Measurements on Superconducting Contacts*

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The contact resistance between crossed wires has been measured as a function of the current at various temperatures in the liquid helium range. Most contacts were stable enough to establish a "diagram of state," i.e., determine curves of constant resistance in I-T space. The following facts have been established:

1. The critical temperature of contacts between clean wires of tin is suppressed by about 0.2°K due to the pressure on the contact.
2. The addition of a copper layer on one or both of the wires reduces the critical currents, but hardly influences the critical temperature. This can be understood if one assumes that the density of the superconducting electrons decreases in the copper layer, thus producing an increase in the penetration depth. At layer thicknesses of several hundred angstroms the penetration depth becomes large compared to the contact radius, and the critical current *vs* temperature curve approaches that of a very thin wire.
3. Contacts between tin and copper wires below the critical temperature and at low currents show a constant resistance, which rises sharply at a critical current. Graphs of this quasi-critical current as a function of the temperature have been obtained.
4. Clean contacts between tin and indium usually behave as one would expect from the foregoing. In one case out of three, however, the resistance was strongly dependent on the current at a temperature as high as 4.2°K. Plotting the low-current values of the resistance as a function of the temperature showed a behavior as if one of the contact materials had a critical temperature of 5 to 6°K. A search of the literature revealed that the Sn-In system has two intermetallic compounds. The compound In_6Sn_2 has been prepared, and was found to have a critical temperature of about 5.5°K. Since the contacts were closed at 4.2°K and no possibility of transfer of metal from one side to the other existed, it must be assumed that the proximity of the tin to the indium is sufficient to produce partial superconductivity by way of long-range correlation of the electronic wave functions.

I. INTRODUCTION

It has been known for quite some time[1–3] that the contact resistance between two metals is reduced if one side becomes superconducting, and vanishes at sufficiently low currents and temperatures if both sides are superconducting. It has been pointed out[3,4] that measurements on contacts with barriers as well as of electron emission should be capable in principle of giving information about the electronic states in a superconductor.

Previously contacts have been investigated which had their naturally grown oxide layers[1,3] or which had insulating barriers produced by evaporation. These contacts were usually not very stable, so that the results are rather scant. Nevertheless it was possible to obtain an approximate "diagram of state," i.e., lines of constant resistance in I-T space (I is the current and T the temperature).

The experiments to be described here deal mainly with contacts between crossed wires of tin of which either one or both were electroplated with various thicknesses of copper. Great care was taken in order to avoid additional barriers, although an adsorbed gas layer was certainly always present.

Most contacts were stable enough so that the temperature could be lowered from 4.2°K to well below the critical temperature of tin (3.72°K) and raised again to 4.2°K without any appreciable change in the characteristics of the contact. Thus, it was possible to obtain "diagrams of state" for quite a number of contacts and to study how they are affected by the thickness of the copper layer.

The experiments have been extended to contacts between clean tin wires and to contacts between wires of tin and copper. Furthermore, a few contacts between wires of tin and indium have been investigated.

II. EXPERIMENTAL ARRANGEMENT

The cryostat, the circuitry, and the mount for the contacts were the same as used earlier (see reference 3, Figs. 1 and 2). The temperature was maintained constant within a millidegree by an automatic control described elsewhere.[5]

The temperature was determined from the vapor pressure above the liquid helium with a standard U-type manometer, using the 1955_E vapor pressure scale.[6] This simple manometer was probably not quite accurate enough at the lowest temperatures (about 1.4°K) which might therefore be in error by as much as 0.02°K. All other temperatures were rounded off to the nearest 0.01°K. No corrections were made for the hydrostatic pressure head.

III. EXPERIMENTAL PROCEDURE

The tin wires of 0.5-mm diameter were extruded from "high purity" tin of the Vulcan Detinning Company, for which the manufacturer lists a purity of 99.98%. The residual resistivity of extruded wires of this material has been measured in connection with other experiments (see reference 5) and is of the order of 1.2×10^{-4} of the ice-point resistivity.

* Work supported by a contract of the Office of Naval Research.
[1] R. Holm and W. Meissner, Z. Physik **74**, 715 (1932).
[2] I. Dietrich, Z. Physik **133**, 499 (1952).
[3] F. Bedard and H. Meissner, Phys. Rev. **101**, 26 (1956).
[4] Bedard, Meissner, and Owen, Phys. Rev. **102**, 667 (1956).
[5] Hans Meissner, this issue [Phys. Rev. **109**, 668 (1958)].
[6] Clement, Logan, and Gaffney, Phys. Rev. **100**, 743 (1955); see their "Note added in proof."

Commercial copper wire of 0.5-mm diameter was used for the copper wires.

The indium wires of 0.5-mm diameter were extruded from 99.97% pure indium of the Indium Corporation of America. Their residual resistivity is about 4×10^{-4} of the icepoint resistivity, as determined in the course of other experiments.[7]

The tin wires were cleaned by electrolytic polishing in a solution of 20 parts perchloric acid and 70 parts acetic acid, washed in distilled water and alcohol. If they received a copper layer, they were immediately plated in a solution of 20-g KCN, 8-g NH_4OH, 14-g $Cu(C_2H_3O_2)_2$ in 1 liter of water.

It was found by plating for an hour and weighing, that a current of 2 ma deposits on a wire of 0.5-mm diameter and 3-cm length a copper layer of 100 A thickness in 8.3 sec. All copper thicknesses were then calculated from the plating time. After plating the wires were again washed in distilled water and alcohol. The copper wires were only cleaned by scraping with a pen knife.

The indium wires were cleaned by electrolytic polishing in a solution of 2 parts methyl alcohol and 1 part concentrated nitric acid. The solution was cooled with dry ice. The wires were also washed afterwards in distilled water and alcohol.

