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John Clarke
April 1968

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April 1968

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ABSTRACT

The resistance of thin-film lead-copper-lead junctions has been studied with the lead in the superconducting state. The junctions will sustain a supercurrent up to a certain critical value above which a voltage appears, rising smoothly from zero as the current is increased. The effect of a magnetic field upon the critical current has demonstrated that the sandwiches behave phenomenologically as Josephson junctions. The critical current rises rapidly as the temperature is lowered, decreases exponentially with increasing thickness of copper and increases with increase of the mean free path of the copper. A simplified version of the de Gennes theory of the proximity effect has been used to account quantitatively for this behaviour. The experiments show that the coherence length of the paired electrons in the copper increases as the temperature decreases, implying that thermal fluctuations govern the decay of the pairs. From the value of the decay length, the interaction parameter in copper is estimated to lie between +0.06 and +0.14. The properties of these junctions are compared with those of junctions with insulating barriers.

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I. INTRODUCTION

When a non-magnetic normal metal (N) is in good electrical contact with a superconductor (S), Cooper pairs are able to diffuse into the normal metal which then exhibits some superconducting properties. In addition, the presence of the normal metal tends to lower the Cooper pair density in the superconductor in the vicinity of the contact. This phenomenon has become known as the proximity effect.

Two classes of experiments have been used in the investigation of the effect. In the first, the transition temperatures of superposed thin films of normal and superconducting materials were measured (for example, Hilsch 1962). It was found that provided the superconductor was not too thick, its transition temperature was significantly lowered by the presence of the normal metal or even became too small to be measured. This effect demonstrated the quenching of the superconductivity by the adjacent normal metal. In the second type of experiment, an oxide layer was grown over the "normal" side of a NS sandwich and a second normal metal deposited on top of the barrier so as to form a tunnelling junction. The single-particle $i-v$ characteristics of this junction were related to the electronic density of states at the surface of the sandwich. It was found that the excitation spectrum contained a gap (for example, Adkins and Kington, 1966), clearly demonstrating the existence of paired electrons in the normal metal.

These experiments have shown that the ordering of the electrons may extend into a normal metal adjacent to a superconductor over distances of at least several hundred Angstroms. This fact leads us to propose a third type of experiment in which a normal metal is sandwiched between two superconductors. Provided that the normal metal is not too thick, we

might expect to be able to pass a supercurrent between the two superconductors through the normal metal. Preliminary experiments of this kind have been undertaken by Meissner (1958, 1960) who measured the resistance between two copper-plated tin wires which were pressed together at right angles. He showed that at low temperatures, the contacts were resistanceless for thin enough layers of copper. However, because of the uncertainties in the contact area and the thickness of the oxide layer between the metals, the results of these experiments are difficult to interpret quantitatively. The present paper describes a series of detailed experiments on thin-film lead-copper-lead sandwiches. It was discovered that under appropriate conditions the sandwiches were able to sustain a supercurrent but that a voltage appeared across the junction once a certain critical current was exceeded.

There are of course obvious analogies between this system and a Josephson junction (Josephson 1962) consisting of an insulator separating two superconductors. However, we shall not assume a priori that we are presently concerned with Josephson-type behaviour but prefer to first describe the experimental properties of the sandwiches and then to discuss the possible theoretical interpretations of the results.

An outline of these results was given at the Conference on the Electronic Properties of Thin Films held at Orsay, France, in June 1967 and will appear in the conference proceedings. This paper also contains a review of the proximity effect experiments mentioned above.

II. THE PREPARATION OF SPECIMENS

Choice of materials

The choice of the superconductor and normal metal from which specimens are to be made is of the highest importance. It is essential to select two metals which do not form intermetallic compounds (Chiou and Klockholm 1964, 1966) and whose mutual solubility is very low so that the diffusion of one into the other is negligible (Rose-Innes and Serin 1961). In addition, if the prepared specimen has to be exposed to the atmosphere prior to the experiment, the materials should not form a very active electrochemical cell, the action of which tends to oxidize the more anodic metal at the interface (Hauser et al. 1966). The superconductor is required to have a convenient transition temperature and the normal metal to be non-magnetic. Finally, it must be possible to evaporate both materials without too much difficulty.

There are in fact remarkably few pairs of materials which satisfy these criteria: Lead and copper were eventually chosen as the superconductor and normal metal, respectively. The solid solubility of lead in copper is not higher than 0.29 wt.% above 600°C and of copper in lead, less than 0.007 wt.% (Hansen 1958). In order to test the quality of a lead-copper interface, experiments were performed to estimate its equivalent scattering length. Pb-Cu-Pb sandwiches were prepared by applying a small quantity of lead to each side of an annealed copper sheet with a soldering iron. The sheet was typically 1/10 mm in thickness, much too thick to support a supercurrent, with a resistance ratio of 5,000. The resistance of the sandwich was measured with the aid of a superconducting voltmeter (Clarke 1966) and the interface resistance calculated by

subtracting out the bulk resistance of the copper. It was found that the "boundary resistance" was equivalent to roughly one mean free path of copper. This result indicates that boundary disorder and diffusion of lead into copper are not too serious. Similar experiments performed on In-Cu-In specimens yielded an interface resistance two orders of magnitude greater, presumably because of the much higher solubility of indium in copper. Thus Cu/In specimens are unsuitable for proximity effect experiments.

It was intended to reduce the effects of boundary disorder and interdiffusion in the proximity effect specimens by using materials of very short mean free path. This also has an advantage from a theoretical viewpoint in that the electronic motion is governed by a relatively simple diffusion equation. It was initially hoped to "dirty" the materials by alloying about 3% of aluminium with the copper and 10% of bismuth with the lead; both alloys would then have a mean free path of about 100\AA at helium temperatures. Unfortunately, minute traces of bismuth dissolved in copper have a very powerful embrittling effect (Voce and Hallows 1947) and it was found that Cu/Al-Pb/Bi specimens tended to be unreliable mechanically. There appears to be no other suitable material which adequately shortens the mean free path of lead and it was finally decided to use pure lead as the superconductor. The copper alloy was retained as the normal metal and contained 3.35% wt of aluminium. All three metals were 99.999% pure.

A number of authors (for example, Hauser et al. 1964, Hilsch 1962) have obtained specimens with short mean free paths by evaporating their films at low temperatures. This technique also has the advantage of greatly reducing the interdiffusion of superposed layers. However,

Bassewitz and Minnigerode (1964) have demonstrated that copper deposited on a substrate at 77°K has a density of about two-thirds the bulk value. Consequently, the results of experiments on films prepared and kept at low temperatures, although they may provide excellent evidence for the existence of the proximity effect, cannot be used to deduce information on the bulk properties of the materials. The specimens used in the present experiments were all prepared at room temperature so that the evaporated films had densities of approximately the bulk values (Bassewitz and Minnigerode 1964).

Specimen configuration

Each specimen was prepared by evaporating successively onto a water-cooled 3"x1" glass slide a 7000Å strip of lead (0.2 mm wide), a disc of copper/aluminum alloy (5 mm diameter) and a second strip of lead at right angles to the first (see Fig. 1). The specimen area was thus defined by the overlap of the two lead strips, to within an error of the order of the ratio of the copper thickness to the width of the lead strip (not greater than 1/2%). Six specimens of various thicknesses were connected in series on each slide and the voltage across all six monitored. The critical current of each junction was determined by passing a current between the appropriate pair of lead strips and noting the value at which a voltage first appeared; for reasons which will emerge later, the current was divided equally between the ends of each lead strip, as is indicated in Fig. 1.

That part of the copper film which is not included in the junction contributes a conductance in parallel with it. This conductance will not

affect the magnitude of the critical current and an order-of-magnitude calculation indicates that the attenuation of the voltage produced when the junction becomes resistive is negligible.

Determination of mean free path

For each film evaporated, a strip was deposited simultaneously on the same glass slide. The residual resistivity (ρ) of each film was estimated from the resistance of the appropriate strip measured in a four-terminal arrangement at low temperatures. The mean free path (ℓ) was then estimated from the relations $(\rho\ell)^{-1} = (9.4 \pm 0.7) \times 10^{10} \Omega^{-1} \text{cm}^{-2}$ for lead and $(\rho\ell)^{-1} = (15.4 \pm 0.4) \times 10^{10} \Omega^{-1} \text{cm}^{-2}$ for copper (Chambers 1952). The lead had a mean free path of about 10,000Å, much greater than its coherence length, ξ_S , so that it was in the clean limit. The copper alloy was in the dirty limit, having a mean free path of 100Å.

This method of determining the mean free path in the Cu/Al films has some disadvantages. The measurement was not made on the actual material of the junction and the two films may have had a different structure, being deposited on different substrates. In addition, the resistance was measured longitudinally rather than transversely; if the films were at all non-uniform, as seems likely in practice, this measurement gives an incorrect value. The uncertainties in the mean free path, perhaps as high as $\pm 20\%$, were the largest in the whole experiment.

The evaporations

The specimens were made in a Varian Associates V12 ultra high vacuum system, which was baked out for 24 hours prior to the evaporations. The pressure typically rose from 10^{-10} torr to 10^{-8} torr during the course of the evaporations, despite careful outgassing of the boats and materials.

The time interval between successive evaporations was usually about 20 sec, essentially the time required to rotate the mask changer, and it is thought that much less than a monolayer of oxide formed at the SN interface during this period. The lead was evaporated from an electrically heated molybdenum boat. The Cu/Al films were prepared by evaporating to completion tiny pellets dropped into a tantalum boat from a drum operated by a magnetic feed-through from outside the evaporator. It was hoped that this technique would produce alloy films of greater homogeneity. The glass slide onto which the materials were evaporated was firmly clamped to a water-cooled copper block to prevent any temperature rise due to heat radiated from the boats. The thickness of each film was estimated from the change in resonant frequency of a quartz crystal onto which the material was simultaneously deposited (Haller and White 1963). The absolute thickness of the films (assuming bulk values of density) was thought to be accurate to $\pm 5\%$ and the relative thicknesses to $\pm 2\%$.

When the evaporations were complete, the slide was quickly transferred to the cryostat, wired up, and cooled to nitrogen temperatures in an atmosphere of helium; the total exposure to the atmosphere was about 30 min. The various leads were attached to the films by means of small pellets of indium, with the exception of the superconducting wires connected to the superconducting voltmeter, for which 50/50 Pb/Bi alloy was used. This material has a higher transition temperature than pure lead so that measurements could be continued right up to the transition temperature of the specimens.

Asymmetry of specimens

The method of specimen preparation just described may give rise to some degree of asymmetry (Hauser et al. 1964, 1966; Hilsch and Hilsch 1964). A certain amount of oxide will inevitably develop on a freshly-deposited metal before the next layer is evaporated. The oxide growth-rate on lead is greater than on copper, so that there may be different amounts of oxide at each SN interface. It is hoped that the high vacua used rendered this effect relatively unimportant. The second cause of asymmetry is the dependence of the morphology of a thin film on the nature of its substrate; lead deposited on copper may nucleate in a different way from lead on glass. However, since the lead was in the clean limit, slight differences in its structure should not introduce serious discrepancies.

3. EXPERIMENTAL APPARATUS

The cryostat

The cryostat was designed to operate between 1.2°K and 8°K and a longitudinal section of the lower part is shown in figure 2. A thin-walled 3 cm diameter copper-nickel tube, sealed at the lower end, supported a vacuum can in the helium bath. The specimen slide, mounted on a copper plate, was lowered into the tube on a thin stainless steel pipe attached to a gas-tight plug at the top of the 3 cm tube. An expanded polystyrene plug separated the slide from the voltmeter which was maintained at the temperature of the helium bath by the exchange gas (helium at a pressure of about 5 torr) in the 3 cm tube. The plug was not gas-tight but inhibited convection of the exchange gas. The temperature of the slide was raised above that of the bath by means of a heater mounted on the

reverse side of the copper plate and measured by a germanium thermometer (Texas Instruments, Model 340, Type 106), calibrated against a second standardized thermometer.* It is thought that the temperature measurement was accurate to $\pm 20^\circ\text{K}$. The current in the heater was controlled by a servomechanism deriving its input from an Allen-Bradley resistance thermometer to give a stability of $\pm 3\text{m}^\circ\text{K}$ and a time-constant of a few seconds.

