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Nb-Nb Thin Film Josephson Junctions*

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ABSTRACT

We report a simple technique for the construction of thin film Josephson tunnel junctions in which both electrodes are of sputtered Nb. A thin (8Å-20Å) layer of Cu evaporated over the first oxidized Nb film prevents the formation of supershorts between the Nb films. The junctions are extremely rugged, can be stored and cycled indefinitely, and do not require protection against condensation of water vapor. Critical currents in the range $1\mu A < I_c < 10mA$ may be achieved with an accuracy of ±20%. The dependence of junction characteristics on temperature, oxidation time, and Cu thickness is described in detail, and a brief discussion is given of anomalous features in the I-V characteristics. The junctions are particularly suited to the fabrication of practical devices such as SQUIDs.

I. INTRODUCTION

A number of authors[1-6] have reported the successful fabrication of Nb-NbO_x-Pb Josephson tunnel junctions with reasonably predictable characteristics. These junctions can be stored indefinitely at room temperature and cycled repeatedly to liquid helium temperatures, indicating that the NbO_x barrier is very stable. However, while the Nb thin film strips have high mechanical strength, adhere strongly to glass substrates, and are very resistant to corrosion, the lead strips share none of these desirable characteristics. The Pb must be well protected against oxidation, which is particularly rapid in the presence of water vapor, and must be handled with considerable care to avoid peeling or abrasion. Nb-NbO_x-Nb junctions would appear to overcome the drawbacks associated with the use of Pb while still preserving high junction transition temperatures. The purpose of these experiments was to develop a simple and reliable procedure for constructing thin film Nb-Nb junctions with predictable and reproducible critical currents.

Although there is little discussion in the literature, it is apparently well known that the deposition of a second Nb strip directly on a first, thermally oxidized Nb film usually produces a superconducting short. Laibowitz and Cuomo[7] have successfully produced Nb-NbO_x-Nb junctions by epitaxially growing single-crystal Nb films on single crystal Al_2O_3 substrates and forming a barrier by sulphuric acid anodization. They were chiefly concerned with quasiparticle tunneling and mention only briefly the Josephson critical current. Laibowitz and Magadays[8] have utilized the good oxide forming characteristics of Al to construct composite Nb-Al-Al_2O_3-Nb junctions, with Al film thickness in the range 400Å to 1600Å. Josephson tunneling is possible due to the proximity of the normal metal film to the superconducting Nb. In our technique, an insulating barrier is formed by evaporating a few Ångstroms of Cu over an oxide barrier thermally grown on the first Nb film. The Ångstrom thick Cu layer reliably prevents the Nb films from superconductively shorting, although the physical mechanism involved is not fully understood.

II. FABRICATION TECHNIQUES

The first step in junction fabrication is the dc sputter deposition of Nb strips 2000Å thick and 150μm to 400μm wide on a soda glass slide using a Sloan Corporation S-300 Sputtergun.[5] The sputter chamber is initially evacuated to 10^{-6} Torr, and sputtering is performed in argon at 9×10^{-3} Torr. Commerically available Sputtergun targets of 99.9% purity are used. The substrate is initially at room temperature and experiences a temperature rise of about 50°C during the two-minute sputtering process. Sputtering is terminated either by switching off the discharge or by rapidly moving a shutter over the sample. Either procedure avoids the gradual decrease in the sputtering rate at the surface of the Nb strip which occurs if the discharge current is slowly reduced to zero. Immediately after the deposition of the first strip, the sample is removed from the sputterer and thermally oxidized in air at temperatures between 25°C and 130°C for times of 1 min to 20 min.

The sample is then transferred to an evaporator where a thin layer of Cu is deposited on the oxidized strips in the regions where junctions are to be formed. Evaporation occurs at pressures below 5×10^{-5} Torr with the sample well shuttered until the evaporation rate stabilizes at approximately $1\text{Å}\,\text{sec}^{-1}$. Typical Cu thicknesses, as measured by a crystal monitor, range from 8Å to 20Å and are controlled by opening and closing the shutter. Immediately after evaporation, the chamber is vented to air or backfilled with dry N_2. The sample is transferred to the sputtering system for the deposition of a final Nb cross strip of dimensions similar to those of the first. Since the purity of the first atomic layers of Nb is here expected to be most crucial, the sample is well shielded during an initial presputtering period of 1 min to remove adsorbates from the sputter target and to provide some measure of Nb gettering of residual chamber gases.

The masks used in sputtering are machined from Al. There is some evidence that the Nb films are slightly contaminated with Al sputtered from the masks. This contamination is reduced by allowing Nb to accumulate on the masks. Elaborate cleaning of the glass substrates produces results no better than those obtained by washing the slides in laboratory detergent, rinsing them thoroughly in hot tap water, and quickly blowing them dry with a strong jet of oil-free nitrogen to avoid leaving a residue on the substrate. There is no necessity for handling the substrates with plastic gloves, and excessive vacuum chamber cleanliness and long pump down times do not appear to improve junction quality. The total time required to construct a sample slide of eight junctions is typically 1h. Transition temperatures of the Nb films lie between 8K and 8.5K.

