FLUXOID CONSERVATION BY SUPERCONDUCTING THIN FILM RINGS

Thesis by

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ABSTRACT

A torque method for measuring the persistent current in superconducting rings has been used to investigate the conservation of the fluxoid originally predicted by F. London. The fluxoid through a superconducting ring is the action integral of the canonical angular momentum of the superconducting electrons taken around the ring. This is comprised of two parts, one describing the mechanical angular momentum of the electrons and the other describing the magnetic flux trapped by the ring. The mechanical angular momentum depends on the penetration depth (λ) and therefore on temperature. If the fluxoid is conserved, temperature variations should alter the balance between the mechanical and electromagnetic angular momenta. As a consequence, the amount of trapped flux, and hence the persistent current, should vary with temperature even though the ring remains at all times entirely within the pure superconducting state with zero resistance. Very thin films of tin have shown experimentally a decrease in persistent current with increasing temperature and an increase with decreasing temperature which agrees with that to be expected on the basis of the fluxoid conservation predicted by London.

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I. INTRODUCTION

A number of pure metals, at temperatures below a certain characteristic critical value, exhibit no electrical resistivity and almost perfect diamagnetism. In the absence of magnetic fields the transition of these metals into the " superconducting " state is of second order with a nearly discontinuous change in the specific heat.

A number of theories have been proposed to describe the electrodynamic and thermodynamic behavior of superconductors. Particularly valuable contributions were made by Gorter and Casimir in 1934 (1), by F. and H. London in 1935 (2); and later, in the early 1950's, by Ginzburg and Landau (3), by Fröhlich (4), and by Pippard (5). With the exception of Fröhlich's work these theories were of a phenomenological nature with no specific microscopic model as a basis. In 1948 F. London (6) pointed out that the success of certain of these phenomenological theories in interpreting the experimental data indicated some of the features to be expected in a successful microscopic quantum picture of the superconducting state. The first generally successful microscopic theory was formulated in 1957 by Bardeen, Cooper and Schrieffer (7) and is in accord with the outline which London had suggested. The BCS theory has now been greatly extended and has met with notable success.

The possibility of inducing persistent circulating currents in superconducting rings is one of the most intriguing and least understood consequences of the vanishing of electrical resistivity in superconductors. Such a persistent current loop possesses a magnetic moment and "traps" a magnetic flux equal to the product of the self-inductance of the ring and the persistent current. The current carrying state is by no means the ground state of the system but it is nevertheless extraordinarily stable. The earliest investigations of persistent currents were directed toward use of this stability as a means of establishing an upper limit on the possible resistivity of the superconducting state. This approach, with certain modern technical refinements, is still being used for this purpose. (8)

Much recent interest has centered on investigation of the suggestion first made by London, that the "fluxoid" or action integral of the canonical angular momentum of the superconducting electrons taken around the ring, should be both conserved and quantized. The fluxoid contains one term in the mechanical angular momentum of the electrons and one in the magnetic flux trapped by the ring. In typical experiments performed with superconducting rings, the mechanical angular momentum term constitutes only a very small perturbation on the much larger magnetic flux term. It is interesting to note that the situation in atoms is just the converse with the magnetic flux acting as the small perburbation (Zeeman effect). Several investigators (9, 10) using cylinders for which the angular momentum term would be expected to be negligible, have observed an apparent quantization of the magnetic flux trapped by these cylinders. These experiments have not yet shown directly the existence of quantum states with quantum number greater than about 5. Both the BCS and Schafroth (11, 12) boson theories as well as the London theory indicate that the <u>fluxoid</u> is the conserved quantity and each of these theories demands that variations in the ring temperature alter the portions attributable to the trapped flux and to the mechanical angular momentum of the electrons.

A consequence of such a redistribution, which will be discussed in more detail in part II, is that the trapped flux and hence the persistent current in a ring should vary with temperature even though the ring remains in the zero-resistance pure superconducting state at all times. None of the theories yet indicate the detailed mechanism by which this redistribution takes place, but each predicts within its own approximations specific changes in the flux as a function of temperature. The present investigation was undertaken to determine whether these consequences of the conservation of the fluxoid can be observed and, if so, to what extent the observations conform to the predictions of theory.

II. ANALYSIS AND DISCUSSION OF THE FLUXOID PROBLEM

In 1948 F. London (6) showed that according to his phenomenological theory of superconductivity, a superconducting ring should conserve the quantity which he called the fluxoid.

London noted further that the usual quantum mechanical assumption that the wave function of the superconducting charge carriers is single valued implies that the fluxoid is quantized. More recently several authors (12 - 16) have treated the problem of the flux trapped by a hollow superconducting cylinder using a variety of approaches to obtain the electron or electron-pair wave function. As London pointed out, however, the results concerning the flux depend principally on the assumption that the charge carrier wave function is single valued. These various approaches yield a result which also may be obtained merely by application of the Bohr-Sommerfeld quantum condition to the charge carriers. This is simply

$$\oint \underset{\sim}{p} \cdot d\underset{\sim}{\ell} \equiv \text{Fluxoid} = kh \,, \qquad (1)$$

where p is the canonical momentum of the charge carriers in a magnetic field, k is an integer and h is Planck's constant. This canonical momentum is given by

$$P = m^* \underset{\sim}{v} + e^* \underset{\sim}{A} \,, \qquad (2)$$

where m^* and e^* are the mass and charge of the charge carrying units, $\underset{\sim}{V}$ is the charge carrier drift velocity and $\underset{\sim}{A}$ is the magnetic vector potential. The BCS theory and the weight of the experimental evidence indicate that the charge carrying units are in fact electron pairs. If we now insert Eq. 2 into Eq. 1 we may write

$$\oint m^* \underset{\sim}{v} \cdot d\underset{\sim}{\ell} + e^* \oint \underset{\sim}{A} \cdot d\underset{\sim}{\ell} = kh , \qquad (3)$$

where the quantum number k denotes the number of nodes in the electron pair wave function. Noting that $\oint \underset{\sim}{A} \cdot d\underset{\sim}{\ell} = \varphi$, the magnetic flux trapped by the ring, and also that V is constant around the ring for a persistent current, we may evaluate the integrals, obtaining the relation

$$\frac{m^*}{e^*}(2\pi r)v + \varphi = \frac{kh}{e^*} , \qquad (4)$$

where r is the ring radius. Consideration of the parameters describing the superconducting system will now permit us to write Eq. 4 in a more useful form.

If we denote by n_{se} the effective number density of electron pairs, then the current density is given by $j = n_{se}e^*v$ and the characteristic penetration depth, λ, of the magnetic field parallel to the surface of a superconductor is given (17) by the relation

$$\lambda = \left[\frac{m^*}{\mu_o n_{se} e^{*2}}\right]^{1/2} . \qquad (5)$$

If we now consider specifically the case of cylindrical films thin compared to the penetration depth, then the current density will be essentially constant over the film thickness and the total current will be given by $I = w\delta j$. Here w is the axial length of the film cylinder and δ is the film thickness. The experiments verifying the assumption that j is uniform also over the ring length will be discussed in Appendix B. The magnetic flux (φ) trapped by the ring is related to the persistent current (I) by the expression $\varphi = LI$ where L is the self-inductance of the ring. The very thin films under discussion closely approximate a current sheet for which the inductance is given by the relation* (18)

$$L = \frac{\mu_o \pi r^2 K}{w} , \qquad (6)$$

where K is Nagaoka's "constant". K is actually a tabulated function of the ratio w/r and accounts for the finite aspect ratio of the film cylinder. The relations for λ, L, I and φ may now be inserted in Eq. 4 to yield

$$\left(\frac{m^*(2\pi r)w}{\mu_o n_{se} e^{*2} \pi r^2 w \delta K} \right) \varphi + \varphi = \frac{kh}{e^*} . \qquad (7)$$

This relation may be simplified and solved for the flux to give

$$\varphi = LI = \frac{kh/e^*}{\left[1 + \frac{2\lambda^2}{Kr\delta} \right]} . \qquad (8)$$

The above simple analysis is essentially in the form originally suggested by London. It is assumed here that all of the individual electron-pair wave functions have exactly the same number of nodes, or that an average quantum number k may be used. This latter

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*Mercereau and Crane (21) have experimentally verified that Eq. 6 is correct to within a very few percent for rings of the sort used here.

assumption is implicit in treatments employing a self-consistent field approach to these problems. The treatment of this many-body problem by other than a self-consistent method appears grossly impractical but, even more to the point, such a microscopic individual particle treatment may not be meaningful since the physical quantities of interest, persistent currents and trapped flux, are macroscopic, collective properties. The situation in which all the pair quantum numbers are the same corresponds physically to a displacement of the entire Fermi surface without distortion by an amount $\underset{\sim}{q}$ equal to the wave number associated with the collective motion of the current-carrying electrons.

Equation 8 indicates the way in which the mechanical angular momentum of the current-carrying superelectrons acts as a perturbation on the magnetic flux through a superconducting ring. For the simple one electron atom, one may derive an expression quite analogous to Eq. 8 which indicates for that case the small perturbation (Zeeman effect) on the angular momentum due to the magnetic flux. The difference in emphasis is due in part to the difference in the number of electrons involved and in part to the difference in their orbit radii. Equation 8 has been derived by a number of authors (12 - 16) who have discussed the problem on the basis of a variety of microscopic models. The different treatments yield results concerning the trapped flux which differ only in their prediction of the specific properties of the penetration depth. Equation 8 is a consequence of the basic electrodynamic properties of the superconducting state and does not depend on the specific microscopic model used to arrive at these properties.

The penetration depth is a function of temperature through its dependence on the effective number density of superconducting electron-pairs, n_{se}. Since in the normal state the metal will be completely penetrated by a magnetic field it is reasonable to expect that the penetration depth in the superconducting state increases with temperature and becomes very large near the transition temperature (T_c). This is a well known phenomenon experimentally and in general the observed temperature dependence of λ is described quite satisfactorily by the phenomenological London-Gorter-Casimir theory which predicts

$$\lambda^2(t) = \frac{\lambda_o^2}{(1 - t^4)} \approx \frac{\lambda_o^2}{4(1 - t)} \quad \text{near } T_c, \tag{9}$$

where $t \equiv T/T_c$ is the reduced temperature and $\lambda_o \equiv \lambda\ (t = 0)$. This temperature dependence may be inserted in Eq. 8 to yield

$$\varphi = \frac{kh/e^*}{\left[1 + \frac{2\lambda_o^2}{Kr\delta(1-t^4)}\right]} \tag{10}$$

The BCS theory predicts a dependence of λ on the mean free path for electrons in the superconductor (17). Only the qualitative features of this dependence will be mentioned here. The correlation which leads to ideal superconductive behavior extends over a coherence length (ξ_o) which is characteristic of the material and which is of order 10^{-4} cm in pure bulk samples. When the mean free path (ℓ) of electrons in a superconductor is reduced below this value by impurity scattering or by surface scattering in a thin film, then the number density of electrons which are effective in shielding a magnetic field is also decreased. From Eq. 5 it may be seen that this decrease in n_{se} is reflected in an increase in the value of λ_o^2 to be used in Eqs. 9 and 10. In all of the thin films used in this work this correction to the bulk value of λ is significant since the film thickness $\delta << \xi_o$, in all cases.

Variation of Trapped Flux with Temperature

If we make the conventional assumption that all the pairs are in the same state, Eq. 10 may be expected to hold and the quantum number k should be an adiabatic invariant. Under these circumstances Eq. 10 predicts a very interesting behavior for the trapped flux and hence the persistent current as the temperature is slowly varied. It may be seen from Eq. 9 or from the extensive experimental literature on the subject that the penetration depth increases with increasing temperature and becomes very large near T_c. Thus Eq. 8, or its simple elaboration Eq. 10, predicts that the "quantum" unit of flux is a function of temperature and in particular decreases with increasing temperature. When a ring traps flux it "chooses" a value of the quantum number k. It should be made clear that in all of these experiments the quantum numbers k associated with the trapped flux and current are very large, generally of order 10^5 to 10^7. Individual quanta are not resolved by the present experimental technique although the predicted temperature dependence for an individual quantum is the same as that for the larger systems. It should also be noted that the quantum hypothesis is not essential to the derivation of the temperature dependence of trapped flux which is indicated by Eq. 10. Conservation of the canonical angular momentum is to be expected also for a non-dissipative classical system.

On the other hand, since the stability of persistent currents is now generally attributed to local minima in the free energy which appear as one prediction of the sort of quantum analysis given above, the quantum approach seems more appropriate. Since individual quanta are not resolved in the present experiments, however, the results cannot give direct evidence on the quantization aspect of the fluxoid.

If a ring traps flux at a temperature $t \ll 1$ and is subsequently warmed to temperatures near $t = 1$, the trapped flux and thus the persistent current should decrease. This may be expected to occur even though the ring remains at all times entirely within the pure superconducting state with zero resistance. The experimental investigation of this predicted phenomenon and the associated thermodynamic reversibility has been the principal goal of the present research project.

Many of the basic problems associated with critical persistent currents are discussed in detail in Appendix B. Readers wishing to familiarize themselves with these problems may prefer to read Appendix B before proceeding further.

Near T_c the relation between the critical persistent current and the persistent current curve which decreases according to Eq. 10 is also interesting. Both curves of course go to zero at T_c. Both the analytical expressions and the experimental data for critical persistent currents indicate that in this temperature region $I_c \sim (T_c - T)^{3/2}$. Thus the slope of the critical persistent current curve is zero at $T = T_c$. On the other hand, Eq. 10 predicts for a fluxoid curve an approach to the $I = 0$ axis with a finite slope at $T = T_c$. If this were actually to occur, the fluxoid curve would have a current value larger than the critical value for temperatures very close to T_c. Since this cannot happen we must expect that in an experiment on fluxoid conservation the fluxoid curve should undergo an abrupt change in slope as it meets and thereafter follows the critical persistent current curve.

Under the usual conditions in experiments on flux trapping by superconducting cylinders, the quantity $2\lambda_o^2/Kr\delta$ is so small that the temperature effect would only be significant for temperatures so close to T_c that actual observation of the effect would be exceedingly difficult. It is possible however, to take advantage of the increase in penetration depth for small mean free paths which was discussed above.

This was done in these experiments by using very thin tin films some of which were additionally doped with indium to enhance the impurity scattering. These films were condensed at liquid nitrogen temperature to produce a nearly amorphous film structure so that grain boundary scattering might also aid in reducing the mean free path. As will be discussed later, it is possible with these methods to produce values of the quantity $2\lambda_o^2/Kr\delta$ in the range from 0.05 to 0.2.

Fluxoid Conservation- Experimental Approach

In a current-temperature diagram for a ring, the phase boundary between the true superconducting state which is coherent all the way around the ring and the intermediate state is delineated by the curve of critical persistent current as a function of temperature. Since the experiments on fluxoid conservation must be made in the completely superconducting region, the determination of the critical persistent current as a function of temperature is a required initial step. In the course of the present work, critical persistent current curves have been obtained for a wide variety of rings including both bulk and thin film rings. A typical thin film curve is shown below in Fig. 5. A discussion of the measurement of critical persistent currents and some results not specifically associated with the fluxoid experiments will be discussed in more detail in Appendix B.

The principal percentage variation of the trapped flux and persistent current predicted by Eq. 10 should occur at temperatures quite close to the transition temperature although at somewhat lower temperatures the use of higher currents may actually make the total change larger. It may be seen from Fig. 5 that in the temperature range near T_c the critical current is relatively small. In order to remain safely within the pure superconducting state the persistent current with which the fluxoid is to be investigated must remain small compared to the critical current at any temperature reached. Furthermore, in the extremely thin films for which the quantity $2\lambda_o^2/Kr\delta$ is conveniently large, the total critical persistent current itself is quite small. The sensitivity problems which this situation raises will be discussed in the next chapter.

In a typical fluxoid experimental run a small magnetic flux, such that the associated persistent current is much less than critical, is trapped as the ring is cooled well below the transition temperature. The persistent current is then measured carefully as a function of temperature as the ring is warmed to temperatures near T_c. In the remainder of this dissertation a current versus temperature curve so generated will be referred to as a "fluxoid curve." In this experimental method it is the torque due to the current which is actually measured and the relevant critical phenomena also involve primarily the current. As a consequence, a shift in emphasis from the trapped magnetic flux to the persistent current consistent with the flux will be noted. The two are essentially equivalent however (recall that $\varphi = LI$).

A quite different approach has been used by Mercereau and Crane (29) to observe variations in the persistent current in rings whose temperature oscillates rapidly. In their experiment the ring comprises one wall of a cylindrical second sound resonant cavity and a closely coupled sensing coil is used to detect variations in the persistent current which correspond to the temperature oscillations produced by the second sound. The maximum second sound amplitude is about 10^{-3} K^{o} and this occurs at a temperature well below the transition temperature of the tin film which they used. As a consequence, the maximum observed signal corresponds to a variation in the trapped flux of order one part in 10^{6}, or about one "quantum" of flux. They observed no decay in the persistent current, indicating that for these small thermal oscillations the very small changes in the persistent current were reversible. This ac method is unfortunately extremely sensitive to any variation of the mutual inductance between the sensing coil and the persistent current such as might arise from fluctuations in the measurement geometry or from changes in the current distribution within the ring.

The present experiments are insensitive to the details of the current distribution in the ring and are carried out under conditions more closely approximating equilibrium. The present method also permits much larger temperature variations and hence allows the investigation of fluxoid conservation under more extreme perturbations.

