some rather more subtle or refined and complicated techniques than have thus far been employed.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the help in discussion, and otherwise, received from a number of persons. J. R. Nelson was largely responsible for the monochromator and for much of its maintenance as well as initiating some of the early measurements. Stimulating discussions with L. G. Parratt and A. W. Overhauser of this Laboratory and with L. Apker, E. Taft, and H. Phillip of the General Electric Research Laboratory have been valuable. Thanks are also due these people of the General Electric Laboratory for the short loan of an electrometer and some samples of salts on which they also had made measurements.

PHYSICAL REVIEW VOLUME 113, NUMBER 6 MARCH 15, 1959

"Thin-Film" Experiment with Bulk Superconductors*†

HANS MEISSNER AND RICHARD ZDANIS
The Johns Hopkins University, Baltimore, Maryland
(Received October 9, 1958)

Hollow cylindrical samples of indium have been prepared by an extrusion process and equipped with both a center wire and a concentric tube. The critical current has been determined for the case in which the current passes through the sample and returns through the concentric tube. It has been found that this critical current is identical with that of solid samples. The same determination has been made for the case in which the current returns through the center wire. In this case the critical current is about 80 to 90% of that for solid samples. A field was produced by the center wire, the respective current returning through the concentric tube. The resistance of the sample was measured with a small measuring current, also returned through the concentric tube, as a function of the field produced by the center wire. The critical field thus determined depends on the value of the measuring current. These findings can be interpreted in the following way: If the magnetic field of the current through the center wire is dominating, the current in the sample will flow in a very thin layer at the outer surface of the sample. This constitutes a "thin film experiment" which does not require a thin metal film. The resistance measurements have been supplemented by measurements of the circular flux on a sample of larger size.

I. INTRODUCTION

IN the course of the investigation of the paramagnetic effect in superconductors,[1–7] it became apparent (see reference 7) that a number of differences between the present theory (see references 1 and 2) and the experimental results could be resolved if the mean value of the magnetic field between the superconducting domains would be different from the bulk critical field. A calculation of this field in the light of any of the existing theories of superconductivity[8–12] requires a knowledge of the size of the superconducting domains. The principles governing the size of the domains are unfortunately not yet known. Good fortune[13] brought to the attention of the authors an arrangement which seems to be of interest in this connection. The arrangement consists of a sample in the shape of a hollow cylinder, provided with a wire through the center and a concentric tube (see Fig. 1). The current can be passed through the sample and returned either through the center wire (this connection is denoted as "wire") or

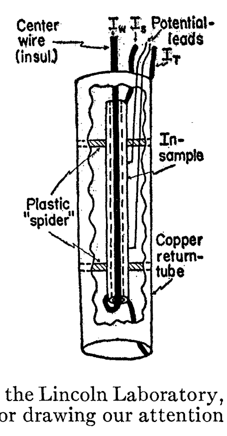

FIG. 1. Hollow cylindrical sample for resistance measurements.

* Supported by a grant of the National Science Foundation.
† A preliminary account was given in the *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, 1957*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), Paper 9-5.
1 Hans Meissner, Phys. Rev. **97**, 1627 (1955).
2 Hans Meissner, Phys. Rev. **101**, 31 (1956).
3 Hans Meissner, Phys. Rev. **101**, 1660 (1956).
4 A. H. Fitch and Hans Meissner, Phys. Rev. **106**, 733 (1957).
5 Hans Meissner, Phys. Rev. **109**, 668 (1958).
6 Hans Meissner and Richard Zdanis, Phys. Rev. **109**, 681 (1958).
7 Hans Meissner, Phys. Rev. **109**, 1479 (1958).
8 F. London, *Superfluids* (John Wiley and Sons, Inc., New York, 1950), Vol. 1.
9 M. v. Laue, *Theory of Superconductivity* (Academic Press, Inc., New York, 1952).
10 V. L. Ginsburg, *Abhandlungen aus der Sowjetphysik* (Verlag Kultur and Fortschritt, Berlin, 1951), Vol. II, p. 135, also Fortschr. Physik **1**, 169 and 333 (1950).
11 A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953).
12 Bardeen, Cooper, and Schrieffer, Phys. Rev. **108**, 1175 (1957).

13 We are indebted to Dr. D. A. Buck of the Lincoln Laboratory, Massachusetts Institute of Technology, for drawing our attention to arrangements of this type.

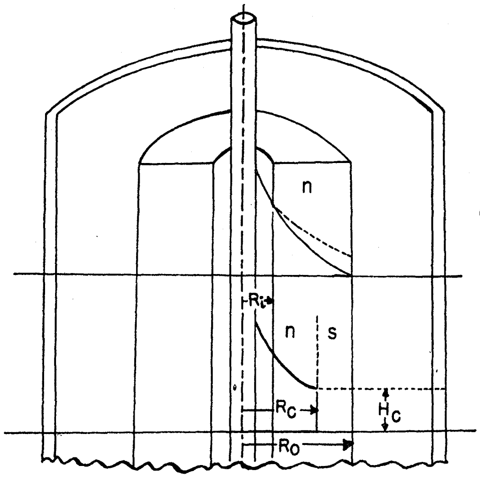

FIG. 2. Field distribution for "wire" connection (a) in the normal conducting state, (b) in the partially superconducting state.

through the concentric tube (denoted as "tube"). The center wire can also be used to provide a magnetic field, the current returning through the concentric tube, while a small measuring current is passed through the sample and also returned through the concentric tube (this connection is denoted as "field").

In "tube" connections the conditions are similar to those of the usual observations of current transitions in wires (see references 3, 5, and 6). The magnetic field is a maximum at the outer surface, and the sample enters the mixed state when the field at the outer surface exceeds the critical value.

In "wire" connections the magnetic field at the outer surface (or at least its mean value) is always zero. If the magnetic field exceeds the critical value at the inner surface a normal conducting core is formed, surrounded by a superconducting sheath (see Fig. 2). As the current is increased further the boundary between the core and the sheath gradually moves outward until it finally comes close to the outer surface. During this stage the current through the sample always flows resistanceless on the inside of the superconducting sheath, shielding it from the magnetic field.

If the current is increased such that the boundary reaches the outer surface, the current transport ceases to be resistanceless. For still larger currents, part of the current has to go through the inner, normal conducting core, reducing the magnetic field produced by the center wire and maintaining its value at the current layer approximately equal to the critical field.

In "field" connections (the measuring current kept in opposition to the current in the center wire), zero resistance is observed until the current in the center wire produces a field of critical value at the outer surface of the sample. If this value is exceeded, resistance appears and rises rapidly with increasing current in the center wire. Normal conductivity is reached when the total field at the surface, i.e., the field produced by the center wire minus the field produced by the sample current (since the two are in opposition) exceeds the critical field.

Also of interest is the case of various intermediate conditions where the sample current is returned partially through the center wire and partially through the concentric tube. The type of transition observed in this case apparently depends on the ratio of the currents through wire and tube.

With a considerably larger sample, similar to the ones used in reference 3, it is possible to measure the circular flux. For the "tube" connection the flux measurements are identical with those of reference 3. With "wire" connection it is possible to show how the phase boundary moves outward with increasing current.