III. JUNCTION CHARACTERISTICS

Typical I-V and differential resistance characteristics for a junction made with an oxidation time of 2 min at 125°C and a Cu thickness of 8Å are shown in Fig. 1. The primary features of interest are the existence of a Josephson critical current and the rapid rise in quasiparticle current that occurs near 2.3mV at 4.2K. Also apparent are a structure near 0.7mV labeled A, consisting of a slight current rise followed by a conductance minimum, and a "knee", or region of high differential resistance, just above the rapid current rise.[1-6,9] When the junctions are cooled from 4.2K to 1.5K the current rise at 2.3mV steepens slightly and moves to roughly 2.5mV. The return to ohmic behavior above the knee moves to higher voltages in a similar manner, while the differential resistance of the knee increases by at least two orders of magnitude and may become negative, with consequent switching effects when the junction is current-biased. The zero voltage conductance decreases by a factor of about 15 on cooling. These features are quite reproducible under a wide range of junction parameters.

There are substantial excess quasiparticle currents at voltages below that of the rapid current rise, an effect commonly observed in junctions having one Nb electrode.[1-9] The excess currents below 0.7mV diminish rapidly with temperature while those above 0.7mV are less dependent on T. As discussed by Rowell and Feldman,[10] such excess currents preclude a detailed theoretical analysis. However, the rapid rise in current is undoubtedly associated with the sum $(\Delta_1+\Delta_2)$ of the superconducting energy gaps in the two Nb films. If we take the voltage at which dI/dV is a maximum as a measure of $(\Delta_1+\Delta_2)$ (J. M. Rowell, private communication), then for the junction of Fig. 1 we obtain $(\Delta_1+\Delta_2)/e\approx 2.3$mV. This value is several tenths of a mV lower than that expected[1-9] for an ideal Nb-NbO$_x$-Nb junction with the same transition temperature (8.5K), suggesting that the gap in one or both of the Nb strips is slightly suppressed. If we associate the weak current rise at 0.6mV with a gap difference $(\Delta_1-\Delta_2)$, we find $\Delta_1\approx 1.4$mV and $\Delta_2\approx 0.9$mV at 4.2K. This procedure may not be wholly justified: Although the structure labeled A contains a component whose position decreases[11] a few tenths of a millivolt when the junction is cooled to 2K, it also has components that appear as conduction minima[10] and depend in a complicated manner on temperature. Nonetheless, the value of Δ_1 deduced from this procedure is comparable with values reported elsewhere [1-9] and likely corresponds to the gap in the lower Nb strip. The smaller value of Δ_2 may then be interpreted as the gap in the upper film, which is reduced by the presence of the Cu layer.

This interpretation is consistent with the observation that the voltage at which dI/dV is a maximum depends strongly on the Cu thickness. As shown in Fig. 2, this voltage decreases from 2.4mV for 8Å of Cu to 1.4mV (approximately Δ_{Nb}) for 50Å of Cu. Concurrently, the structure A moves to higher voltages, while the deduced values of Δ_1 remain at 1.4±0.2mV. The gap depression by a relatively thin layer of Cu is too large to be explained by a simple proximity effect model. It is more likely that the Cu, or gases adsorbed on the Cu, act as an impurity in the surface of the second Nb strip, thus producing a gapless surface layer. This effect may also be responsible for the knee[4] above $(\Delta_1+\Delta_2)$ and the large excess currents below $(\Delta_1+\Delta_2)$.[1,4,10]

IV. CRITICAL CURRENTS

I_c can be varied predictably from less than 1μA to about 10mA by controlling the oxidation time of the first Nb strip and the thickness of the Cu layer. The reproducibility is about ±20% for samples made at different times, and about ±3% for junctions made simultaneously and in close proximity. Figure 3 shows the dependence of I_c on Cu thickness for an oxidation time of 5 min at 125°C. I_c decreases by two orders of magnitude as the Cu thickness is increased from 10Å to 20Å. Although the Cu film is undoubtedly non-uniform across the junction area, the critical current varies smoothly with the average Cu thickness as measured on a crystal monitor. For a given thickness of Cu, the critical current decreases by roughly one order of magnitude as the oxidation time at 125°C is increased from 2 min to 20 min, as shown in Fig. 4 for a 15Å Cu layer.

The temperature dependence of the critical currents for various thicknesses of Cu is shown in Fig. 5. For an 8Å Cu layer, the variation in I_c over the temperature range 1.2K to 4.2K is weak and close to that predicted for an ideal oxide barrier tunnel junction. For Cu thicknesses of 10Å and 15Å, the temperature dependence of I_c is more pronounced. This dependence is not understood, but it may be associated with the increase in gaplessness as the Cu thickness is increased. Figure 5 also includes data for two samples (oxidation time 2 min) with much thicker and presumably more uniform Cu layers. The strong temperature dependence of I_c is primarily due to the increase in the pair decay length in the Cu ($\sim \hbar v_F/k_B T$) as the temperature is lowered and is reminiscent of that observed in SNS Josephson junctions.[12]

The junction critical currents modulate to zero in the presence of an external magnetic field applied parallel to one of the Nb strips, usually with several modulation periods observable. Figure 6 shows a typical Frauenhofer-like diffraction pattern. Although the modulation is not precisely of the form[4] $|(\sin x)/x|$, its general shape implies that the supercurrent has a reasonably uniform distribution across the junction and is not carried by a small number of microshorts. From the modulation period of approximately 0.5G and the Nb strip width of 375μm we deduce a value of 500Å for the penetration depth in the Nb films, assuming it to be equal in each film. Schwidtal[4] found values varying between 674Å and 1215Å, depending on the film purity. The bulk value[13] is about 400Å.