All contact wires were installed immediately after their preparation. The Dewar vessel was then closed, evacuated, filled with dry helium gas, and precooled by means of an inserted well which was filled with liquid nitrogen. The well was then removed and liquid helium was siphoned in. The contacts were closed only after the siphon was removed and everything was ready for the run. The time which elapsed between the electrolytic polishing and the closing of the contacts was $1\frac{1}{2}$ to 2 hours.

The contact load was always 60 g. However, in some runs the hinges (see reference 3, Fig. 1) froze to the extent that the contact remained completely open. It must therefore be taken into consideration that friction in the hinges might occasionally reduce the contact load. This seems to be the case in some of the contacts.

It should be noted that the electrical circuitry is such that the potential across the contact cannot exceed a few tenths of one volt, even if the contact is open. Since the experiments were always started with the lowest current, the potential difference was in practice even much smaller. Any possibility of a formation of a bridge due to coherer action is therefore excluded (see reference 3, p. 30).

The resistance of the contact was measured by reversing a known current through it and observing the deflection of a galvanometer connected to the potential leads of the contact (see reference 3, Fig. 2).

If the contacts were asymmetric, rectification was sought for by observing the deflection at all three positions of the current switch: I—0—II. In position I the (conventional) current goes always from the plated to the unplated wire or from the indium to the tin wire respectively. (In position II the current goes in the opposite direction.)

A Leeds and Northrup K2 potentiometer was kept in the galvanometer circuit all the time, and was used for the calibration of the galvanometer and for compensation of thermal emf's. The galvanometer (L and N type HS No. 2284-c) was used with a telescope and a scale at 5-m distance and had a sensitivity of 3.7×10^8 mm/v (for reversal) which could be shunted down by factors of 10^{-1}, 10^{-2}, and 10^{-3}.

IV. MEASUREMENTS ON CONTACTS BETWEEN BARE TIN WIRES

Figure 1 shows plots of resistance *vs* current for contacts *A* and *B*. The resistance is independent of the current down to 3.62°K, which is below the critical temperature of tin (3.72°K). At lower temperatures, the contacts are superconducting at low currents, the resistance reappearing above a critical current. When the temperature was returned to 3.81°K (curve 7) the resistance of the contacts returned quite precisely to the initial value.

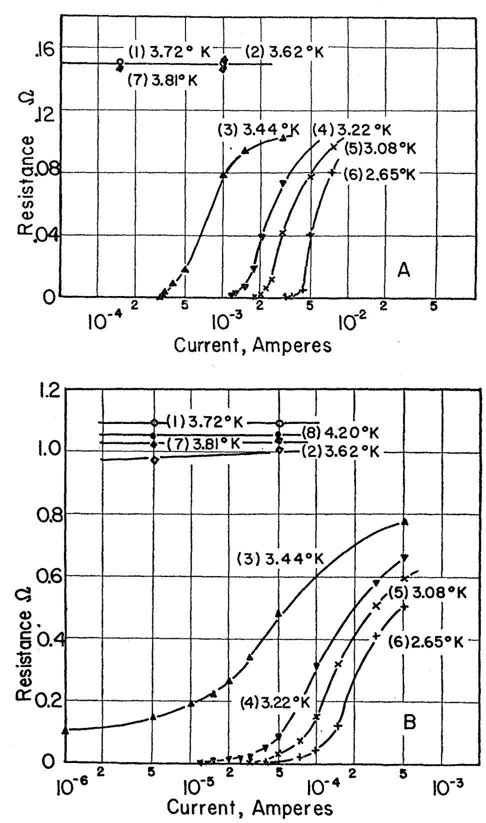

FIG. 1. Plots of resistance *vs* current for the contacts *A* and *B* between bare tin wires. The numbers on the curves give the sequence and the temperature of the measurements.

[7] Hans Meissner and Richard Zdanis, preceding paper [Phys. Rev. **109**, 681 (1958)].

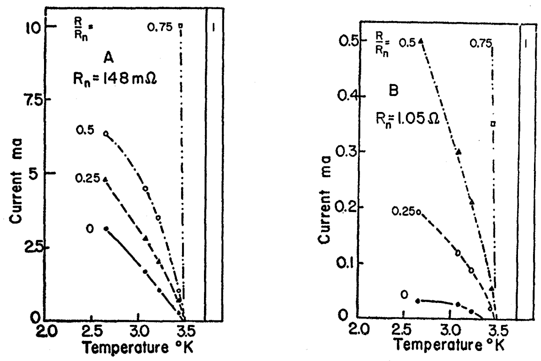

FIG. 2. Diagrams of state for the contacts A and B between bare tin wires. The curve $R=0.25R_n$ is by definition the critical field curve.

Figure 2 shows plots of constant resistance in the I-T space. Any one of the curves $R=0$, $R=0.25R_n$, or $R=0.5R_n$ (R_n=normal resistance) can be taken as the "critical current" curve, depending on the definition which one prefers for the critical current. For a long straight wire all three curves would ideally coincide (see reference 5, Fig. 4). We shall take here $R=0.25R_n$ as the "critical current" for the following reasons: $R=0$ always includes "tails" (see contact B) and can therefore be distorted. $R=0.5R_n$ sometimes seems to be subject to influences other than the first reappearance of the resistance. The thus-defined "critical current" curve for contacts A and B is very similar to that of a solid wire, with the exception that the critical temperature is about 3.53°K instead of 3.72°K, which it usually is for tin. This shift is due to the pressure on the contact. Since the rate of change of the critical temperature with pressure is known to be[8] $dT_c/dp=-4.58\times10^{-5}$ °K/atmos, the pressure on the contact can be calculated. As can be seen from Table I, this pressure is always about 5×10^3 kg/cm², which has to be compared with a value of 5.5 to 11×10^3 kg/cm² which Holm and Meissner[9] found by direct measurement of the flow pressure of tin at 4.2°K.