Among the cases discussed, those in which the lowest value of the magnetic field in the sample occurs at the outer surface are of special interest. It is certain in these cases that all superconducting domains must be in a thin sheath at the outer surface. This presents a much simpler problem than that of the paramagnetic effect, where the distribution of the superconducting domains has to be evaluated from the theory. If one can understand the factors governing the thickness and structure of the thin superconducting sheath, one can hope to obtain the knowledge necessary to estimate the size and shape of the superconducting domains in the paramagnetic effect and in the pure current transitions.

II. EXPERIMENTAL ARRANGEMENT

(a) Cryostat and Measuring Equipment

The cryostat and the high current connections were the same as described in reference 3. The temperature was automatically held constant to within about a millidegree, as described in reference 5. The temperature was evaluated from the vapor pressure above the liquid, no correction being made for the hydrostatic pressure head. The earth's magnetic field was compensated by a

TABLE I. Data on indium samples.

Sample No.	O.d. (mm)	I.d. (mm)	Length (mm)	$R_{0°C}$ (milliohm)	$10^4 r_0$ [i]	$R_{0\,min}$ [j] / $\langle R_0\rangle_{Av}$
XVI[a]	1.49	1[g]	46.5[h]	3.96	2.54	0.86
VII[b]	1.94	1[g]	39[h]	1.49	2.04	0.85
VIII[b,c]	1.94	1[g]	39[h]	1.49	2.04	0.90
X[b]	3.04	1[g]	55.2[h]	0.738	1.88	0.93
XIV[d]	12.7	6.4	80	0.0588	1.09	0.97
XVIII[b,e]	1.94	1[g]	54.8[h]	2.058	1.90	0.65
XIX[b,f]	1.94	1[g]	46.2[h]	1.816	2.42	0.90

[a] Extruded as 1.94-mm o.d., 1-mm i.d. and then o.d. reduced by drawing on a steel mandrel.
[b] Extruded.
[c] Same sample as VII except center wire more accurately centered.
[d] Vacuum grown on a graphite core, sample contained a few large crystals and was electropolished.
[e] Center wire in eccentric position ($R_{0\,min}=0.635$ mm).
[f] Sample has longitudinal slot 0.15 mm wide.
[g] Nominal diameter.
[h] Length between potential taps. The samples themselves were about 10 mm longer.
[i] r_0 is the residual resistance ratio.
[j] $R_{0\,min}/\langle R_0\rangle_{Av}$ is a measure for the eccentricity of the center wire. $R_{0\,min}$ is usually taken to be 0.1 mm smaller than $\langle R_0\rangle_{Av}=\frac{1}{2}$ o.d.

pair of Helmholtz coils to a value of less than 3×10^{-3} amp/cm. The measuring equipment, i.e., potentiometer, galvanometer and flux meter, was the same as described in references 3 and 5. It should be noted that the resistance was always measured by observing the deflection of the galvanometer when a known current through the sample was reversed. For very small deflections of the fluxmeter a photoelectric reading device was used which is described elsewhere.[14]

(b) Samples

The resistance measurements described in references 5 and 6 indicated that indium, unlike tin, gives good results in extruded form as well as in the form of a single crystal. This fact encouraged us to use hollow samples extruded from 99.97% pure indium of the Indium Corporation of America for the resistance measurements of this investigation. They were not electrolytically polished, since this would have been possible only for the outer surface, thus creating an artificial difference between the inner and outer surface. The discussion will show, however, that the inner surface is hardly of importance, and that an electrolytic polish, even if possible only for the outer surface, might have been desirable. The center wire in these samples was an 0.8-mm lead (Pb) wire which was insulated with vinylite lacquer. Since lead is superconducting under the conditions prevailing, no heat is developed in the center wire. The lacquer layer was made thick enough so that the center wire fits snugly into the hole of the sample. It is estimated that the axis of wire and sample usually coincided within 0.1 mm. The potential leads were attached with indium-tin solder at about 5 mm from the ends of the sample. The current leads (from lead) were soldered to the ends of the sample. Since the potential taps are at a distance of 2–3 diameters from the ends and since no longitudinal magnetic fields were used, it is assumed that the measurements on these samples are relatively free from disturbances by the ends. For a sketch of the mounted sample, see Fig. 1. The samples were mounted vertically allowing free access of the liquid helium. One sample (No. XVIII) had a center wire which was purposely placed in a very eccentric position in order to check the effects of misalignment of the center wire. Another sample (No. XIX) was provided with a longitudinal slot so that no doubly connected superconducting domains could be formed. All other data will be found in Table I. The sample No. XIV for the flux measurements was vacuum grown on a graphite core which was removed later, as with the tin samples in reference 3. Both the inside and outside of this sample were electrolytically polished by the method described in reference 6. The current and potential leads were attached as shown in Fig. 2 of reference 3. A toroidal search coil was wound around this sample with a total of 554 turns of No. 40

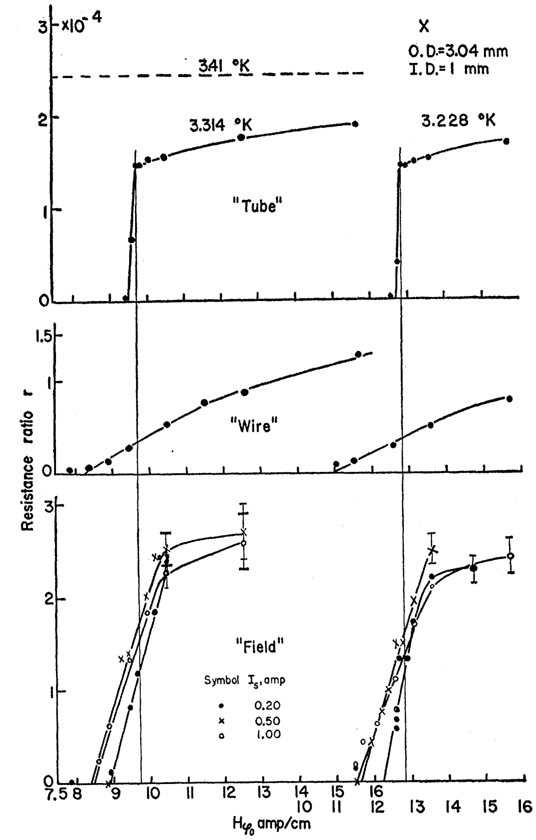

FIG. 3. Dependence of the resistance ratio r on the magnetic field $H_{\varphi 0}$ for sample No. X, for "tube" connection (top), "wire" connection (center), and "field" connection (bottom).

wire. The center wire consisted of a brass tube of $\frac{1}{16}$-in. i.d., $\frac{1}{8}$-in. o.d. which was filled with lead. The degree of filling was checked by weighing, insuring at least the absence of larger bubbles. The center wire was centered to within 0.2 mm by insulating spiders machined on a lathe. All other data can be found in Table I.