The magnetic field in the cryostat could be reduced to less than 5 mG by means of two Helmholtz pairs and a mu-metal can outside the nitrogen dewar. A lead can around the specimen chamber, in weak thermal contact with the helium bath, could be used to freeze in "zero field" by warming it to above its transition temperature and cooling it down again. A solenoid wound on the 3 cm tube enabled fields of a few gauss to be applied to the junctions.

The voltmeter

When the current through a junction exceeded the critical value, the voltage developed was detected by means of a superconducting galvanometer in series with a resistance of $10^{-7} \Omega$ (Clarke 1966). The sensitivity was 10^{-13} V and the time-constant about 0.3 sec.

* The writer gratefully acknowledges the loan of a calibrated germanium thermometer by Dr. C. R. Barber of the National Physical Laboratory, Teddington, Middlesex.

4. EXPERIMENTAL RESULTS

i-v characteristics

The i-v characteristic of a typical sandwich, measured with a current source, is shown in figure 3. The voltage developed when the critical current was exceeded was small; this is to be expected because of the very low resistance of the metal barrier. Figure 4 shows on a larger scale the region around the critical current; there was no voltage step but rather a continuous rise in voltage as the current was increased from the critical value.

The estimated resistance of the barrier was $7 \times 10^{-7} \Omega$ and the measured differential resistance at currents well above the critical current approximately $5 \times 10^{-7} \Omega$.

Critical current measurements

The measurements made of critical current* are presented in figures 5-10. With the exception of the last two graphs, each point represents an average value of critical current obtained from two specimens prepared simultaneously. The agreement well below the transition temperature of the sandwiches (T_{CNS}) was usually to within $\pm 10\%$ but near T_{CNS} , the discrepancy was often much greater. This was attributed to the presence of strains set up in the films by differential contraction during cooling which affected the transition temperature.

Figure 5 shows the variation of critical current density with temperature for twelve thicknesses of Cu/Al. The estimated mean free path of the latter is seen to vary by as much as $\pm 30\%$ from specimen to

* We denote measured critical current by i_c and critical current density by I_c .

specimen; the importance of this fact is illustrated by the apparent interchange of the curves obtained from the two pairs of specimens of greatest thickness. It also seems more than likely that variations in mean free path of the same order will exist across the copper film of individual specimens. This would probably explain why some of the curves do not seem to follow the general trend and, in particular, why two of them intersect. The curve for the specimens of thickness $3,520 \text{ \AA}$ seems particularly anomalous and has in fact been neglected in the subsequent analyses. We can give no other reason for its unusual behaviour. Figure 6 further demonstrates the importance of the mean free path of the copper in determining the critical current; even though it is thinner, the specimen with the shorter mean free path has a lower critical current.

Figure 7 illustrates an experiment performed to investigate the effect of an oxide barrier at one of the SN interfaces. Two pairs of specimens were prepared, the first under standard conditions. The second pair differed in that the evaporator was opened to the atmosphere for 15 mins prior to the deposition of the upper lead strip, so that the junctions were identical to the first pair except for the presence of an oxide layer at one of the copper-lead interfaces. As we might expect, the critical current density of the second pair was greatly reduced. No attempt was made to investigate this effect systematically but the considerable attenuation of the proximity effect by a thin oxide layer between the two metals is clearly demonstrated.

Figure 8 illustrates a geometric effect of considerable practical significance. The critical current was measured for two pairs of specimens prepared simultaneously but with different cross-sectional areas.

The critical current density was then calculated assuming a uniform current flow through each junction. It appears that this assumption is valid for low values of critical current but not for higher values. Figure 9 demonstrates a related effect. The critical current of one specimen was measured firstly with the current applied to one end only of each lead strip and secondly in the symmetric manner described in section 2. We see that the results differ appreciably at higher values of critical current.

Finally, figure 10 shows the dependence of critical current on a magnetic field applied parallel to one of the lead strips. The general form of the result was typical although the extent of the linear region at low values of field depended enormously upon the parameters of the specimen, as will be discussed.

5. DISCUSSION AND INTERPRETATION OF RESULTS

We shall briefly discuss the possible mechanisms for the flow of supercurrent through the junctions. The first possibility is that the current is carried by lead filaments which extend right through the barrier and that the critical current is just that of these superconducting shorts. Secondly, the sandwiches may behave phenomenologically as Josephson junctions in that the supercurrent is related to the difference in the phase of the order parameter across the junction, ϕ , by an equation of the form (Josephson 1962, 1964, 1965):

$$I = I_c \sin \phi \quad . \quad (1)$$

The critical current occurs when phase coherence across the junction is broken. Finally, the critical current may be defined by the maximum supercurrent that the middle of the Cu/Al film, where the ordering is weakest, is able to sustain, so that the breakdown is something like a thermodynamic phase change.

The dependence of the critical current upon magnetic field strongly suggests that equation (1) is applicable, since no other mechanism could give this type of behaviour. As we shall see below, the self-field limiting properties of the junction add further support to this assumption. The dependence of critical current upon magnetic field and upon the mean free path of the normal metal rules out the possibility of superconducting shorts. In addition, if the supercurrent were carried by lead bridges through the barrier, the critical current would flatten out as the temperature was lowered whereas a rapid increase is in fact observed.

The only evidence in favour of the thermodynamic behaviour is the shape of the i - v characteristic. One possible explanation of the continuous development of the voltage is to suppose that superconductivity ceases just in the middle of the barrier as the critical current is exceeded and that this normal layer widens as the current is further increased. However, on this model it is difficult to explain why the measured resistance of the junction is comparable with the estimated resistance of the copper unless one invokes a high boundary resistance between the normal and superconducting phases, which in view of our earlier measurements seems unlikely. We shall not discuss further the finite-voltage regime but clearly a detailed investigation is required.

We shall assume henceforward that the junctions exhibit Josephson behaviour in so far as (1) is applicable and first discuss the phenomenological aspects, namely the behaviour in a magnetic field and self-field limiting, on this assumption. We shall then describe the theory which relates the critical current of the junctions to their various parameters and finally analyse our data in the light of this theory.

Self-field limiting

It is convenient to discuss first the results presented in figures 8 and 9. It has been shown (Anderson 1964, Ferrell and Prange 1963, Josephson 1964) that if the junction area is large or the current density high, the self-field generated by the current is not negligible and gives rise to a Meissner effect within the junction. Unfortunately, a quantitative description for the present specimen configuration is hardly tractable but we shall very briefly discuss the one-dimensional situation. Our main purpose is to determine which of our results are seriously self-field limited.

Consider the one-dimensional junction shown in figure 11. The junction is supposed to be infinitely long in a direction perpendicular to the page; the width of the junction is w . Consider first the situation

where the current is applied at A and C. The current will flow uniformly through the junction if w is small compared with the junction penetration depth given by (Anderson 1964, Ferrell and Prange 1963, Josephson 1964)

$$\lambda_J = \left(\frac{\hbar c^2}{16\pi(a+\lambda)eI_c} \right)^{1/2} \quad (2)$$

λ is the penetration depth of the superconductor, $2a$ the thickness of the barrier and I_c the critical current density. Taking the values $(a+\lambda) \sim 5,000 \text{ \AA}$ and $I_c \sim 1 \text{ Amp cm}^{-2}$ we find $\lambda_J \sim 1/5 \text{ mm}$. On the other hand, if $w \gg \lambda_J$, the current will be confined by its self-field to the edge of the barrier nearer AC and the effective current-carrying width of the junction will be $2\lambda_J$ (Ferrell and Prange 1963). Note that the current distribution is not exponential.) If we instead divide the current input equally between A and B and the output equally between C and D, the effective junction width becomes $4\lambda_J$. Assuming that a similar result holds for the two-dimensional case, we can immediately understand qualitatively the results of figure 9.

For the one-dimensional case with a symmetrical input, it is clear that self-field limiting should become significant when $\lambda_J \leq w/4$. We should expect roughly the same result for the two-dimensional situation and figure 8 shows that the larger of the two pairs of specimens becomes self-field limited (critical current density reduced below true value by about 10%) when $\lambda_J \sim w/4$. We shall therefore assume that critical current for which $\lambda_J > w/4$ are not significantly self-field limited. For the size of specimen used and a copper thickness of $5,000 \text{ \AA}$, this

criterion corresponds to a critical current density of about 15 \AA cm^{-2} . All subsequent measurements were taken with the symmetrical current input.

As we shall see, the fact that the two-dimensional self-field limiting problem has not been solved exactly is unfortunate since not all of the data can be interpreted quantitatively.

The effect of an external magnetic field

It was predicted by Josephson (1962) and demonstrated by Rowell (1963) that the magnetic field dependence of the critical current for a one-dimensional junction which was not self-field limited was of the form

$$I_c(\Phi) \propto \left| \frac{\sin(\pi\Phi/\Phi_0)}{\pi\Phi/\Phi_0} \right|. \quad (3)$$

$\Phi = 2(a+\lambda)wH$ is the flux threading the junction (at right angles to the page in figure 9) and Φ_0 the flux quantum. Similar results were obtained for the Pb-Cu-Pb specimens for which $\lambda_J > w/4$.

Figure 10 shows the behaviour for a specimen which was self-field limited. When the field is increased from zero, it is screened out from the junction and the critical current decreases linearly; this is just the Meissner effect. Let us consider again the one-dimensional symmetrical case. If the current is confined to the two edges of the junction, the circulating current generated by the applied field, $cH/(4\pi)$, will be added to the current at one edge and subtracted from that at the other. The critical current will be reached when the greater of the edge currents has its maximum value, the other current then being smaller by $cH/(2\pi)$. Thus the total critical current per unit length is of the form

$$I_c(H) = 4\lambda_J I_c - \frac{cH}{2\pi} \quad (4)$$

Now Josephson (1964, 1965) has shown that the flux will completely penetrate the junction (as in a type II superconductor) at a field

$\pi H_{c1}/2$, where

$$H_{c1} = \left(\frac{16\kappa I_c}{\pi e(a+\lambda)} \right)^{1/2} = \frac{16 \lambda_J I_c}{c} \quad (5)$$

From (4) and (5) we see that $\pi H_{c1}/2$ is just that field required to reduce the critical current to zero. For fields greater than $\pi H_{c1}/2$ the behaviour reverts to that described by (3). However, it may be energetically favourable for the flux to penetrate at a field less than $\pi H_{c1}/2$, since in general this value will not correspond to an integral number of flux quanta. In this case the oscillatory behaviour will begin at a field below $\pi H_{c1}/2$. This problem has been discussed in detail by Owen and Scalapino (1967).

The behaviour is more complicated when we have a two-dimensional junction, as in the present case.* The main problem is to deduce a value

* A further complication arises in that the current distribution across the lead strips is by no means uniform. The high current density at the edges will probably distort the current flow through the junction.

of I_c , the true critical current density. We shall assume a highly idealized model of the current distribution in a square junction in which the current flows uniformly in a peripheral strip of width $2\lambda_J$. This is obviously a reasonable approximation if $w \gg \lambda_J$ but of less certain validity in the present case; the main objection is the neglect of the high current distribution at the corners. On this model, the

effective area of the junction is $8\lambda_J(w - 2\lambda_J)$ so that $I_c = i_c [8\lambda_J(w - 2\lambda_J)]^{-1}$. Combining this relation with (2) we find $\lambda_J = \frac{1}{2} \pi w [h + \pi e i_c (a + \lambda) / c^2]^{-1}$ 2.7×10^{-3} cm for the specimen of figure 8. Thus $I_c \approx 105$ A cm⁻² and $w/\lambda_J \approx 8.5$. Inserting the values of λ_J and I_c in (5) gives $\pi H_{c1} / 2 \approx 0.7$ G, which, bearing in mind the crudeness of the model, must be considered in acceptable agreement with the observed value of 0.77 ± 0.02 G.

If, instead, we assume a uniform current distribution, we find $I_c = 75$ A cm⁻². This error of 28% should be compared with the experimentally estimated error of about 20% at $w/\lambda_J = 8.5$ in figure 8. It appears that our approximations are of the right order and, in addition, that the self-field limiting correction increases relatively slowly with w/λ_J .

The period of oscillation at fields greater than $\pi H_{c1} / 2$ is 1.95 ± 0.2 G cm², one flux quantum, to within the experimental error. The additional part of an oscillation obtained when the field is reduced to zero again is due to flux trapping in the barrier.