The normal state resistances (R) of our junctions lie in the range 0.1Ω to 100Ω. The maximum Josephson critical current is always less than that predicted for an ideal oxide barrier tunnel junction[4,14] of the same normal state resistance by a factor of typically 6. The low values of I_cR are to some extent caused by the conduction mechanism which is responsible for the excess currents and which shunts the junctions to lower resistances. Impurity induced gaplessness and mechanical strains are also likely causes for reduced values of I_c.[15] Nb-NbO_x-Pb junctions constructed in the same apparatus have values of I_cR no greater than one-half the theoretical maximum.

V. JUNCTION RELIABILITY

The Nb-Nb junctions have extremely stable storage and cycling characteristics, which we attribute to the stability of the NbO_x. After several initial thermal cyclings, during which the critical currents usually rise about 10%, the junction characteristics exhibit no significant changes, at least after 50 thermal cyclings over a period of 6 months. The junctions require no protective coatings. They may be removed from the cryostat, exposed to the atmosphere, and replaced in the cryostat, even though a layer of water and ice forms in the process. The junctions can withstand high currents and current pulses (>300mA) without destruction and usually without flux trapping. Flux trapping from pulsed magnetic fields is significantly less in the Nb-Nb samples than in unshunted $Nb-NbO_x-Pb$ junctions.

VI. DISCUSSION

Of the many explanations[10,16] advanced to account for excess currents in tunnel junctions, microshorts and gaplessness appear to be the only possibilities consistent with the magnitude of the observed effects. The rapid decrease in the zero bias conductance of our junctions with decreasing temperature would appear to be inconsistent with a microshort model[4]. Also, we have made Nb-Nb junctions <u>without</u> Cu by presssing together, in liquid helium, two thermally oxidized Nb strips deposited on separate glass slides. These junctions have I-V characteristics similar to those shown in Fig. 1, with critical currents suppressed by a field of about 1G. Microshorts are less likely to be present in this configuration, since oxide defects are less likely to overlap in a geometry employing two separately oxidized strips than for Nb sputtered directly onto a single oxidized film. Although we cannot rule out the possibilities that abrasion could damage the surface oxide and thereby produce microshorts, we observe no irreversible behavior in breaking and reforming the pressed film junctions under helium. We tend to believe that gaplessness at the Nb surface most likely accounts for the excess currents and that Cu alone is not entirely responsible for this effect.

The role of the Cu in preventing superconductive shorting between the Nb films is not well understood. We have investigated the composition of the junctions in the vicinity of the interface using simultaneous ion etching and Auger analysis. The presence of the Cu layer is just detectable. The total oxygen content of the barrier region is essentially independent of Cu thickness, suggesting that the Cu serves primarily to stabilize an existing oxide against destructive penetration by freshly sputtered Nb. There are several mechanisms by which this might occur. The Cu could preferentially fill point defects in the thermal oxide of the first Nb strip. SEM studies of the oxidized Nb film, with or without Cu, show an essentially featureless surface on a 100Å scale. However, it is not clear whether this technique would reveal oxide defects. Alternatively, the Cu could alloy uniformly onto a defect-free oxide surface to form a composite barrier that is not penetrated by the second Nb film. In this case, we speculate that without Cu, shorting occurs when the second Nb strip is deposited because of a chemical reduction of the final surface oxide phase, Nb_2O_5, an insulator, to non-insulating oxide phases stable at lower oxygen concentrations. In particular NbO, which is a conductor, has been proposed as a source of excess currents.[2] Laibowitz and Cuomo[7] have successfully produced oxide barriers that are not destructively reduced by the deposition of a second Nb strip. However, it is unclear whether anodization produces a defect free surface or whether the insulating oxide layer is simply too thick to be fully reduced. J. Matisoo (private communication) has suggested that the Cu layer might absorb the kinetic energy of the incident Nb atoms that would otherwise penetrate the oxide layer.

We have also constructed junctions with similar characteristics using Ångstrom layers of Ti, Cr, and Pb in place of Cu. The range of acceptable thicknesses is much narrower than for Cu. However, the possibility of using a high melting point material, such as Ti, should be investigated more fully, since there are some indications[17] that high temperature outgassing just prior to deposition of the second electrode is effective in reducing excess currents.

We have fabricated dc SQUIDs using Nb-Nb junctions shunted with Ti underlays to eliminate hysteresis in the I-V characteristic. A thick oxide layer may be grown on the Ti and Nb by immersing the films in hydrogen peroxide. A Pb ground plane can then be evaporated directly over the structure with no further insulating layer. The resistance between the Ti, Nb and the Pb is typically 100Ω to 1000Ω for an overlap area of 1 cm^2.

Depite the existence of substantial excess currents and the lack of a good physical model of the barrier structure, the predictable characteristics, simplicity of preparation, and durability of Nb-Nb junctions make them attractive candidates for applied devices such as SQUIDs and for maintaining and comparing standards of EMF.