III. RESISTANCE MEASUREMENTS WITH CURRENT RETURN THROUGH CONCENTRIC TUBE

The upper part of Fig. 3 shows a plot of the resistance ratio $r=R(I,T)/R(0,0°\text{C})$ as function of the field $H_{\varphi 0}=I_T/2\pi R_0$ for the sample No. X (3.04-mm o.d., 1-mm i.d.) for two temperatures and "tube" connection. (I_T is the current returned through the tube and R_0 the outer radius of the sample.) This plot has to be compared with Fig. 4 of reference 5 or Fig. 2 of reference 6, which show similar plots for solid samples. One can see that the resistance rises abruptly if $H_{\varphi 0}$ exceeds the critical field. Figure 4 shows a plot of the critical field H_c derived from resistance measurements with "tube" connection on most of the hollow samples. It gives a critical temperature of 3.400±0.005°K and an initial slope of $(dH_c/dT)_{T_c}=-118$ amp/cm°K$=-14.85$

[14] Hans Meissner, Am. J. Phys. **25**, 639 (1957).

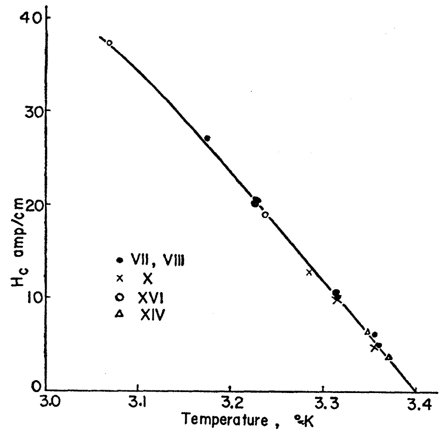

FIG. 4. Critical field curve as derived from measurements with "tube" connection.

oe/°K. The small difference between this curve and the one found for solid samples in reference 6 is probably due to the fact that the temperatures were not corrected for hydrostatic pressure head. This especially seems to affect the initial slope which depends on the accuracy of small temperature differences.

At the critical field the resistance ratio attains a value r_c. Experimentally it has been found that for solid samples r_c is always larger than $0.5r_n$, the theoretical value. (See references 5 and 6, r_n is the resistance ratio in the normal conducting state.) Table II shows the values of r_c and r_c/r_n found for the various samples. r_n has been corrected for the variation of the resistivity with temperature, but not for its variation with the magnetic field, since the latter variation was less than 1%. The table also lists the values of ρ_i, the ratio of the inner radius R_i to the outer radius R_0 of the sample and the average of r_c/r_n for each value of ρ_i. One can see that r_c/r_n clearly depends on ρ_i. This dependence can be understood in the following way: the sample is in a mixed state similar to that shown in reference 1, Fig. 5.

TABLE II. Values of r_c/r_n for hollow samples.

Sample No.	Temp (°K)	$10^4 r_c$	$10^4 r_n$	r_c/r_n	$\rho_i = R_i/R_0$	$\langle r_c/r_n\rangle_{Av}$
X	3.355	1.45	2.41	0.60	0.33	0.61
	3.314	1.46	2.39	0.61		
	3.284	1.45	2.38	0.61		
VII	3.314	1.14	2.55	0.45	0.50	0.46[b]
	3.228	1.14	2.51	0.45		
VIII	3.355	1.87	2.79	0.67[a]	0.50	
	3.314	1.12	2.77	0.41		
	3.228	1.29	2.73	0.47		
	3.174	1.42	2.70	0.53		
VIII	3.360	1.37	2.80	0.49	0.50	
	3.283	1.24	2.76	0.45		
	3.231	1.20	2.73	0.44		
XIV	3.370	0.75	1.63	0.46	0.50	
	3.348	0.73	1.62	0.45		
XVI	3.232	0.86	3.01	0.29	0.67	0.30
	3.068	0.87	2.94	0.30		

[a] Smeared transition curve, value discarded in average.
[b] Average over all measurements with $\rho_i = 0.5$.

As in the discussion of the circular flux there, we will assume that the bulk of the sample is in the same state as a solid cylinder would be at the same temperature, subject to the same current. Since we are not interested in a superimposed longitudinal magnetic field H_{z0}, we will now refer to the simpler treatment by London (reference 8, p. 120), rather than to references 1 and 2. We will further restrict ourselves to the case where the magnetic field at the surface is just equal to H_c, that is no normal conducting sheath has been formed yet.

The current density depends on the radius R, as in reference 8, p. 121, Eq. (1):

$$J_z = H_c/R. \quad (1)$$

(We use the rationalized mks system. H is the mean magnetic field averaged over the normal conducting regions.) The current through the bulk of the material is therefore

$$I_1 = \int_{R_i}^{R_0} 2\pi H_c dR. \quad (2)$$

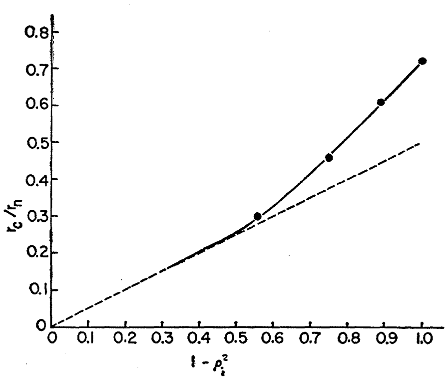

FIG. 5. Dependence of r_c/r_n on $1-\rho_i^2$. For a solid sample of comparable diameter r_c/r_n has a value of $r_c/r_n = 0.72$. Dashed curve according to Eq. (7).

In addition there is a layer current of magnitude

$$I_2 = 2\pi R_i H_c, \quad (3)$$

at the inner surface which brings the magnetic field from $H=0$ on one side to $H=H_c$ on the other side of the surface. According to our assumptions the current is just critical, which gives [similar to reference 8, p. 122, Eq. (3)] for the critical value of the mean electric field E_{zc}

$$\sigma_n E_{zc} = H_c/R_0. \quad (4)$$

Defining the critical resistance per unit length by

$$\Omega_c = E_{zc}/I_c, \quad (5)$$

where I_c is the sum of I_1 and I_2, and observing that the resistance per unit length in the normal conducting state is given by

$$\Omega_n = [\sigma_n \pi (R_0^2 - R_i^2)]^{-1}, \quad (6)$$

one finds[15]

$$r_c/r_n=\Omega_c/\Omega_n=\tfrac{1}{2}(R_0^2-R_i^2)/R_0^2=\tfrac{1}{2}(1-\rho_i^2). \quad (7)$$

Figure 5 shows a plot of r_c/r_n *vs* $1-\rho_i^2$. It can be seen that the experimental points do not follow the straight line given by Eq. (7). Complete agreement was not quite to be expected, since for a solid indium sample ($\rho_i=0$) of comparable diameter it has been found that $r_c/r_n=0.72$ rather than 0.5 (see reference 6). The experimental points indeed join this value rather smoothly.