It appears that we can satisfactorily explain the general features of the behaviour of the junction in a magnetic field although a complete quantitative description is impossible without a knowledge of the current distribution in the self-field limited situation. However, the qualitative behaviour is of greater importance because of the information which it gives us about the nature and behaviour of the junctions.

Theoretical considerations

We now discuss the theory of the proximity effect which is relevant to the understanding of the experimental data.

According to the B.C.S. theory of superconductivity (Bardeen et al. 1957), a metal may be characterized by the parameter NV , where N is the density of states at the Fermi surface and V the effective electron-electron interaction. The criterion for the metal to be a superconductor is $V > 0$. Morel and Anderson (1962) have pointed out that V is essentially a point interaction so that in a system consisting of two different superposed metals, we can suppose that the interaction parameter changes instantaneously on the boundary. It is the fact that the condensation amplitude of the superconducting electrons cannot change abruptly at the boundary, but only over a distance of the order of a coherence length, which gives rise to the proximity effect.

In the situation where NV varies spatially, the B.C.S. theory is no longer applicable. The degree of order becomes a function of position and may be expressed as the condensation amplitude, $F(\underline{r}) = \langle \Psi^\dagger(\underline{r}) \Psi(\underline{r}) \rangle$ where $\Psi^\dagger(\underline{r})$ is an electron annihilation operator (see, for example, Landau and Lifshitz, 1959). $|F(\underline{r})|^2$ is then essentially the probability of finding a Cooper pair at \underline{r} , that is, it represents the superfluid density. The pair potential, $\Delta(\underline{r}) = V(\underline{r}) F(\underline{r})$ is analogous to the energy gap, Δ , of the B.C.S. theory.

The de Gennes-Guyon theory of the proximity effect (de Gennes and Guyon 1963, de Gennes 1964) uses the Gor'kov self-consistent integral equation (Gor'kov 1959) to calculate the spatial variation of $\Delta(\underline{r})$:

$$\Delta(\underline{r}) = \int d^3\underline{r}' K(\underline{r}, \underline{r}', T) \Delta(\underline{r}'), \quad (6)$$

where K is the kernel with a range (coherence length):

$$\xi(T) = \left(\frac{\hbar v_F \ell}{6\pi kT} \right)^{1/2} \quad (7)$$

v_F is the Fermi velocity and ℓ the electronic mean free path. Equation (6) expresses the cooperative nature of the effect (just as the B.C.S. integral equation does) in that pairing at a point r is related to that at all other points r' . The theory is valid strictly only for a dirty system ($\ell \ll \xi$) near its transition temperature, where $\Delta(r)$ is small.

De Gennes (1964) has used (6) to study the variation of $F(x)$ across a NS sandwich. He derives the following boundary conditions on the $F(x)$ at the interface ($x=0$):

$$\frac{F_N(0)}{N_N} = \frac{F_S(0)}{N_S} \quad (8)$$

and

$$v_{FN} \ell_N \left. \frac{dF_N(x)}{dx} \right|_0 = v_{FS} \ell_S \left. \frac{dF_S(x)}{dx} \right|_0 \quad (9)$$

Equation (8) implies that the fraction of electrons paired is conserved across the interface. This is so only if boundary scattering may be completely ignored. However, as we have seen earlier, the scattering resistance of the boundary is equivalent to not more than one mean free path so that (8) will be sufficient true in practice provided the condition $\ell/\xi \ll 1$ is valid in both materials.

For the case in which each film is much thicker than a coherence length, de Gennes obtains explicitly the spatial variation of $F(x)$. In the normal metal ($0 > x > d_N$), far from the interface (in the "one-frequency approximation," $k_N x \gg 1$), $F(x)$ has the form:

$$F_N(x) \propto \exp(-k_N x), \quad (10)$$

where

$$k_N^{-1} = \left(\frac{\hbar v_{FN} \ell_N}{6 \pi k T} \right)^{1/2} \left(1 + \frac{2}{\ln(T/T_{CN})} \right)^{1/2} \quad (11)$$

represents the depth of penetration of pairs into the normal metal. It should be emphasized that (10) is not true for $x \lesssim k_N^{-1}$ in which region the contribution of the superconductor to the kernel in (6) is substantial. In the superconductor ($-d_S < x < 0$) the condensation amplitude is considerably depressed near the boundary by the presence of the normal metal over a distance of the order of the Ginzburg-Landau coherence length, ξ_{GL} .

De Gennes (1964,1966) has continued his analysis to derive the current carrying capacity of a SNS junction. However, we shall describe a highly simplified model which is adequate to enable us to interpret most of our results.

We calculate first $|F_N(x)|$ in an NS sandwich by finding its value at the boundary, $|F_N(0)|$, and then using (10). We suppose that $T_{CN} = 0$ so that $k_N^{-1} = \xi_N$ and choose $N_N = N_S$ and $\xi_N = \xi_S$ so that both F and dF/dx are conserved at the interface. The simplest possible approximation to the variation of $|F_N(x)|$ near the boundary is shown in figure 12, and from it we see that $|F_N(0)| = |F_0| \xi_N / \xi_{GL}$ in the limit $\xi_N / \xi_{GL} \ll 1$ which is valid near T_{CS} , $|F_0|$ being the condensation amplitude in the bulk superconductor. Thus from (10) we have $|F_N(x)| = (\xi_N / \xi_{GL}) |F_0| \exp(-x/\xi_N)$. We now return to the SNS junction and assume that in the weak-coupling limit $a/\xi_N \gg 1$ the currents which can flow are so small that they affect only the phase and not the amplitude of the order parameter. If the two boundaries are at $|x| = a$ and ϕ_a and ϕ_{-a} are the values of $\arg F$ at $x = a$ and $-a$ respectively,

we have that

$$F_N(x) = |F_0| (\xi_N / \xi_{GL}) \{ \exp[(x-a)/\xi_N + i\phi_a] + \exp[-(x+a)/\xi_N + i\phi_{-a}] \}. \quad (12)$$

The supercurrent is calculated from the expression $I \propto (F^* \partial F / \partial x - F \partial F^* / \partial x)$, giving

$$I(T) = A |F_0(T)|^2 [\xi_N(T) / \xi_{GL}^2(T)] \exp[-2a/\xi_N(T)] \sin \phi. \quad (13)$$

A is a temperature-independent constant and $\phi = \phi_a - \phi_{-a}$. We see that $I(T)$ is independent of x as required and obtain $I_c(T)$ by comparing (13) with (1). Our expression for $I_c(T)$ is similar to that obtained by de Gennes (1964) except in that he replaces ξ_N by k_N^{-1} for the more general case $T_{CN} > 0$.

It should be remarked that in the present experiments the superconductor was in the clean limit so that there is some doubt as to the applicability of (13). Probably the immediate effect is to introduce a multiplying constant into the boundary condition (8) which will not affect the general form of (13).

Finally, it is of interest to compare (13) with the result for an insulating junction (SIS). In this case there is no depression of the order parameter in the superconductor by the insulator and the tunnelling probabilities are independent of temperature. Near T_{CS} , the critical current then has the form (Josephson 1965):

$$I_c(T) = A' |F_0(T)|^2. \quad (14)$$

Temperature dependence of $I_c(T)$ near T_{CS}

For temperatures close to T_{CS} , $\xi_{GL}(T)$ and $F_0(T)$ vary as $(1-t)^{-1/2}$ and $(1-t)^{1/2}$ respectively, where $t = T/T_{CS}$. Thus near T_{CS} for a given thickness of Cu/Al the critical current may be written in the form:

$$I_c(T) \simeq B(1-t)^2, \quad (15)$$

where $B(a)$ is a constant. This result should be contrasted with that for a SIS junction for which $I_c \propto (1-t)$ near T_{CS} (from (15)). This qualitative difference is a significant one and (15) may be regarded as an important test of the functional form of the de Gennes boundary conditions.

Before embarking upon an analysis of the data, we should bear in mind that in the present specimens the superconductors have a finite thickness, namely $7,000 \text{ \AA}$. When the temperature is sufficiently high for ξ_{GL} to become comparable with this figure, the order parameter will be depressed by the copper throughout the lead and as the temperature is raised further, $I_c(T)$ will drop off more rapidly than predicted by (15). Assuming $\xi_{Pb}(T) \simeq 0.5 \xi_0 (1-t)^{-1/2}$ near T_{CS} and $\xi_0 \simeq 830 \text{ \AA}$ (Bardeen and Schrieffer 1961) we find that $\xi_{Pb}(T) = 7,000 \text{ \AA}$ at a temperature about 0.025°K below T_{CS} , so that we might expect an appreciable effect over a temperature range of perhaps two or three times this value.

Another difficulty arises in trying to estimate the range of temperature implied by "near T_{CS} ". The de Gennes theory is valid for small Δ and in the local limit, that is, $(1-t)^{1/2} \ll 1$, but in practice it is often found that neither of these conditions is very stringent. The approximate temperature dependences of $\Delta_{Pb}(T)$ and ξ_{GL} are valid to a few percent for about 0.5°K below T_{CS} and over this range the variation of the exponential term makes a contribution of comparable magnitude but opposite sign. It would seem reasonable to expect (15) to hold to a few percent down to about 6.6°K although this fact is apparently somewhat fortuitous.

Figure 13 shows the variation of $I_c^{1/2}(T)$ with T for five thicknesses

of Cu/Al. We see that the dependence is sensibly linear until the lead ceases to exhibit bulk behaviour, after which the critical current falls off more rapidly as predicted. The junctions are not significantly self-field limited with the exception of the 2,000 Å film, for which the linear behaviour must be regarded as somewhat fortuitous. The extrapolations of the linear regions should intersect the temperature-axis at the transition temperature of the superconductor. The rather large spread, about $\pm 0.1^\circ\text{K}$, is attributed to strains in the lead films set up by differential contraction during the cooling of the slide.

Despite the difficulties in interpreting the data, it is felt that these results do suggest that the boundary conditions on F and dF/dx are of the correct form. We cannot, of course, make any statement about constant factors in (8) and (9).

One other useful piece of information may be extracted from figure 13. We have seen that the value of A in equation (13) is strongly dependent upon the amount of oxide at the SN interface and it is therefore of interest to see how much A varies from specimen to specimen. We may rewrite (13) in the form

$$I_c^{1/2} = C(T_{CS} - T) \exp(-k_N a), \quad (16)$$

where C is a new constant and we have replaced ξ_N by k_N^{-1} . We calculate the value of C for each specimen from the slope of $I_c^{1/2}$ against T , the known value of a and the value of k_N at 6.9°K for the appropriate mean free path. The value of k_N is derived from the data in a later section. No correction has been made for the mean free path dependence of C , which is proportional to $\ell^{1/4}$; the correction is never greater than 5%. The values obtained for C

are shown in Table I.

Table I: Values of C for five different specimens					
Thickness of Cu/Al (Å)	2000	2250	3010	3250	3790
C ($\text{Å}^{1/2} \text{ cm}^{-1} \text{ } ^\circ\text{K}^{-1}$)	430	960	710	1180	840

The value of C for the thinnest specimen, for which there is significant self-current limiting, should be discounted. The remaining values indicate that $A(\propto C^2)$ is constant to within about $\pm 50\%$. This result is hardly very satisfactory but is perhaps as good as can be expected. The fact that the lead was in the clean limit implied that A was highly sensitive to slight boundary irregularities and the use of a dirty superconductor may well greatly reduce the spread in the value of A.*

* The values of C are of course strongly dependent upon the value of k_N chosen; we shall see that the error in k_N is about $\pm 10\%$. We have here used the mean value of k_N whereas the minimum value makes A constant to within $\pm 30\%$. However, there seems no a priori justification for preferring the latter value of k_N .

Temperature dependence of $I_c(T)$ at low temperatures

Although (13) is strictly true only near T_{CS} it might be expected to hold approximately at much lower temperatures. Now between 1°K and 4°K , ξ_{GL} and Δ_{Pb} ($\propto F_0$) are nearly constant and k_N^{-1} changes by a factor of only 2. Consequently the exponential term dominates the temperature dependence of $I_c(T)$ and we may write

$$I_c(T) \approx D \exp(-GT^{1/2}), \quad (17)$$

where D and G are positive constants.