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REFERENCES

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1. J. E. Nordman, J. Appl. Phys. 40, 2111 (1969); J. E. Nordman and W. H. Keller, Phys. Lett. 36A, 52 (1971).
2. L. O. Mullen and D. B. Sullivan, J. Appl. Phys. 40, 2115 (1969).
3. R. Graeffe and T. Wiik, J. Appl. Phys. 42, 2146 (1971).
4. K. Schwidtal, J. Appl. Phys. 43, 202 (1972).
5. P. K. Hansma, J. Appl. Phys. 45, 1472 (1974).
6. Paul Rissman and Thomas Palholmen, Solid St. Electronics 17, 611 (1974).
7. R. B. Laibowitz and J. J.Cuomo, J. Appl. Phys. 41, 2748 (1970).
8. R. B. Laibowitz and A. F. Mayadas, Appl. Phys. Lett. 20, 254 (1972).
9. M. L. A. MacVicar and R. M. Rose, Phys. Lett. 25A, 681 (1967); 26A, 510 (1968).
10. J. M. Rowell and W. L. Feldmann, Phys. Rev. 172, 393 (1968).
11. B. N. Taylor, J. Appl. Phys. 39, 2490 (1968).
12. J. Clarke, Proc Roy. Soc. A308, 447 (1969).
13. R. A. French, Cryogenics 8, 301 (1968).
14. P. W. Anderson, in Lectures on the Many-Body Problem, edited by E. R. Caianiello (Academic, New York, 1964), Vol. 2.
15. J. J. Hauser, Phys. Rev. 164, 558 (1967).

16. I. Giaever, H. R. Hart, Jr., and K. Megerle, Phys. Rev. 126, 941 (1962); B. N. Taylor and E. Burstein, Phys. Rev. Lett. 10, 14 (1963); C. J. Adkins, Phil. Mag. 8, 1051 (1963); G. B. Donaldson, Solid State Commun. 5, (1967); J. R. Schrieffer and J. W. Wilkins, Phys. Rev. Lett. 10, 17 (1963); J. L. Levine, Phys. Rev. 155, 373 (1967); J. Millstein and M. Tinkham, Phys. Rev. 158, 325 (1967); M. A. Woolf and F. Reif, Phys. Rev. 137, A577 (1965); J. J. Hauser, Physics 2, 247 (1966); L. Y. L. Shen and J. M. Rowell, Phys. Rev. 165, 566 (1968).

17. P. Townsend and J. Sutton, Phys. Rev. 128, 591 (1962); L. Y. L. Shen and J. M. Rowell, Phys. Rev. 165, 566 (1968).

FIGURE CAPTIONS

Fig. 1. I-V and differential resistance characteristics at 4.2K and 1.5K of a typical Nb-Nb thin film junction with an oxidation time of 2 min at 125°C and a Cu thickness of 8Å.

Fig. 2. Voltage of maximum dI/dV as a function of Cu thickness.

Fig. 3. I_c versus Cu thickness for a thermal oxidation period of 5 min at 125°C.

Fig. 4. I_c versus oxidation time at 125°C for 15Å Cu.

Fig. 5. I_c versus temperature for several values of Cu thickness.

Fig. 6. Modulation of I_c by an external magnetic field. The dotted curve is $|(\sin x)/x|$ fitted at $H_{ext} = 0$ and at the first minimum.