IV. RESISTANCE MEASUREMENTS WITH CURRENT RETURN THROUGH CENTER WIRE

The middle part of Fig. 3 shows a plot of the resistance ratio r as a function of the magnetic field $H_{\varphi 0}=I_w/2\pi R_0$ which the current through the center wire

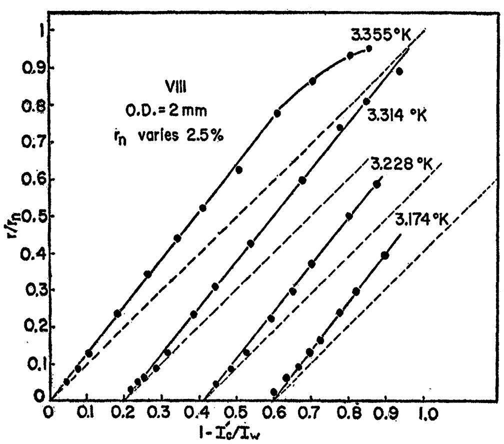

FIG. 6. Dependence of r/r_n on $1-I_c'/I_w$ for sample No. VIII in "wire" connection. Each curve has been displaced to the right by an amount of $1-I_c'/I_w=0.2$. Dashed curves have a slope of unity. The small temperature variation of r_n (about 2.5%) has been neglected.

alone would produce at a radius R_0. The value of ρ_i for this sample is $\rho_i=0.33$. The field which the center wire produces at the inner surface of the sample is accordingly $3H_{\varphi 0}$. One can see that the first appearance of the resistance occurs at values of $H_{\varphi 0}$ somewhat smaller than H_c (defined by the break in the r *vs* $H_{\varphi 0}$ curve in the "tube" connection) but not as low at $\frac{1}{3}H_c$. The latter fact is in agreement with the discussion in the introduction (see also Fig. 2).

The details of the transition follow from the requirement that for $I_w>2\pi R_0H_c$ an electric field E_z is set up sufficient to pass enough current through the normal conducting core to always keep the magnetic field at R_0 down to H_c. This electric field is apparently given by

$$2\pi R_0H_c=I_w-\int_{R_i}^{R_0}\sigma_nE_z\,2\pi RdR. \quad (8)$$

[15] A conflicting statement made in the discussion in reference 3 is wrong.

TABLE III. Constants for Eq. (10a).

Sample No.	Temp (°K)	a	I_c'/I_c	$R_{0\,\min}/\langle R_0\rangle_{Av}$
VII	3.314	0.38±0.04	0.84±0.02	0.85
	3.228	0.42±0.06	0.81±0.01	
VIII	3.360	0.16±0.30	0.75±0.08	0.90
	3.355	0.29±0.02	0.80±0.01	
	3.314	0.25±0.08	0.83±0.01	
	3.283	0.11±0.15	0.80±0.01	
	3.231	0.35±0.10	0.82±0.01	
	3.228	0.27±0.05	0.84±0.02	
	3.174	0.33±0.05	0.85±0.04	
X	3.355	0.11±0.10	0.83±0.03	0.93
	3.314	0.14±0.03	0.83±0.04	
	3.284	0.24±0.22	0.82±0.02	
XIV	3.370	0.0 ±0.63	0.97±0.10	0.97
	3.348	0.08±0.06	0.97±0.04	
XVI	3.237	0.16±0.08	0.89±0.01	0.87
	3.068	0.08±0.04	0.89±0.01	
XVIII	3.355	0.80±0.2	0.65±0.05	0.65
	3.232	0.65±0.1	0.69±0.05	
XIX	3.356	0.17±0.02	0.89±0.02	0.90
	3.232	0.05±0.02	0.92±0.02	

Observing that the normal resistance per unit length is given by Eq. (6), and defining the resistance per unit length in the intermediate state by

$$\Omega=E_z/I_w, \quad (9)$$

we find

$$r/r_n=\Omega/\Omega_n=1-I_c/I_w, \quad (10)$$

with

$$I_c=2\pi R_0H_c. \quad (11)$$

Comparison between Fig. 3 and Eq. (10) shows that the rise of resistance starts at a current $I_c'<2\pi R_0H_c$. Plotting r/r_n as a function of $1-I_c'/I_w$ gives in Fig. 6 curves with a slope larger than unity. The linear portion of the curves can be represented by an equation of the type

$$r/r_n=\Omega/\Omega_n=(1+a)(1-I_c'/I_w). \quad (10a)$$

Table III lists the values of a and the values of I_c'/I_c for all curves measured. It should be mentioned that I_c was usually determined by measuring curves with "tube" connection before and after other curves at this temperature were measured. Any error, made in the evaluation of the temperature, therefore, cannot influence the ratios I_c'/I_c. The constancy of the temperature was usually so good that I_c was reproduced to better than 1%.

If the center wire is in a slightly eccentric position, the boundary between the superconducting sheath and the normal core reaches the outer surface at a current $I=2\pi R_{0\,\min}H_c$, where $R_{0\,\min}$ is the smallest distance between the center of the center wire and the outer surface of the sample. For comparison with the values of I_c'/I_c, the estimated values of $R_{0\,\min}/\langle R_0\rangle_{Av}$ are also listed in Table III.

The outstanding result of the measurements with "wire" connection is the reduction of critical current and field from the usual values I_c and H_c to I_c' and H_c' [the latter is defined in a way similar to Eq. (11)].

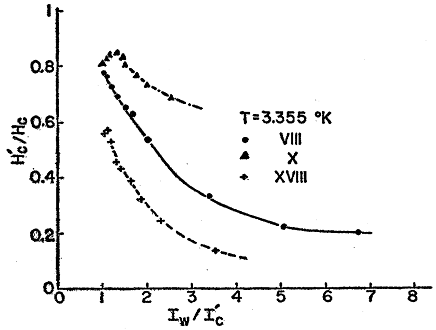

FIG. 7. Dependence of the magnetic field H_c' on the inside of the current sheath on I_w/I_c' for samples No. VIII, X, and XVIII, and "wire" connection.

Such a reduction actually is to be expected for the current transition of thin films (see reference 9, p. 115). On the other hand, one might suspect that the reduction is caused by an eccentric position of the center wire. Considering the electrodynamics only, one would come to the conclusion that an eccentric position causes an increase in the critical current since the magnetic field of an eccentric wire at the surface is lower on one side of the sample than that of a well-centered wire. This side should stay superconducting to larger currents than in the ideal case.[16] Small changes in the accuracy of the position of the center wire seemed indeed to confirm this point of view. In order to further check the effect of eccentricity, sample No. XVIII was prepared with a completely eccentric center wire (see Table I). *This sample showed a very marked further decrease in the critical current for "wire" connection* (see Table III). Moreover Table III shows that the values of I_c'/I_c are quite close to the estimated values of $R_{0\,\min}/\langle R_0\rangle_{\text{Av}}$. It seems that the first resistance appears as soon as the boundary between the superconducting sheath and the normal material reaches the surface of the sample on one side.‡ At this point the superconducting sheath ceases to be doubly connected.

Sample No. XIX was made up with a well-centered center wire and provided with a longitudinal slot 0.15 mm wide so that the superconducting sheath would never be doubly connected.[17] As Table III shows, there was no reduction in the critical current beyond that of other well-centered samples.

At the critical current I_c' the field at the inside of the current layer is about 85% of H_c. At larger currents it decreases further. This reduction can be calculated from the data. Assuming σ_n to remain unchanged, one obtains from Eqs. (6), (8), and (9)

$$H_c'=(I_w/2\pi R_0)(1-\Omega/\Omega_n). \qquad (12)$$

[16] This point of view was taken at the time of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, 1957.