III. EQUIPMENT AND PROCEDURES FOR CURRENT MEASUREMENT

The magnitude of the persistent current in a superconducting ring may be readily determined by mounting the ring as the "moving coil" in a galvanometer system. Since no electrical leads to the ring are needed, the suspension system is much simpler than those in conventional galvanometers. In these experiments Helmholtz coils were used to compensate for the earth's magnetic field during all runs and the residual field was always less than 2×10^{-3} gauss. The measuring magnetic field B_m was generally of order 0.1 gauss and was produced by a set of coils located outside the helium dewar for convenience. The current in the measuring field coils could be reset to about 0.5% from run to run, but the uncertainty in the absolute magnitude of the measuring field was approximately 5%.

In these experiments the ring in which persistent current was to be measured was suspended in a liquid helium bath by a quartz torsion fiber of known torsion constant K. The equilibrium position which the fiber and ring assume in the absence of a magnetic field was then arranged so that the normal to the ring plane, $\underset{\sim}{n}$, was perpendicular to the measuring field $\underset{\sim}{B}_m$ shown in Fig. 1a as seen from above.

A ring carrying a total persistent current, I, has a magnetic moment $\underset{\sim}{M} = I\underset{\sim}{A}$ where A is the area enclosed by the ring. This relation holds for the total current regardless of the distribution of the current within the length of the ring. When the small field $\underset{\sim}{B}_m$ is applied, the ring assumes a new equilibrium position as in Fig. 1b in which the torque $\underset{\sim}{M} \times \underset{\sim}{B}_m$ is just balanced by the restoring force

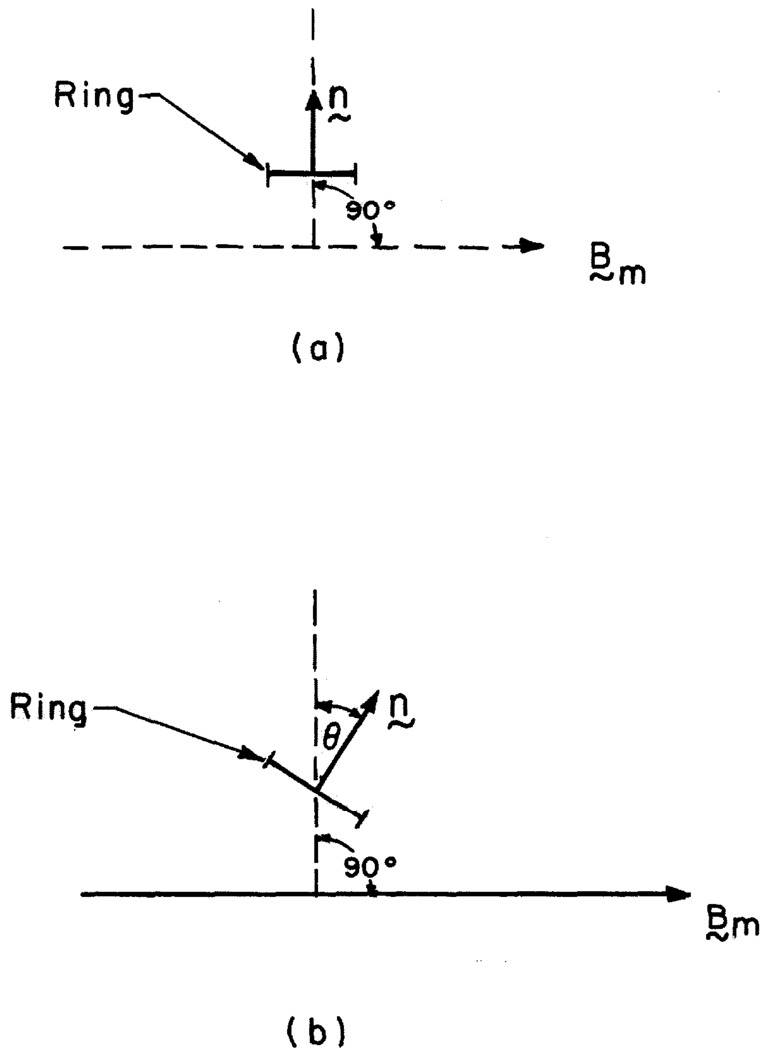


Fig. 1

$-K\theta$ due to the fiber. Thus in terms of the equilibrium deflection angle θ_e, one deduces that the equilibrium persistent current in the ring is given by

$$I_e = \frac{K\theta_e}{\underset{\sim}{A} \times \underset{\sim}{B}_m} = \frac{K\theta_e}{AB_m \cos\theta_e} . \qquad (11)$$

The equilibrium current in the ring while it stands at the angle θ_e in the field $\underset{\sim}{B}_m$ is actually somewhat less than the current which was originally trapped (see part 4 for a detailed discussion of the trapping process). The difference is due to the counter or Lenz's law current induced by the component of the measuring field perpendicular to the plane of the ring. This induced counter current is simply

$$I_{ind} = \frac{\underset{\sim}{A} \cdot \underset{\sim}{B}_m}{L} = \frac{AB_m}{L} \sin\theta_e , \qquad (12)$$

where L is the inductance of the ring. The current I_t, originally trapped is therefore

$$I_t = I_e + I_{ind} = \frac{K\theta_e}{AB_m \cos\theta_e} + \frac{AB_m \sin\theta_e}{L} . \qquad (13)$$

Experimental Details

In these experiments a strip of the superconductor material was evaporated uniformly onto a rotating quartz tube which was maintained at liquid nitrogen temperature. The resulting ring-substrate combination was raised to room temperature for mounting and was then suspended in the helium dewar by a quartz torsion fiber about 2 to 5 microns in diameter. The suspension system consisted of a rotatable central shaft which permitted adjustment of the zero field equilibrium position and a concentric glass tube and chamber (Fig. 2) which serves both as a bearing for the central shaft and as protection for the fiber and ring against the turbulence associated with the boiling of the helium during the pumpdown period. In measuring the deflection angle, a light beam produced by a standard 35 mm slide projector was reflected from a mirror attached to the substrate to a translucent scale graduated in millimeters which was taped to the outer wall of the nitrogen dewar. With this method angles could be readily measured to about 0.1 degrees.

The torsion constants of the fibers used were determined in the usual manner by measuring the torsional oscillation period using a test cylinder of easily calculable moment of inertia in place of the sample ring and substrate*. The uncertainty in the torque constant (K) is approximately 2%. Typically useful values of K lay in the range $10^{-8} > K > 10^{-12}$ newton meters.

The self-inductance of the thin film rings used in these experiments may be calculated with sufficient accuracy by assuming that the current distribution is uniform in the film and by using the inductance relation for a current sheet previously given. The length and diameter of these rings may be very accurately determined by using a microscope fitted with a calibrated eyepiece scale. The film thickness was determined by weighing a test film evaporated at the same time the ring was deposited. This weighing method has a precision of about 10% and within these limits yields results consistent with residual electrical resistivity measurements of thickness made on the same test films.

*The torsion constant K was computed from the relation $K = 4\pi^2 I/T^2$ in which T is the period of the suspended system and I is the moment of inertia of the test cylinder.

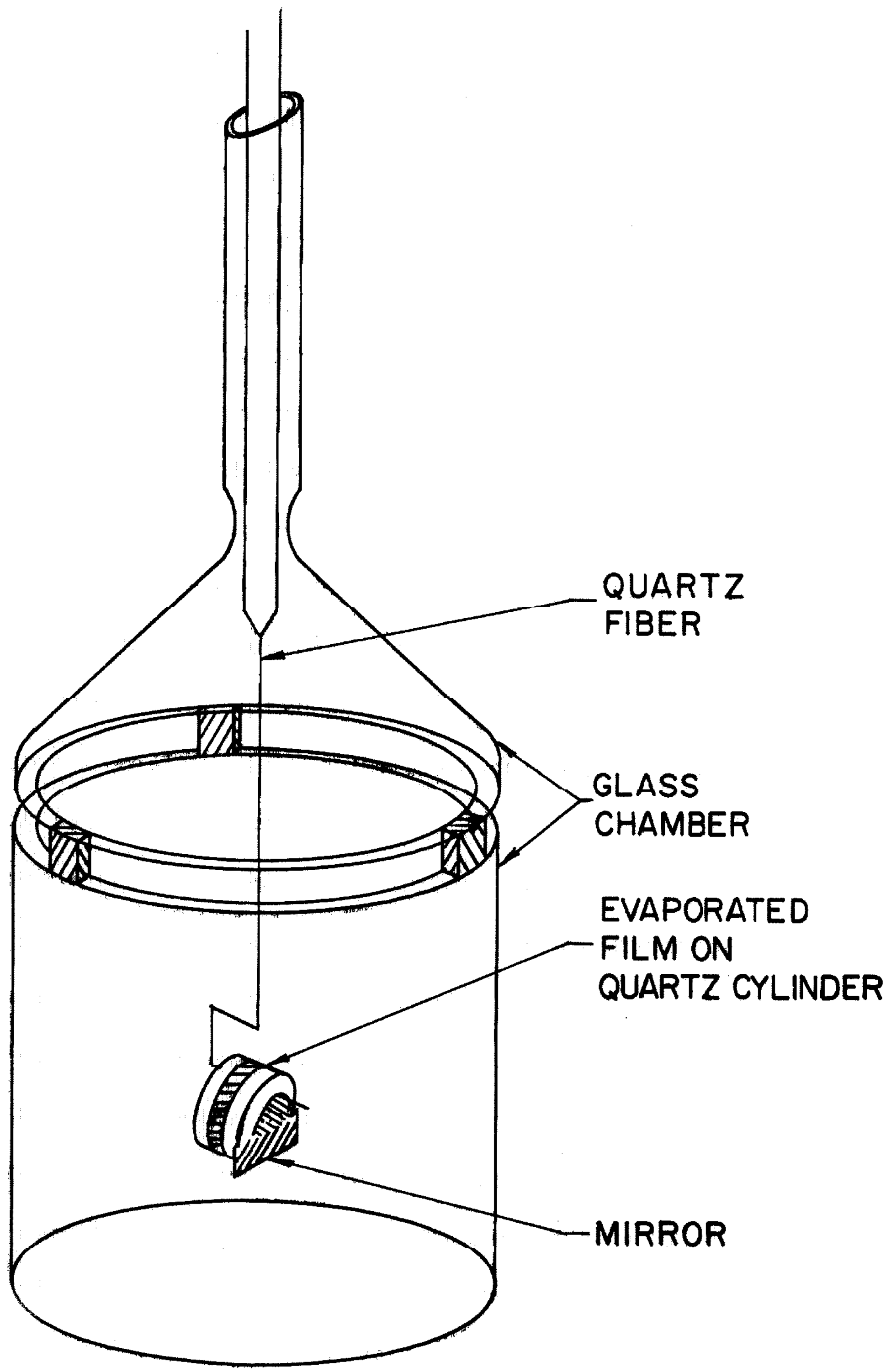

QUARTZ
FIBER
GLASS
CHAMBER
EVAPORATED
FILM ON
QUARTZ CYLINDER
MIRROR

Fig. 2

In the critical current measurements discussed in Appendix B, the temperature was determined from vapor pressure measurements made with a standard mercury manometer system. The manometer system yields a precision of about 1 millidegree over the range of interest. The absolute value of the temperature, however, is probably uncertain by about 10 millidegrees. For the experiments on fluxoid conservation, carbon resistance thermometers calibrated against the mercury manometer during pump-down were used to measure the temperature near the ring. (See typical R vs. T curve in Fig. 3.) Within a given run these resistors (1/4 watt Allen Bradley resistors dipped in glyptal) give readings which are reproducible to the precision with which the calibration manometer may be read. Cycling to room temperature can shift the entire calibration curve by 10 to 20 millidegrees, however, and for this reason the resistor is recalibrated after each cycle during the early stages of the following run.

To meet the exacting sensitivity requirements for carbon resistance thermometry at temperatures above about 2 $^{\circ}$K a convenient method* was devised for comparing very precisely the thermometer resistance with a standard decade resistance box and a precision Helipot. The General Radio 1650 A Impedance Bridge, arranged in the ac resistance connection, is used as the comparator as shown in Fig. 4. [This transistorized bridge has available for such measurements 100 times the sensitivity needed to obtain its advertised accuracy of 1%.] In operation the bridge dial is set to some value and the bridge is then

*Subsequently to this work several commercial units designed to do essentially the same job have appeared on the market.

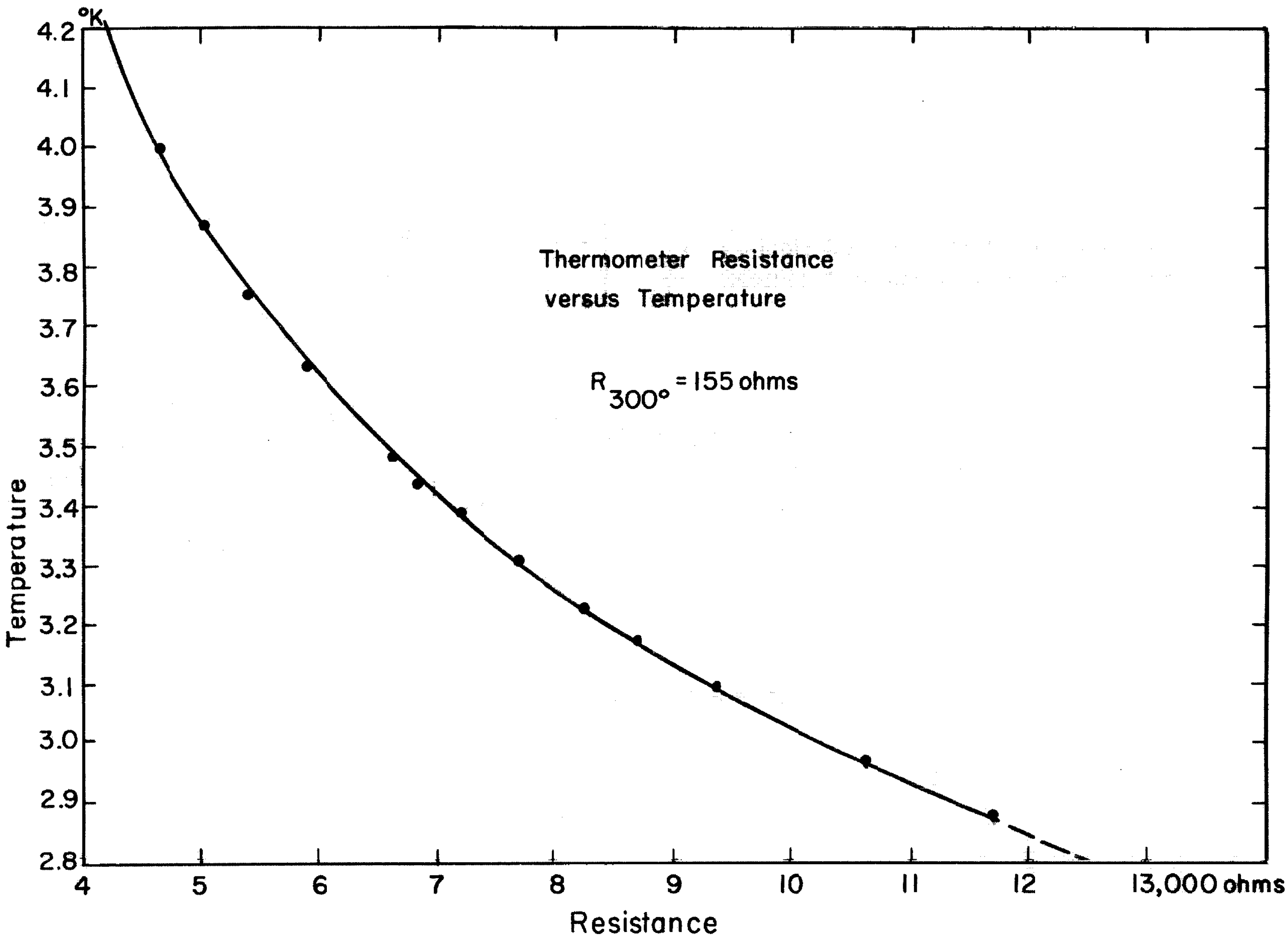
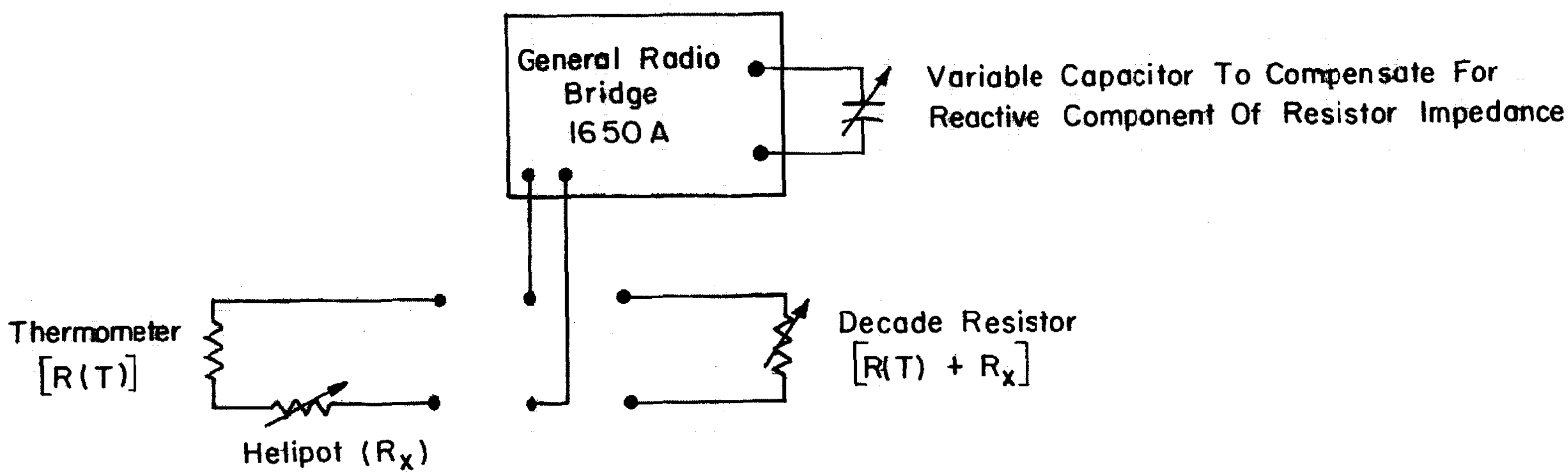

°K
4.2
4.1
4.0
3.9
3.8
3.7
3.6
3.5
3.4
3.3
3.2
3.1
3.0
2.9
2.8
Temperature
4
5
6
7
8
9
10
11
12
13,000 ohms
Resistance
Thermometer Resistance
versus Temperature
R 300° = 155 ohms

Fig. 3



Comparator Circuit for Resistance Thermometry

Fig. 4

balanced by adjusting the external decade resistor. During this process the sensitivity is such that one can, for example, detect a change of 1 ohm in a total of 15,000 ohms.