As long as the diameter of the contact area is large compared to the penetration depth of tin ($\delta=5.0\times10^{-6}$ cm), the critical current curve should have the same temperature dependence as the critical field curve,[10] that is, in first approximation.

$$I_c/I_{c0}=H_c/H_{c0}=1-(T/T_c)^2, \tag{1}$$

where I_{c0} and H_{c0} are the values of the critical current and the critical field at absolute zero.

The ratios of the critical currents at two temperatures have been calculated and compared with the values expected from Eq. (1) (see Table I). The value of T_c is hereby chosen for best over-all fit. One can see that the agreement is reasonable considering that it is based on relatively crude assumptions.

The contact area can be calculated in two ways.

(1) The "load-bearing area" can be calculated from the pressure on the contact and the known force on it.

(2) The "current-bearing" area can be calculated from Silsbee's hypothesis which connects the critical current with the critical field.

The results in both cases differ widely. For the load-bearing radius one obtains values of a few times 10^{-4} cm, and for the current-bearing radius, values of a few times 10^{-6} cm. For contacts A and B, the latter values are in direct disagreement with the temperature dependence of the critical current, which requires that the current-bearing radius be larger than the penetration depth. We shall therefore omit discussions of the contact area almost entirely and see what conclusions can be obtained just from the temperature dependences and general behavior of the contacts.

TABLE I. Data on contacts A, B, and 1–9.

Contact	Copper thickness A	Normal resistance milliohms	Critical current in ma I_1 2.65°	I_2 3.08°	I_3 3.22°	I_4 3.44°K	I_1/I_2 exp.	theor.	I_1/I_3 exp.	theor.	I_1/I_4 exp.	theor.	T_c °K	P 10^3 kg/cm²
A	0+0	148	4.8	2.8	2.0	0.65	1.71	1.75[a]	2.40	2.45[a]	7.38	7.0[a]	3.55	3.6
B	0+0	1050	0.19	0.12	0.088	0.017	1.58	1.92[a]	2.16	2.71[a]	11.2	15[a]	3.51	4.4
1	0+100	148	0.90	0.25	···	0.030	3.60	2.50[b]	···	···	30	(30)[c]	3.515	4.4
2	0+100	5.1	40	14	···	0.23	2.85	2.65[b]	···	···	174	(174)[c]	3.464	5.5
3	0+700	1.68	15	3.0	1.6	0.18	5.00	2.55[b]	9.38	4.7[b]	83	(83)[c]	3.477	5.2
4	0+1000	4.0	2.5	1.2	0.35	···	2.10	2.78[b]	7.12	(7.1)[c]	···	···	3.44	5.9
5	500+500	80	0.28	0.090	···	0.005	3.11	2.53[b]	···	···	56	(56)[c]	3.489	4.9
6	0+2000	18.5	1.40	0.60	···	0.12	2.34	2.11[b]	···	···	11.2	(11.2)[c]	3.596	2.7
7	1000+1000	1.44	0.35	0.043	···	···	8.14	4.35[b]	···	···	···	···	3.30	···
8	2500+2500	0.90	···	···	···	···	···	···	···	···	···	···	···	···
9	0+5000	2.7	0.23	0.11	···	0.001	2.10	2.65[b]	···	···	230	(230)[c]	3.458	5.6

[a] From Eq. (1). T_c fixed for best over-all fit.
[b] From Eq. (5). T_c fixed to match I_1/I_4 or I_1/I_3, respectively.
[c] Matched values.

[8] Nils L. Muench, Phys. Rev. **99**, 1814 (1955), see Table I.
[9] R. Holm and W. Meissner, Z. Physik **74**, 736 (1932).
[10] D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, 1952), pp. 10 and 64.