A graph of $\ln I_c$ against $T^{1/2}$ is shown in figure 14. For $T < 2^\circ\text{K}$, there would seem to be some evidence for the validity of (17). However, much of the data is subject to self-field limiting and the higher values of critical current should be somewhat increased. Thus the apparently linear behaviour at the lower temperatures, with the exception of the 6,500 Å specimen, may be spurious. If (17) were approximately true below a certain temperature, the linear behaviour would begin at the same temperature for all specimens; in fact the "knee" in the curves occurs at a variety of temperatures and is probably governed partly by the onset of self-field limiting. Nevertheless, it appears that there is some evidence in favour of (17) and clearly the behaviour is quite different from that of a SIS junction for which $I_c(T)$, which is proportional to Δ^2 , tends to a finite limit at low temperatures.

The dependence of I_c on the mean free path of the Cu/Al

Unfortunately, no systematic study was made of the dependence of the critical current on the mean free path of the Cu/Al and the only results are those presented in figure 6. It is obvious that the longer mean free path has given rise to a much higher critical current, as we should expect, since the coherence length, ξ_N , is much longer. However, it is not possible to directly compare the two curves as the mean free path of the upper one (220 Å) is too high for the alloy to be in the dirty limit; equation (7) is no longer applicable and we have no means of estimating ξ_N . It is clear that a proper investigation of the effect of mean free path is required.

The dependence of I_c on the thickness of the Cu/Al film

According to equation (13), at constant temperature, I_c varies as $\exp(-Ml^{-1/2}a)$, where M is a positive constant. Unfortunately, the test of this result is complicated by the variation of the mean free path of the Cu/Al alloy from specimen to specimen.

Figure 15 shows the variation of $\ln I_c$ with thickness of Cu/Al at three different temperatures. A correction has been applied for those specimens in which the mean free path differed markedly from the average value (different specimens are represented at different temperatures so that the average mean free path is not necessarily the same). The observed value of I_c was corrected for mean free path assuming the validity of (13). It should be pointed out that in this respect we have not justified the formula in the present work, but in fact the correction was made to only three points. At each temperature,* a linear regression line has been fitted to the data points. The fit may be considered acceptable.

* For the results at 7°K we have simply taken the value of critical current for each specimen at a measured temperature of 7.0°K. Now as is evident from Figure 13 there is considerable variation in transition temperature (T_{CS}) from specimen to specimen and since the $(T_{CS}-T)^2$ term dominates near T_{CS} , it might be more appropriate to measure the critical current for each specimen at a fixed value of $(T_{CS}-T)$. In fact, this procedure gives a value of k_N^{-1} remarkably close to the value obtained but with a greater error. It appears that within limits rather less than the error attached to the value of k_N^{-1} , the two procedures give the same value. At the lower

temperatures, where the exponential term dominates, it is more appropriate to use the values of critical current at the measured temperatures.

Self-field limiting is not important for the results at 7.0°K, except possibly for the thinnest specimen, but certainly is important for the higher values of critical current at the two lower temperatures. However, the correction at 100 A cm⁻² is about 20-30%, which on a logarithmic scale may not be too significant and in any case is probably smaller than the random errors in the observed values due to the variation of A from specimen to specimen.

The value of NV in copper

From Fig. 15 we may obtain an estimate of k_N^{-1} at 7.0°K and by comparing it with ξ_N deduce a value for T_{CN} in copper (or more strictly, Cu/3% Al) using (11). We restrict ourselves to the data at 7.0°K since it is not self-field limited and the theory is strictly applicable at this temperature. We estimate ξ_N (from (7)) by making use of a relation between lv_F and $\rho\gamma$, where γ is the electronic specific heat constant of copper. In a cubic or polycrystalline metal, $\rho = 12\pi^3\hbar/(Se^2\bar{l})$ and $\gamma = k^2S(\overline{1/v_F})/(12\pi\hbar)$ (Pippard 1960); \bar{l} and $(\overline{1/v_F})$ are average values over the Fermi surface whose area is S. Combining these expressions and assuming that $[(\overline{1/v_F})]^{-1} = \overline{v_F}$ we obtain $\overline{1/v_F} = \pi^2k^2/(e^2\gamma\rho)$. The value of γ used to calculate ξ_N , 6.9×10^{-4} joules/mole/deg.² (Corak et al. 1955), is that for pure bulk copper. We find $\xi_N = 260 \text{ \AA}$ and $k_N^{-1} = 310 \pm 30 \text{ \AA}$, giving $T_{CN} = 6 \times 10^{-2}$ °K with possible limits (one standard deviation) of 3×10^{-5} °K and 4×10^{-1} °K. The corresponding value of NV, derived from the B.C.S. relation $T_c = 1.14\theta_D \exp[-1/(NV)]$ with

$\theta_D = 344^\circ \text{K}$ (Corak et al. 1955) is 0.11 with limits of 0.06 and 0.14. It is evident that the experimental errors associated with the value of T_{CN} are enormous and that it should not be taken too seriously.

Table II compares the present value of NV in copper with those obtained

Table II: Values of NV and the corresponding T_{CN} obtained by various authors for copper

Author	Method	$(\text{NV})_{\text{Cu}}$	Estimated Limits	Corresponding $T_{\text{CN}} \text{ } ^\circ \text{K}$
Hilsch* (1962)	T_{CNS} of NS	+0.05	+0.10	1.5×10^{-2}
			-0.06	7×10^{-7}
			--	--
Hauser et al. (1964)	T_{CNS} of NS	+0.09		6×10^{-3}
Minnigerode (1966)	T_{CNS} of NS	+0.116		7×10^{-2}
Adkins and Kington (1966)	Tunnelling	-0.01	+0.06	2×10^{-5}
			-0.10	--
Present work	Supercurrents in SNS	+0.11	+0.14	4×10^{-1}
			+0.06	6×10^{-2} 3×10^{-5}

by other authors. The experimental errors are large and in addition it seems doubtful whether the theory is sufficiently advanced to allow a meaningful value of NV to be deduced from the data. For example, Jacobs (1967) has pointed out that in the vicinity of the SN interface, where the kernel in (6) contains contributions from both materials, it is

* See de Gennes (1964).

not clear as to how one should treat the cut-off of the electron-phonon interaction. He has showed further that different methods of performing the cut-off in the superconductor gave rise to markedly different values of NV for copper; this is a consequence of the fact that lead, which was used as the superconductor in all the experiments, is strong-coupling.

A particular difficulty arises in the thick-film limit which is relevant to the present results. The experiment measures k_N^{-1} which is very insensitive to the value of T_{CN} when T/T_{CN} is large, as is obvious from (11); consequently small experimental discrepancies give rise to large errors in the estimate of NV . Nevertheless, it seems likely that a study of SNS sandwiches using a weak-coupling superconductor and at very low temperatures may enable a much more accurate estimate of NV to be obtained than is possible by other means.

The coherence length in copper

Table III compares the measured and estimated values of $\xi_N(T)$ at three different temperatures. At the two lower temperatures, the observed value of k_N^{-1} has been divided by $\{1 + 2/[\ln(T/T_{CN})]\}^{1/2}$ to obtain ξ_N , taking $T_{CN} = 6 \times 10^{-2} \text{ K}$.

Table III: Variation of ξ_N with T

T(°K)	\bar{l} (Å)	ξ_N (Å) measured	ξ_N (Å) estimated
7.0	100	260±25	260
5.5	100	275±20	295
3.0	135	470±35	465

ξ_N rises with decreasing temperature as suggested by (7), indicating that thermal fluctuations determine the decay length of the electron pairs. The form of (7) seems likely to be valid provided $kT > \Delta$ and, since Δ is very small indeed in copper, the exponential term in (13) may well remain accurate down to very low temperatures.

6. DISCUSSION

The purpose of these experiments has been to examine the general features of SNS sandwiches and it is felt that we now have a reasonable overall description of their properties. It is clear that the junctions behave as Josephson junctions although there are marked quantitative and qualitative distinctions between them and junctions with insulating barriers. For example, the insulator in a SIS junction is typically 10-20 Å thick with a resistance of 1Ω whereas the copper barrier may be 5000 Å thick with a resistance of $10^{-7}\Omega$. This very low resistance implies that the voltage produced when the critical current is exceeded may be as low as 10^{-10} V compared with 10^{-3} V for SIS junctions. Qualitatively, the most striking difference is in the temperature dependence of the critical current.

The main sources of error in the experiments have been the irreproducibility of the Cu/Al alloy and the difficulty of estimating the corrections for self-field limiting. Possibly an improved evaporation technique of sputtering would yield greater uniformity of mean free path. The self-field limiting problem could be most easily overcome by making the lead strips narrower; with the aid of masks in contact with the substrate it should be possible to evaporate junctions of 5×10^{-3} cm square for which the self-field limiting would be negligible below about 250 A cm^{-2} .

Junctions in which the Cu/Al alloy is replaced by other metals are also worthy of study. In the case of aluminum, for example, k_N^{-1} would become very large as the temperature was lowered. Thus we could not only make a more thorough test of equation (13) but also use relatively thick films of aluminium whose properties could be more carefully controlled. The possibility of using a magnetic material such as iron also seems interesting at first sight. However, for a mean free path of 50 \AA , $k_N^{-1} \sim 6 \text{ \AA}$ (Hauser et al 1966) and it is doubtful if continuous films of iron thin enough to pass a supercurrent could ever be prepared. However, if we used instead a dilute magnetic alloy, k_N^{-1} would still be relatively long; a study of such junctions might well yield valuable information on the scattering of electrons by localized magnetic moments.

Finally a study of SNS junctions at very low temperatures is not only of intrinsic interest as in addition it may well yield a useful tool for the accurate determination of very low transition temperatures. The coherence length in the normal metal would be very long so that it should be possible to use thick layers with relatively well-defined properties.

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FIGURE CAPTIONS

- Fig. 1 Specimen configuration, showing a "symmetrical" current input to specimen 3; the voltage is measured across all six specimens.
- Fig. 2 Longitudinal section of cryostat.
- Fig. 3 The i-v characteristic of a typical SNS junction.
- Fig. 4 An enlarged portion of the characteristic of Fig. 3.
- Fig. 5 Critical current density (I_c) against temperature (T).
(Figures refer to thickness of Cu/Al and those in parenthesis to its mean free path.)
- Fig. 6 Critical current density (I_c) against temperature (T), showing its dependence on the mean free path of the barrier.
- Fig. 7 Critical current density (I_c) against temperature (T) showing the effect of an oxide layer at one NS interface.
- Fig. 8 Critical current density (I_c) against temperature (T) for two junctions of different areas, showing self-field limiting (I_c calculated as measured current divided by junction area).
- Fig. 9 Critical current density (I_c) against temperature (T) showing the effect of symmetric and asymmetric current inputs.
- Fig. 10 Magnetic field dependence of the critical current.
- Fig. 11 Section of 1-D tunnelling junction. Two superconductors, AB and CD, are separated by a barrier of thickness $2a$ and width w . The junction is infinitely long in a direction perpendicular to the page.
- Fig 12 Simplest possible model to give value of condensation amplitude ($|F|$) at SN interface.

- Fig. 13 $I_c^{1/2}$ against T near transition temperature of junction.
(Figures refer to thickness at Cu/Al and those in parenthesis to its mean free path.)
- Fig. 14 $\text{Log } I_c$ against $T^{1/2}$ at low temperatures. (Figures refer to the thickness of Cu/Al and those in parenthesis to its mean free path.)
- Fig. 15 $\text{Log } I_c$ against thickness of barrier.