Leaving the bridge setting unchanged one then substitutes the thermometer-helipot combination for the decade resistor and rebalances the bridge with the Helipot. The difference between the decade box resistance and the Helipot resistance gives the value of R(T) desired. This method, while limited by the 1% accuracy inherent in the decade box, is capable of excellent precision and settings are reproducible to about 1 or 2 ohms in 15,000 ohms. The method has the two further advantages that long term stability of the bridge detector system is not needed and the measuring system dissipates only a few microwatts in the thermometer.

Application to Critical Current Measurement

A number of investigators have measured critical currents in thin films by detecting the onset of a potential difference across the film as the current is increased, with the film in a 4-terminal connection. Ginzburg and Shalnikov (27) achieved perhaps the best results by this method, using cylindrical films with axial current in order to minimize magnetic field edge effects which can cause difficulty in flat films. With such methods involving external current sources a number of difficulties exist. Chief among these is the fact that they do not measure a property of the true, coherent superconducting state. Since these methods depend on the presence of a detectable resistance and since the transition has a finite width, the first observations occur only after the superconductor has been driven out of the pure superconducting state by the current. The curve which is actually obtained gives only the variation with temperature of the minimum detectable resistance in the intermediate state. An extrapolation to the superconducting state then necessarily depends on some assumed model. Another uncertainty is the possibility that Joule heating at the normal-superconducting contacts will initiate normal front propagation which switches the film into the normal state at currents less than the true critical value. At lower temperatures, where the critical current is larger, the Joule heating accompanying the transition is often sufficient to cause irreversible changes in the film structure. There are also sensitivity problems near the transition temperature. In the work of Ginzburg and Shalnikov these latter two effects limited the useful region to a temperature range $0.02\ K^o < (T_c - T) < 0.4\ K^o$.

The determination of critical currents for thin films by means of persistent currents in rings avoids the difficulties encountered with the use of a current source, as discussed above, and furthermore permits measurements over a wider range of temperatures. In the present critical-current measurements the ring was placed in a magnetic field B_t of a few gauss perpendicular to the plane of the ring, and then cooled well below the critical temperature. This field was then slowly removed leaving trapped by the ring a flux due to the critical current. A small measuring field of the order of 50 to 100 milligauss was then applied as discussed previously and the magnetic moment (and hence the persistent current) was determined as a function of temperature as the ring warmed up toward its transition temperature (see fig. 5, for

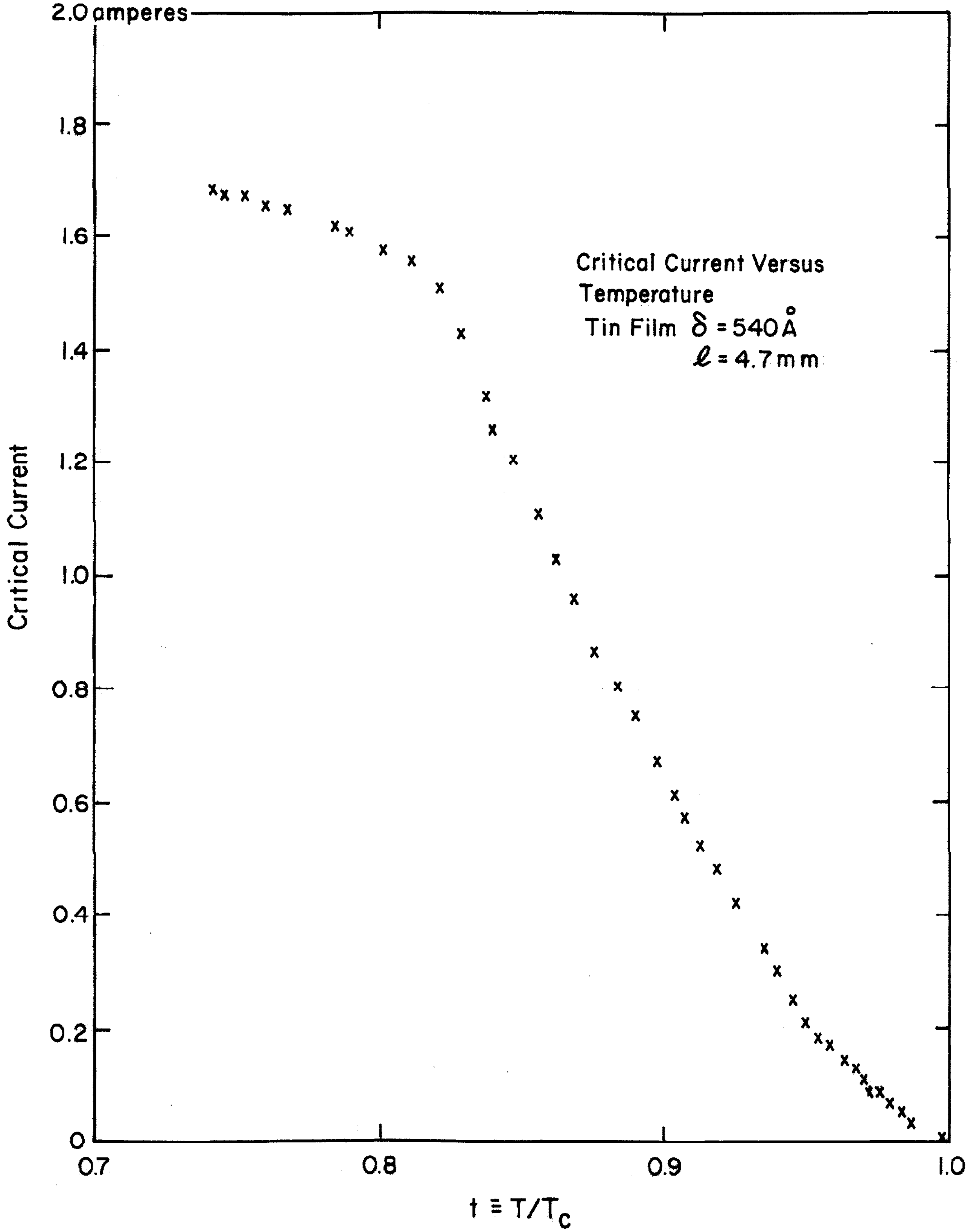

2.0 amperes
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
Critical Current
Critical Current Versus
Temperature
Tin Film δ = 540 Å
ℓ = 4.7 mm
0.7
0.8
0.9
1.0
t ≡ T/T_c

Fig. 5

example). When the critical persistent current is reached by increasing the temperature the ring is not driven "normal" with loss of the entire persistent current. Rather, the current decays only slightly to a sub-critical value.

Noise and Sensitivity

When measuring very small persistent currents such as those associated with films as thin as 100 to 200 Å, it is necessary to use fibers of such sensitivity that noise induced by bubbling or convection in the liquid helium can become a serious problem. Several weeks of futile experiments verified how serious this difficulty may become when the torsion constant of the fiber is less than about $K = 3 \times 10^{-10}$ newton meters.

Considerable experimentation made it apparent that it would be impossible to observe the temperature dependence of the trapped flux with reasonable precision unless some way could be found to reduce drastically the noise induced by boiling and convection of the helium I. A successful method for achieving this noise reduction was finally devised. The method depends on two facts: the liquid helium in a bath with a positive temperature gradient upward is stable and no convection currents will arise; and the low thermal conductivity of helium above the λ point makes the establishment of such a positive gradient quite simple. In establishing a positive gradient the bath is first pumped down to some lower temperature T_{min}. During this operation the temperature gradient is negative and convection stirs the liquid, maintaining the bath at an essentially uniform temperature throughout. The pumping line is then closed off and warm helium gas is introduced until the pressure in the dewar is again raised to one atmosphere. The surface of the liquid helium is then at 4.2 ^{o}K but well below the surface, in the region of the ring and fiber, the temperature is only slowly rising from T_{min} toward 4.2 ^{o}K. The actual pressure at the ring level is thus essentially one atmosphere while the vapor pressure of the helium at that level is much less. Hence no boiling occurs. Since vapor pressure thermometry will no longer serve, the carbon resistance thermometers discussed above were used to measure the temperature in the region of interest near the ring.

The presence of a temperature gradient in the helium bath would at first sight appear to render suspect the "temperature" of the ring since different parts of the ring would be at different temperatures. For this reason the actual temperature gradient under typical conditions was investigated using three resistance thermometers in the arrangement shown in Fig. 6, with a spacing of 2 cm, twice the normal ring diameter. The results of this experiment, shown in Fig. 7, indicate that the temperature difference across the ring diameter is approximately 0.010 ^{o}K. The actual uncertainty in the ring temperature from run to run may be expected to be much less than this 10 millidegree maximum temperature differential. We may conclude that a temperature gradient adequate to suppress the noise is still so small that it does not invalidate any of the temperature measurements. The physical problem of the current behavior in the presence of this temperature gradient has been dispensed with by means of the tacit assumption that overall control is exerted by the warmest point, at the top of the ring. In practice the temperature is

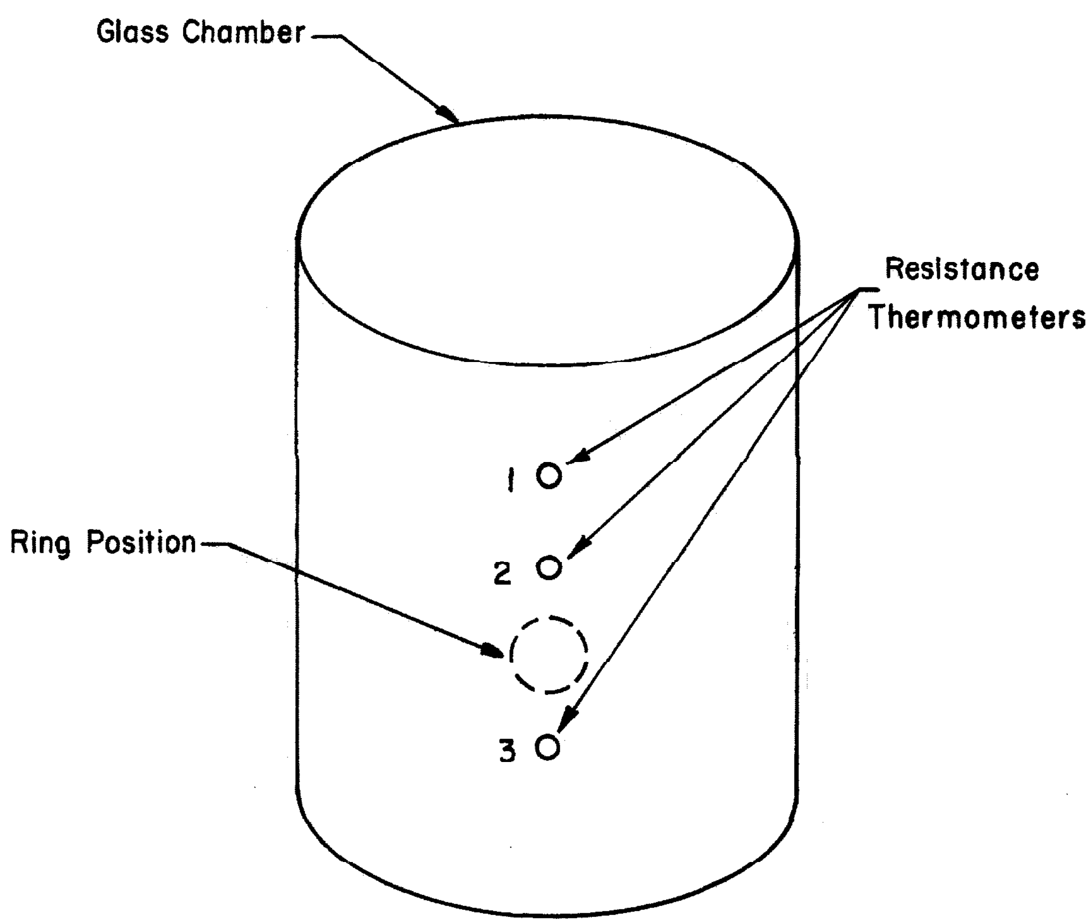


Fig. 6

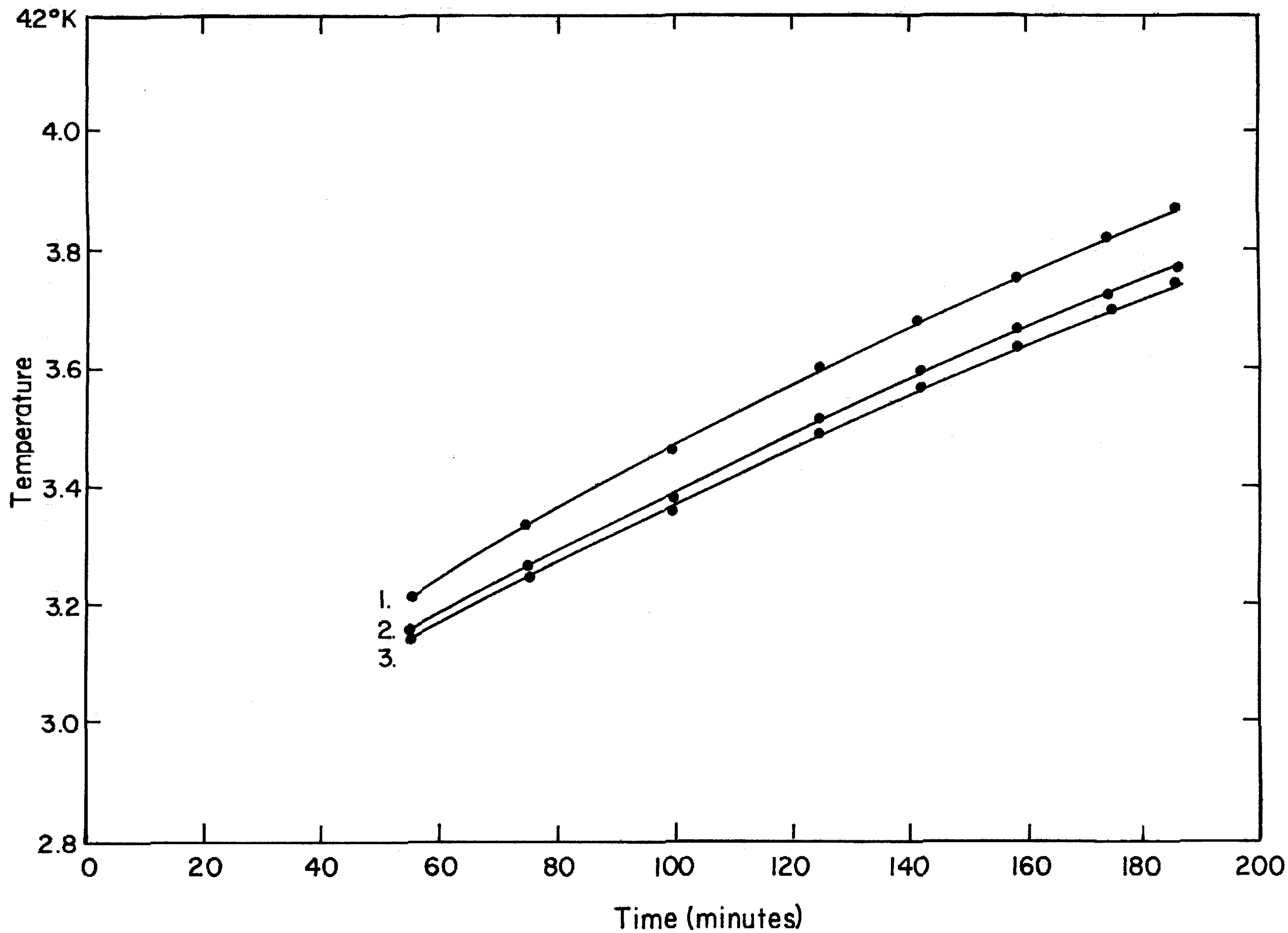

42°K
4.0
3.8
3.6
3.4
3.2
3.0
2.8
Temperature
0
20
40
60
80
100
120
140
160
180
200
Time (minutes)
1.
2.
3.

Fig. 7

measured at this level.

Although the number density of superconducting electrons will be a function of position around the ring, no conversion of electrons between the superconducting and normal states need occur. This is in distinct contrast to the process which takes place when the temperature of the entire ring is raised or lowered. The case of electric current flow around a superconducting ring in a temperature gradient is quite analogous to the case of non-dissipative fluid flow in a toroidal pipe of varying cross section. The moving fluid "piles up" in the regions which have a larger static capacity but the flow remains continuous.