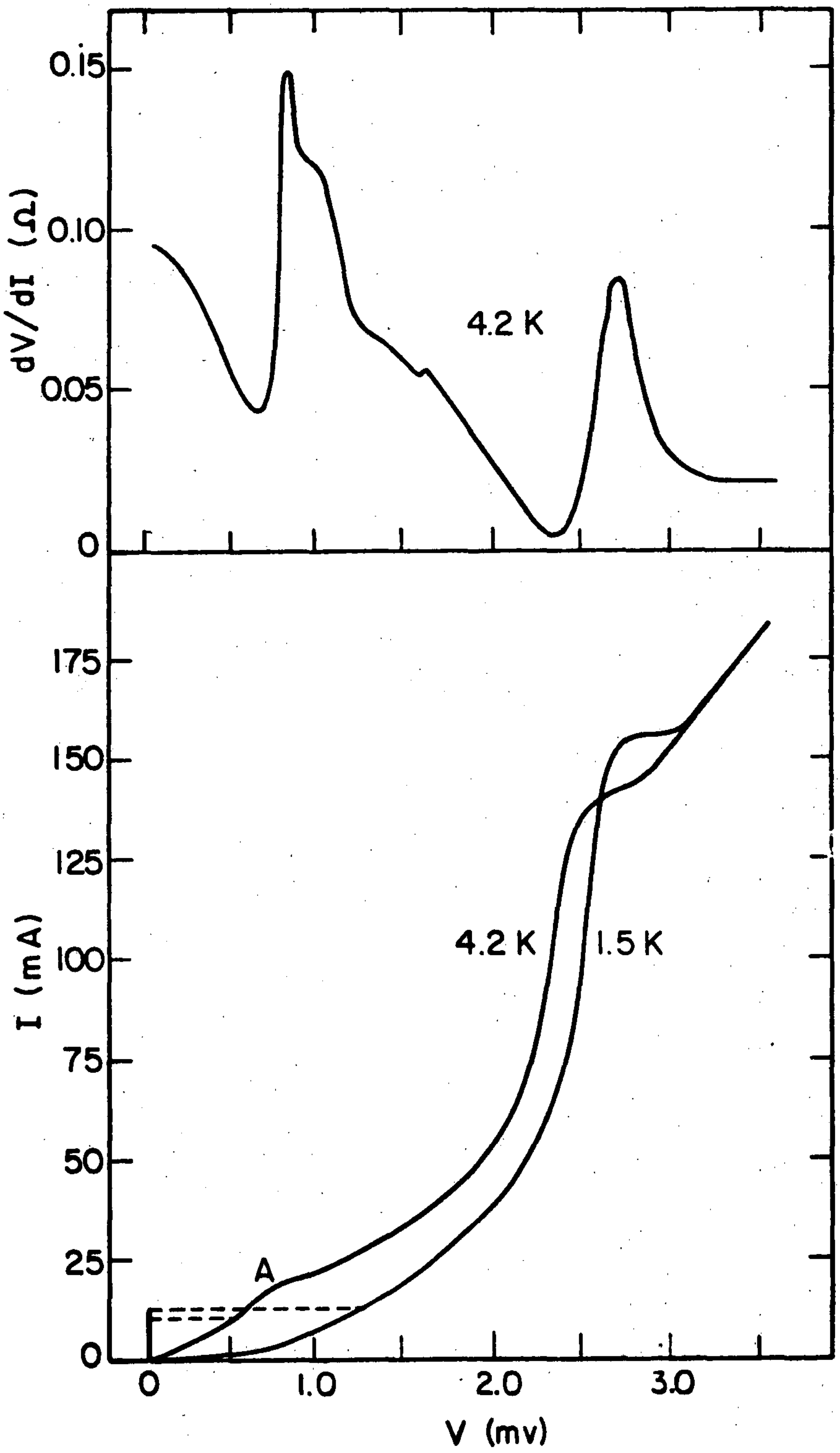

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Fig. 1

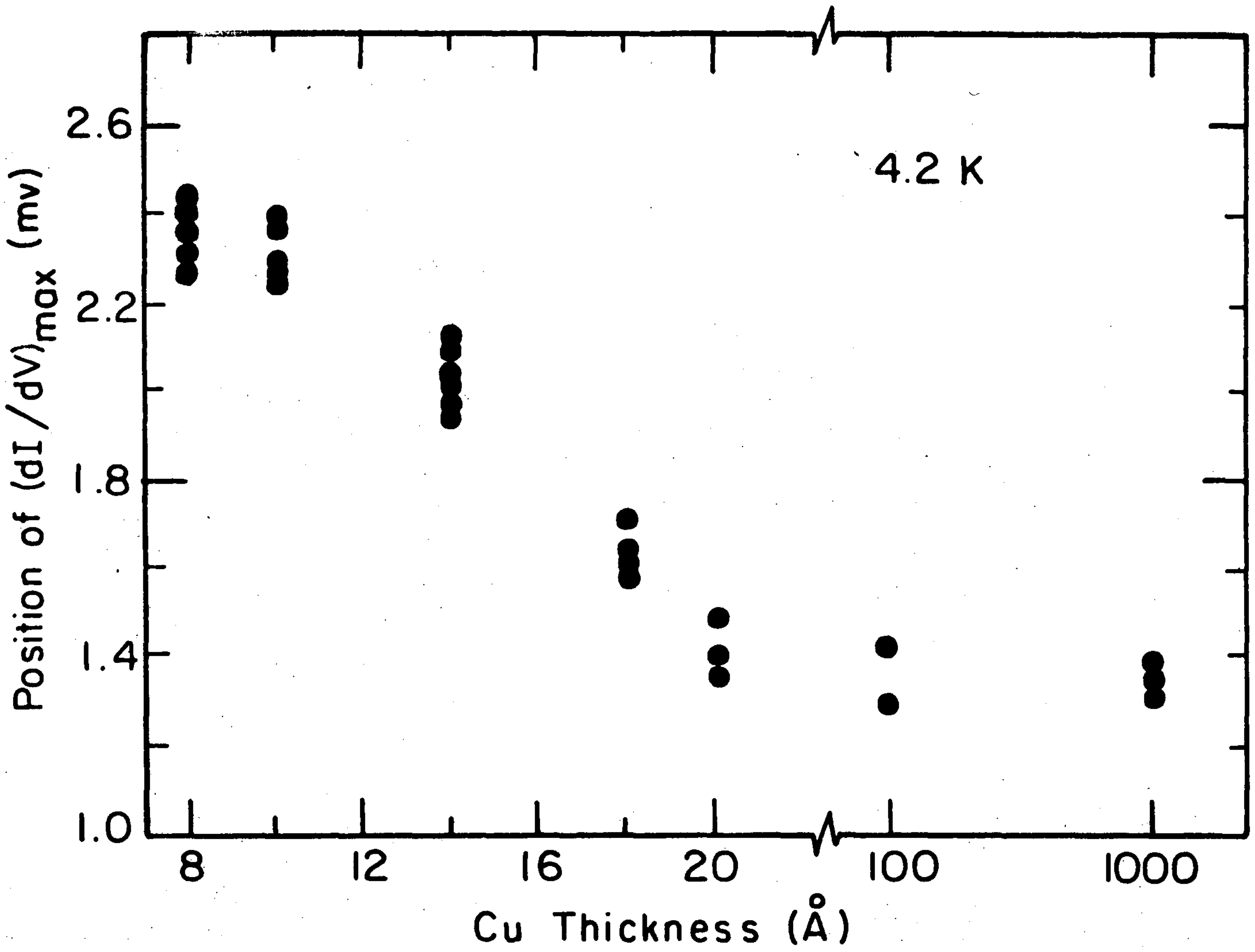