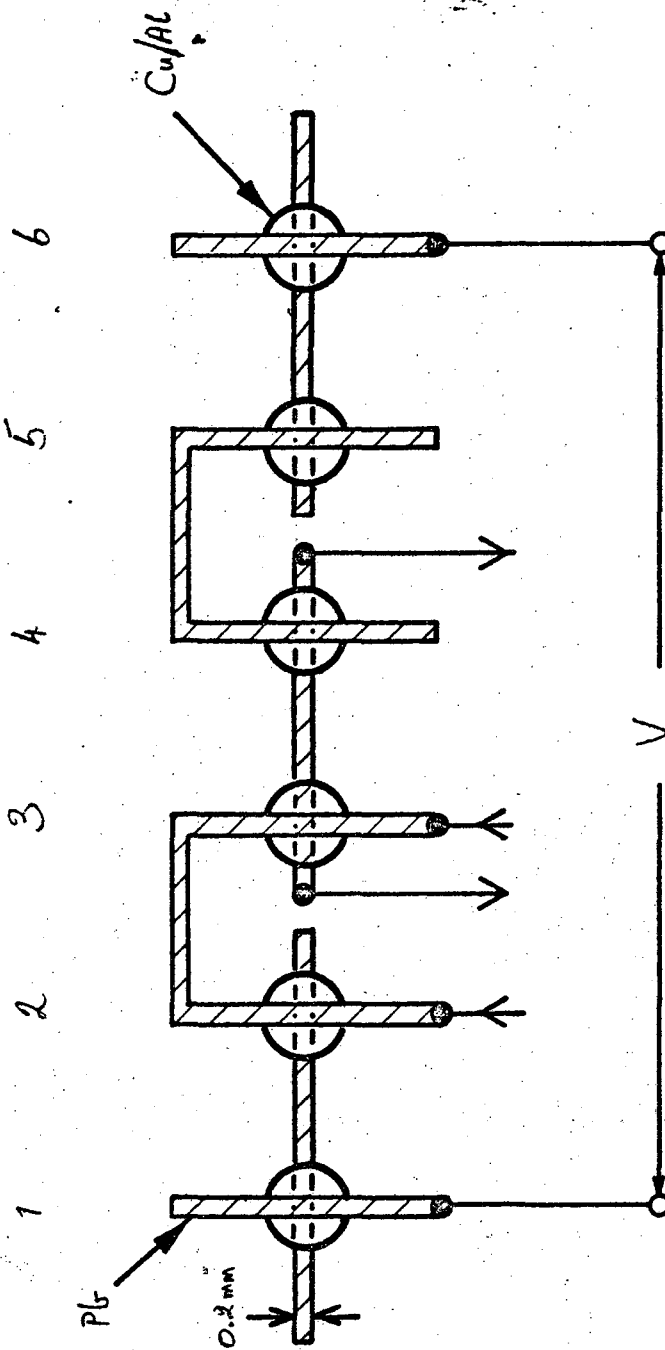
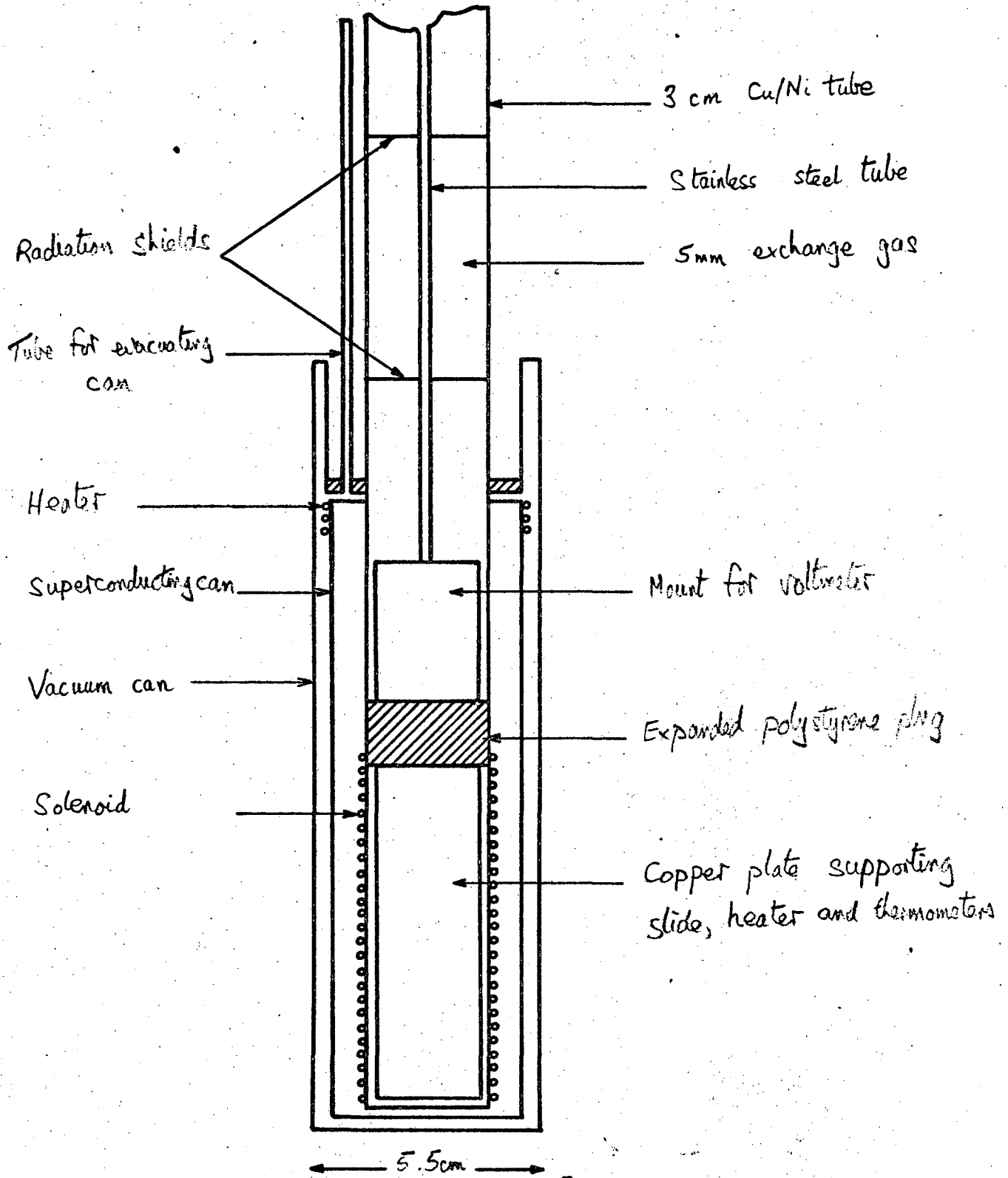


Fig. 1

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Fig. 2.