Most of the gain achieved by this noise reduction technique was spent for increased fiber sensitivity. Thus in spite of the general effectiveness of this method of noise reduction, it remains true that the principal contribution to the uncertainty in a current measurement is the inability to read the equilibrium deflection angle with sufficient precision. The deflection angle itself can be read under quiescent conditions to about 0.1° over any part of the scale. For typical deflections ranging from about 1° up to 60° this is an average uncertainty for a single reading of about 1%. The residual random noise usually increases this to an average potential error of about 5% and this is the major factor limiting the overall precision of a single reading. In actual experiments a number of successive readings are averaged at each temperature to produce a single point on the curve.

At one stage of this work it was hoped that the random noise introduced by the liquid helium could be largely eliminated by choosing to work with a material, e. g. aluminum, whose transition temperature lies below the λ point. Temperature measurement and control would also be simplified by the essentially isothermal nature of the helium II bath. Unfortunately, however, the noise reduction achieved by going below the λ point is not nearly as great as the loss in critical persistent current level in going from tin to aluminum. This approach, therefore, failed to solve the noise-sensitivity problem.

IV. RESULTS OF FLUXOID CONSERVATION EXPERIMENTS

Considerations of Sample Preparation

For any given temperature (t) the percentage reduction in persistent current from the low temperature value will depend on the magnitude of the quantity $2\lambda_o^2/Kr\delta$. While it is true that the denominator of the Eq. 10 will become large for any value of $2\lambda_o^2/Kr\delta$ if one goes sufficiently close to T_c, the sensitivity and noise problems discussed above limit the closeness with which T_c may be approached while remaining in the superconducting state with a readily measurable current. It is therefore essential to maximize the value of the quantity $2\lambda_o^2/Kr\delta$ by appropriate choice of the properties of the superconducting ring to be used. By so doing the observability of the effect may be greatly enhanced.

Due to the combined features of the current measurement method, manipulation of the ring radius (r) is not very useful in increasing the observability of the persistent current variations which are predicted by Eq. 10. While smaller radii will give large values of $2\lambda_o^2/Kr\delta$ it must be remembered that for a given persistent current capability* the magnetic moment and hence the torque on which the measurement interaction relies is proportional to r^2. Thus, in order to get useful deflections for smaller rings, one must use more sensitive fibers with their correspondingly larger noise figures as discussed in Part III. One does not gain by increasing the radius since the larger

*In Appendix B the critical current density is shown to be quite independent of length and film thickness over wide ranges of the parameters.

moment of inertia of the larger substrate may cause the torsional oscillation period of the system to become unwieldy. Furthermore, with periods of several minutes the correlation of an averaged set of readings with a specific temperature is less meaningful since the temperature is changing significantly on this time scale.

Variation of the length of the ring (w) is likewise not very useful in increasing the observability of the effect. While lengthening the ring increases the useful magnetic moment the corresponding change in Nagaoka's "constant" (K) largely offsets this gain. In the light of these considerations compromise values of both w and r were chosen to facilitate the evaporation and handling of the ring-substrate combinations. The rings used in these fluxoid experiments were all 1 cm in diameter and had lengths in the range from 0.3 to 1.5 mm.

The major gain in the observability of the effect is achieved by using very thin films produced in a manner tending to promote extraordinarily large penetration depths. It was mentioned earlier that the microscopic BCS theory and Pippard's prior non-local theory predict that the penetration depth increases as the electron mean free path is made shorter. The essential correctness of their prediction is now supported by a considerable body of experimental evidence (see Ref. 25, for example). Since the mean free path is influenced by both impurity and boundary scattering, what is needed are very thin impure films. The properties of tin with indium as an impurity have been studied rather extensively in bulk samples by Pippard and others. From measurements at high frequency it is known that for concentrations of up to nearly 3% indium the primary change in the superconducting properties involves the penetration depth, which increases quite markedly as the mean free path is decreased by the indium. There is also considerable evidence that decreasing the film thickness increases the penetration depth (21, 22, 25, 26). The present work involves films which are at the same time thinner than those previously investigated and contain up to 2% indium as an impurity.

During the early portion of this work little attention was paid to specific control of impurities during the rather crude evaporation process then used. The tin used was "reagent" chemical grade* containing 0.002% lead. One ring (δ = 310 A) evaporated at that time, which will be discussed in more detail shortly, appeared to exhibit a very large penetration depth in the fluxoid experiments. This large value is probably due to selective evaporation of the Pb impurity from the large evaporation source. Thus the Pb concentration in the film may have been greatly enhanced over the value initially present in the source material.

Later more effort was made to control the impurity level in the films by using 99.999% pure tin† and by alloying the films with 0.5 to 2.0% of similarly pure indium.‡ The relative concentrations were determined by evaporating carefully weighed charges to completion. By putting into the source evaporation boat only the amount required for the film, no enhancement of impurity concentration can occur due to selective evaporation rates. While it is true that the indium or lead

*Baker Chemical Co., Phillipsburg, New Jersey.

†Supplied by A. D. MacKay, Inc., 198 Broadway, N.Y. 38, N.Y.

‡Indium Corp. of America, Utica, N.Y.

will be almost completely deposited as a layer before any of the tin reaches the substrate, the diffusion of the impurity throughout the film takes place rapidly at room temperature even in films 1500 A thick.* By using this approach, superconducting alloy films as thin as 65 Å have been produced. The films used in this work were evaporated onto liquid nitrogen cooled substrates at pressures generally of order 5×10^{-6} mm of mercury. The survival rate of such films appears to be considerably enhanced if, while the substrate is still cold and very shortly after the evaporation, argon gas is admitted to the vacuum chamber and the system is permitted to warm up slowly thereafter.

Flux Trapping Methods

In the previous discussions of experimental techniques, no details have been given about the process by which the flux is trapped and the persistent current thereby set up. Certainly the most straightforward method for trapping current† in a superconducting ring is that mentioned briefly in Part III in connection with critical current measurement. In this approach a magnetic "trapping" field (B_t) is applied perpendicular to the plane of the ring, the ring is cooled down through the transition to a temperature T_1, and the field B_t then removed. The ring will then trap a persistent current AB_t/L where A and L are the area of the ring hole and the self-inductance as usual. It will, of course, not be possible to trap a persistent current larger than the

*W. H. Meiklejohn - private communication. This assumption was confirmed by x-ray analysis of films evaporated in the same way used here.

†Since the limiting feature in the trapping process for thin films involves the current rather than the flux the shift in emphasis from flux to current "trapping" seems useful here.

critical value, so the maximum useful trapping field is given by

$$(B_t)_{opt} = \frac{LI_c(T_1)}{A} \quad . \tag{14}$$

The use of a larger trapping field will merely result in the dissipation of energy in the ring as the trapping field is removed. Whereas for fields $B_t < LI_c(T_1)/A$ there is no dissipation, for fields even slightly larger there is dissipation whose magnitude increases linearly with the rate at which the field is reduced. The dissipation associated with this process can in fact raise the ring temperature (T_R) considerably above that of the bath. If this occurs the current finally trapped $i_c(T_R)$ may be significantly less than $i_c(T_1)$. This difficulty was encountered in some of the early work on critical currents which is discussed in Appendix B. In that work a wirewound potentiometer was first used to control the trapping field and to reduce it at the lower temperature T_1. This method did not prove to be sufficiently slow or smooth to retain the lower temperature (T_1) and thus avoid the above difficulty. The stray field of a bar magnet was later substituted and trapping accomplished more successfully by physically removing the magnet.

A second approach has often proved to be more convenient when the ring orientation is not readily fixed during pump-down. If the ring goes superconducting in zero field and a magnetic field of magnitude less than $(B_t)_{opt}$ (see Eq. 14) is applied perpendicular to the ring plane, the ring will carry a current just sufficient to prevent any flux from penetrating the central hole. If the field is increased beyond this value the excess will leak into the ring hole. When this has happened the ring will contain a flux due to the excess field and will still carry a shielding current $i_c(T_1)$ (except for thermal loss as discussed above). If the trapping field is now removed the flux which leaked in will remain trapped. This description of the process should be valid until B_t exceeds the value $2LI_c(T_1)/A$, whereupon the same thermal problems previously discussed will occur again. Using this method, it has often been possible to trap currents quite close to the critical value.

For the fluxoid conservation experiments a persistent current considerably less than critical is usually desired. Furthermore, when using the more sensitive suspension fibers which are necessary it becomes unfeasible to fix the ring orientation during the pump-down period while the helium is boiling violently. The resulting rapid fluctuations in the relative orientation of the ring and the applied field B_t make precise control of the effective perpendicular component of the field essentially impossible. This considerably complicates the problem of trapping a current which is a specified fraction of the critical persistent current. It is possible, however, to trap small fractions of the critical persistent current by using the second technique mentioned above at a time and temperature during pump-down when the critical current has approximately the value one desires to trap.

Fluxoid Measurements and Data Analysis

In the experimental investigation of fluxoid curves one is in practice limited in the useful number of points which can be taken on a given curve. It is necessary to read the extremes of the omnipresent fluctuations in the position of the light spot in order to obtain averages for determining the equilibrium position. Usually the extremes of at least 6 to 10 swings are recorded and averaged for each point shown on curves such as those in Fig. 8. Thus, since the half-period of a typical fiber-ring-substrate system is usually about 25 to 40 seconds, the measurement of a single point may take several minutes. In order for the current-temperature relation to make sense these recorded points must be separated in temperature by more than the change with time which occurs during an actual reading process.

An additional consideration limits somewhat the temperature range over which the fluxoid curves may be taken. The temperature difference which establishes the positive temperature gradient can never exceed about 2 K^o because when the surface comes quickly to 4.2 ^oK with the addition of the warm helium gas, the cooler portions of the bath come just as quickly to 2.17^oK due to the rapid rate at which thermal equilibrium is established below the λ point. The turbulence and fluid motion associated with the violent boiling during the pump-down period persists for some time after the positive temperature gradient has been established. This residual fluid motion can cause an apparent shift in the zero-field equilibrium position of the ring. Since the warm-up rate is determined by factors which are not easily controlled such as the heat leak through the dewar walls and the helium bath level, waiting for the decay of this fluid motion limits the temperature range in which good current measurements can be made to $t \gtrsim 0.7$. This is not a serious limitation, however, since most of the change in the persistent current occurs for values of $t > 0.7$.

Fluxoid curves were investigated experimentally for several tin rings representing different compromises in the choice of the parameters just discussed. Very pure tin films were examined at thicknesses down to $\delta = 350$ Å, and alloy films containing up to 1.9% indium as an impurity were examined at thicknesses down to 110 Å. The tin film of poorly known impurity content and thickness $\delta = 310$ Å is also included. This film appears from these measurements to have an extraordinarily large penetration depth, probably due to the lead impurity, and it exhibits the expected form of current decrease quite dramatically.

In the figures which follow, showing persistent current as a function of temperature, the solid curves are plots of Eq. 10 with the quantity $2\lambda_o^2/Kr\delta$ chosen to yield the best fit for all the fluxoid runs on a given ring. Eq. 10 may be rearranged in a form which simplifies this analysis somewhat. If we define $C \equiv 2\lambda_o^2/Kr\delta$ and $y \equiv 1/(1-t^4)$ then Eq. 10 becomes

$$I(t) = \frac{I_o}{1 + Cy} , \tag{15}$$

from which we may write

$$\frac{1}{I(t)} = \frac{C}{I_o} y + \frac{1}{I_o} . \tag{16}$$

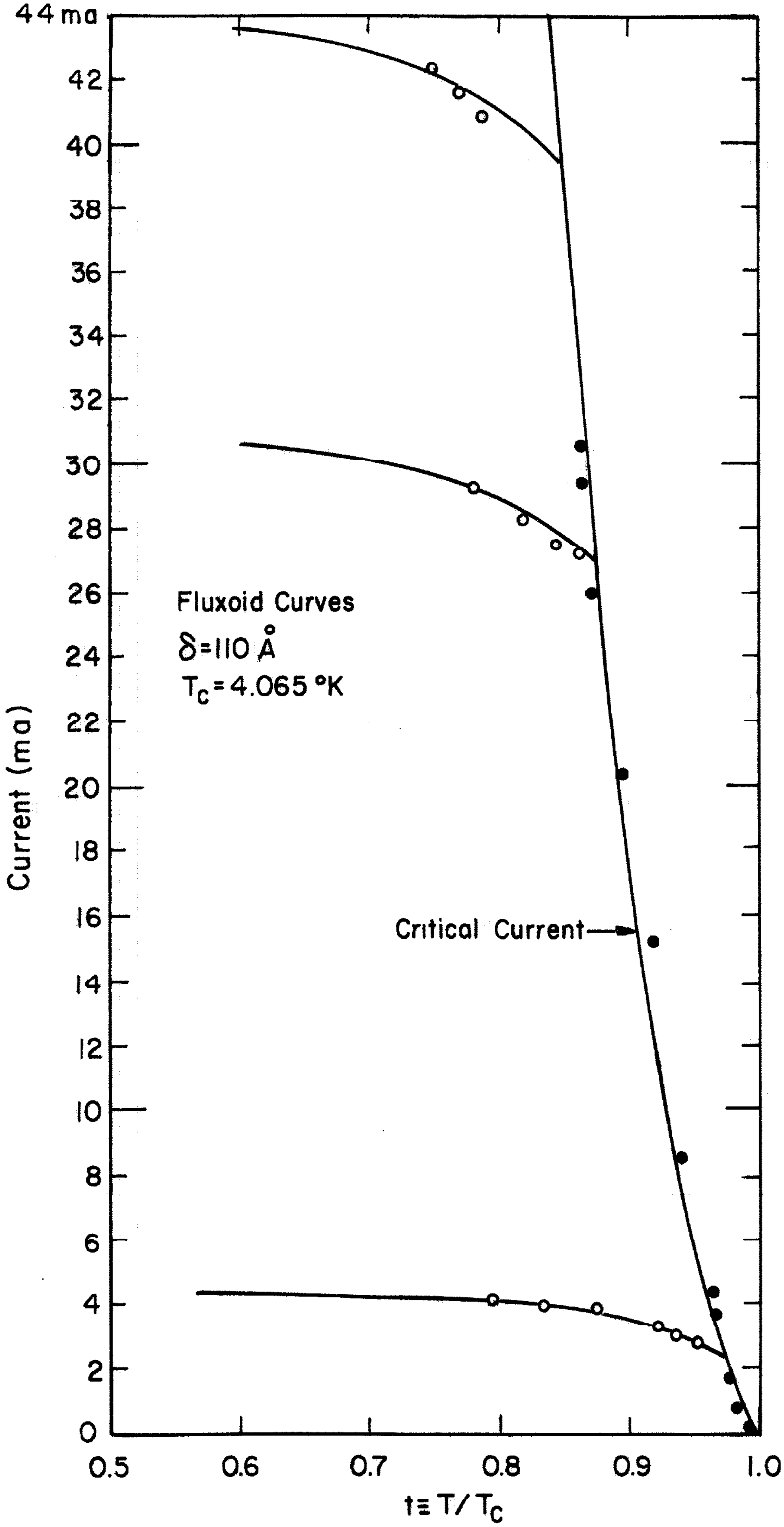
If we now replot the fluxoid data in the form $1/I(t) \equiv z$ versus y (a typical curve is shown in Fig. 9) we should get a straight line of slope C/I_o and z intercept $z_o = 1/I_o$. Then clearly

$$C = \frac{\Delta z}{\Delta y} \left(\frac{1}{z_o} \right) . \tag{17}$$

This procedure was carried out for a number of fluxoid runs on each ring and the resulting values of C averaged to obtain the single value used in fitting all of the fluxoid curves shown for a given ring. For each individual fluxoid curve, the value of I_o is then chosen by fitting the curve at one temperature, usually at $t = 0.8$.

Figure 8 shows curves from a series of fluxoid runs made over a period of several days on the 110 Å tin plus 1.9% indium ring. As predicted by the analysis, the greatest percentage decrease in the persistent current occurs near T_c and this region of rapid variation can only be reached for low persistent current values. While the percentage change is far less at lower temperatures, much higher persistent current values may be used while still satisfying the criterion of remaining entirely within the pure superconducting state at all times. The actual observable decrease in current is in fact somewhat larger for this latter case. The particular nature of the experimental method is such that the precision in the measurement of current values is not appreciably less for large currents than for small. Thus it is very nearly as easy to measure a change of 1 ma in 80 as it is to measure a change of 1 ma in 3. This is true primarily because the measuring field is fixed and the reading error is independent of deflection. The critical current shown is the composite result of a number of runs made before and after each of the fluxoid runs.