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Fig. 2

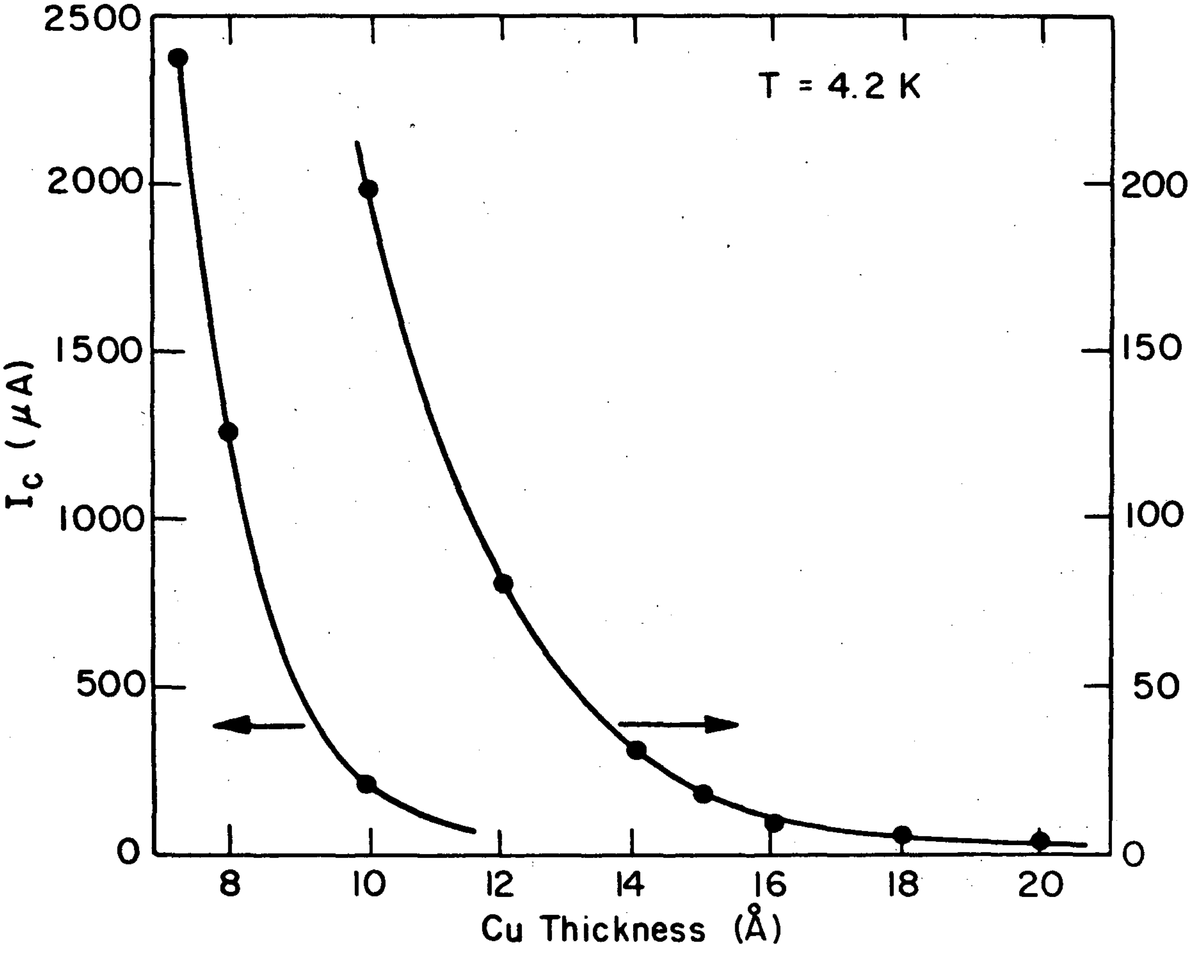

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Fig. 3

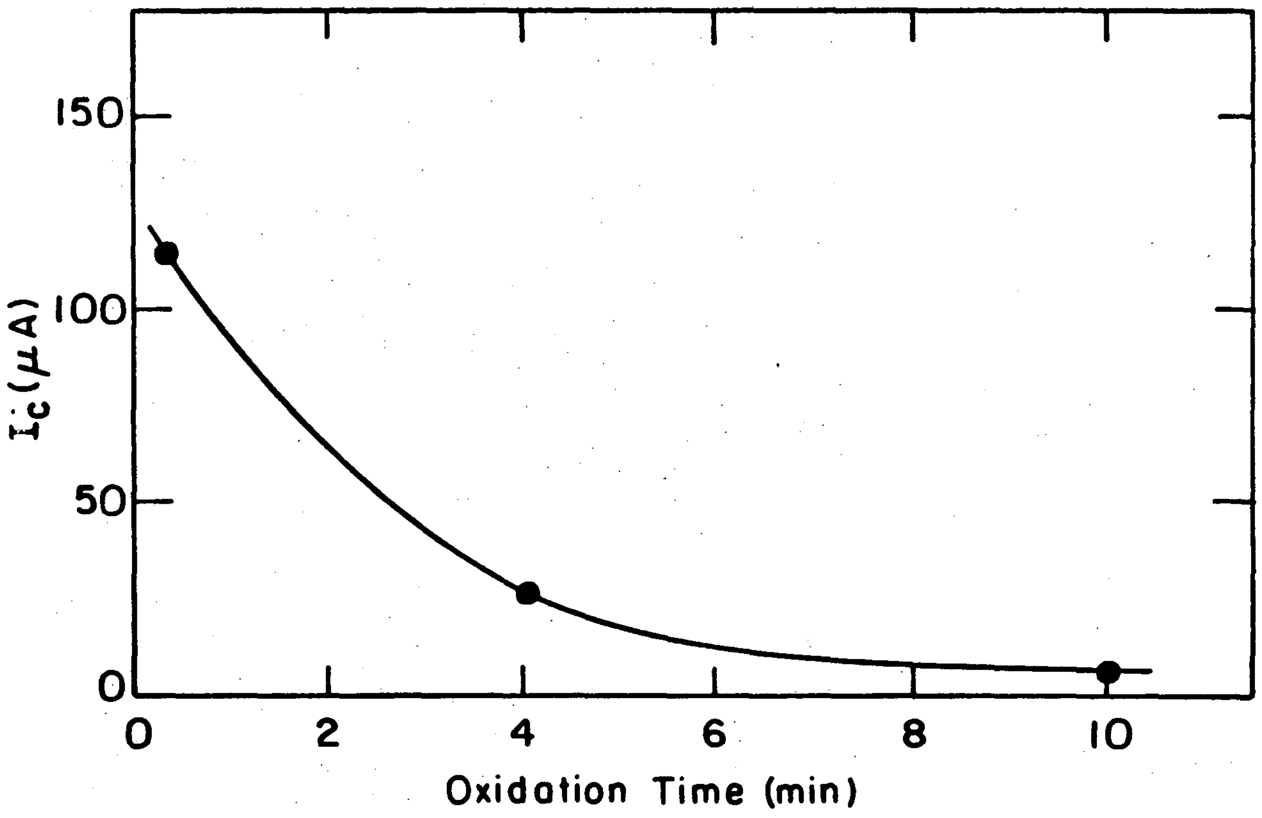