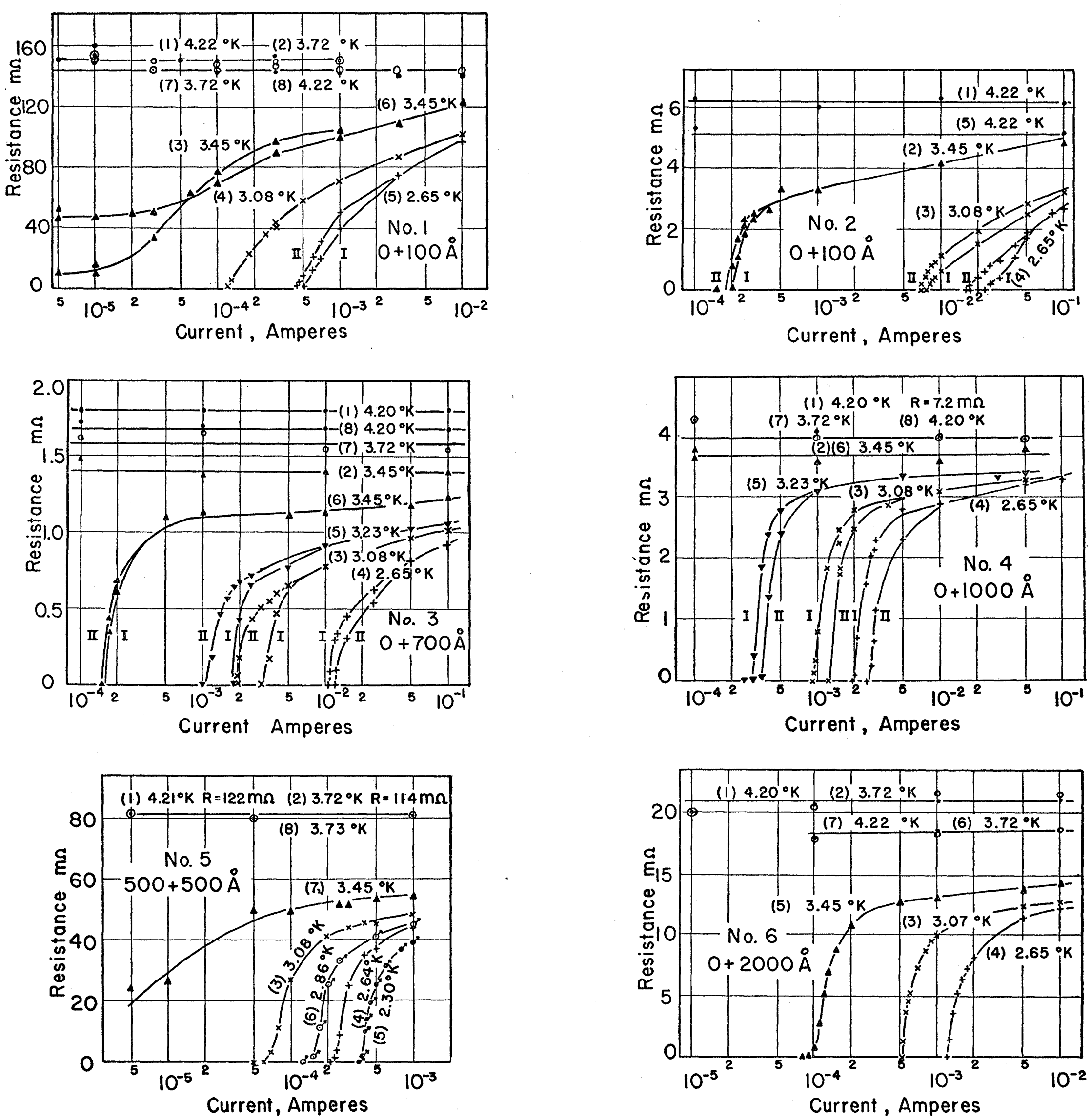

FIG. 3. Plots of resistance *vs* current for the contacts between copper plated tin wires Nos. 1–6. The numbers on the curves give the sequence and the temperature of the measurements. The notation 0+100 A refers to the thicknesses of the copper plating on the two contact members. Roman numerals refer to the current direction, I being from plated to unplated wire.

V. MEASUREMENTS ON CONTACTS BETWEEN COPPER PLATED WIRES

Figures 3 and 4 show plots of resistance *vs* current for contacts Nos. 1 to 12. All necessary explanations can be found in the captions of the figures. Most of the contacts were quite stable, although Nos. 4, 5, and 10 did show an initial shift, and Nos. 8 and 12 shifted after several curves had been measured.

All diagrams look somewhat similar, if properly scaled, though the current and resistance ranges vary widely.

There are additional measurements on two contacts which did not become superconducting even at 1.45°K. Contact No. 13 was plated with 5000+5000 A (i.e., 5000 A on each of the wires) and had at 4.2°K a resistance of 2.4 milliohms. At 1.45°K the resistance had dropped to about 1.74 milliohms at high currents, and to somewhat less at the lowest current of 50 μa. As the temperature was returned to 4.2°K, the resistance increased to 2 milliohms.

Contact No. 14 (10 000+10 000 A) initially had a resistance of 18 milliohms which, after the first measurements, dropped to 10 milliohms and stayed there even at 1.45°K and 20 μa. When the temperature was increased to 4.2°K the resistance went up to 10.7 milliohms.