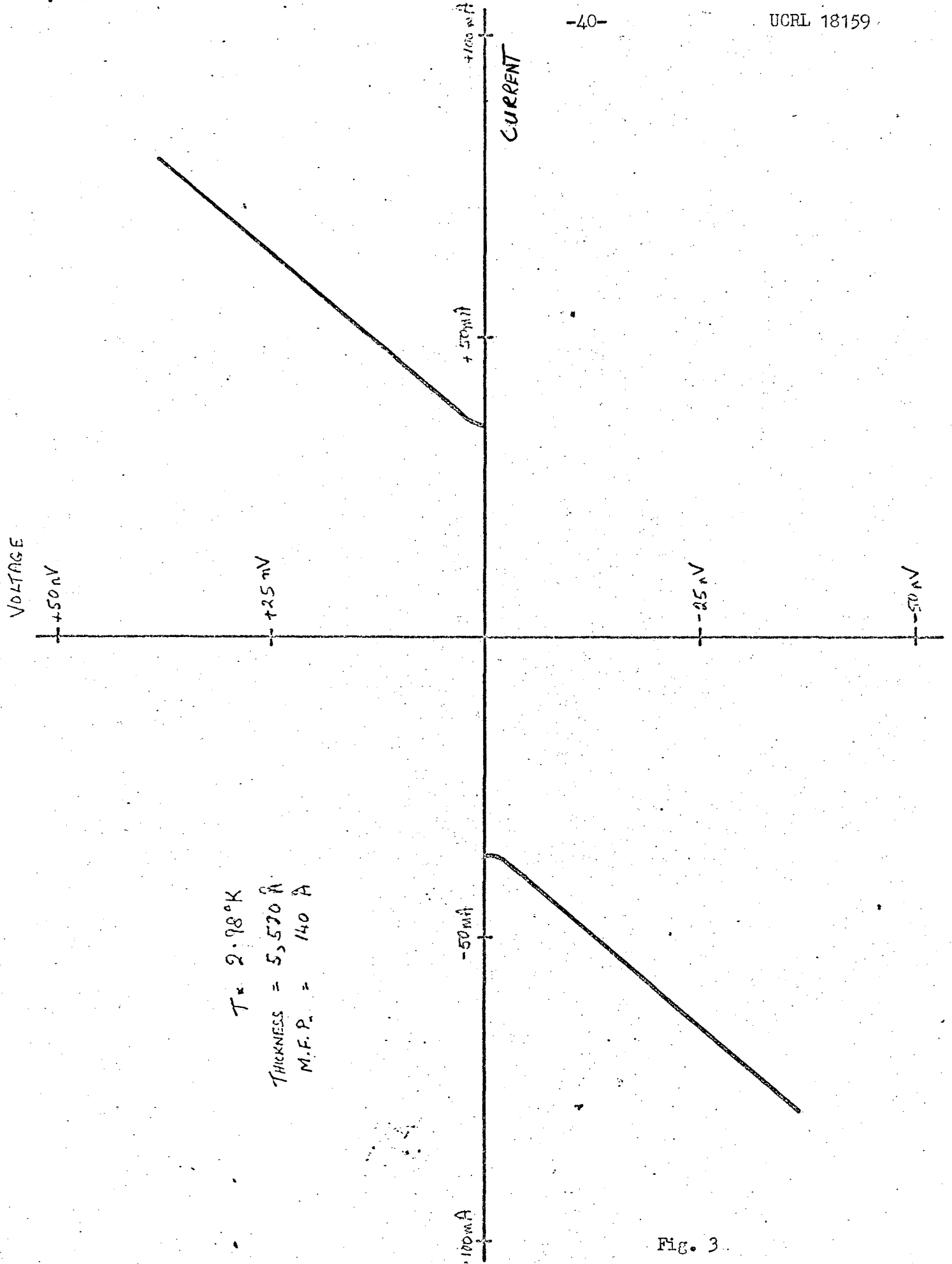


Fig. 3

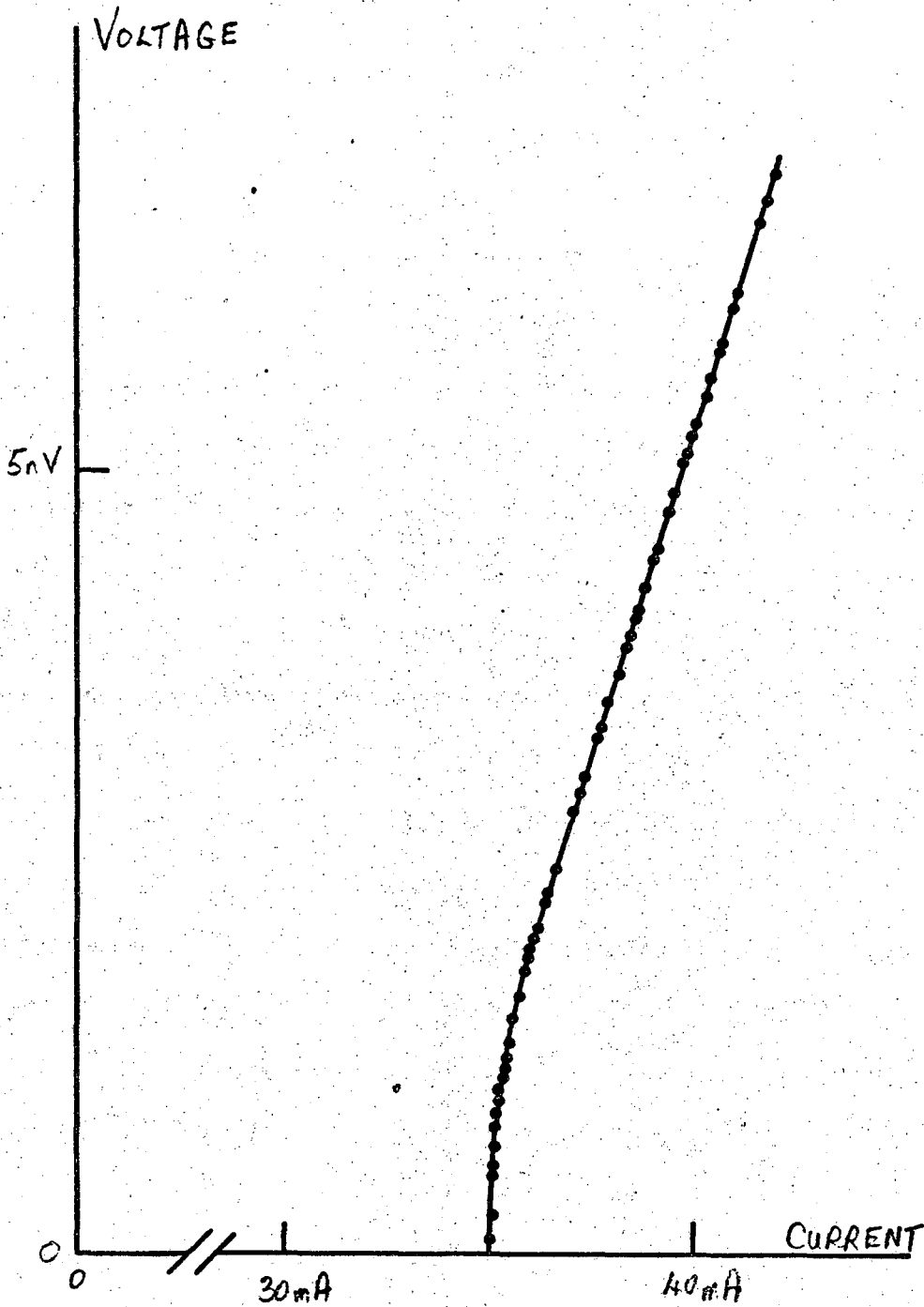


Fig. 4

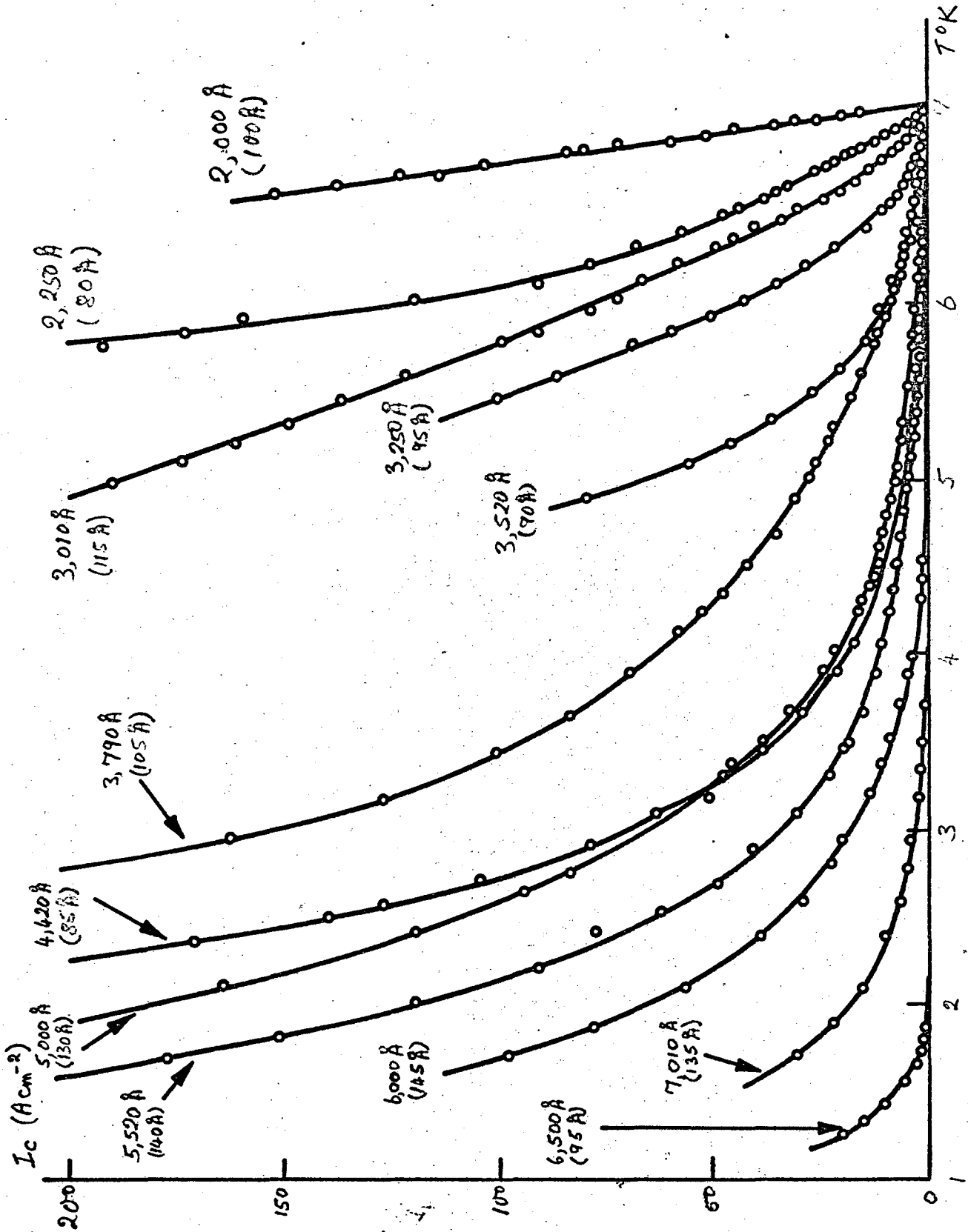


Fig. 5

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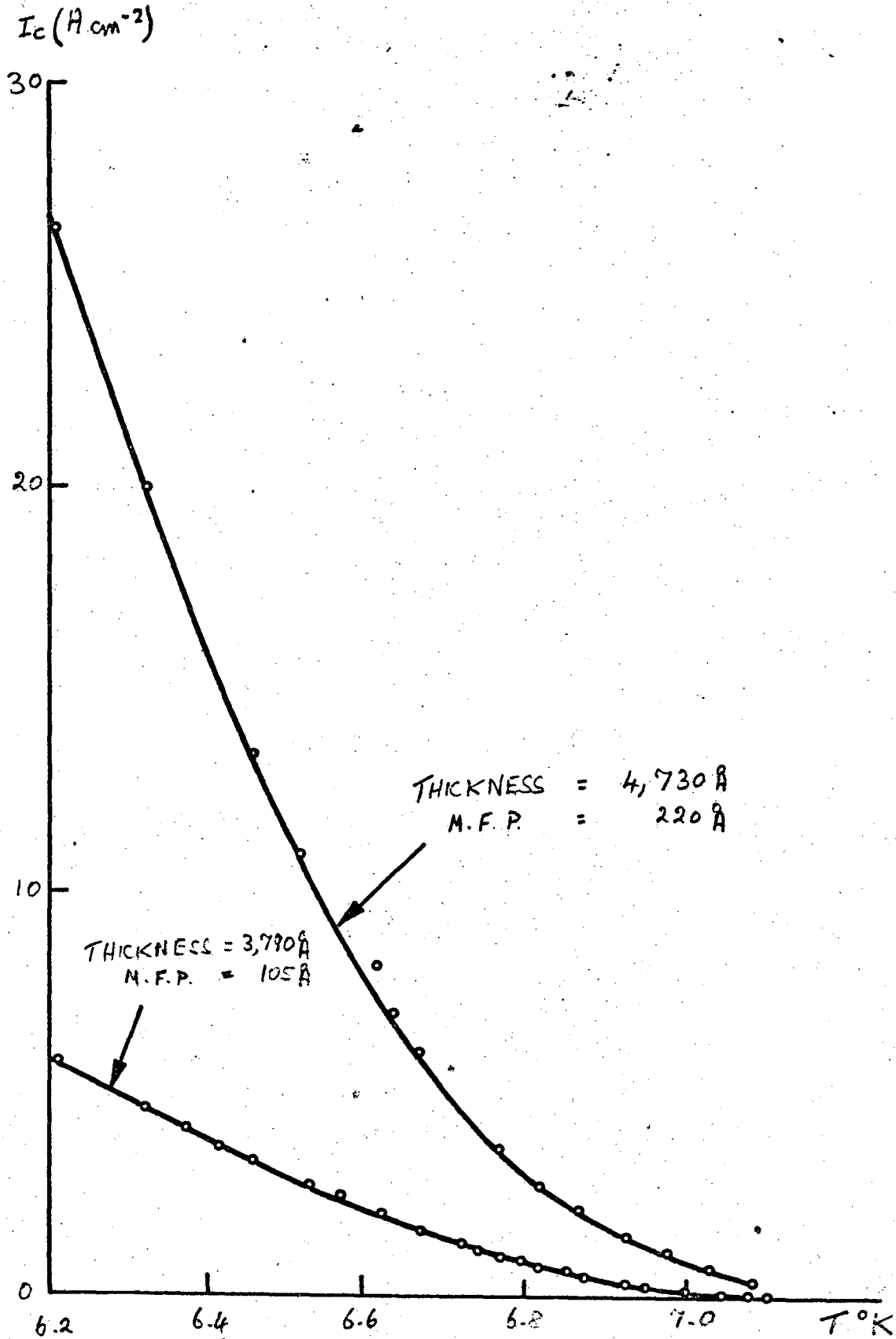