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Fig. 4

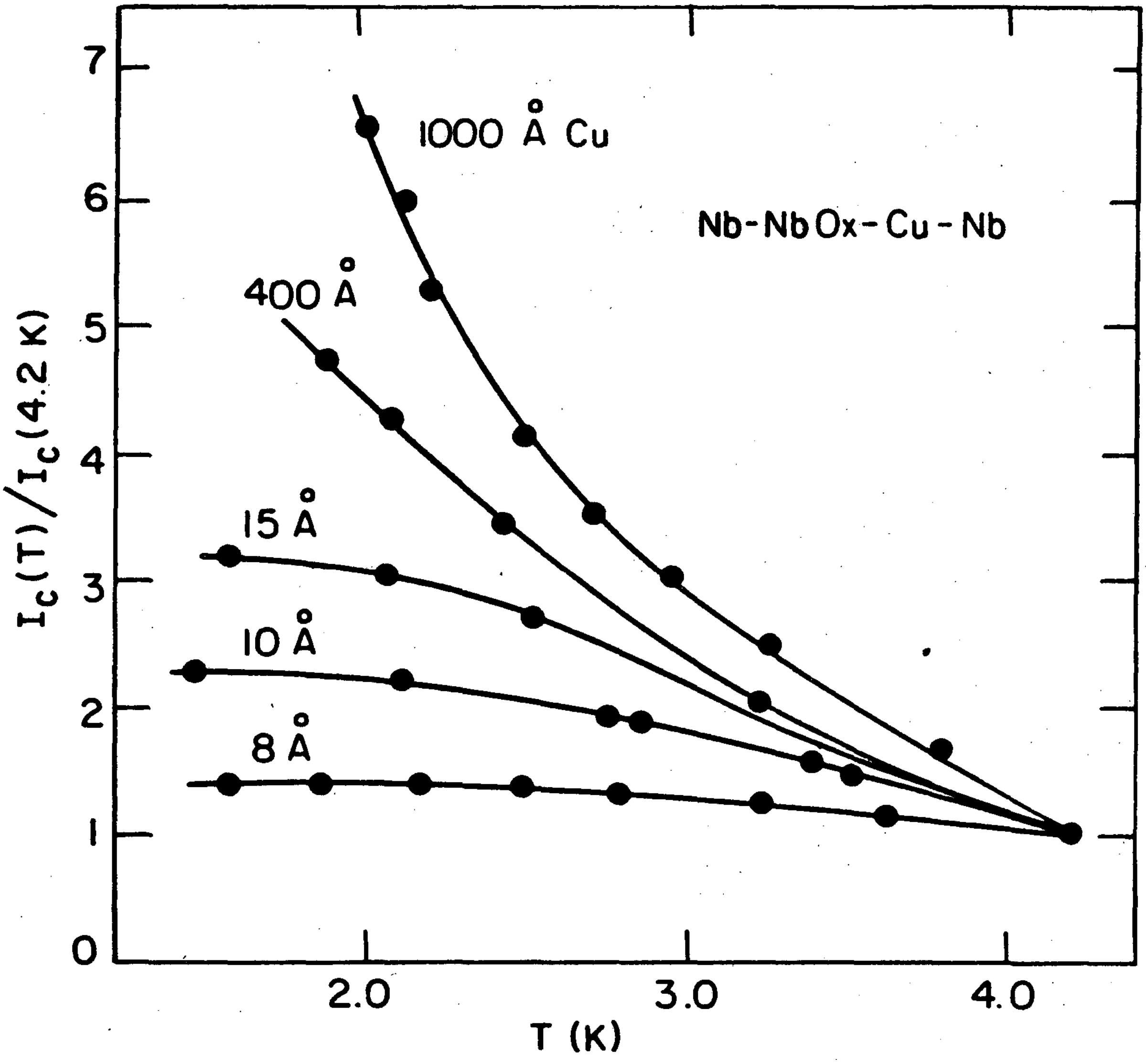

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Fig. 5