‡ *Note added in proof*—See also J. W. Bremer and V. L. Newhouse, Phys. Rev. Letters 1, 282 (1958).

[17] The authors are indebted to Professor Max Dresden for suggesting this modification.

Figure 7, a plot of H_c'/H_c *vs* I_w/I_c' for the samples No. VIII, X, and XVIII, shows the reduction in H_c' at larger currents. The curve for sample No. VIII shows the largest reduction found for any of the curves of well-centered samples. Sample XVIII, however, gives still lower values. The curve for sample X is more typical, in magnitude of the change as well as the occurrence of a "bump" close to I_c'. This "bump" arises from deviations of the data from a straight line in Fig. 6. These deviations are visible in all but the 3.355°K curve for sample VIII. The values of H_c'/H_c are extremely sensitive to such deviations. Equation (10a) and Eq. (12) give for H_c'/H_c:

$$\frac{H_c'}{H_c}=(1+a)\frac{I_c'}{I_c}-a\frac{I_c'}{I_c}\frac{I_w}{I_c'}. \qquad (13)$$

Although the data seem to fit Eq. (10a) reasonably well, the values of H_c'/H_c calculated from the data show a dependence as given by Eq. (12) only in a very approximate way.

It should be noted that the curves of H_c'/H_c *vs* I_w/I_c' are rather independent of the temperature.

It is also possible to calculate the radius R_c at which the field in the sample is just critical. This radius increases as I_w is increased. For sample No. VIII and $T=3.355$°K, the radius R_c is 0.243 mm smaller than R_0 at $I_w=I_c'$, while at $I_w=6.75\ I_c'$ the radius R_c is only 0.049 mm smaller than R_0. This is not in contradiction to the values of H_c', since at large values of I_w/I_c' the derivative dH/dR at $R=R_0$ becomes very large.

As long as magnetoresistance can be neglected, there is no reason to suppose that the effective conductivity should be less than σ_n. If the core would not be completely normal conducting the effective conductivity would be larger than σ_n and more current would go through the core. This leads to still lower values of H_c'. The actual values of H_c' therefore can never be larger than those shown in Fig. 7. The layer current, of course, decreases with H_c'.

V. RESISTANCE MEASUREMENTS WITH A CIRCULAR FIELD PROVIDED BY THE CENTER WIRE

It was suspected that the reduction of the critical current for the "wire" connection was due to the current through the current sheath and that one would observe the bulk critical field if a small measuring current was used. The center wire was used to provide the magnetic field, this current returning through the concentric tube. The measuring current was passed through the sample such as to reduce the magnetic field produced by the center wire and also returned through the concentric tube. It turned out that a noticeable reduction of the critical field occurred even at measuring currents smaller than one ampere. This made these measurements very difficult, since the potential differences were frequently of the order of 10^{-8} volt. Therefore the error limits are considerably larger than those of the other measurements. The bottom part of Fig. 3 shows plots of the resistance ratio as function of the field $H_{\varphi 0}$ provided by the current in the center wire. The critical fields of these measurements are indeed in between those for the "wire" connection and those for the "tube" connection. (Only in one of the measurements with the eccentric sample, No. XVIII, has a critical field larger than that for "tube" connection been observed.) As mentioned above, I_c has been determined for each temperature before and after all these curves were measured, insuring freedom from inaccuracies in the determination of the temperature.

One might expect that the value of H_c'/H_c depends on the surface density of the layer current, that is on I_s/d, where I_s is the sample current and d the diameter of the sample. (Note that the division by π has not

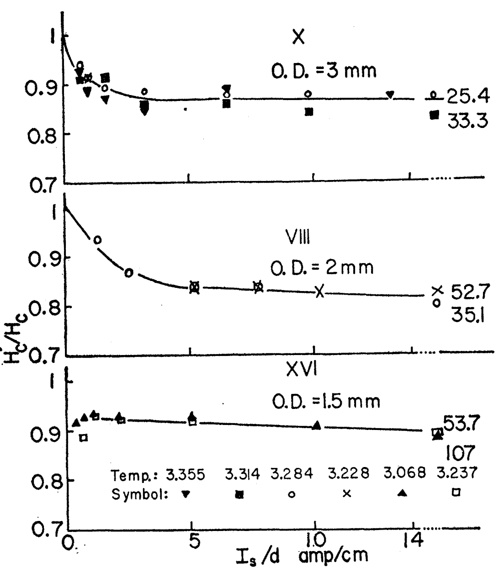

FIG. 8. Dependence of the critical field H_c' on the inside of the current sheath on the surface density of the current in the sheath. Note that the division by π has not been carried out in the values shown on the abscissa.

been carried out.) Figure 8, where H_c'/H_c is plotted *vs* I_s/d, shows that the curves thus obtained seem to be independent of the temperature but vary from sample to sample. For the samples No. X and VIII the curves seem to approach a value of $H_c'/H_c=1$ at low values of the surface density of the current; however, this is not so certain for sample XVI. It should be noted that this sample has a wall thickness of only 0.25 mm.

The last points at the right of Fig. 8 are points obtained from the "wire" connection, that is the case where the measuring current just equals the field current. The respective values of I_s/d have been marked on the figure.

Attempts have been made to fit the curves of Fig. 8 (with proper normalization) into the gap between $I_w/I_c'=1$ and $I_w/I_c'=0$ of Fig. 7. It appears that they

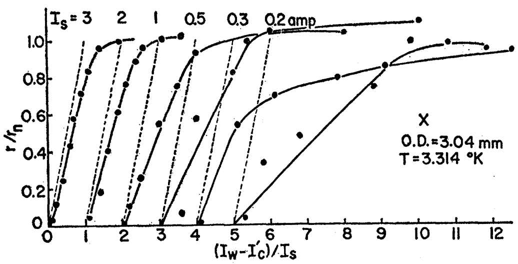

FIG. 9. Dependence of r/r_n on $(I_w-I_c')/I_s$ for various values of the measuring current I_s. The curves have been displaced by amounts of $(I_w-I_c')/I_s=1$. The dashed curves have a slope of unity.

do not join the curves of Fig. 7 smoothly. A reason for this behavior might be that, except for the point at $I_w/I_c'=1$, there is always an electric field for all points of Fig. 7 but not for the points of Fig. 8.

It is of some interest to consider the details of the transitions in "field" connection. Following the treatment in the last section, one will conclude that the resistance starts appearing if $R_c=I_w/2\pi H_c$ reaches the value $R_c=R_0$. Part of the sample current I_s will then flow through the normal conducting core, reducing the magnetic field to H_c at the inside of the current layer. The necessary electric field will be given by Eq. (8). However, the resistance per unit length is now defined by

$$\Omega=E_z/I_s. \qquad (14)$$

Using this definition, we find

$$r/r_n=\Omega/\Omega_n=(I_w-I_c)/I_s, \qquad (15)$$

which is valid for $I_c \leqslant I_w \leqslant I_c+I_s$.

Figure 9 shows a plot of r/r_n as a function of $(I_w-I_c')/I_s$ for sample No. X, where I_c' is the point of the first appearance of the resistance. Contrary to the "wire" connection (see Fig. 6) the actual slope is now smaller than that given by Eq. (15), even when I_c is replaced by I_c'. The slopes of the experimental curves decrease with decreasing measuring currents.