Figure 10 shows similar data on the $\delta = 150$ Å tin plus 1.0% indium ring. Again the solid analytical curves are plotted for a single value of $2\lambda_o^2/Kr\delta$ determined by the method discussed previously. The experimental runs on the $\delta = 310$ Å ring were carried out much



44 ma
42
40
38
36
34
32
30
28
26
24
22
20
18
16
14
12
10
8
6
4
2
0
Current (ma)
Fluxoid Curves
δ=110 Å
Tc = 4.065 °K
Critical Current
0.5
0.6
0.7
0.8
0.9
1.0
t ≡ T/ Tc

Fig.8

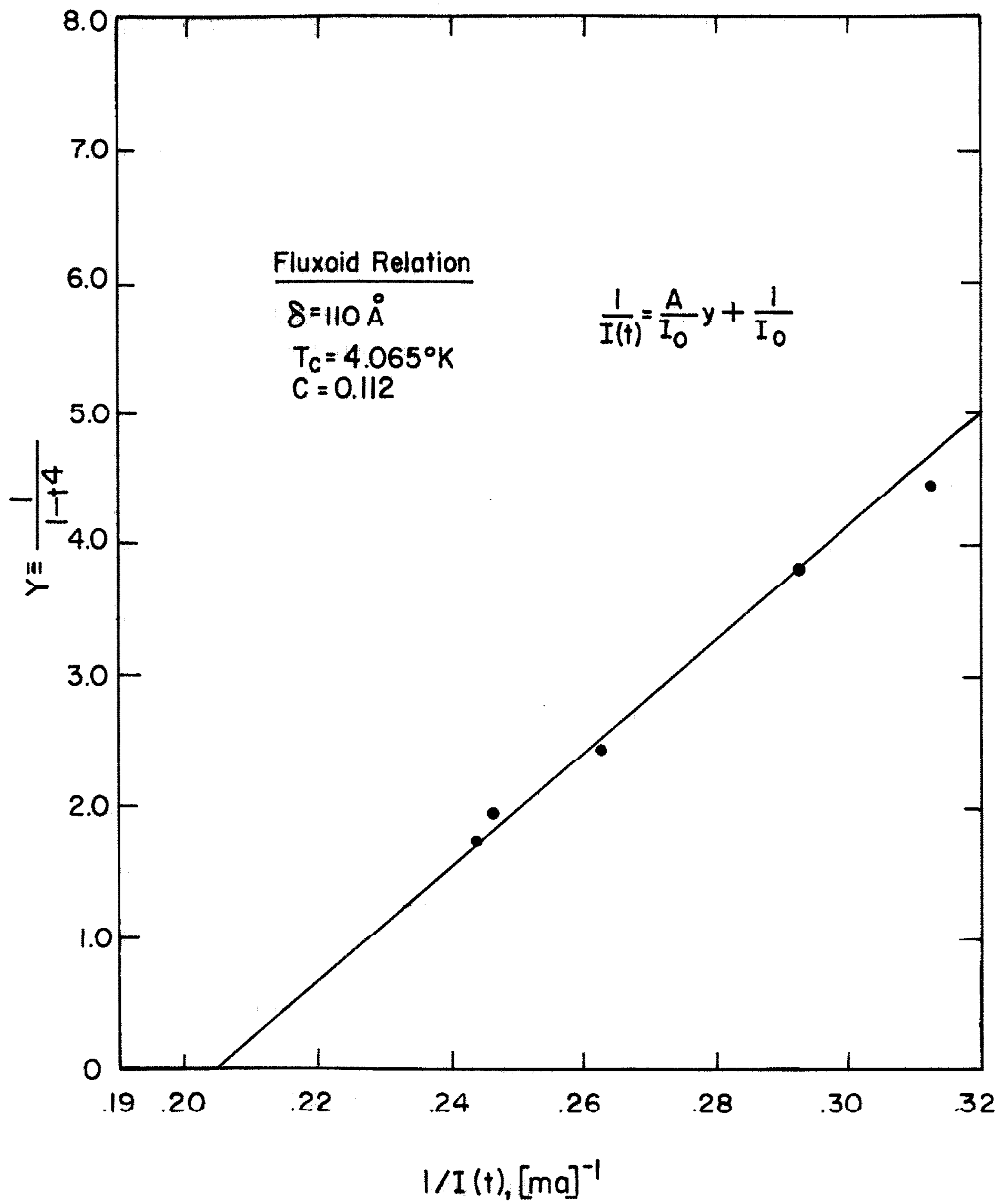

8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
.19
.20
.22
.24
.26
.28
.30
.32
Fluxoid Relation
δ = 110 Å
Tc = 4.065°K
C = 0.112
1/I(t) = A/I0 y + 1/I0
Y = 1/(1−t4)
1/I(t), [ma]-1

Fig. 9

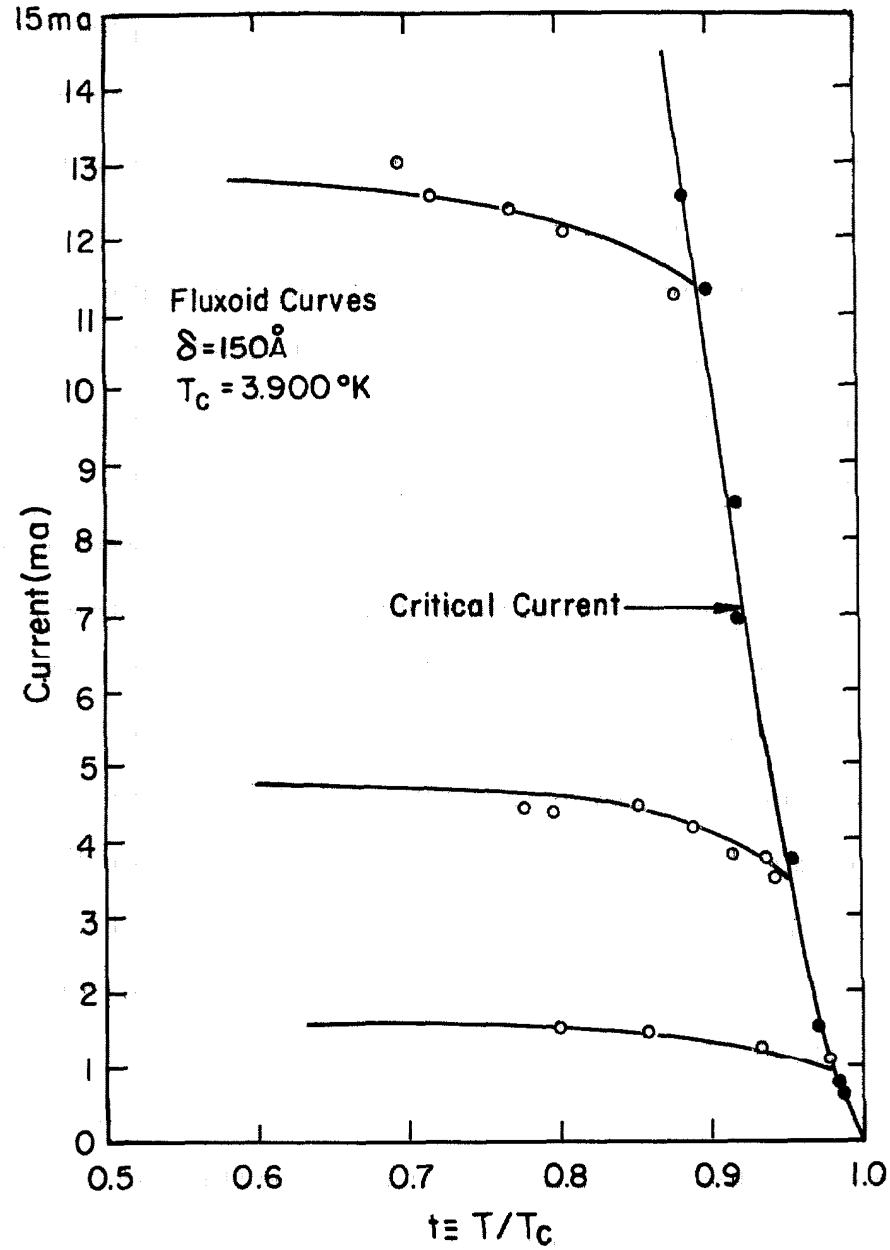
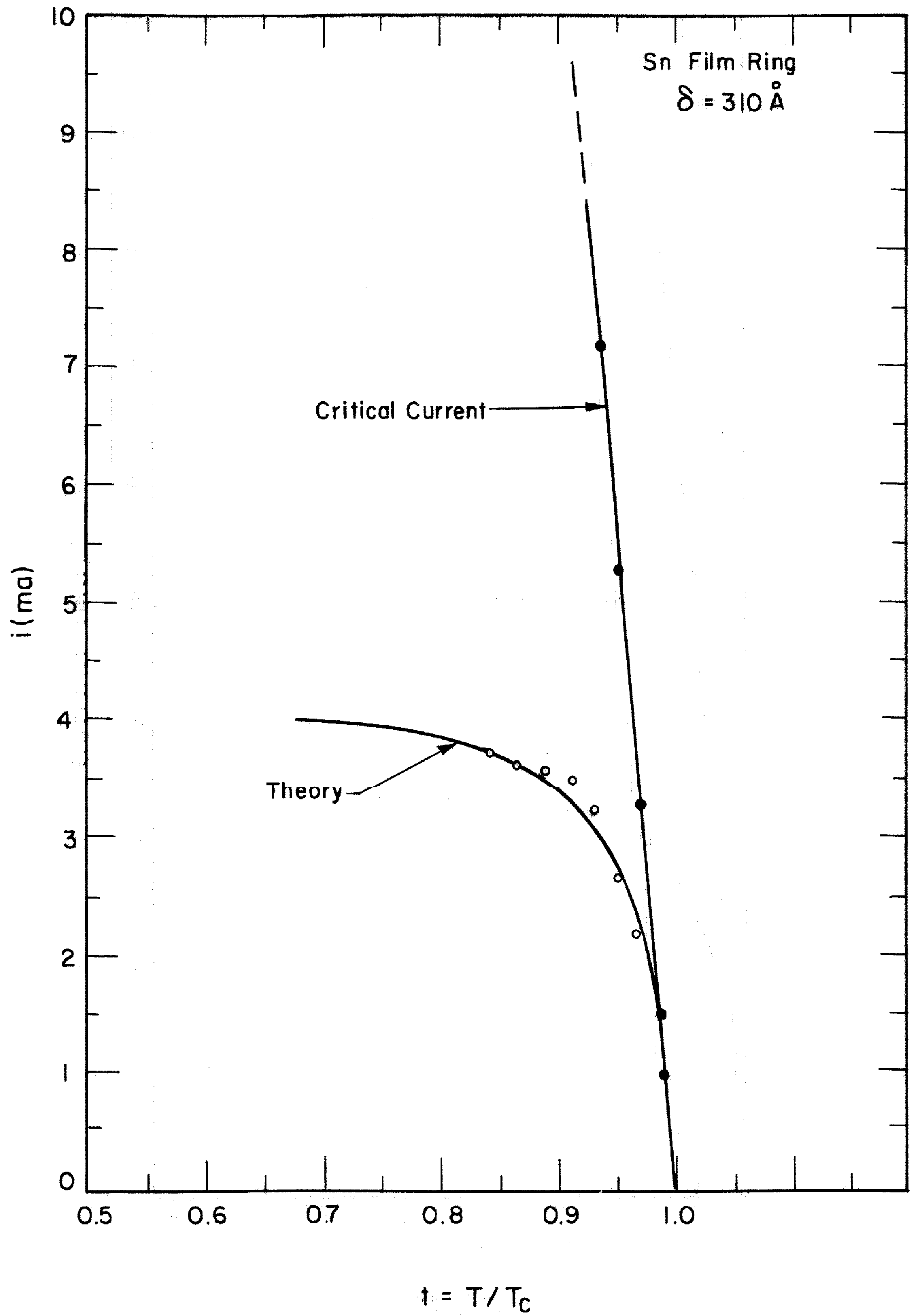

15ma
14
13
12
11
10
9
8
7
6
5
4
3
2
1
0
Current (ma)
Fluxoid Curves
δ=150Å
Tc = 3.900 °K
Critical Current
0.5
0.6
0.7
0.8
0.9
1.0
t≡ T/Tc

Fig. 10

earlier in the project and were all taken at relatively low current values. Representative curves for this ring are shown in Figs. 11a and 11b which are plotted separately to avoid confusion since these two curves nearly overlap.

Figure 12 shows two fluxoid curves for a $\delta = 350$ Å tin film evaporated to completion directly from the 99.999% Sn. In this film the current decrease is quite small and is almost completely masked by the scatter in the data. It is thus not very meaningful to attach numbers to the coefficient C which would be obtained by the above fitting process. A probable upper limit on λ may be obtained, however, from a curve fit such as that shown.

The sample parameters and experimental values of C for these rings are summarized in Table I. At this time there is no other published source of experimental data on penetration depths even in pure films as thin as the 110 Å and 150 Å films used here. Penetration depth data on alloy films is not available even for considerably thicker films. However, data of Mercereau and Crane (21) on thin films of pure tin show a thickness dependence of the penetration depth in relatively good agreement with the theoretical work on mean free paths by Douglass (22). If the theoretical curves of Douglass are extrapolated beyond the values he considers, to the shorter mean free paths associated with very thin and impure films, the results yield values of $\lambda_0 \approx 6500$ Å. In view of the uncertainty of this extrapolation process and the notorious difficulty of producing good thin films of specified properties, this value agrees remarkably well with the penetration depths shown in Table I for the 110 Å and 150 Å films. The data of Mercereau and Crane on a pure tin



Sn Film Ring
δ = 310 Å
Critical Current
Theory
i (ma)
0
1
2
3
4
5
6
7
8
9
10
0.5
0.6
0.7
0.8
0.9
1.0
t = T/Tc

Fig. 11a

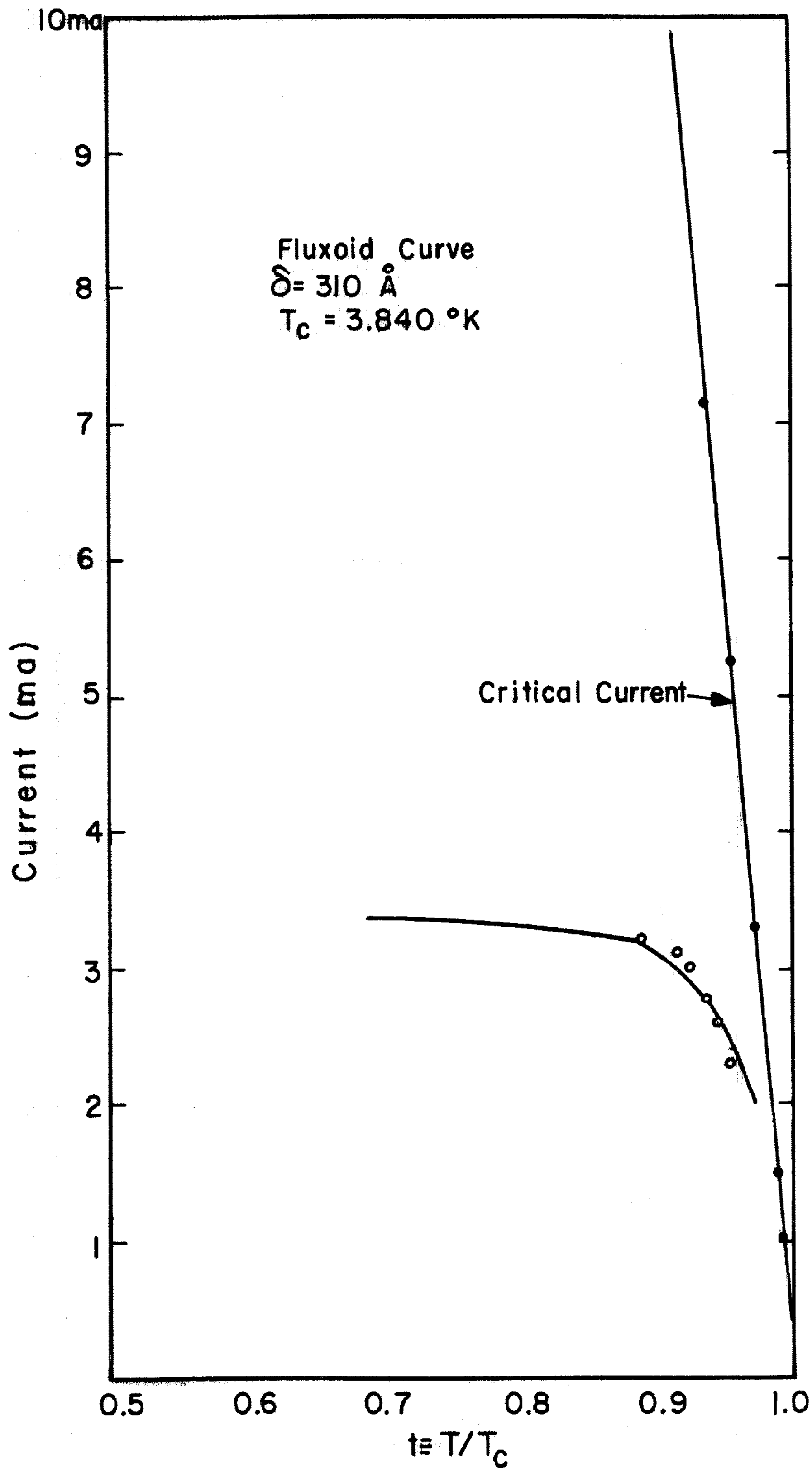

10ma
9
8
7
6
5
4
3
2
1
Current (ma)
Fluxoid Curve
δ= 310 Å
Tc = 3.840 °K
Critical Current
0.5
0.6
0.7
0.8
0.9
1.0
t≡ T/Tc