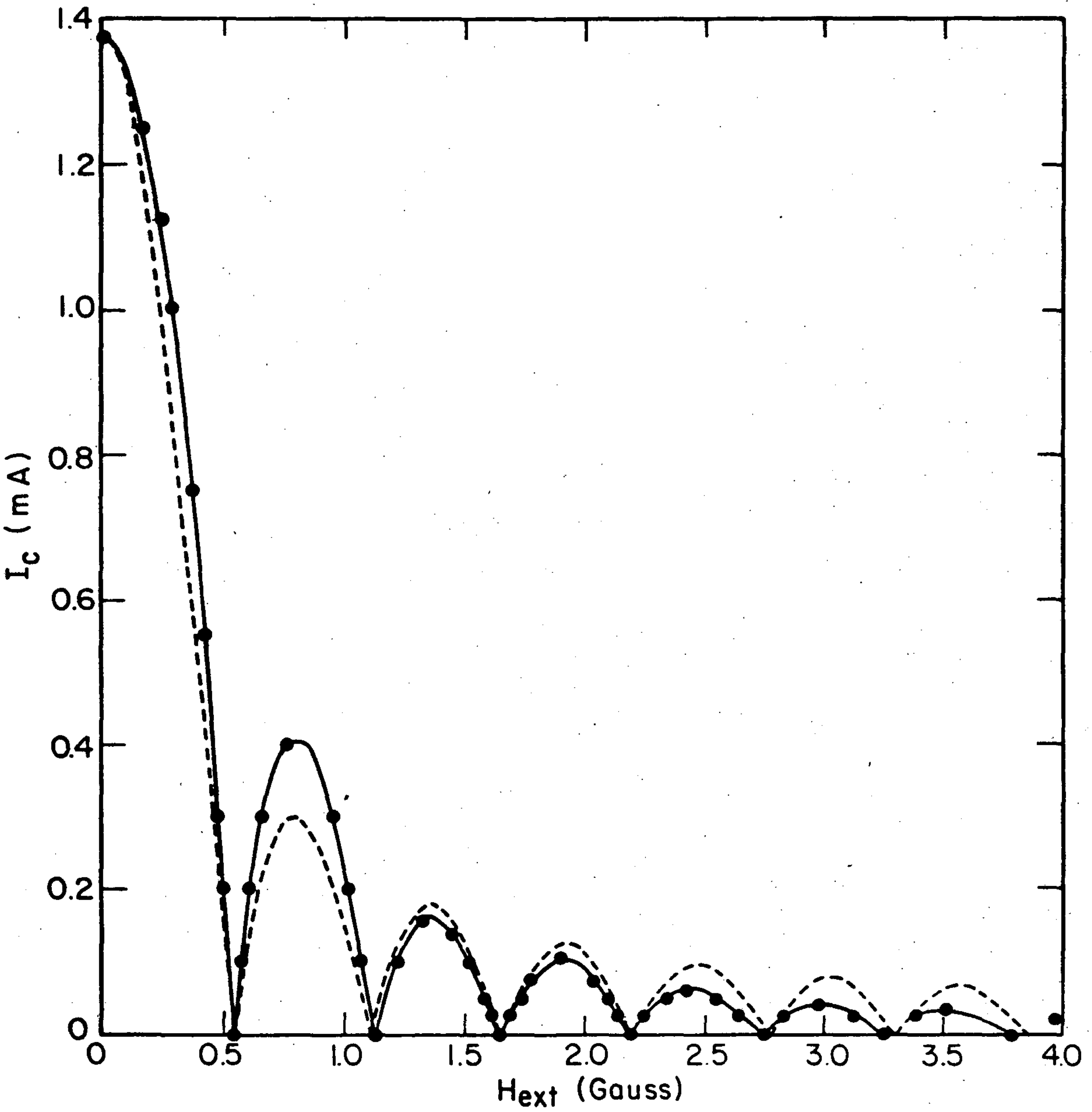

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Fig. 6

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