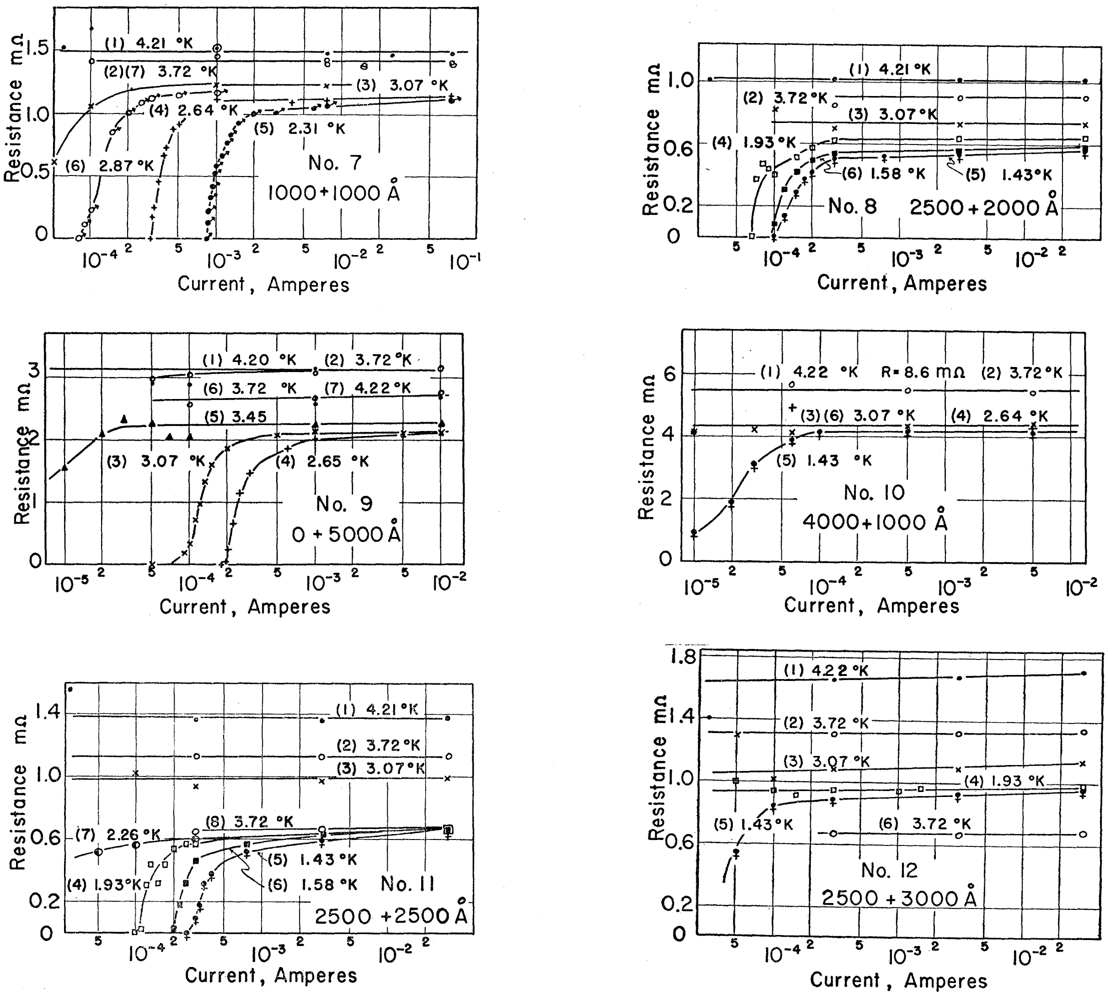

FIG. 4. Plots of resistance *vs* current for the contacts between copper plated tin wires Nos. 7–12. The numbers on the curves give the sequence and the temperature of the measurements. The notation 0+5000 A refers to the thicknesses of the copper plating on the two contact members.

It is believed that contact No. 13 (5000+5000 A) would probably become superconducting at very low temperatures and currents, while contact No. 14 (10 000+10 000 A) would not become superconducting under any conditions.

Diagrams of state for contacts 1 through 9 are shown in Fig. 5. These diagrams are, if properly scaled, all very similar. Note, however, that the temperature scale on contact No. 8 has been shifted.

Taking again the curve $R=0.25R_n$ as the "critical current" curve, one observes that all critical temperatures are at least depressed to about 3.5°K. In contacts Nos. 7 and 8, which have heavy copper plating on both contact members, the critical temperature is still lower.

The temperature dependence of the critical current curves is different from that of the contacts between bare wires. This can be understood if one assumes that the penetration depth δ is large compared to the radius a of the contact area.

The critical field is in this case not equal to the bulk critical field, but is given by[11]

$$H_{cI}=H_{c\,\mathrm{bulk}}[-J_1(ia/\delta)]/J_0(ia/\delta), \tag{2}$$

where J_0 and J_1 are the Bessel functions of zeroth and first order.

If $a\ll\delta$, Eq. (2) reduces to

$$H_{cI}=H_{c\,\mathrm{bulk}}a/\delta, \tag{3}$$

which with the temperature dependence of H_c [Eq. (1)] and the temperature dependence of δ (see reference 10, p. 143),

$$\delta=\delta_0[1-(T/T_c)^4]^{-\frac{1}{2}}, \tag{4}$$

gives for the temperature dependence of the critical current

$$I_c/I_{c0}=H_{cI}/H_{cI0}=[1-(T/T_c)^2][1-(T/T_c)^4]^{\frac{1}{2}}. \tag{5}$$

This gives indeed a critical current curve which has

[11] M. v. Laue, *Theory of Superconductivity* (Academic Press, Inc., New York, 1952), p. 115.