It seems that for this discrepancy an explanation can be found similar to that offered in reference 7 for the extension of field transitions to fields $H>H_c$. The inner core of the sample is subject to a rather strong magnetic field. Thin superconducting filaments can persist to some degree in external fields larger than H_c if they are not subject to sizable current. Increase of the measuring current (but still keeping its contribution small compared to the field of the center wire) makes the transitions sharper, more closely approaching the ideal conditions of Eq. (15).

Mathematically one can take the presence of the filaments into account by assuming an apparent conductivity σ' larger than the normal conductivity σ_n for the core. The first rather than the latter should be used in the integral of Eq. (8). This leads to

$$r/r_n=[\sigma_n/\langle\sigma'\rangle_{\text{Av}}](I_w-I_c)/I_s, \qquad (15a)$$

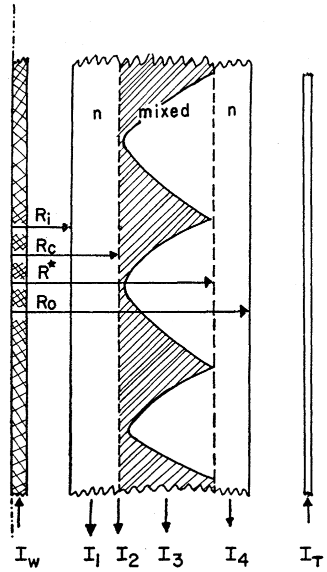

FIG. 10. Intermediate state of a hollow cylinder if part of the current is returned through the center wire and part through the concentric tube. The shaded areas give the density of the superconducting domains but not necessarily their shape.

where $\langle\sigma'\rangle_{Av}$ is a suitable average since the apparent conductivity would probably still depend on the radius.

The following picture emerges from these considerations. At the first appearance of resistance in the transitions with "field" connection, there seem to be thin superconducting filaments embedded in a normal conducting matrix. The density of these filaments increases toward the surface of the sample. It becomes questionable whether, under these conditions H_c' still can be considered as the critical field of a thin, current-carrying, superconducting film.

VI. TRANSITIONS WITH CONSTANT RATIO OF THE CURRENTS PASSED THROUGH CENTER WIRE AND CONCENTRIC TUBE

Measurements of this type have been performed only with samples No. VII and No. VIII and were later abandoned since they did not seem to contribute anything to the knowledge of the fundamentals. However, the few data which were obtained will briefly be communicated here to complete the picture.

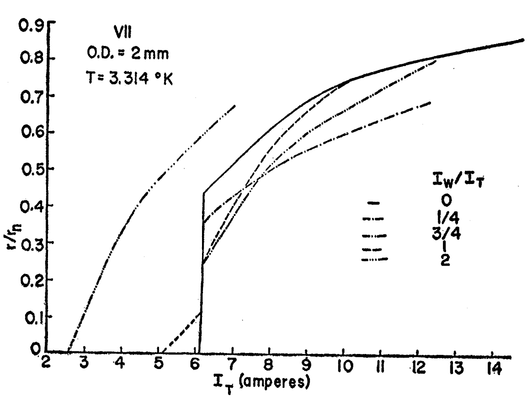

FIG. 11. Dependence of the resistance ratio r on the value of the "tube" current I_T for various ratios of I_w/I_T. Measured points were omitted for clarity.

The part I_w of the current which returns through the center wire cannot cause resistance unless it exceeds a value of $2\pi R_0 H_c$. The same also holds for the part I_T which returns through the concentric tube. A mixed state, similar to that of a solid wire subject to a current is set up only if $I_w/I_T<1$. If this is the case, the sample will have the structure shown in Fig. 10. A normal conducting core caused by the magnetic field of the center wire extends from the inner radius R_i to a radius R_c.

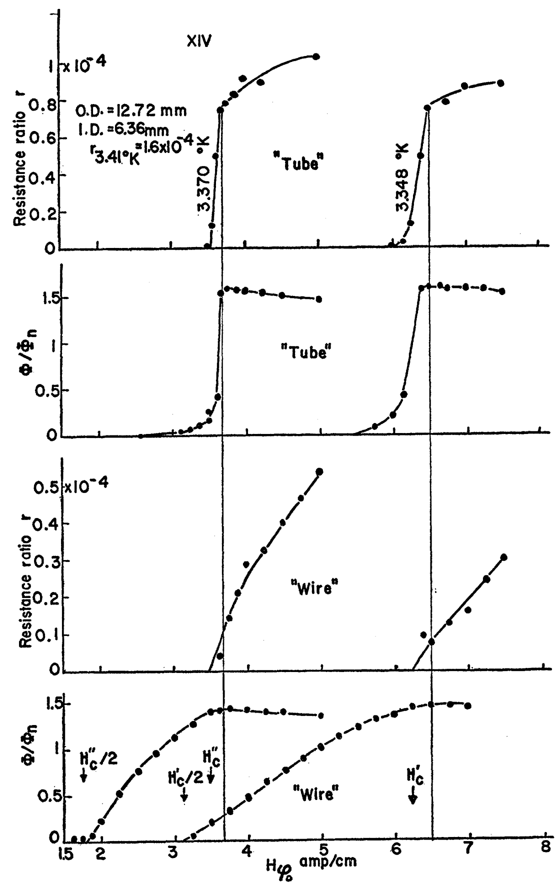

FIG. 12. Dependence of the resistance ratio r and the normalized flux Φ/Φ_n on the circular magnetic field $H_{\varphi0}$ for "tube" and for "wire" connection. Sample XIV.

From R_c to a radius R^* will be a mixed state. The mean magnetic induction at the radius R_c as well as at the radius R^* will be equal to $\mu_0 H_c$ but will be in opposite directions at the two radii. One can follow the calculation given above in Sec. III, Eqs. (1)–(7), closely, properly modifying the boundary conditions.

It is intersting to note that a total current of almost $2I_c$ can be passed through the sample without causing resistance if it is arranged that $I_w=I_T$.

The curve $I_w/I_T=1$ in Fig. 11 shows something very striking. Theoretically it should not have the "tail." As shown in Sec. IV above, the effective critical fields H_c' for the "wire" connection are smaller than those for the "tube" connection. The *effective* value of I_w/I_T of the curve $I_w/I_T=1$ is therefore *larger than* 1. This curve therefore represents in a way a simultaneous measurement of both critical fields.

VII. MEASUREMENTS OF THE CIRCULAR FLUX WITH CURRENT RETURN THROUGH CONCENTRIC TUBE

The two upper diagrams of Fig. 12 show plots of the resistance ratio r and of the normalized circular flux Φ/Φ_n (Φ_n=flux in normal conducting state at same current) as function of the circular magnetic field $H_{\varphi 0}$ for "tube" connection. Φ/Φ_n has been measured with a flux meter as described in reference 3, making corrections for the leakage flux similar to those in Eq. (7) of reference 3.