Fig. 11b

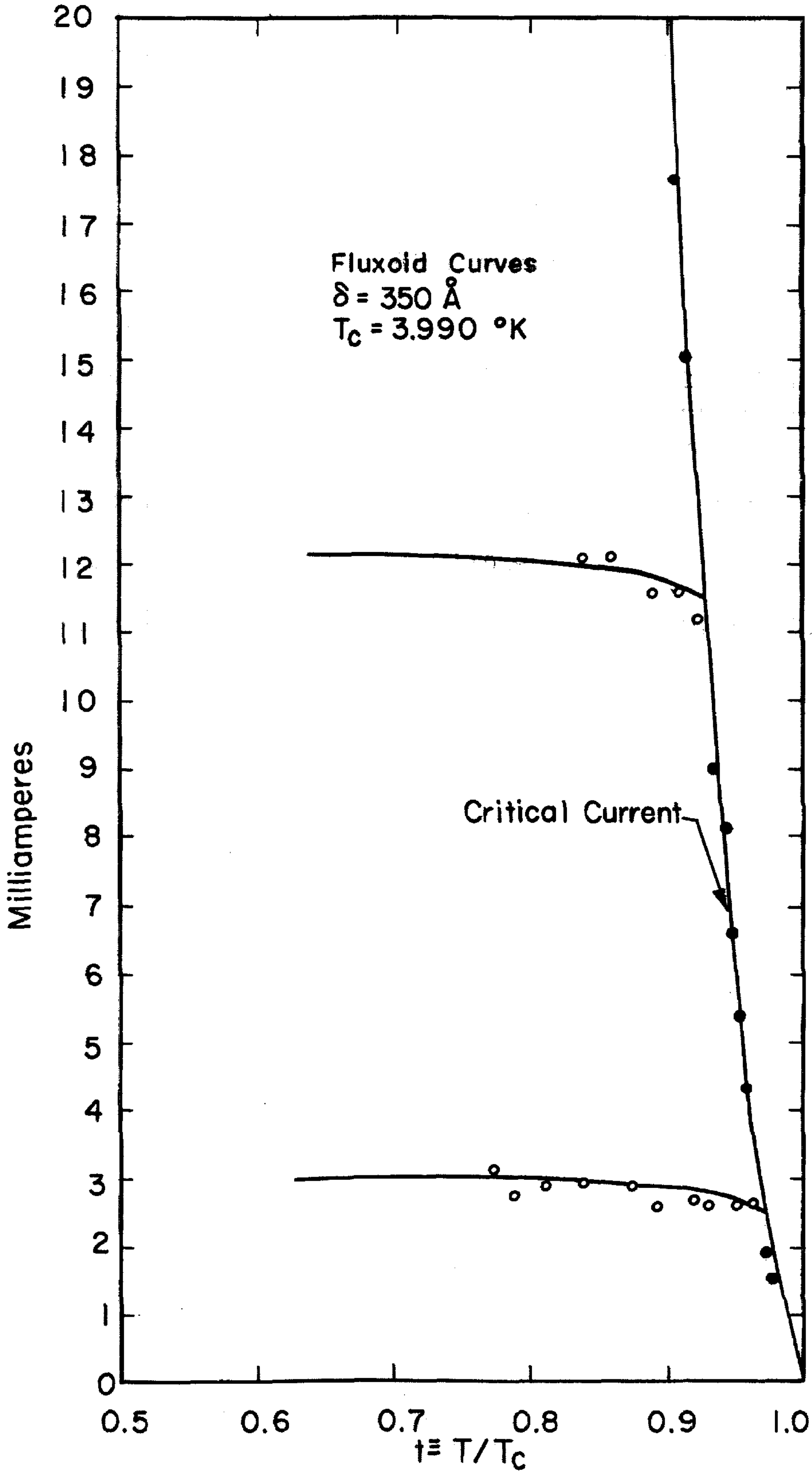

20
19
18
17
16
15
14
13
12
11
10
9
8
7
6
5
4
3
2
1
0
Milliamperes
Fluxoid Curves
δ = 350 Å
Tc = 3.990 °K
Critical Current
0.5
0.6
0.7
0.8
0.9
1.0
t≡ T/Tc

Fig. 12

Table I

Ring Parameters

Impurity Concentration	Film Thickness	$2\lambda_o^2/Kr\delta$	λ_o
~ 0	350 Å	$0 \rightarrow 0.015$	<4500 Å
1.0% indium	150 Å	0.082	7500 Å
1.9% indium	110 Å	0.12	7700 Å
up to 0.3% lead	310 Å	$0.12 \rightarrow 0.15$	~8400 Å

film with $\delta = 350$ Å yield a penetration depth well within the limits indicated in the table for the present $\delta = 350$ Å ring.

Another physical phenomenon which had not been observed prior to the present work is the existence of trapped magnetic flux in rings whose thickness is very much less than the magnetic field penetration depth. Though for these rings the field penetration is substantially complete the flux does not leak out. Currents only about 10% less than the critical value were observed to be persistent for periods as long as 10 hours in rings for which $\lambda/\delta = 20$ or more. Persistent currents have been observed in an alloy film with $\delta = 65$ Å which corresponds to an average thickness of the order of 15 to 20 atom layers.

Reversibility of the Change in the Persistent Current

After the observation of the expected decreases in persistent current due to fluxoid conservation it became of prime importance to investigate the possibility of reversing the process. Thus if the number k in Eq. 10 is a good quantum number under perturbations in the number of superconducting electrons (the basic effect of changing the temperature), then one must expect that the persistent current carried by a superconducting thin film ring will actually increase as the ring is cooled down, even in the absence of any applied magnetic field.

On the basis of a classical particle concept of the electron-pairs carrying the current it is somewhat difficult to visualize the mechanism by which the reconversion from mechanical to electromagnetic angular momentum takes place. For a ring with one fluxoid quantum trapped it seems clear that, since one either has the entire quantum or none of it, the current decrease predicted by Eq. 10 must be reversible. The situation seems somewhat less clear-cut for the case of 10^5 to 10^7 quanta. However, since even the case with a single quantum involves a macroscopic current with a very large number of electron-pairs taking part, the situation is basically the same as with the large quantum numbers.

Temperature Control and Thermal Overshoot

Because of the presence of the positive temperature gradient in the helium bath during the fluxoid measurements, it is not at all a simple matter to control experimentally the maximum temperature which the ring may reach in the warm-up which occurs during the fluxoid measurement process discussed previously. If it is desired to reduce the temperature following the measurement of a fluxoid curve one must pump on the helium bath. The boiling and convection associated with this pump-down process will tend to mix the warmer helium in the upper levels of the bath with the cooler helium at the ring level. It is therefore possible for a considerable temperature overshoot to occur, although the amount of the overshoot from run to run may well depend quite sensitively on the specific details of helium level, pumping speed and the temperature at which pumping begins. Because it is necessary always to maintain a positive temperature gradient during the actual measurement of persistent current, it is not possible to take a series of datum points as the temperature is lowered. The best one can hope to do is to pump back down to some low temperature, reestablish the positive temperature gradient and then observe whether the low temperature current significantly exceeds the lower current values measured on the original fluxoid curve.

Even if the phenomenon is fundamentally reversible, the thermal overshoot may lead to the sequence of events depicted in Figure 13a. Here the fluxoid curve is followed to point A as the ring warms up. At this point pumping is started. The thermal overshoot, however, drives the ring into the critical state at point B and on down the critical current curve, thereby changing the quantum number k as energy is dissipated in the ring. Once the maximum temperature is reached at point C, the ring cools down and even if the persistent current does increase, it presumably does so corresponding to its new quantum number $k' < k$. An observed value such as at D may thus be much less than the value last observed at point A. Concern over this potential difficulty, and preoccupation with the effort to use aluminum films below the λ point to avoid this problem, led to a significant postponement of the effort to use the above method to investigate the question of reversibility.

A series of experimental runs have been made to investigate the reversibility question by the method just discussed and by one other approach which will be described first. The process discussed above for trapping small currents may, with care, be used in a way which sets quantitative upper limits on the amount which can be trapped. The approach is illustrated in Fig. 13b. From an initial temperature of 4.2 K, the bath is cooled down slowly and at an accurately known temperature t_o one attempts to trap the entire critical current $i_c(t_o)$ using the second method discussed previously. If the current decrease phenomenon is irreversible, $i_c(t_o)$ would constitute the upper limit on

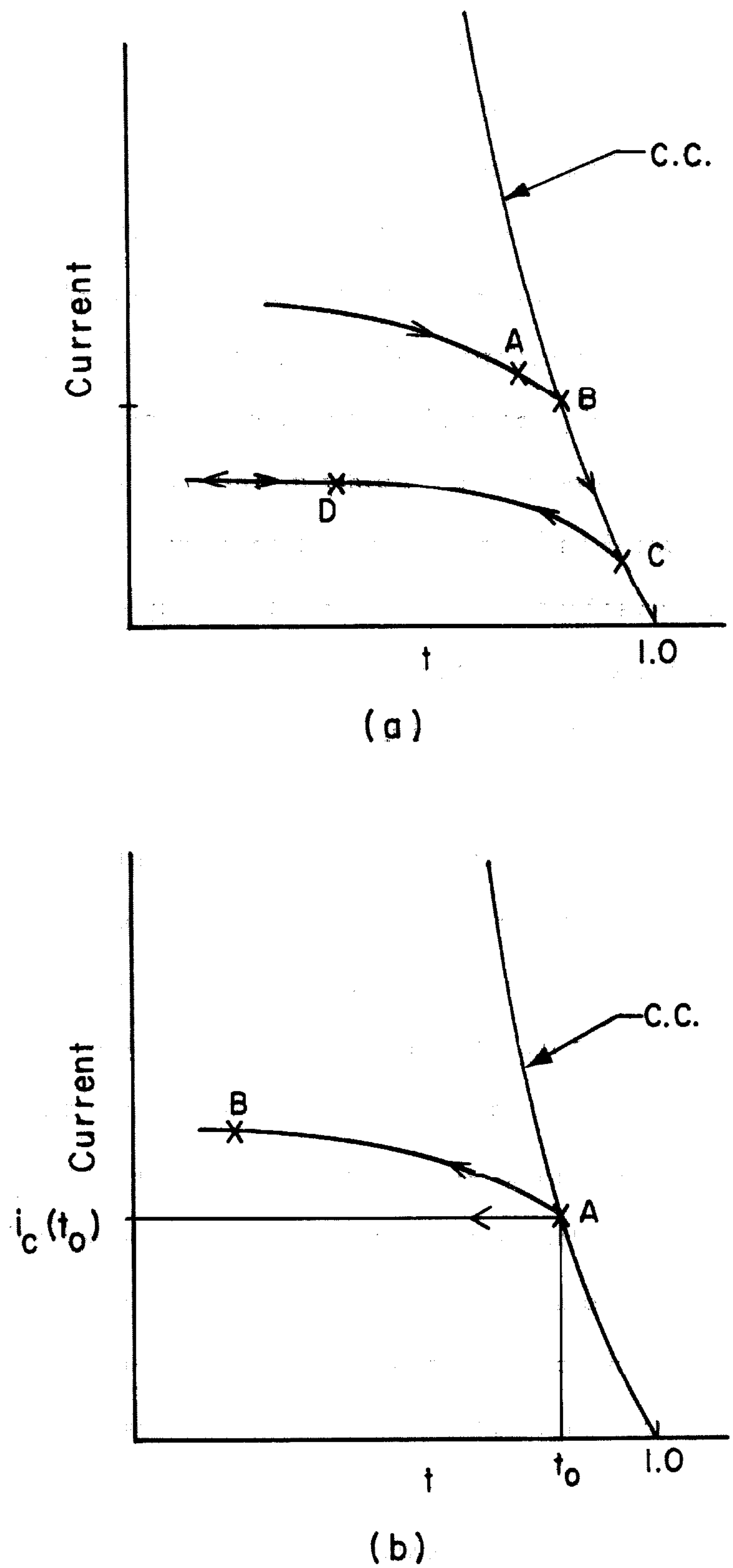


Fig. 13

the current which could ever be observed even at very low temperatures. On the other hand, if the phenomenon is reversible the upper limit at any given lower temperature is set by the fluxoid curve passing through $i_c(t_o)$ at $t = t_o$. A measured low temperature value lying significantly above the irreversible upper limit is thus to be considered an indication that the phenomenon is reversible. While a number of runs yielded values well below both limits due to incomplete trapping, Fig. 14 shows an instance for the $\delta = 110$ Å ring in which the low temperature current lies distinctly above the "irreversible upper limit" as indicated, although it is still below the "reversible upper limit." An ordinary fluxoid curve was observed during the warm-up for this run. The measurement of t_o was made just after the initial trapping in order to be clearly on the safe side with respect to the temperature reading since the critical current curve is quite steep at this temperature.

In spite of the anticipation of thermal overshoot problems a number of attempts were also made to cool down smoothly after a normal fluxoid experiment. Several of these runs confirm the presence of thermal overshoot amounting to several tenths of a degree by exhibiting significantly reduced persistent current values after pump-down to low temperatures. The results of a run made with a quite low helium level in the dewar and for which the pump-down was initiated at temperatures more than 0.2 K^o distant from the critical current curve are shown in Fig. 15. In this experimental run the point denoted by a cross was measured after pump-down, following the recording of the three circled points on the original fluxoid curve. It should be recalled that all points are necessarily taken with the positive temperature gradient established

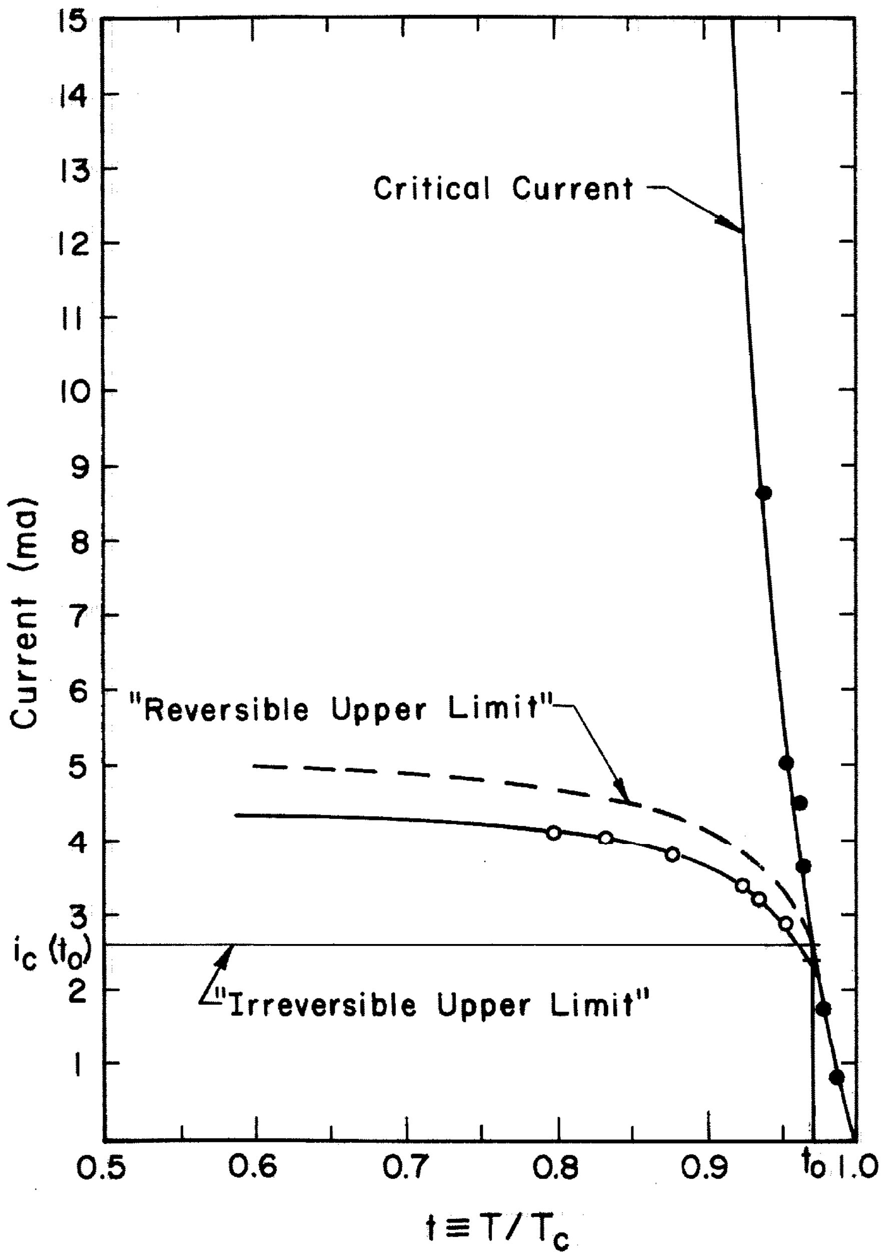

15
14
13
12
11
10
9
8
7
6
5
4
3
2
1
Current (ma)
Critical Current
"Reversible Upper Limit"
$i_c (t_0)$
"Irreversible Upper Limit"
0.5
0.6
0.7
0.8
0.9
t_0
1.0
$t \equiv T/T_c$

Fig. 14

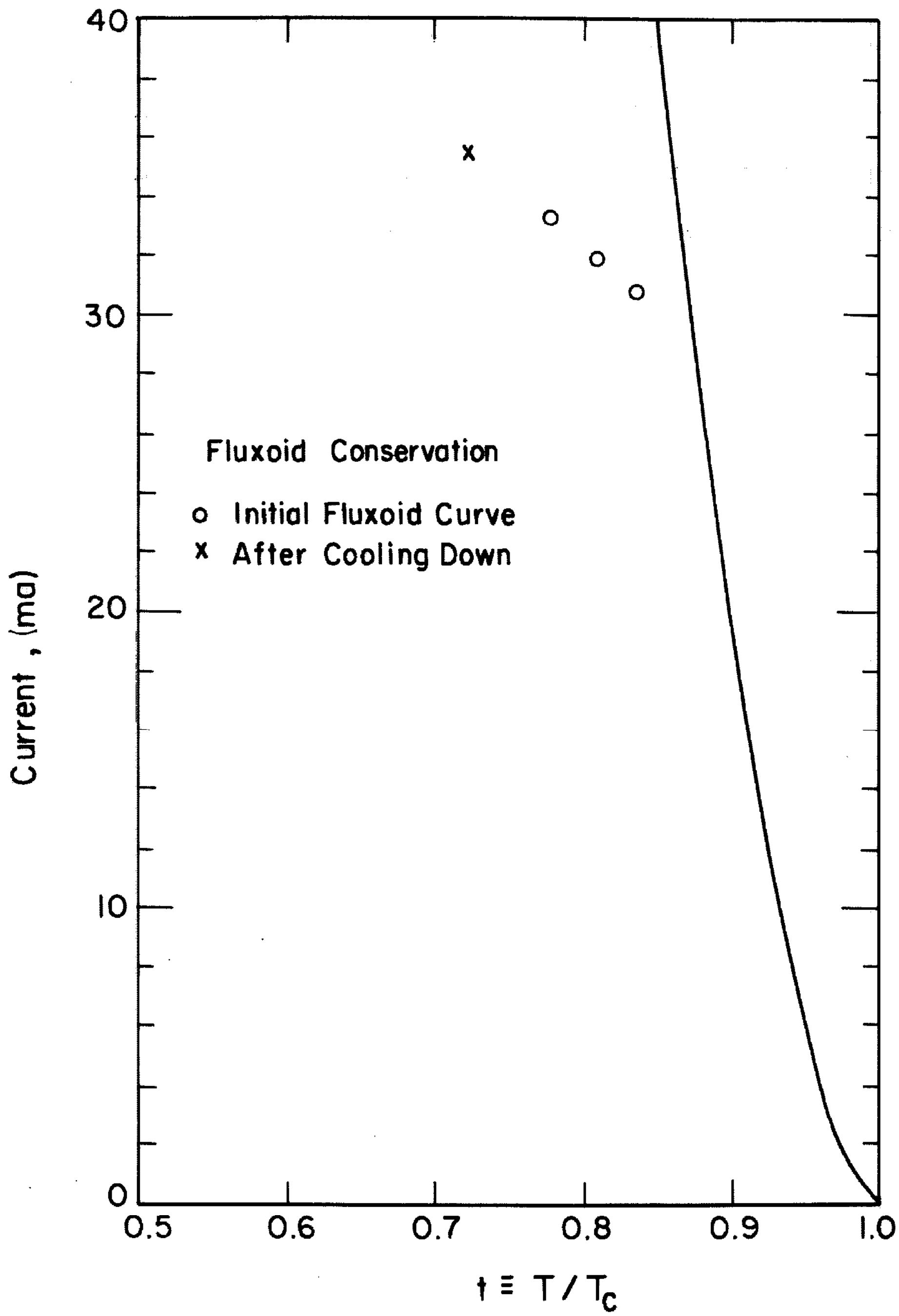

40
30
20
10
0
Current , (ma)
0.5
0.6
0.7
0.8
0.9
1.0
t ≡ T / T_c
Fluxoid Conservation
o Initial Fluxoid Curve
x After Cooling Down

Fig. 15

and hence yield "warm-up curves." The point shown with the cross lies well above the three points taken on the initial fluxoid curve and is definitely well beyond the error limits associated with the point and the original curve. The fluxoid curve could not be followed again in this particular run since by this time the helium level was quite low. Although the initial fluxoid curve for this run is not particularly smooth the observed increase in persistent current upon returning to lower temperatures is quite clear cut and the evidence for reversibility is quite definite.

