and from (6) and (7) we find:

$$\begin{aligned} J_y &= (\sigma_1 - \sigma_2) \sin \alpha \cos \alpha E_z \\ J_z &= (\sigma_1 \sin^2 \alpha + \sigma_2 \cos^2 \alpha) E_z \end{aligned} \quad (10)$$

where

$$\sigma_1 = \sigma_n / \xi_1 ; \quad \sigma_2 = \sigma_n / \xi_2 \quad (11)$$

From equations (4), (5), (8), and (11) we have:

$$\frac{\kappa}{\sigma_2} - 1 = \left(\frac{\ell}{d} - 1 \right) \left(1 - \frac{B}{\mu_n H_c} \right) \equiv C \left(1 - \frac{B}{\mu_n H_c} \right) \quad (12)$$

Since $\sin \alpha = H_y / H_c$ and $\cos \alpha = H_z / H_c$ we can rewrite equation

(10), using equation (12), as:

$$J_y = \frac{\mu_0 H_c}{B} C \left(1 - \frac{B}{\mu_0 H_c} \right) \frac{H_y H_z}{H_c^2} \sigma_n E_z$$

$$J_z = \frac{\mu_0 H_c}{B} \left[C \left(1 - \frac{B}{\mu_0 H_c} \right) \frac{H_z^2}{H_c^2} + 1 \right] \sigma_n E_z \quad (13)$$

Now we return to cylindrical coordinates where $J_z \rightarrow J_z$ and $J_y \rightarrow J_r$. The magnitude of $\vec{H}$ is H_c , thus we can write:

$$H_c^2 = H_r^2 + H_z^2 \quad (14)$$

From the Maxwell relation: $\vec{\nabla} \times \vec{H} = \vec{J}$, we have

$$J_z = \frac{1}{r} \frac{\partial}{\partial r} (r H_r); \quad J_r = - \frac{\partial H_z}{\partial r} \quad (15)$$

Thus if we differentiate (14) with respect to r and use (15) to eliminate the derivatives, the result is:

$$H_z J_r / H_r + H_r / r = J_z \quad (16)$$

Substitution of (13) into (16), thereby eliminating the currents, yields:

$$B = \left(\mu_0 H_c \sigma_n E_z r / H_r \right) \quad (17)$$

For $r = a$ (i.e., at the surface), $B = \mu_0 H_c$ and $H_r = H_a$; therefore:

$$a = H_a / \sigma_n E_z \quad (18)$$

and:

$$B = \mu_0 H_c r H_r / (a H_r) \quad (19)$$

As in equation (2) we define the effective maximum permeability as:

$$\tilde{K}_m = \frac{2\pi}{\pi a^2 \mu_0 H_{20}} \int_0^a r B_z(r) dr$$

which by virtue of (19) becomes:

$$\tilde{K}_m = \frac{2}{a^2} \int_0^a \frac{H_z}{H_{20}} \frac{H_z}{H_y} \frac{r}{a} r^2 dr \quad (20)$$

In order to find H_z and H_y we must solve equations (15).

We rewrite them now using (13), (14), (18), and (19):

$$\frac{dH_z}{dr} = C \left(\frac{H_y}{r} - \frac{H_z}{a} \right) \left(1 - \frac{H_y^2}{H_c^2} \right) \quad (21)$$

$$-\frac{dH_y}{dr} = C \left[\frac{(H_c^2 - H_z^2)^{1/2}}{r} - \frac{H_y}{a} \right] \frac{H_z (H_c^2 - H_z^2)^{1/2}}{H_c^2} \quad (22)$$

For the rest of the theory Meissner assumes that C is a constant, giving as his reason that the superconducting particles always have the same field (H_0) at their surfaces. Before discussing these equations, let us rewrite them in terms of dimensionless variables:

$$\varphi = \frac{H_y}{H_{20}}; \quad \varphi_0 = \frac{H_{y0}}{H_{20}}; \quad \chi = \frac{H_z}{H_{20}}; \quad \rho = \frac{r}{a}; \quad \frac{H_c}{H_{20}} = (1 + \varphi_0^2)^{1/2} \quad (23)$$

so that:

$$\frac{d\varphi}{d\rho} = \varphi_0 C \left[\frac{\varphi}{\varphi_0 \rho} - 1 \right] \left[1 - \frac{\varphi^2}{1 + \varphi_0^2} \right] \quad (21a)$$

$$-\frac{d\chi}{d\rho} = C \left[\left(\frac{1 - \chi^2}{1 + \varphi_0^2} \right)^{1/2} \frac{1}{\rho} - \frac{\varphi_0}{(1 + \varphi_0^2)^{1/2}} \right] \left[1 - \frac{\chi^2}{1 + \varphi_0^2} \right] \chi \quad (22a)$$

$$1 + \varphi_0^2 = \chi^2 + \varphi^2 \quad (14a)$$

$$\tilde{K}_m = 2 \int_0^1 (\varphi_0/\varphi) \chi_{\rho^2} d\rho \quad (20a)$$

Only one of the two equations (21a), (22a) need be solved, the other quantity is found from (14a). Hence $\tilde{K}_m = \tilde{K}_m(\varphi_0)$ only. The boundary conditions are: $\rho = 0$, $\varphi = 0$; $\rho = 1$, $\varphi = \varphi_0$.