The flux curve as well as the resistance curve rises rather abruptly at a critical field, r to a value r_c and

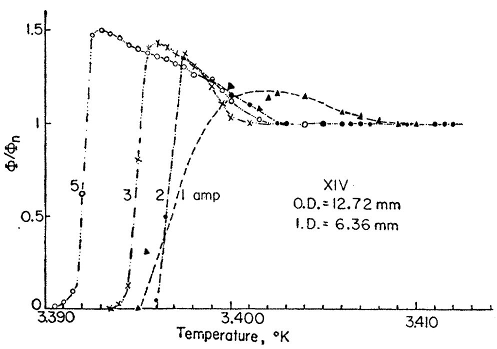

FIG. 13. Dependence of the normalized flux Φ/Φ_n on the temperature for various currents in "tube" connection. Sample XIV.

Φ/Φ_n to a maximum, which we will denote, as earlier, by $\tilde{K}_{m\varphi}$.

The measurements shown are at temperatures about 30 and 52 millidegrees below the critical temperature of indium. In order to have values comparable to those of reference 3, measurements still closer to the critical temperature were desirable. It was decided to cover this range by measuring the flux as function of the temperature for fixed current as in reference 3. These measurements are shown in Fig. 13.

The curves at 2, 3, and 5 amperes were measured by reading the fluxmeter with telescope and scale at 5 meters distance. At 1 ampere the deflection in the normal conducting state would have been only 27 mm, and in the fully superconducting state 12 mm. Therefore use was made of the photoelectric reading device mentioned above (see reference 14), which is capable of measuring deflections of 1 mm at 1 meter distance with an accuracy of about 2%. As above, the flux was measured by observing the deflection for reversal of the magnetic field. This procedure insures freedom due to

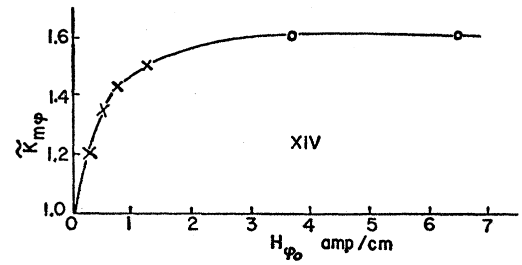

FIG. 14. Dependence of the maximum value $\tilde{K}_{m\varphi}$ of Φ/Φ_n on the value of $H_{\varphi 0}$. Sample XIV. The abscissa could also have been labeled H_c since the maximum value of Φ/Φ_n occurs at $H_{\varphi 0}=H_c$. "Tube" connection.

errors caused by "creeping" of the fluxmeter. However, occasionally the "creeping" went out of the range of the reading device, necessitating readjustment. The latter interfered with the pressure reading and gave rise to the scatter of the points on the 1-ampere curve of Fig. 13. The value of Φ/Φ_n, however, should be rather accurate.

Figure 14 shows a plot of $\tilde{K}_{m\varphi}$ *vs* $H_{\varphi 0}$ which should be compared with Figs. 5 and 6 of reference 3. As in Fig. 5 and 6 of reference 3, the value of $\tilde{K}_{m\varphi}$ drops off sharply at low values of the current, i.e., very close to T_c. The new measurements seem to indicate that $\tilde{K}_{m\varphi}$ approaches a value of $\tilde{K}_{m\varphi}=1$ at $H_{\varphi 0}=0$, i.e., at $T=T_c$. Reasons for this departure at low values of $H_{\varphi 0}$ have been discussed in reference 7.

Figure 14 shows, moreover, that the two temperatures used in the measurements of Fig. 12 are at the "plateau" and that therefore the measurements of Fig. 12 should be free from anomalies due to the closeness of T_c.

VIII. MEASUREMENTS OF THE CIRCULAR FLUX WITH CURRENT RETURN THROUGH CENTER WIRE

The two lower diagrams of Fig. 12 show plots of the resistance ratio r and of the normalized circular flux Φ/Φ_n as function of the circular magnetic field $H_{\varphi 0}$ for "wire" connection. Flux appears in the sample at about a fraction ρ_i of the value H_c of the magnetic field $H_{\varphi 0}$ at which the resistance reappears. For this sample there is very little reduction of the critical field for "wire" connection compared to that of "tube" connection. Theoretically the flux in "wire" connection should appear at a fraction, ρ_i of the critical field for "tube" connection, rather than that of "wire" connection. The measurements slightly favor the theoretical expectation but are not decisive on that point.

The details of the increase of the flux with increasing current can be understood in the following way.

The field distribution in the normal conducting sample is given by

$$H_{\varphi n}=\frac{H_{\varphi 0}}{1-\rho_i^2}\left(\frac{R_0}{R}-\frac{R}{R_0}\right). \qquad (16)$$

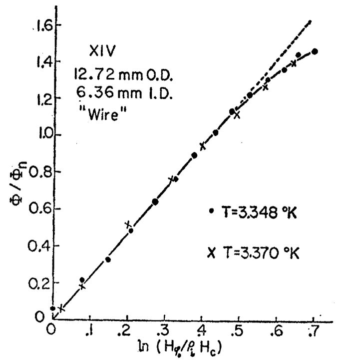

FIG. 15. Dependence of the normalized flux on the $\ln(H_{\varphi0}/\rho_i H_c)$. "Wire" connection, sample XIV. Dashed curve according to Eq. (22).

This gives for the normal flux per unit length

$$\frac{\Phi_n}{\mu_0 H_{\varphi0}}=R_0\left[\frac{1}{1-\rho_i^2}\ln\frac{1}{\rho_i}-\frac{1}{2}\right]. \tag{17}$$

In the partially superconducting state there is a normal conducting core (see Fig. 2) in which the field is given by

$$H_\varphi=H_{\varphi0}R_0/R \quad \text{with} \quad H_\varphi\geq H_c. \tag{18}$$

This core extends to a radius R_c given by

$$R_c=R_0H_{\varphi0}/H_c. \tag{19}$$

Outside of this core the material is fully superconducting and shielded from the field by a layer current at R_c.

The flux per unit length in the partially superconducting state is therefore

$$\Phi/\mu_0H_{\varphi0}=R_0\ln(R_c/R_i)=R_0\ln(H_{\varphi0}/\rho_iH_c). \tag{20}$$

Equation (20) should be valid for $H_{\varphi0}\leqslant H_c$ or at least for $H_{\varphi0}\leqslant H_c'$, where H_c' is the reduced critical field for "wire" connection. If $H_{\varphi0}$ exceeds this value the current transport is no longer resistanceless, part of the current goes through the normal conducting core and the normalized flux decreases again, approaching unity for infinitely large currents.

We find therefore for the range $R_i\leqslant R_c\leqslant R_0$

$$\frac{\Phi}{\Phi_n}=\frac{\ln(H_{\varphi0}/\rho_iH_c)}{[1/(1-\rho_i^2)]\ln(1/\rho_i)-\frac{1}{2}}. \tag{21}$$

For sample No. XIV we have $\rho_i=\frac{1}{2}$ and

$$\Phi/\Phi_n=2.36\ln(H_{\varphi0}/\tfrac{1}{2}H_c). \tag{22}$$

Figure 15 shows the normalized flux plotted as function of $\ln(H_{\varphi0}/\rho_iH_c)$. H_c has been taken from the measurements with "tube" connection. The data obey Eq. (18) very accurately for the larger part of the transition. However, they start deviating before R_c reaches a value $R_c=R_0$. From the point of the departure at $\ln(H_{\varphi0}/\rho_iH_c)=0.49$ one finds, using Eq. (19), $R_c=5.18$ mm, which is 1.18 mm smaller than $R_0=6.36$ mm. This is a very large difference. One might try to explain it with deviations from the theoretical field distributing near the ends of the sample. However, such deviations would have to be very large and really should be visible also at lower values of R_c where the agreement between theory and experiment is perfect.