V. CONCLUSIONS

For the first time persistent currents have been observed in thin film rings for which the film thickness is very much less than the penetration depth. In these films the field penetration is essentially complete and it is thus clear that the Meissner effect is not essential for the stability of persistent currents.

The prediction that the fluxoid through a superconducting ring is conserved has been verified by an investigation of the temperature dependence of the magnetic flux trapped by such rings in the pure superconducting state. The experimental results show clearly both the decrease in trapped flux with rising temperature and the increase in trapped flux with falling temperature which are a direct consequence of fluxoid conservation. Previous experiments carried out to observe flux "quantization" have explored only the properties of the electromagnetic part of the fluxoid since the effects of the mechanical term in those cases were negligible. The present experiments are sensitive to both terms comprising the fluxoid and thus fill in a conspicuous gap in the experimental picture of trapped flux and persistent currents in superconducting rings.

VI. Appendix A

Considerations in the Measurement of Critical Persistent Currents

The basic approach to critical persistent current measurement has already been discussed. There are two potential sources of difficulty with this experimental approach which should be discussed for the sake of completeness although they do not ordinarily affect the results in practice.

Catastrophic Flux Leakage Due to Heating

The decay of the persistent current in the ring as the critical current measurement proceeds is accompanied by the dissipation of energy, $dE = - LI\, dI$ in the material of the ring. The introduction of fluxoid quantization here merely establishes a minimum step for this dissipation. Since the ring and that part of the substrate with which it is in intimate contact have a finite heat capacity C, this energy dissipation will produce a small rise in the temperature of the ring and substrate above the ambient value in the helium bath. If this temperature change $dT = - (LI/C_1)\, dI$ is larger than the dT corresponding to the same current decrease along the critical current curve, then the new current will be larger than the critical value at the new ring temperature. As a consequence the decay will proceed further with no need for an increase in the helium bath temperature. The situation will be unstable in this way until a temperature is reached such that $(dT/dI)_{c.c.} > (dT/dI)_{decay}$. There is an inflection point in the critical current curve for a thin film (see Fig. 5 for a typical curve) and it is upon approaching this point from lower temperatures that such a catastropic current decay will most readily occur. Thus far in these experiments only one such event has been observed. On that occasion the magnitude of the jump was in approximate agreement with the prediction of this simple analysis based on a very rough estimate of the effective heat capacity C.

<u>Leakage Due to Oscillations of the Ring in a Magnetic Field</u>

It has been shown above that the measuring field B_m induces a counter current given by Eq. 12. If the ring position fluctuates, due to thermal noise, convection or boiling in the liquid helium, this counter current will also fluctuate. Let us consider the case in which the current at the equilibrium deflected position has the critical value, I_c, that is

$$I_e = I_t - \frac{AB_m}{L} \sin \theta_e = I_c .$$

If for some reason the ring is deflected to a larger angle (θ) then I decreases below I_c and all is well. If on the other hand the ring is deflected to smaller angles (θ), then (I) will try to increase past I_c. Since it cannot do this the ring must sacrifice some of its zero-field trapped current I_t. Then when equilibrium is again established it will be at a new deflection angle (θ_e') smaller than the original θ_e. This will correspond to a current less than critical since the bath temperature is assumed to have remained constant. Calculation of the deflection loss is straightforward starting from Eqs. 11 and 12. When a fluctuation decreases the angle θ by $\Delta\theta_f$ one finds from Eq. 12 that

$$\Delta I_t = \frac{AB_m}{L} \cos \theta_e \, \Delta\theta_f \, . \tag{18}$$

Initially we have the relation

$$I_e = I_t - \frac{AB_m}{L} \sin \theta_e \tag{19}$$

but after losing ΔI_t we have

$$I'_e = I_t - \Delta I_t - \frac{AB_m}{L} \sin \theta'_e \, . \tag{20}$$

Now, subtracting Eq. 19 from Eq. 20 and using Eq. 11 for I_e, we obtain

$$I_t - \frac{AB_m}{L} \sin \theta_e - I_t + \Delta I_t + \frac{AB_m}{L} \sin \theta'_e = \frac{K\theta_e}{AB_m \cos \theta_e} - \frac{K\theta'_e}{AB_m \cos \theta'_e} \, . \tag{21}$$

Collecting terms we may write

$$\Delta I_t = \frac{AB_m}{L} \left[\sin \theta_e - \sin \theta'_e \right] + \frac{K}{AB_m} \left[\frac{\theta_e}{\cos \theta_e} - \frac{\theta'_e}{\cos \theta'_e} \right]$$

$$= \frac{AB_m}{L} \cos \theta_e \, \Delta\theta_f \tag{22}$$

If we then expand the trigonometric functions in their power series, the lower order terms are given by

$$\frac{AB_m}{L} \cos \theta_e \, \Delta\theta_f = \left[\frac{AB_m}{L} + \frac{K}{AB_m} \right] (\theta_e - \theta'_e)$$

$$- \left[\frac{AB_m}{6L} + \frac{K}{2AB_m} \right] (\theta_e^3 - \theta_e'^3) \tag{23}$$

For small angles the expression simplifies to

$$\Delta\theta_e = \frac{AB_m/L}{\left[\frac{AB_m}{L} + \frac{K}{AB_m}\right]} \Delta\theta_f \qquad (24)$$

If we now compute this coefficient for a typical case for which

$$A = 0.7 \times 10^{-4}\ m^2, \quad B_m = 0.9 \times 10^{-5}\ w/m^2, \quad L = 2 \times 10^{-8}\ h,$$
$$K = 5 \times 10^{-10}\ n.m.\ ,$$

we get the relation

$$\Delta\theta_e = 0.4\ \Delta\theta_f \qquad (25)$$

It follows that, unless the random fluctuations are so large that they would by themselves seriously interfere with correct reading of the equilibrium deflection, the small loss in current due to this effect will be negligible. This appears to be well confirmed by the experimental observations.

It may be possible to make use of this phenomenon in an investigation of the time constant involved in the destruction or establishment of the superconducting state which is coherent all the way around the ring. With the ring carrying its critical current and deflected in a dc measuring field (B_m) it may be possible, by applying an orthogonal ac field, to periodically "superheat" the ring in the sense of the oscillating decay scheme just discussed. Some attempts to explain the negative flux quantization results of Mercereau and Vant-Hull (28) have involved the discussion of a time constant for the establishment of the coherent superconducting state which is longer than 0.2 milliseconds. If such a time constant is appropriate, one might hope to find a threshold frequency above which the decay indicated by Eq. 25 will not occur.

VII. Appendix B

Critical Persistent Current Results

Introduction

It has already been pointed out that quite precise knowledge of the phase boundary between superconducting and normal states is a prerequisite for the fluxoid experiments which have been discussed. This phase boundary is the critical persistent current as a function of temperature. In the early portion of this research therefore, considerable effort was spent in measuring and attempting to understand critical persistent currents in thin film rings. This effort was also desirable from the point of view of optimizing the current measurement procedure and investigating the validity of the assumptions made concerning the uniformity of the current distribution in these rings.

Calculations of the critical current depend in general on an analysis of the free energy difference between the superconducting and normal states as a function of current. For a bulk cylindrical ring, much thicker than the penetration depth the magnetic energy dominates. Silsbee's hypothesis implies that

$$I_c = bwH_c = bwH_o(1 - t^2) \equiv I_o(1 - t^2) , \qquad (26)$$

where b is a dimensionless constant which is nearly equal to one, but which differs from unity in experiments because of irregularities in the surface and shape of actual specimens. The length of the cylinder as usual is w. In Eq. 26, expansion of the $(1 - t^2)$ term near $t = 1$ shows that the critical current in bulk rings should vary linearly with $\Delta T \equiv (T_c - T)$

in this region.

For the case of thin film rings in which $\lambda >> \delta$, the kinetic energy of the electrons rather than the magnetic energy of the excluded field, dominates the free energy difference. The resulting expression for the critical current as calculated on the basis of the Ginzburg-Landau theory (23) is

$$I_c = \left(\frac{2}{3}\right)^{3/2} wH_c \frac{\delta}{\lambda} = \left(\frac{2}{3}\right)^{3/2} wH_o \frac{\delta}{\lambda_o} (1 - t^2)(1 - t^4)^{1/2} . \qquad (27)$$

If the right-hand side of Eq. 27 is expanded near $t = 1$ it predicts that in this region the critical current should be proportional to $(\Delta T)^{3/2}$, a result which is also obtained by the BCS theory.

Measurements

Figure 16 shows the critical current in a bulk indium ring for which $w = 1$ mm, the outer radius is $r = 5$ mm, and $\delta = 0.4$ mm. The solid curve is a plot of Eq. 26 with I_o chosen to fit the data, and the agreement is quite satisfactory. The value H_o is well known from magnetic measurements and the value of b which may thus be determined from the fitted value of I_o is $b = 0.93$ for this ring. When particular care is taken with the measurements very near the critical temperature and the critical current is plotted on an expanded linear scale in this region, one obtains, for bulk rings of different superconductors and widely differing geometries, results of the general form shown for a bulk aluminum ring in Fig. 17. For values of $\Delta T \lesssim 0.02$ Kelvin degrees the critical current in the aluminum ring clearly deviates from the linear behavior expected throughout this range on the basis of Eq. 26. It is at about this temperature that the penetration depth, increasing near T_c, should become equal to the coherence length (ξ_o) for bulk aluminum. The situation $\lambda > \xi_o$ satisfies the criterion for applicability of the local Ginzburg-Landau theory which predicts, as before, $I_c \propto (\Delta T)^{3/2}$. In Fig. 18, the data points for the bulk aluminum ring are replotted on log log scales. The data for two runs indicated respectively by the circles and crosses yield a slope of 1.43 in rather satisfactory agreement with the 3/2 value which might be expected on the basis of the Ginzburg-Landau theory.

Comparisons between various thin film samples are facilitated when the critical persistent current data are plotted in terms of the current density, j_c, rather than the current itself. This has been done in Fig. 19 as a function of reduced temperature for a variety of thin film tin rings. The two curves shown for $\delta = 540$ Å were made on the same ring which was shortened between runs by scraping a length of the film cylinder from the substrate. Within the experimental uncertainties and near T_c, these curves indicate that the average critical persistent current density is independent of both film cylinder length (w) and thickness (δ). Mercereau and Crane (21) have more recently extended these results to length variations by a factor of 100 obtaining values which continue to lie on this seemingly standard curve. This evidence tends to support the assumption that the current density is essentially uniform in these films. Furthermore, at least down into this thickness range, the character of tin films evaporated onto substrates at liquid nitrogen temperature does not appear to change appreciably. Near the critical temperature, the critical persistent current exhibits very closely the $(\Delta T)^{3/2}$ behavior predicted by the theory. Figure 20 is a plot of I_c versus ΔT on log log scales and, as indicated, the slopes give very satisfactory agreement with the predicted value, 1.5. It must be pointed out, however, that the actual magnitude of these critical current densities is not in agreement with the predictions of Eq. 27. Equation 27 and other presently available theories over-estimate j_c by a factor of from 5 to 10.

With the exception of the curve on the shortened δ = 540 Å ring, each of the curves in Fig. 19 exhibits an apparent "saturation" effect at low temperatures. This effect has been observed in every thin film sample in sharp contrast to the observed behavior in bulk rings. Below a certain temperature which is characteristic of the film sample, the persistent critical current, measured in this way, becomes very nearly independent of temperature. The Ginzburg-Landau theory is supposed to be valid only near T_c so that its failure to predict this saturation behavior is not at all surprising. Thus far no theory adequate for the discussion of this effect is available. The curve on the shortened 540 Å ring illustrates the typical results observed when a current less than critical is trapped at low temperatures. When a current less than the full critical value is trapped in a ring for which the quantity $2\lambda_o^2/Kr\delta$ is very small, no change in the current is observed until the critical state is reached as the ring warms up. When the maximum possible current is trapped in this 540 Å ring, a "saturation" curve of the typical form is observed. It is interesting to note that the electron drift velocity is given by $v_d = \mu_o \lambda^2 j_c (e/m)$ and that the value corresponding to the "saturation" j_c is approximately 10^5 cm/sec. This is very nearly equal to the speed of sound in tin. It is perhaps possible that electrons which are urged to move faster than the phonon velocity may in some way radiate "Cerenkov" phonons, thus dissipating energy.

The experimental data on critical persistent currents are quite consistent but no detailed explanation of the magnitude of the current density or of the saturation phenomenon has been found.