Solution of these equations exactly is impossible, and various approximations must be made before the theory can be compared with experiment. For $\varphi_0^2 \ll 1$ (since $\varphi \leq \varphi_0$) (21a) becomes:

$$d\varphi/d\rho = \varphi_0 C [(\varphi/\varphi_0 \rho) - 1] \quad (24)$$

with a solution:

$$\varphi = [\varphi_0/(C-1)] [C\rho - \rho^C] \quad (25)$$

This yields a rather complicated expression for $\tilde{K}_m$:

$$\tilde{K}_m \equiv 2 \int_0^1 [C\rho - \rho^C]^{-1} (C-1) [1 + \varphi_0^2 - (\varphi_0^2/(C-1)^2)(C\rho - \rho^C)^2]^{1/2} \rho^2 d\rho \quad (26)$$

An approximate solution, correct to terms of the order C^{-2} , is:

$$\tilde{K}_m = \frac{2}{3} \left(\frac{C-1}{C} \right)^3 \varphi_0^{-2} [(1 + \varphi_0^2)^{3/2} - 1] - \varphi_0^{-2} \frac{(C-1)^{C+2}}{C^{C+3}} \quad (27)$$

In this approximation $\tilde{K}_m \leq 1$, since the approximation breaks down before $\tilde{K}_m$ reaches unity! However, if C is infinite ($C \gg \varphi_0^{-2}$) then:

$$\tilde{K}_m = (2/3) \varphi_0^{-2} [(1 + \varphi_0^2)^{3/2} - 1] \approx 1 \quad ; \quad \varphi_0 \gg 0 \quad (28)$$

which is the form used in Figure 17.

In fact for $C \rightarrow \infty$, the differential equation (21a) implies that $\varphi = \varphi_0 \rho$ since $d\varphi/d\rho = 0$ at $\rho = 1$. This form of azimuthal field dependence ($\varphi = \varphi_0 \rho$) on current is the same used in Kittel's development and leads to equation (3) or (28) alike. This is the other approximation used for φ , i.e., for $C \rightarrow \infty$, $\tilde{K}_m$ is given by (28) for any $\varphi_0 \geq 0$.

Thus two alternatives are available, first to stay in the limit of small φ with any value of C allowed, or to go to large C and use any value of φ . For large C ($C > 50$), φ may be suitably approximated by:

$$\varphi = \varphi_0 \rho C / (C-1)$$

leading to

$$\tilde{K}_m = \frac{2}{3} \varphi_0 \left(\frac{C-1}{C} \right)^3 \left\{ \left[\frac{1+\varphi_0^2}{\varphi_0^2} \right]^{\frac{3}{2}} - \left[\frac{1+\varphi_0^2}{\varphi_0^2} - \left(\frac{C}{C-1} \right)^2 \right]^{\frac{3}{2}} \right\} \quad (29)$$

This is the form used for most calculations. It reveals that as C decreases, the permeability given by (28) is decreased so that a threshold appears (e.g., for $C \approx 21$, $\tilde{K}_m \leq 1$ for $\varphi_0 \leq 0.6$), and the permeability observed for a given φ is less. One notes that this approximation fails for large φ , i.e., if:

$$\frac{1+\varphi_0^2}{\varphi_0^2} < \left(\frac{C}{C-1} \right)^2$$

Strictly speaking for C finite the only satisfactory recourse is numerical integration for calculation of the reduced fields φ and χ , then Simpson's rule for $\tilde{K}_m$.

This tedious process has prevented adequate comparison of the Meissner theory with experiment.

D. Behavior for Currents Above the Critical Current

The preceding exposition of the Meissner theory for the maximum paramagnetic permeability was given to provide a background for the following remarks concerning the flux and resistance when the current slightly exceeds the critical current. As the current is increased above I_c , the extra flux does not disappear at once, rather it decreases slowly toward the normal state value (see Figure 4). If one assumes, following London, that a sheath of normal metal is set up about the core of intermediate state material, then the decrease of extra flux can be understood.