IX. CONCLUSIONS

This investigation has opened a new and interesting field of superconductivity. The major features of the experiments can be explained by the theory presented above. Just as in the current transitions in wires and in the paramagnetic effect, there are small but significant differences between the measured data and the theory. Part of the differences may be due to imperfect geometry, while part of the differences are due to the structure of the superconducting layer at the surface of the sample. The major interest rests in these latter differences, since they may enable one to obtain knowledge about the structure of this layer.

While the flux measurements are somewhat questionable due to unavoidable disturbances at the ends, the resistance measurements on the thinner samples should be free from such effects, since the potential taps were well removed from the ends of the sample. Previous experience with extruded indium wires together with the sharp transitions observed for "tube" connection and the low values of the residual resistance seem to indicate a good quality of the samples. The only difficulty which arises in the interpretation of the results is the question of the effects of an eccentric position of the center wire. At first sight this seems to complicate matters very much. Nevertheless a few conclusions can be drawn: Since the slotted sample (No. XIX) does not exhibit any further decrease of the critical currents I_c' (for "wire" connection), one can conclude that it is not the change from a doubly connected superconducting sheath to a singly connected domain which causes the reduction of I_c' when the center wire is in an eccentric position.

So far the effects of surface energy have been left out of all the discussions above. If one takes the surface energy into account one will expect that the thin superconducting film will coagulate just as a thin water film on a nonwetting surface coagulates into droplets. Just as the vapor pressure of water droplets is smaller than that of a plane water surface, the critical field (H_c') of the coagulated superconducting domains will be smaller than that of the (almost) plane boundary of bulk conductors. Both of these statements are in agreement with the experimental results, although they do not receive very strong support.

One further conclusion can be drawn: So far almost all determinations of critical currents for thin films[18-23] are in disagreement (an exception is reference 20) with the theoretically expected temperature dependences [see reference 9, p. 114, Eq. (18-11); reference 10, p. 183, Eq. (4.38); and reference 22, Eq. (2)]. In the theoretical calculations it has not been taken into account that the transition might go via a domain structure. (For very thin films the transition usually proceeds almost instantaneously from complete superconductivity to complete normal conductivity; see, however, reference 23.) Such a coagulation into a domain structure (and subsequent complete breakdown of superconductivity) might completely change the criteria to be applied for the breakdown of superconductivity.

It should be noted that von Laue (see reference 9) and Ginsburg (see reference 10) apply different criteria for the breakdown of superconductivity under the influence of an externally supplied current. von Laue's treatment (as well as our simple "droplet" model above) is objectionable because the transition is treated by equilibrium thermodynamics while the thermodynamics of irreversible processes should be used. The treatment by Ginsburg seems to be free of such an objection. Both von Laue and Ginsburg use a local theory of superconductivity, while it is now certain that a nonlocal theory is necessary to properly describe superconductivity.[11,12]

[18] A. L. Shalnikov, Nature **142**, 74 (1938).
[19] A. L. Shalnikov, J. Exptl. Theoret. Phys. (U.S.S.R.) **10**, 630 (1940).
[20] W. F. Brucksch, Jr., and W. T. Ziegler, Phys. Rev. **62**, 348 (1942).
[21] N. E. Alekseevski and M. N. Mikheeva, J. Exptl. Theoret. Phys. (U.S.S.R.) **31**, 951 (1956) [Soviet Phys. JETP **4**, 810 (1956).]
[22] L. A. Feigin and A. I. Shalnikov, Doklady Akad. Nauk S.S.S.R. **108**, 823 (1956) [Soviet Phys. Doklady **1**, 375 (1957)].
[23] W. Buckel and R. Hilsch, Z. Physik **149**, 1 (1957).

X. ACKNOWLEDGMENTS

The authors are indebted to Professor G. H. Dieke for the provision of the liquid helium. They wish to thank the National Science Foundation for supporting this work by a grant. Thanks are also due Mr. R. Sarup for the help with the drawings of this report.

PHYSICAL REVIEW VOLUME 113, NUMBER 6 MARCH 15, 1959

Influence of Solutes on Self-Diffusion in the Face-Centered Cubic Lattice

HOWARD REISS
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received July 16, 1958; revised manuscript received November 13, 1958)

A theory is given for the influence of substitutional solutes on self-diffusion in the face-centered cubic lattice. The theory is limited to cases in which the concentration of solute is low enough so that only one solute atom at a time can interact with a given tracer atom.

Two different kinds of approximation are employed, one in which the processes of association and dissociation of vacancies and solute atoms do not themselves contribute to transport, and one in which they do but the frequency of exchange between solute and vacancy is considered to be infinite.

From data on the diffusion coefficient of the solute as well as on the self-diffusion coefficient in its dependence on solute concentration the ratio of the frequency with which a vacancy exchanges with a solute atom to that with which it exchanges with a host atom in the first coordination shell of a solute can be estimated. This ratio appears to lie between 0.1 and 0.5 for solutes in silver which increase self-diffusion and for which experimental data are available.

An analysis is given which shows that a good estimate of the influence of a given solute on self-diffusion can be made when only the diffusion coefficient of that solute is known.

Finally the effect which Pd in silver has on the self-diffusion coefficient (Pd reduces the self-diffusion coefficient) is calculated on the basis of the theory. Agreement between theory and experiment is satisfactory.

1. INTRODUCTION

A NUMBER of investigations dealing with the influence of solutes on self-diffusion in silver have recently been published.[1-6] Although the experimental work is very thorough no truly detailed theoretical analysis of the data has been given. The present article represents an attempt to supply such an analysis.

Our investigation will be concerned with the more or less correlated motion of three *lattice* particles:

(1) a tracer atom isotopic to the host lattice;
(2) a solute atom occupying a substitutional position in the lattice;
(3) a lattice vacancy.

It will be assumed that the diffusion of both the tracer and the solute involves a vacancy mechanism. In general the vacancy will exhibit different perferences for different sites, e.g., in the neighborhood of a solute atom or otherwise and we shall eventually treat the general situation. However, the problem is very

[1] R. E. Hoffman and D. Turnbull, J. Appl. Phys. **23**, 1409 (1952).
[2] Hoffman, Turnbull, and Hart, Acta Met. **3**, 417 (1955).
[3] Hart, Hoffman, and Turnbull, Acta Met. **5**, 74 (1957).
[4] R. E. Hoffman, Acta Met. **6**, 95 (1958).
[5] E. Sonder, Phys. Rev. **100**, 1662 (1955).
[6] Nachtrieb, Petit, and Wehrenberg, J. Chem. Phys. **26**, 106 (1957).