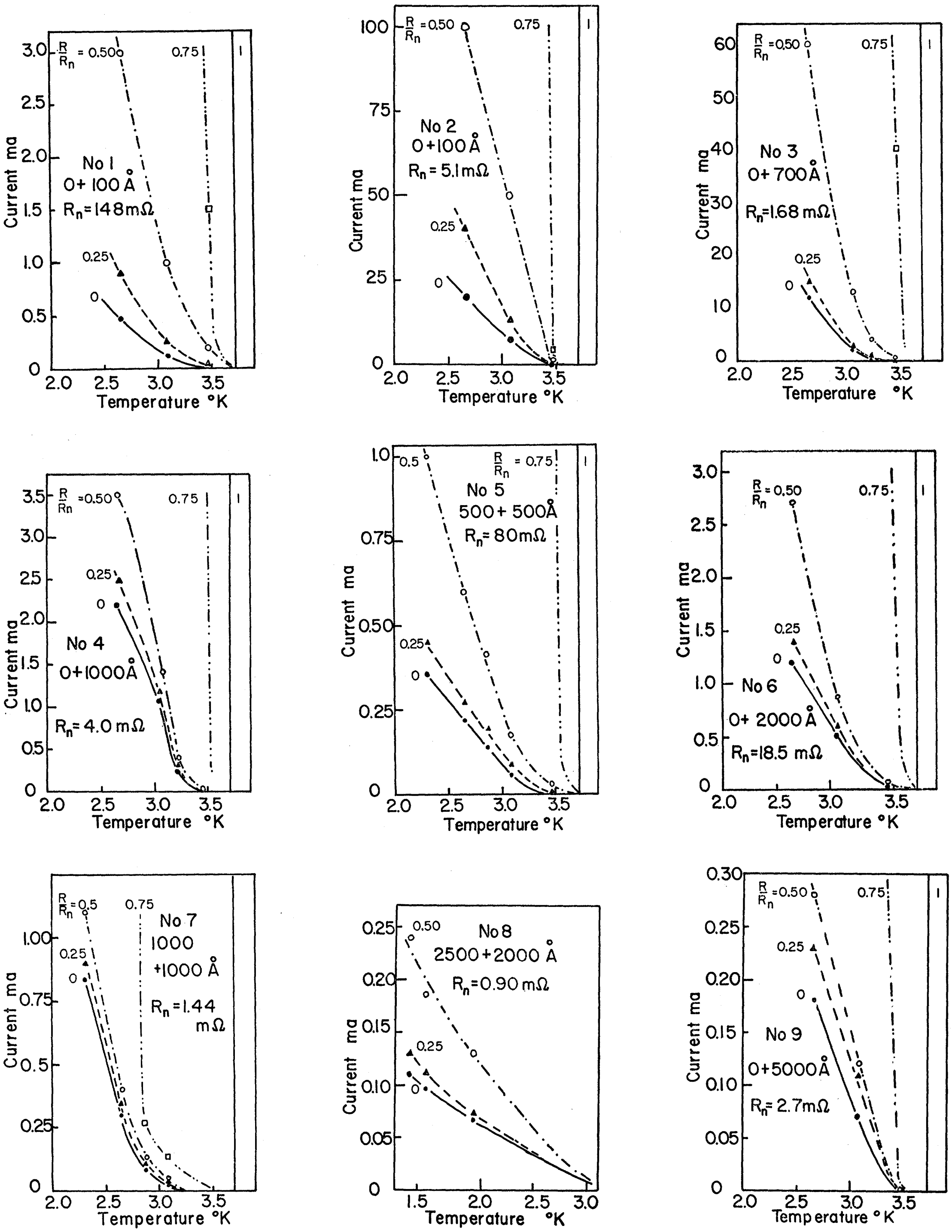

FIG. 5. Diagrams of state for the contacts between copper plated tin wires Nos. 1–9. The curve $R=0.25R_n$ is by definition the critical field curve. Note the shift in the temperature scale of contact No. 8.

zero slope at the critical temperature, in qualitative agreement with the diagrams of Fig. 5.

The quantitative agreement is checked in Table I, by calculating the ratios of the critical currents at two temperatures and comparing this value with the expected value of Eq. (5). T_c is hereby chosen to match the larger of the ratios. In this comparison one should, however, keep in mind that the reduction of Eq. (2) to Eq. (3) may not be justified for the contacts with thin copper layers.

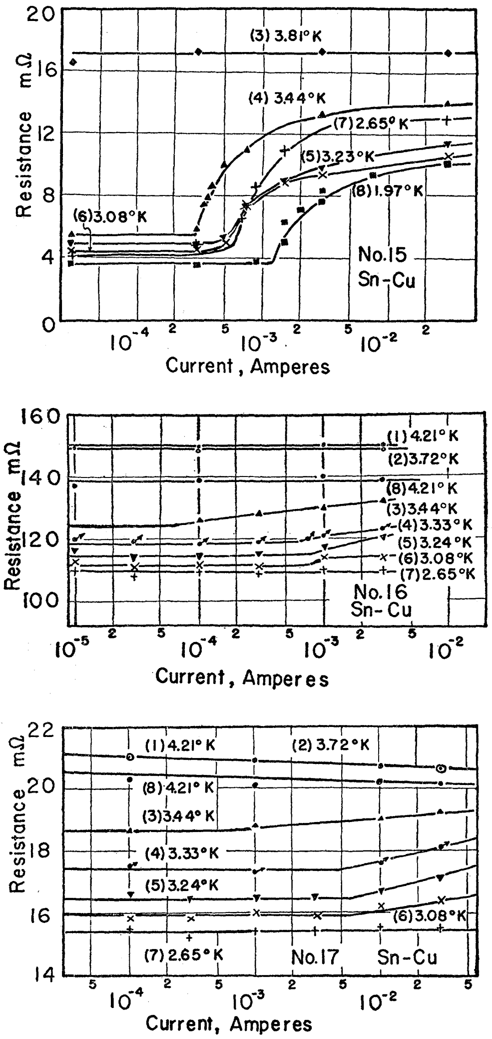

FIG. 6. Plots of resistance *vs* current for the contacts between tin and copper wires Nos. 15–17. Contact No. 15 shifted after curve 2 and after curve 8.

VI. MEASUREMENTS ON CONTACTS BETWEEN TIN AND COPPER WIRES

Figure 6 shows three diagrams where the resistance is plotted as a function of the current for the three tin-copper contacts Nos. 15, 16, and 17. They represent the limiting case where the copper plating on one side is infinitely thick.

The resistance at low currents and temperatures is reduced through the influence of the superconductivity of the tin. It rises at a "quasi-critical current" from this low-current plateau. This quasi-critical current is relatively sharply defined for contact No. 15, and hardly defined at all for contact No. 17. It should be noted that contact 15 originally had a resistance of 384 milliohms which dropped to 17.2 milliohms after curve 2, and dropped to 9 milliohms when the temperature was raised to 4.2°K after curve 8.

Figure 7 shows diagrams of state for contacts Nos. 15 and 16 where the temperature dependence of the quasi-critical current can be seen. The value of the resistance at low currents seems to be slightly temperature-dependent, but only for contact 17 is this dependence strong enough to be clearly larger than small incidental shifts of the resistance. It is plotted in Fig. 8.

VII. MEASUREMENTS ON TIN-INDIUM CONTACTS

Figure 9 shows plots of the resistance *vs* current for the three tin-indium contacts *a*, *b*, and *c*. Contacts *b* and *c* have diagrams of the type which one would expect for contacts between two superconductors with different critical temperatures. Contact *a*, however, behaves as if one contact member had a transition temperature above 4.2°K. This is in agreement with the behavior of the contacts Nos. 12, 13, and 14 of reference 3. All contacts did show some rectification which, however, is indicated for contact *b* only, where it was largest. Superconductivity persists to higher currents if the direction of the (conventional) current is from the tin to the indium.

Diagrams of state of contacts *a*, *b*, and *c* are shown in Fig. 10.