Fig. 6

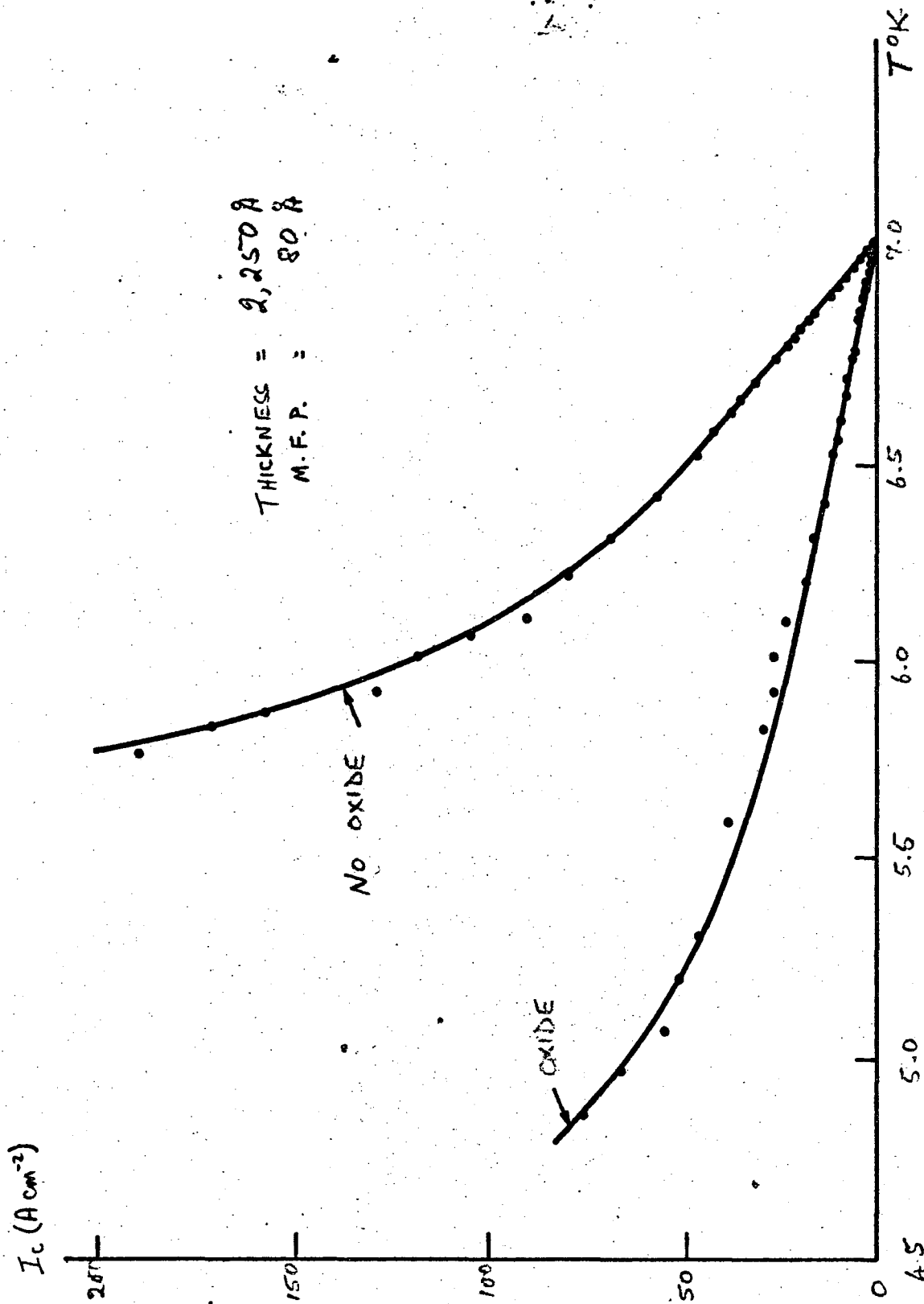


Fig. 7

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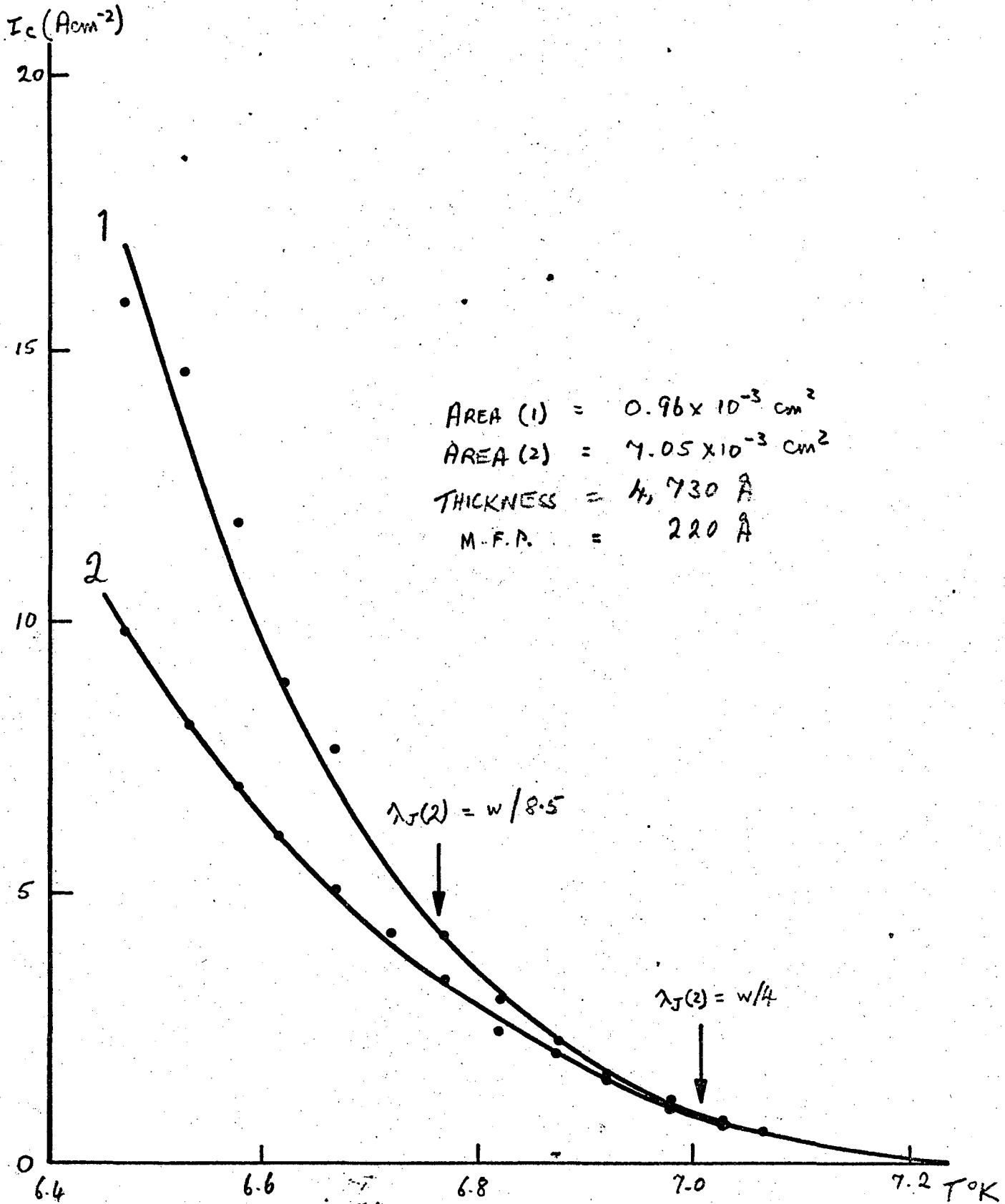


Fig. 8

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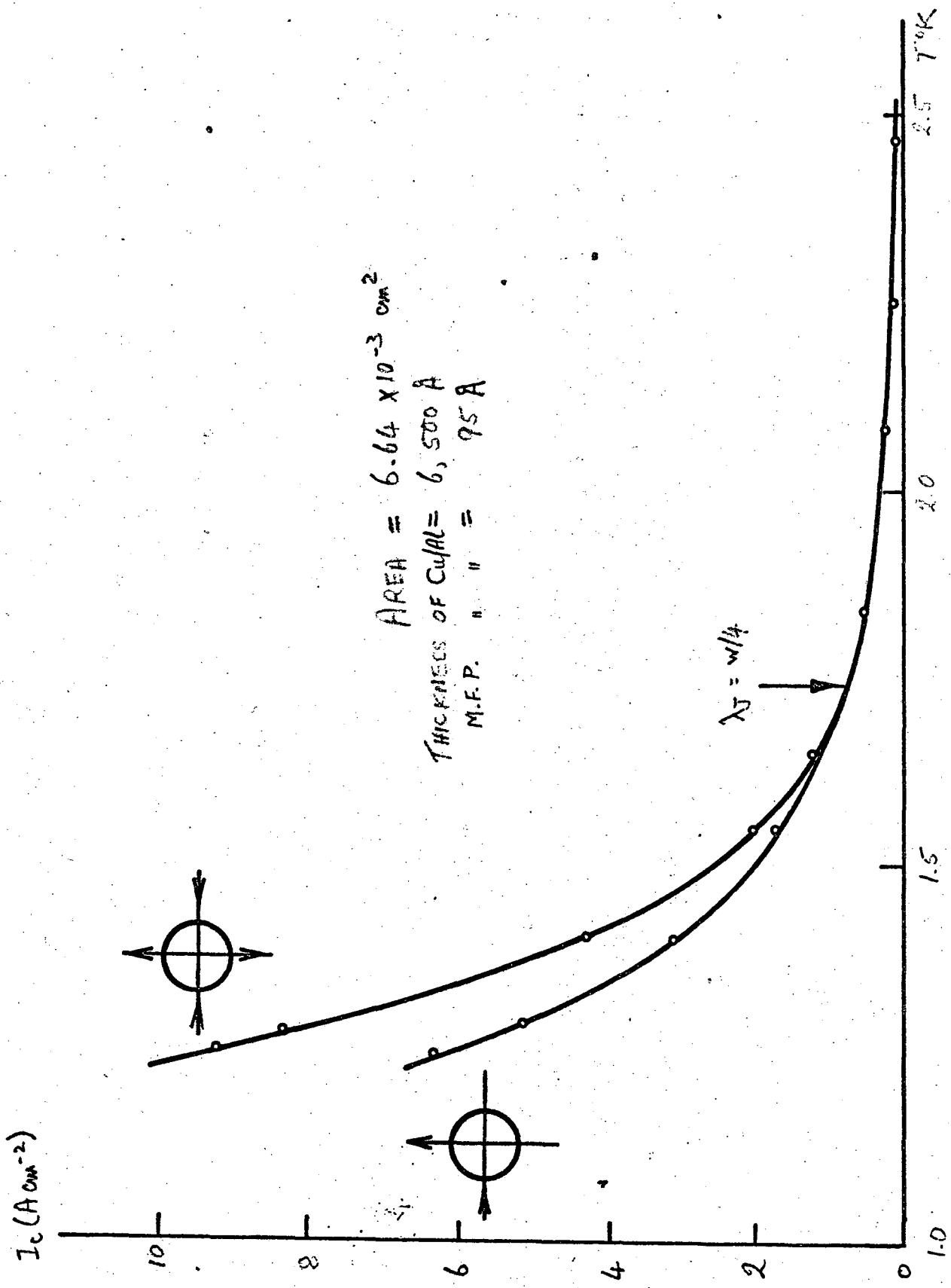
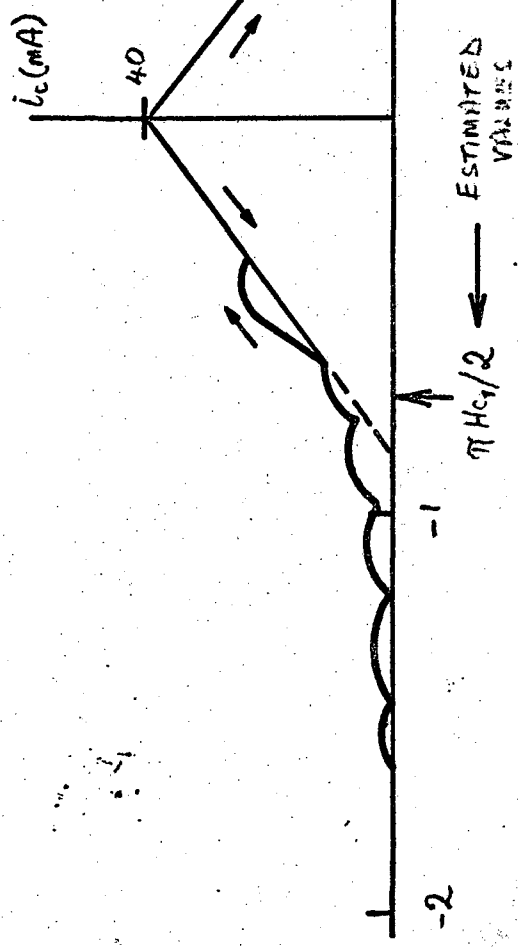


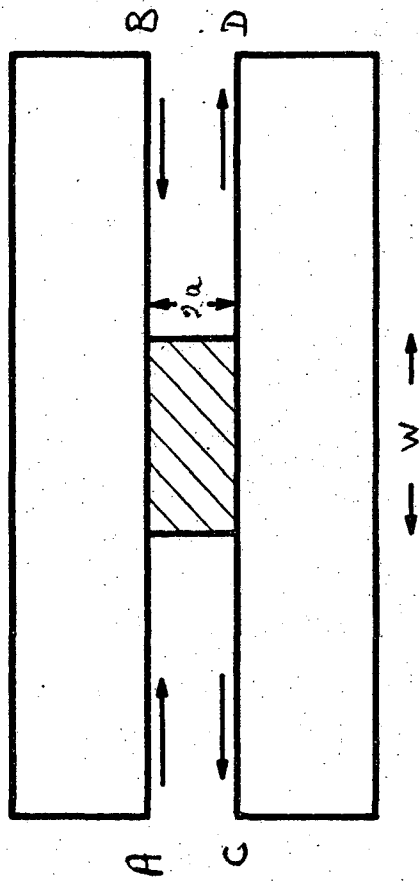
Fig. 9

MEASURED PERIOD = $1.95 \pm 0.2 \text{ Gcm}^2$
 THICKNESS OF Cu/AI = $2,250 \text{ \AA}$
 M.F.P. " " = 80 \AA
 T = 6.3°K
 W/λ_g = 8.5



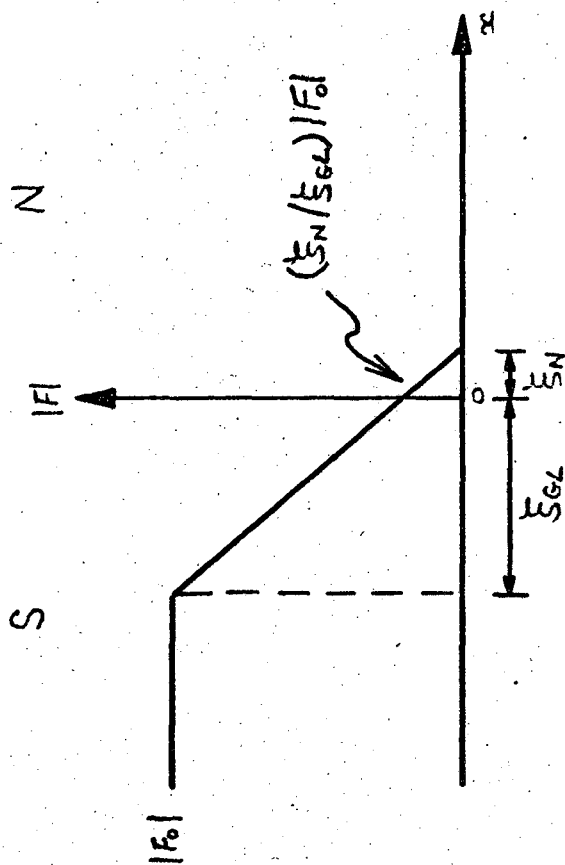
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Fig. 10