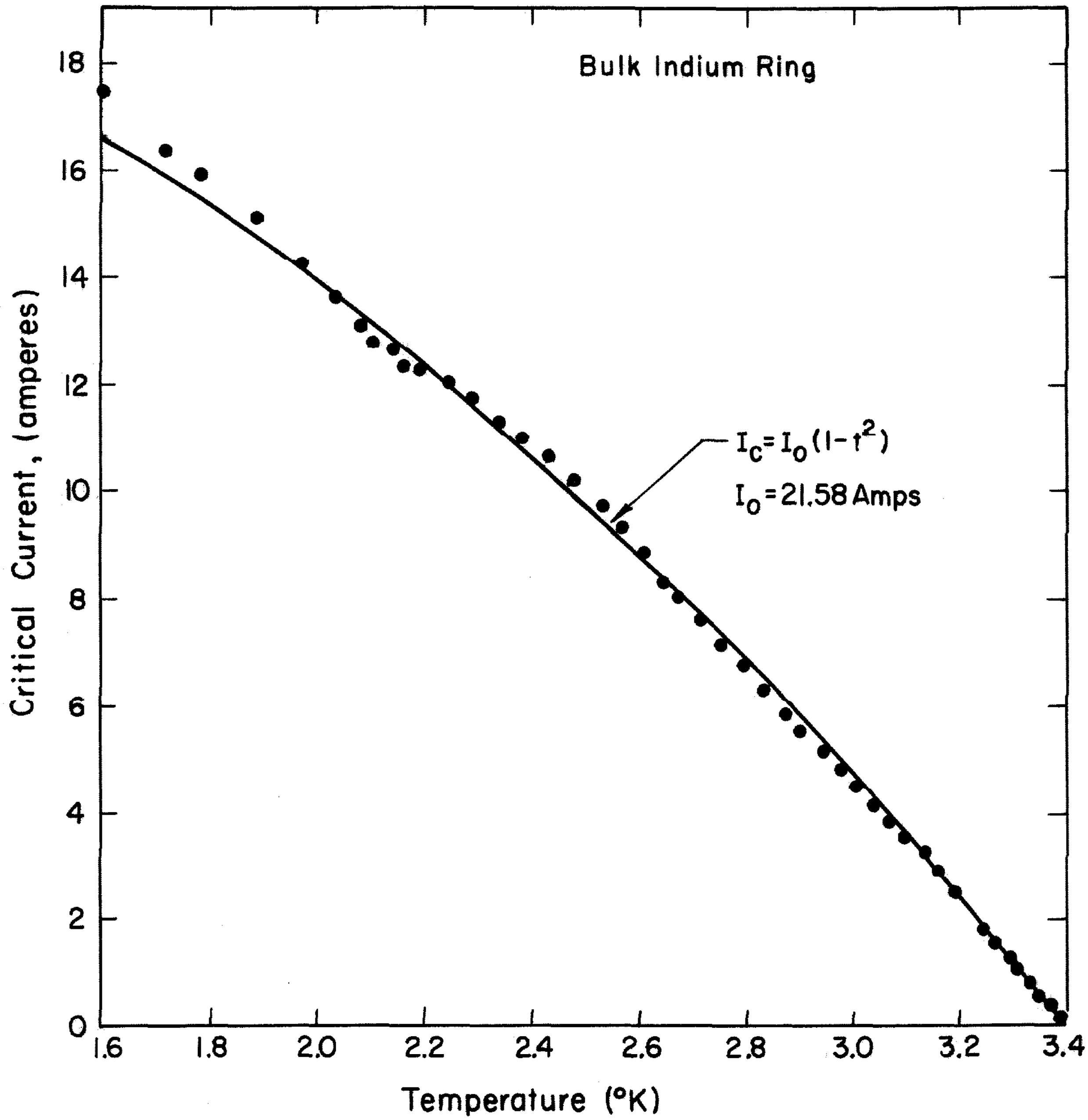
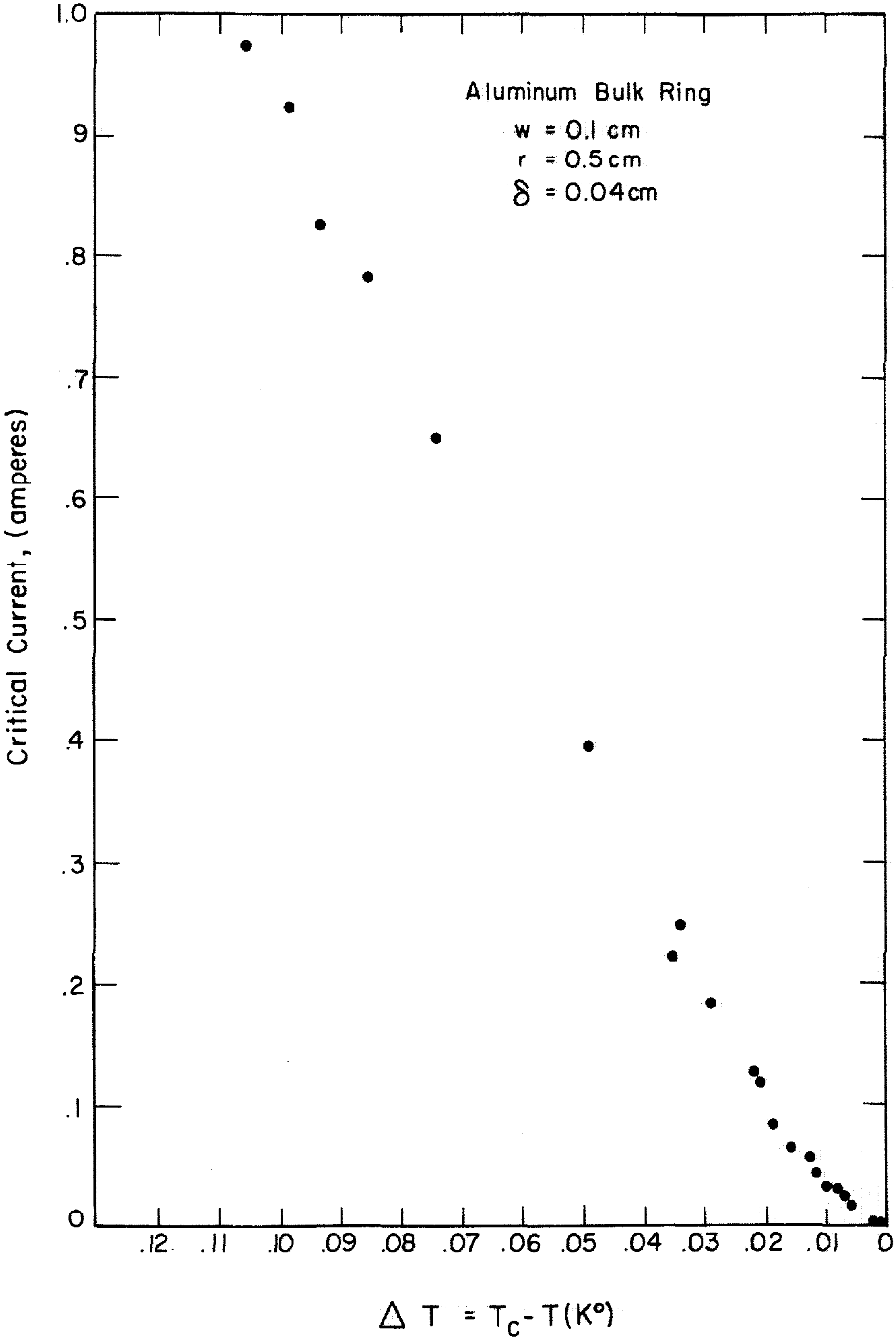
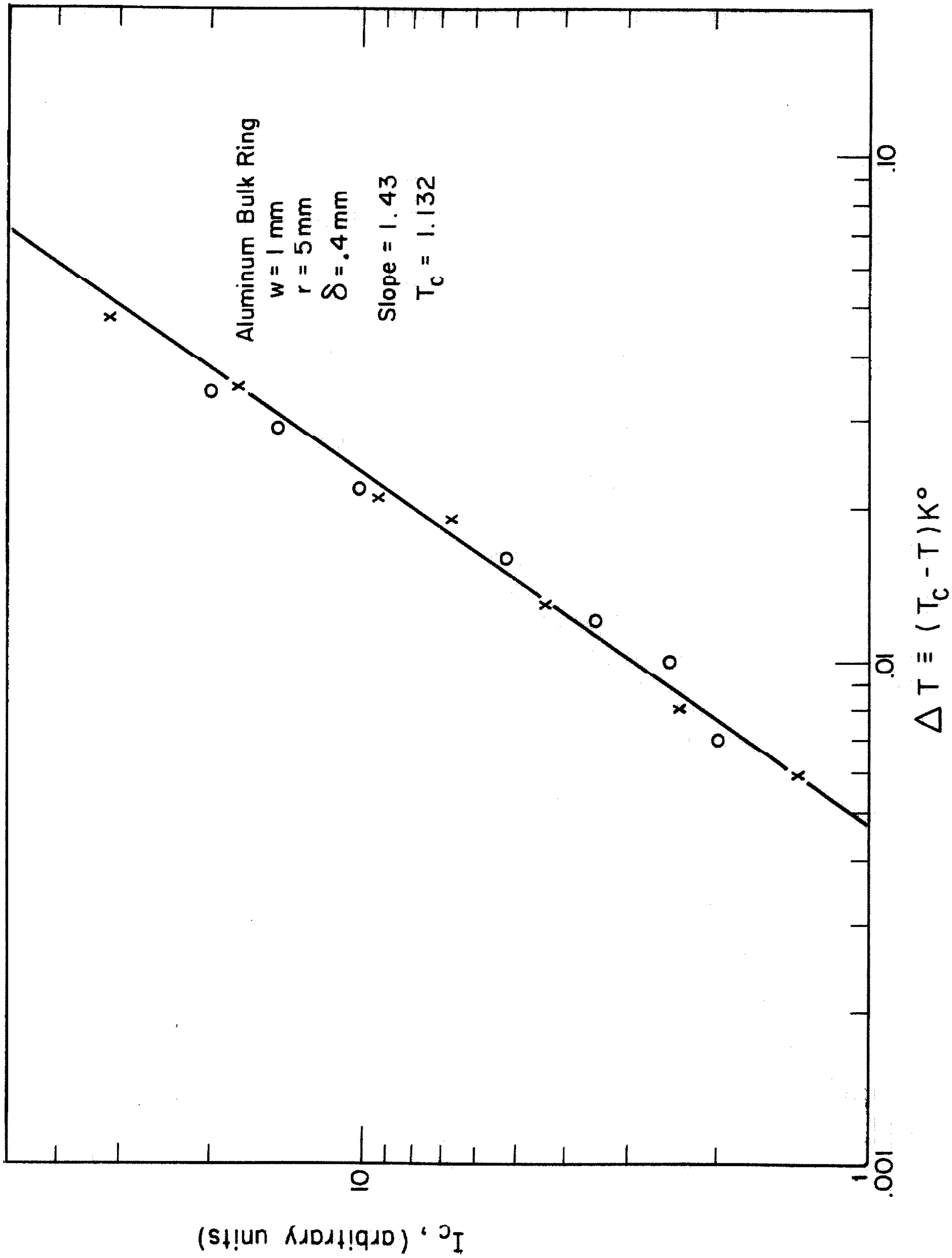

Bulk Indium Ring
$I_c = I_0(1-t^2)$
I_0 = 21.58 Amps
Critical Current, (amperes)
Temperature (°K)
18
16
14
12
10
8
6
4
2
0
1.6
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4

Fig. 16



1.0
9
.8
.7
.6
.5
.4
.3
.2
.1
0
Critical Current, (amperes)
Aluminum Bulk Ring
w = 0.1 cm
r = 0.5 cm
δ = 0.04 cm
.12 .11 .10 .09 .08 .07 .06 .05 .04 .03 .02 .01 0
ΔT = T_C - T(K°)

Fig. 17



Aluminum Bulk Ring
w = 1 mm
r = 5 mm
δ = .4 mm
Slope = 1.43
T_c = 1.132
I_c, (arbitrary units)
10
1
.001
.01
.10
$\Delta T \equiv (T_c - T) K°$

Fig. 16

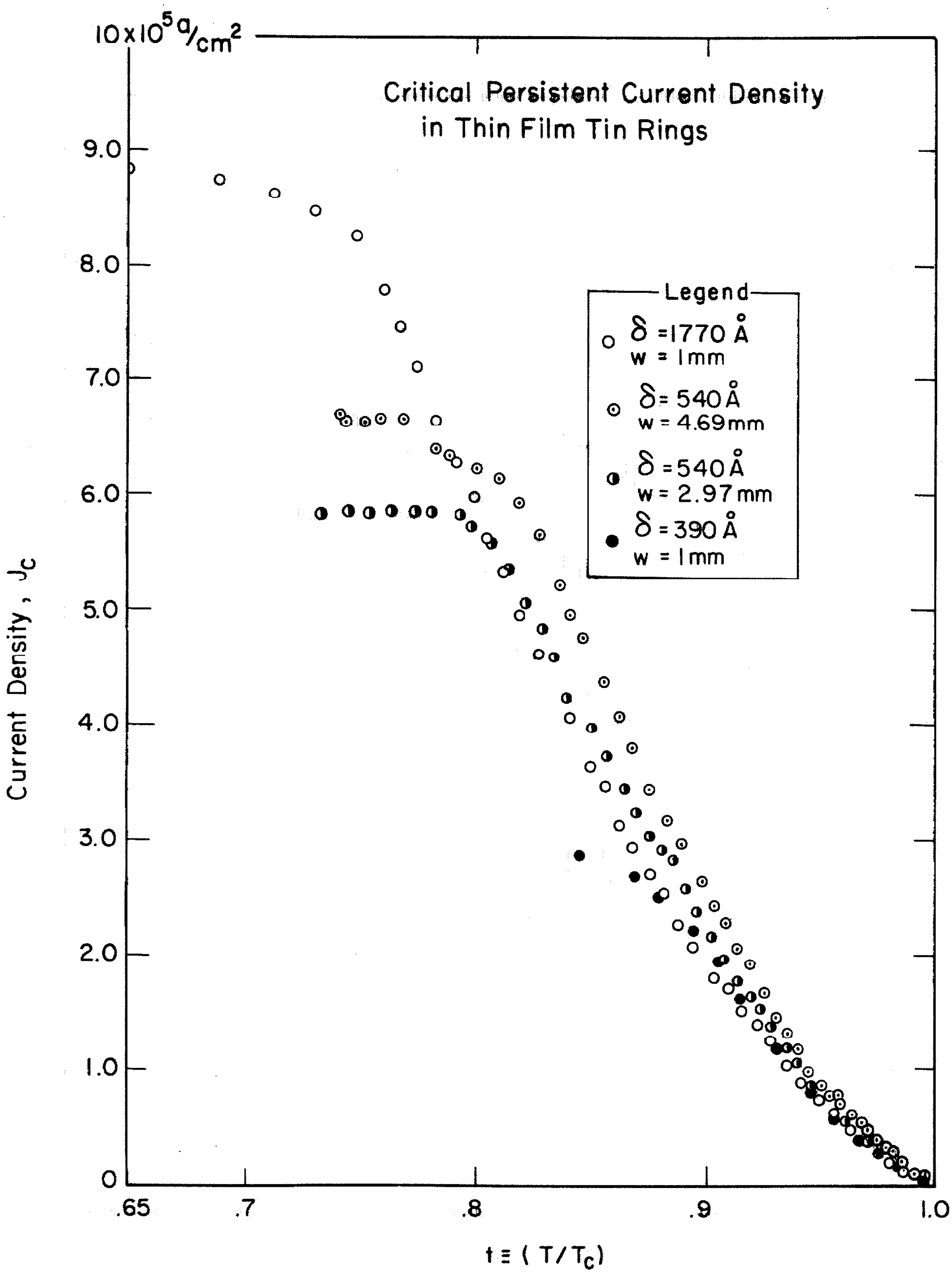
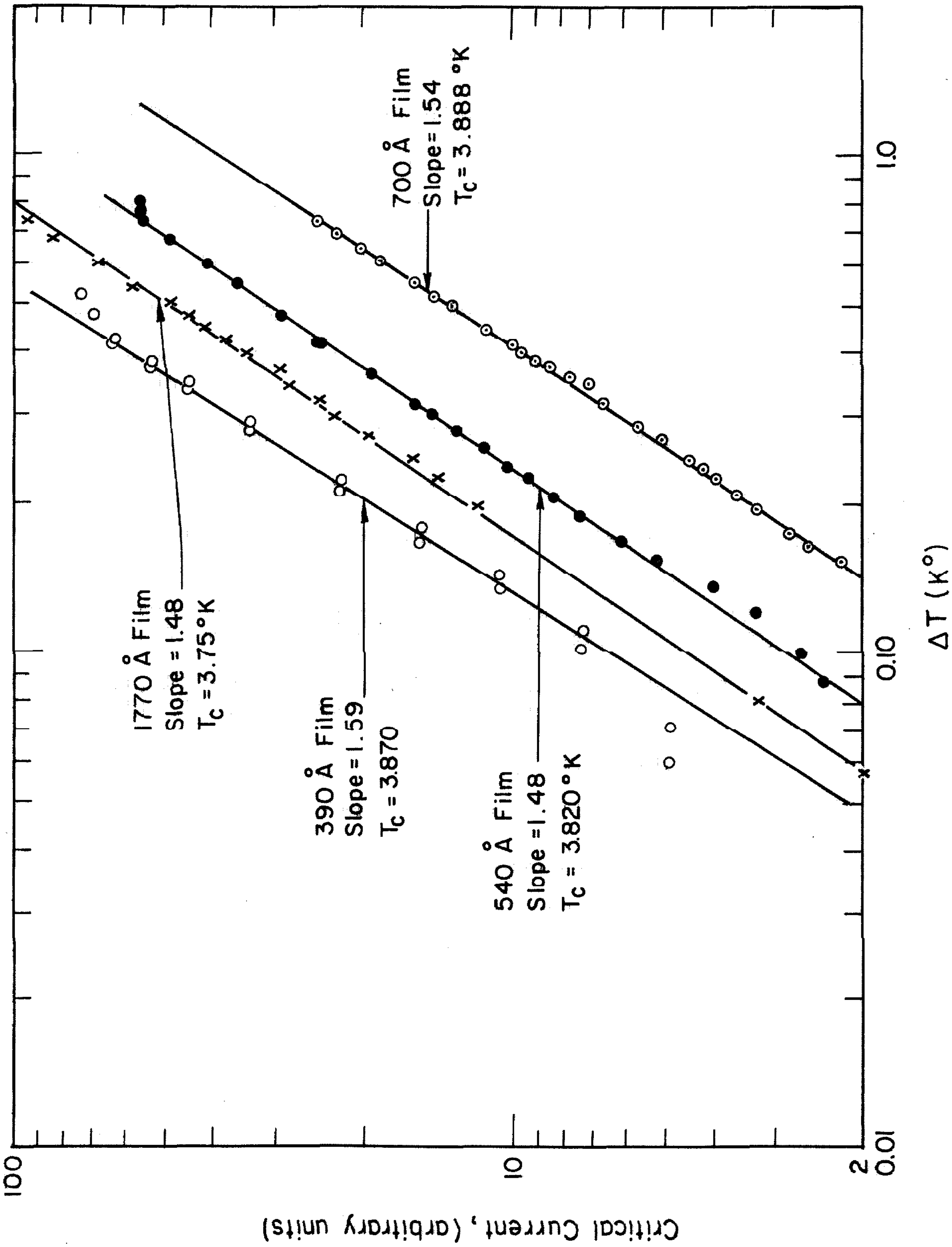

Critical Persistent Current Density
in Thin Film Tin Rings
10x10^5 a/cm^2
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
.65
.7
.8
.9
1.0
Current Density, J_c
t ≡ (T/T_c)
Legend
δ = 1770 Å
w = 1mm
δ = 540 Å
w = 4.69mm
δ = 540 Å
w = 2.97mm
δ = 390 Å
w = 1mm

Fig. 19



700 Å Film
Slope = 1.54
T_c = 3.888 °K
1770 Å Film
Slope = 1.48
T_c = 3.75 °K
390 Å Film
Slope = 1.59
T_c = 3.870
540 Å Film
Slope = 1.48
T_c = 3.820 °K
ΔT (K°)
Critical Current, (arbitrary units)
100
10
2
0.01
0.10
1.0

Fi . 20

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