Figure 11 shows the value of the resistance at low currents of contact *a* as a function of the temperature. The only reasonable curve which can be fitted through the points has a sharp break at the critical temperature of tin. The fact that the curve is still noticeably rising at 4.2°K indicates that the effective critical temperature of this combination must be still much higher, probably around 5–6°K.

VIII. DISCUSSION

The most striking fact which follows from the different temperature dependence of the critical current of plated and unplated wires is the change in penetration depth as compared to the diameter of the contact area. It is extremely improbable that the tin formed superconducting bridges through the copper layers with such regularity, especially since some of the copper layers were quite thick.

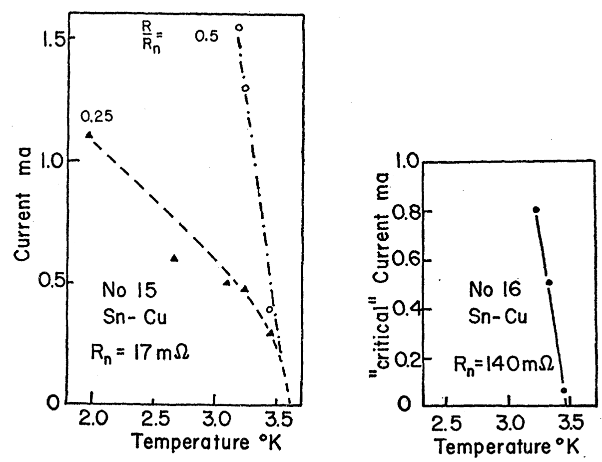

FIG. 7. Diagram of state of contact No. 15 and temperature dependence of "quasi-critical current" of contact No. 16.

It is much more probable, especially in the light of the concept of long-range order (see reference 10, p. 207), that the density of the superconducting electrons does not go abruptly to zero at the boundary of the copper, but gradually decreases with the distance from the tin.

The sketch in Fig. 12 shows the channeling of the current, the range of order extending a distance ξ from the tin and the copper plating on one or both sides. One can see that the long-range order is more effective if only one side is plated, because the point of narrowest constriction of the current is close to the tin. This explains the difference between contacts Nos. 6 and 7, and between contacts Nos. 8 and 9. It also shows that one would expect that the range of order is of about 2000 A $=2\times10^{-5}$ cm, which compares well with a value of 10^{-4} cm for pure tin.

If the density of the superconducting electrons decreases, the penetration depth increases, since both are connected by an equation of the type (see reference 10, p. 181)

$$\delta=(mc^2/4\pi ne^2)^{\frac{1}{2}}, \tag{6}$$

where m and e are the mass and the charge of the electrons of number density n, and c is the velocity of light, all in cgs units.

At sufficiently large copper thicknesses, n will decrease enough so that δ becomes larger than the contact radius, thus giving rise to the different temperature dependence of the critical current.

Along with this change in the temperature dependence goes a decrease of the value of the critical current, since, for $\delta\gg a$,

$$I_c=2\pi aH_{cI}=2\pi aH_{c\,\mathrm{bulk}}\delta/a, \tag{7}$$

which makes the critical current independent of the contact area. The series of the contacts 2, 3, 4, 6, and 9 shows indeed a behavior of this sort, as can be seen from the values of I_1 in Table I. The contact series 5, 7, and 8 also show a behavior of approximately this type. Exceptions are contacts A, B, and 1, which are also marked by their higher resistances. It seems that there are also other reasons for a decrease in the critical current; for instance, multiple contacts can give depressions of the critical current similar to those resulting from the filament structure of impure bulk superconductors (see reference 10, pp. 37–47).

It should be noted that in contrast to the critical current, there is certainly no depression in the critical

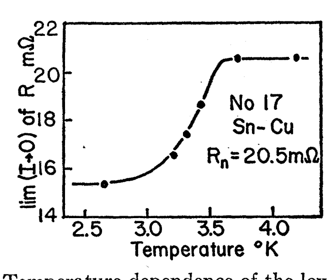

FIG. 8. Temperature dependence of the low-current resistance of contact No. 17.

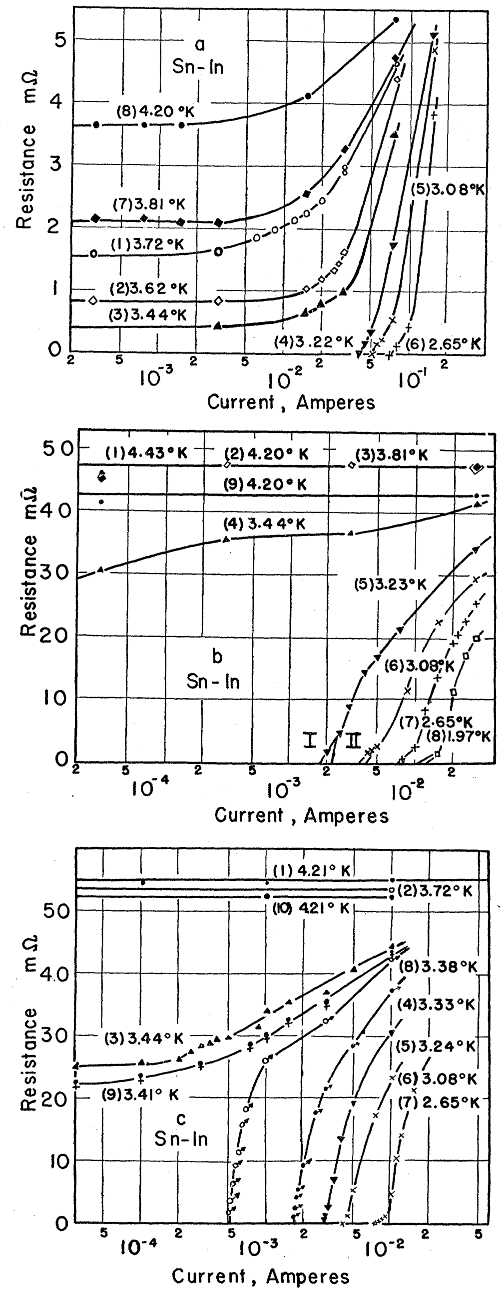

FIG. 9. Plots of resistance *vs* current for the contacts between tin and indium wires designated as a, b, and c. Slight rectification was observed in all cases, but is indicated only on contact b. The current flows in direction I from the indium to the tin.

temperature beyond the pressure effect up to quite large copper thicknesses. Only the contacts 7 and 8 with 1000+1000 and 2500+2000 A plating show a clear further decrease.