The radius of the intermediate state core, R , is defined by:

$$H_{20}^2 + (I_1/2\pi R)^2 = H_c^2 \quad (30)$$

where I_1 is the part of the current flowing in the core.

Thus $I = I_1 + I_2$, and I_2 is the current flowing in the normal sheath. Using (14) and (30), where (14) has been evaluated at $r=a$, one finds:

$$H_r = I_1/2\pi R = H_{rk} \quad \text{or} \quad I_1/R = I_c/a \quad (31)$$

For a normal state conductivity σ_n and an applied electric field E , I_2 is given by:

$$I_2 = 2\pi \int_R^a \sigma_n E r \, dr = \pi \sigma_n E (a^2 - R^2) \quad (32)$$

We now use one of Meissner's results, equation (18), where now the surface of the intermediate state is at $r = R$, thus:

$$R = H_{p_0} / \sigma_n E \quad (33)$$

To find R one combines (31) and (32):

$$I_1 = I_c (R/a) = I - I_2 = I - \pi \sigma_n E (a^2 - R^2)$$

which with (33) yields:

$$\begin{aligned} I_c (R/a) &= I - (\pi H_{p_0} / R) (a^2 - R^2) \\ &= I - (I_c / 2aR) (a^2 - R^2) \end{aligned}$$

This gives a quadratic expression for R :

$$R^2 - (2aI/I_c)R + a^2 = 0$$

with a solution:

$$R = a \left[\frac{I}{I_c} \pm \sqrt{\left(\frac{I}{I_c}\right)^2 - 1} \right] \quad (34)$$

Since $R \leq a$ for $I \geq I_c$ the negative sign is used. Knowing R , one can return to the definition of $\tilde{K}$ (equation (20) above) and note that for $r \geq R$, $H_z = H_{z0}$ and $H_\varphi = (H_{p_0} r/a)$, and for $r \leq R$ we use the Meissner result. From equation (31) the value of $\tilde{K}_m(I_1/R) = \tilde{K}_m(I_c/a)$. In terms of the effective permeability:

$$\begin{aligned} \tilde{K} &= \tilde{K}_m \frac{R^2}{a^2} + 1 \left(\frac{a^2 - R^2}{a^2} \right) \\ &= 1 + (\tilde{K}_m - 1) \frac{R^2}{a^2} \end{aligned} \quad (35)$$

and using the result (34):

$$\tilde{K} = 1 + (\tilde{K}_m - 1) \left[2(I/I_c)^2 - 1 - 2(I/I_c) \sqrt{(I/I_c)^2 - 1} \right] \quad (36)$$

This is the desired expression for the variation of $\tilde{K}$ with current. Note that it depends not only on the model for $\tilde{K}_m$, but also on the London model of a central core of metal in the intermediate state surrounded by a concentric sheath of

normal metal.

At this point a calculation of the resistance variation is simple. From (31) and (32) after R is eliminated by using equation (33):

$$I = I_1 + I_2 = (I_c/a)(H_p/\sigma_n E) + \pi \sigma_n E [a^2 - (H_p/\sigma_n E)^2] \quad (37)$$

$$= I_c^2 / (4\pi a^2 \sigma_n E) + \pi a^2 \sigma_n E$$

To simplify let $\Omega_n = (\pi a^2 \sigma_n)^{-1}$, which is the normal resistance per unit length. Then

$$I = I_c^2 / (4\Omega_n E) + E/\Omega_n \quad (37a)$$

Solution of the quadratic in E gives:

$$E = (\Omega_n I / 2) [1 \pm \sqrt{1 - (I_c/I)^2}] ; \quad I \geq I_c \quad (38)$$

From equation (33):

$$E = H_p / \sigma_n R \geq H_p / \sigma_n a = I_c / (2\pi a^2 \sigma_n) = I_c \Omega_n / 2$$

hence the positive sign is used before the radical in equation (38). The resistance may be defined by $\Omega = E/I$ and the final result is:

$$\Omega = (\Omega_n / 2) [1 + \sqrt{1 - (I_c/I)^2}] \quad (39)$$

This is exactly London's result except that I_c has been defined as: $I_c = 2\pi a (H_c^2 - H_{c0}^2)^{1/2}$, instead of $I_c = 2\pi a H_c$. London also assumed that $H_p = H_c$ in the intermediate state (for any r) but this result is obtained without that assumption. Calculations using a uniform current density, as Kittel does, have not been made since a choice of boundary conditions at $r=R$ (corresponding to equation (18) of Appendix C) is not indicated by the Kittel model. The behavior of resistance and flux for $I \geq I_c$ is dependent on this boundary condition.

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