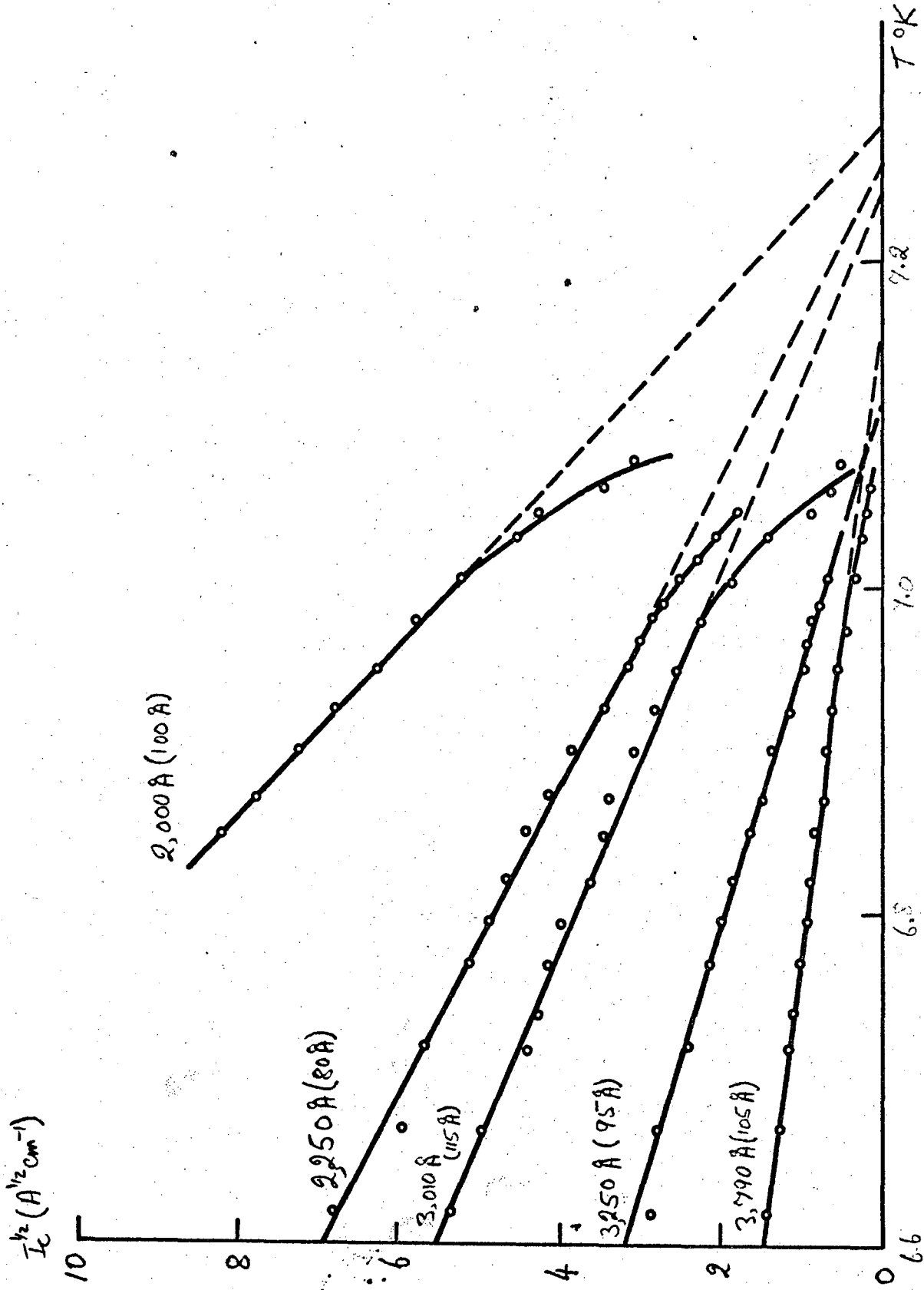
XBL 681 - 1752

Fig. 11



XBL681-1753

Fig. 12



XBL681-1754

Fig. 13

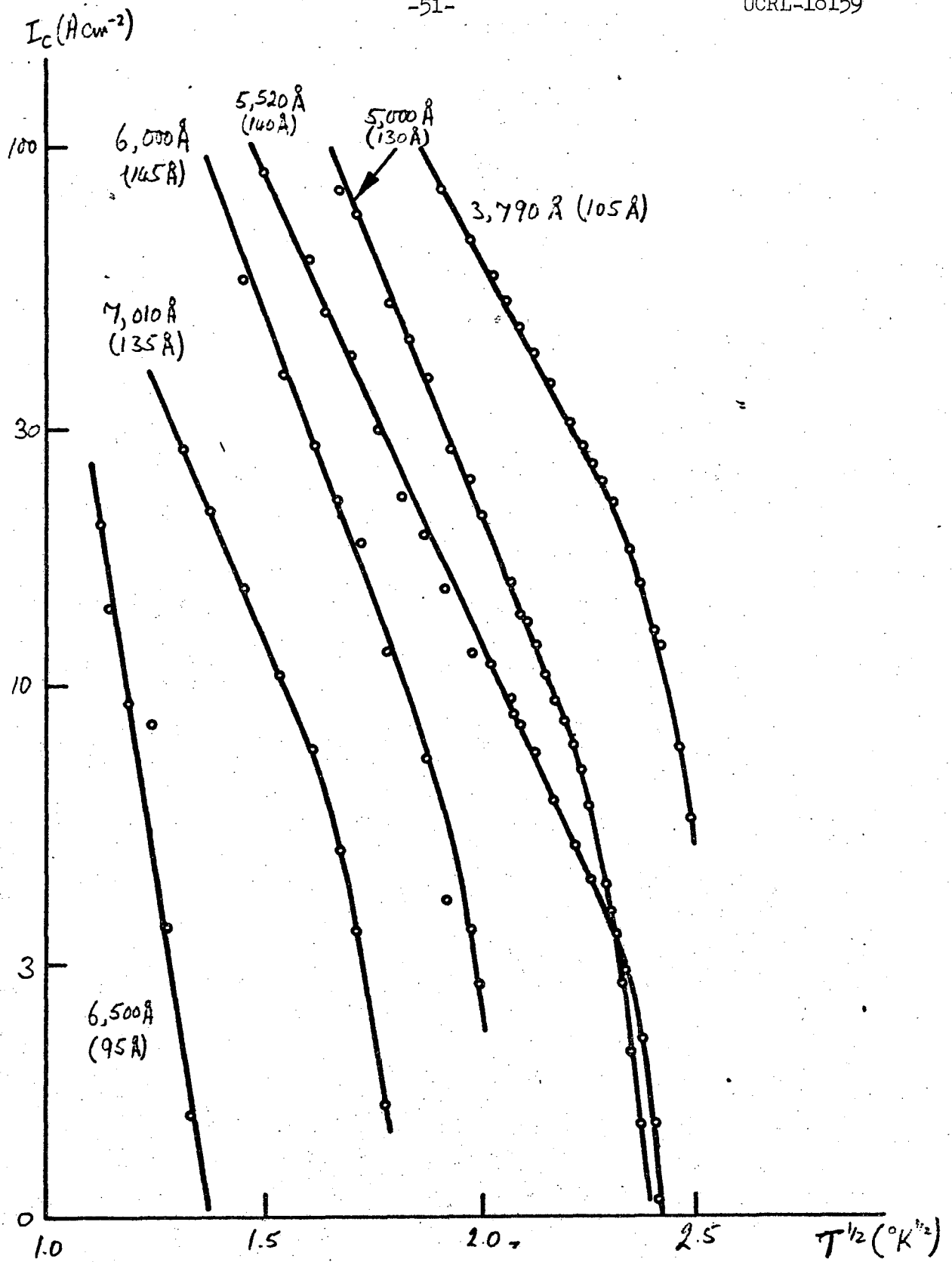


Fig. 14

XBL681-1755

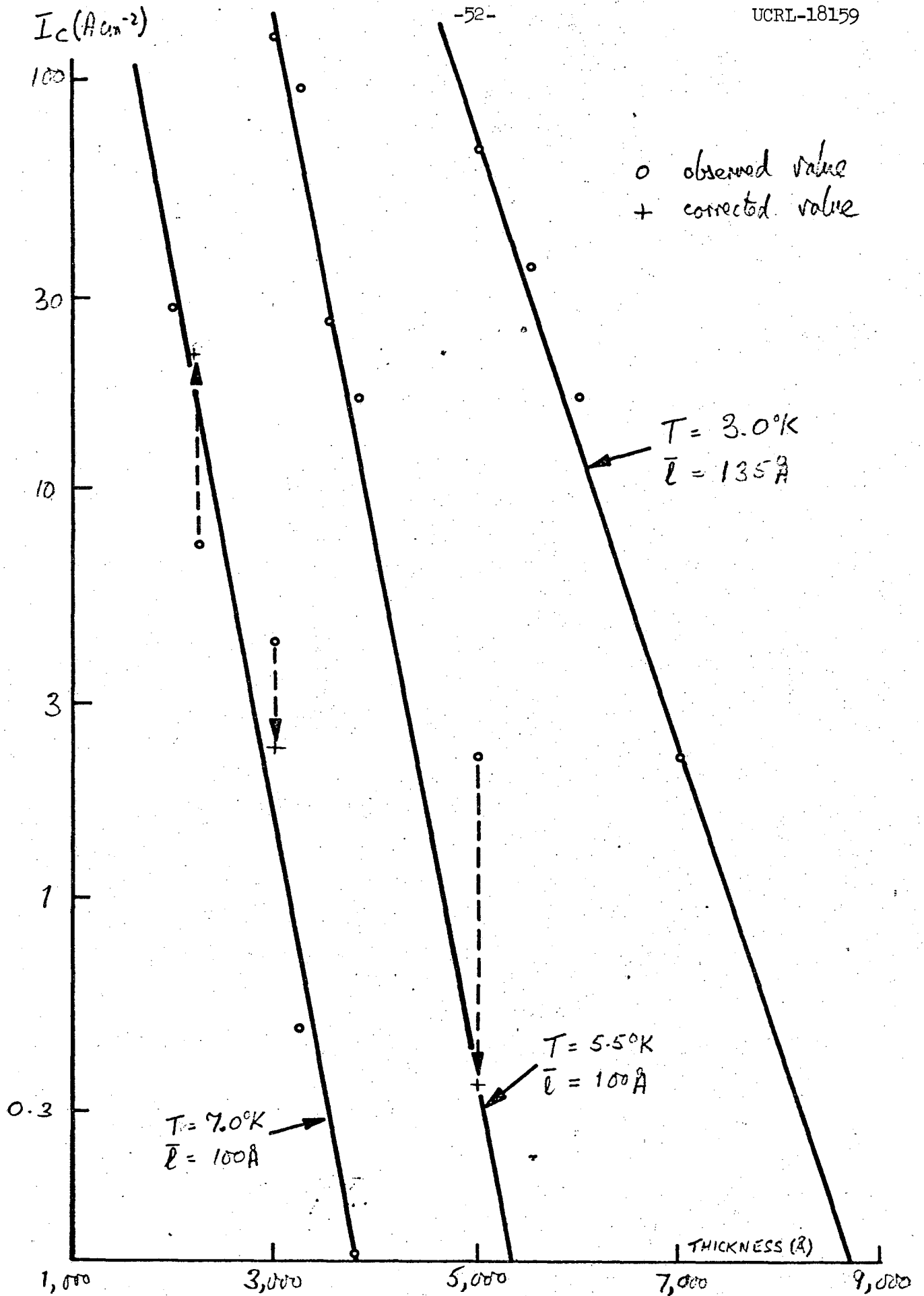


Fig. 15

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