The results on the tin-indium contact a indicate that the combination of tin and indium has a higher critical temperature than either of the metals. A search of the literature revealed[12] that the In-Sn system has two

[12] S. Valentiner, Z. Metallkunde 32, 31 (1940).

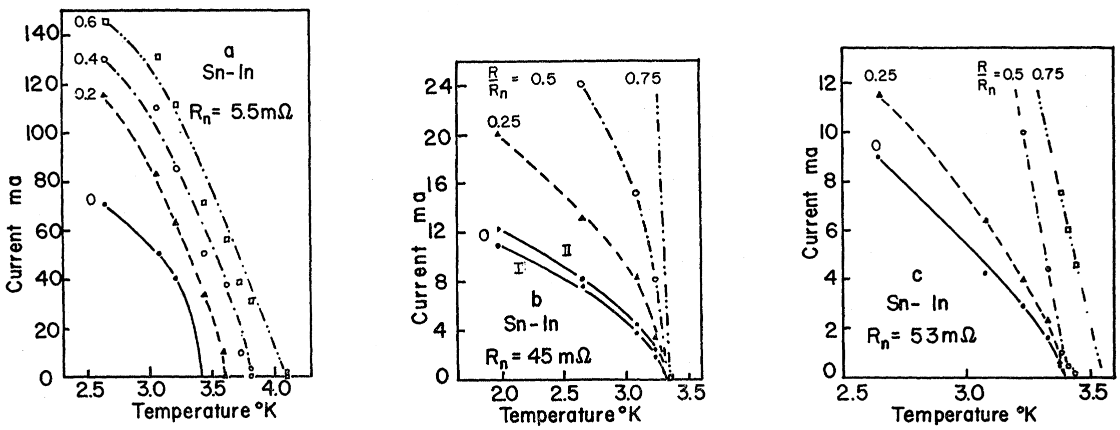

FIG. 10. Diagrams of state for the tin-indium contacts *a*, *b*, and *c*.

intermetallic compounds of the composition In_6Sn_2 and In_1Sn_{15}.

A sample of the composition In_6Sn_2 has been prepared by melting the correct amounts, quickly cooling the mixture and extruding the pellets in an extrusion press to wire of 1-mm diameter. It was found that this wire was still superconducting at 4.2°K and 2.8 amp, the highest current which could be reached with the simple arrangement used. The sample was then mounted together with a carbon thermometer and lowered into the neck of a standard 15-liter storage tank with liquid helium. By appropriately positioning it in the temperature gradient and simultaneously measuring the resistance and temperature, it was found that the critical temperature of this sample was about 5.5°K. Its resistance at 14°K was about 40% of its resistance at room temperature.

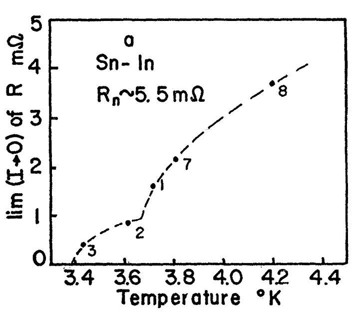

FIG. 11. Temperature dependence of the low-current resistance of the tin-indium contact *a*. The numbers at the points refer to the sequence of the measurements.

IX. CONCLUSIONS

The following conclusions can be drawn from this investigation:

(1) The pressure on the contacts shifts the critical temperature in agreement with the pressure dependence of the critical temperature found for bulk superconductors.

(2) Copper plating reduces the density of the superconducting electrons at the current constriction, thereby increasing the penetration depth and decreasing the critical currents.

(3) Copper plating gives a sizable reduction of the critical temperature beyond the pressure shift only at copper thicknesses of at least 1000 A on each contact member.

(4) If two metals have a combination which is superconducting, a contact formed of these two metals can show a sizable reduction in resistance long before the metals themselves become superconducting.

(5) It is believed that for copper plating on tin, the range of order is about 2×10^{-5} cm. It may be considerably smaller for other combinations, especially since there is some evidence (see reference 3, contact 5) that a combination of copper and tin leads to results similar to those obtained for the combination Sn-In.

(6) The observations on the contacts give strong support to the following statements: the conductivity is not a point function, but depends on the vicinity of the point in consideration. It may have values between the normal conducting values and infinite, i.e., superconducting values. This point of view is in accordance with Pippard.[13]

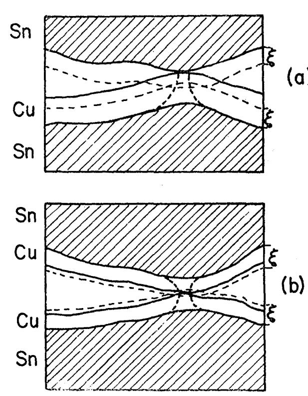

FIG. 12. Schematic diagram of contacts with symmetric and asymmetric copper plating. Note the current constriction indicated and the range of order ξ extending from the tin.

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[13] A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953).