Discussion 52

LYNTON: I didn't quite catch the dependence on thickness that you said the Ginzburg–Landau theory predicted.

J. P. BALDWIN *Mullard Research Laboratories:* An inverse thickness.

LYNTON: According to the equation you had on the board it goes as λ_0/d, where λ_0 is the experimental penetration depth which is never implied to be a constant in the Ginzburg–Landau theory.

BALDWIN: Yes, quite. So that lambda is in fact related to the thickness.

MEISSNER: I would like to remind you that about eight years ago Maxwell and Lutes made quite similar hysteresis observations on tin whiskers. [O. S. Lutes and E. Maxwell, Phys. Rev. **97**, 1718 (1955); O. S. Lutes, *ibid.* **105**, 1451 (1957)].

Superconductivity of Thin Films of Niobium

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The object of this investigation is to determine the critical currents in niobium films as a function of temperature and magnetic field and to study the critical phenomena in detail.

The method is similar to that used by Mercereau and Crane[1] in their study of tin films. Supercurrents are induced in a cylindrical film by a primary coil and measured by a secondary coil. But whereas Mercereau's films have the shape of a short ring, our films are long cylinders surrounded by short coils. This geometry was chosen in order to avoid field concentration at the edges of the film. A screen of high-purity aluminium was necessary for the suppression of the low frequency ripple from a liquid-nitrogen-cooled solenoid which surrounds the cryostat. This solenoid can produce fields up to 15000 Oe. The primary coil is supplied with alternating current of either 18 to 100 cps or 0.05 to 0.3 cps. The voltage picked up by the secondary is integrated either electronically or in a galvanometer–photocell arrangement. This enables one to display the instantaneous flux through the secondary as the y deflection on an oscilloscope. The x deflection is connected to a current shunt in the primary circuit.

The films are produced by evaporation from an electrically heated niobium wire of 1-mm diameter which has the shape of a twisted hairpin and is positioned at the axis of a glass tube of 42-mm i.d. The deposition rate is 0.3 Å/sec and the vacuum of the order of 10^{-10} mm Hg. This corresponds to a purity of the film of 0.1 to 1 at. %. The substrate temperature during the deposition is about $-100°$ C. The film remains under vacuum throughout the measurements. The thickness of the film can be estimated from the increase of the resistance of the niobium wire and can later be determined by quantitative analysis. The normal resistance of the film is monitored by a single layer coil connected to a Q meter. This gives its first response about 20 sec after the start of the deposition indicating that the film becomes coherent when its nominal thickness is about 6 Å. This coil can also be used as an alternative to the usual coil system for measuring critical currents and temperatures on the freshly deposited film before it has been exposed to room temperature. One film was later subjected to further heat treatment.

The properties of two films of different thickness are listed in Table I. The normal resistivity is given in arbitrary units which are different for the two films. The critical currents are expressed in terms of H_{Ic}, the difference of the magnetic field which the critical current produces on the two sides of the film. The values of the critical magnetic field H_u at 4.2°K are obtained by extrapolation from a nearly linear H_u versus T plot obtained at higher temperatures.

For the discussion of the details of the critical current loops we first consider the idealized case of a coil system that is long in relation to its diameter. The radius of the film is R_F and the radius of the secondary is R_s, the field produced by the primary is H_p and the currents in the film are expressed by the magnetic field difference H_I which they produce. The flux Φ through the secondary is expressed in terms of $\phi = \Phi/\pi R_F^2$. It is plotted on the oscillograms as a function of H_p. The normal resistance of the film is so high that at the frequencies used the induced currents are unobservable. Thus in the normal state the ϕ vs H_p curve is a straight line of slope R_s^2/R_F^2. If the film is superconductive, the slope will be $(R_s^2 - R_F^2)/R_F^2$.

[1] J. E. Mercereau and L. T. Crane, Phys. Rev. Letters **9**, 381 (1962).

TABLE I. Properties of the films.

Thickness (A)	Normal resistance at 78°K	290°K	Critical temperature (°K)	H_{Ic} (Oe)	H_U (kOe)	Treatment
440	15	...	7.7	12		Freshly deposited
	10	20	8.6	11	32	Exposed to room temperature
1050	36.6	...	7.02	14		Freshly deposited
	30.6	51.4	7.65	13	30	Exposed to room temperature for 15 days.
	20.1	36.0	8.25	19	29	7 h at 250°C
	18.6	34.4	8.30	17.5	25	2½ h at 450°C
			8.30	14.5	20	6 months at room temperature; accidentally exposed to 25μ prevacuum for 4 h.

If the film current reaches its critical value H_{Ic}, there are several possibilities. In the most simple case, which is realized close to the critical temperature and also at lower temperatures in the presence of a high superimposed magnetic field, the current just stays at its critical value as long as H_p is increasing, and prevents any change in flux when H_p is decreasing. The resulting pattern is a parallelogram the sides of which are parallel to the superconducting and to the normal lines. The distance in the H_p direction of the sides with normal slope is always $2H_{Ic}$, whereas the position of the sides with superconductive slope increases with the amplitude of H_p. The vertical distance of a point on the parallelogram from the "normal" line gives the film current H_I.

The length of the coils used in the experiments is only 2.15 times the distance of the primary from the film. Consequently the film current does not become critical everywhere at the same time. This has the effect to round off the obtuse corners of the parallelogram as can be seen in Fig. 1(a).

A different behavior is observed at external fields below about 3 kOe and at temperatures below 7.7°K [Fig. 1(b)]. As soon as the film current reaches a

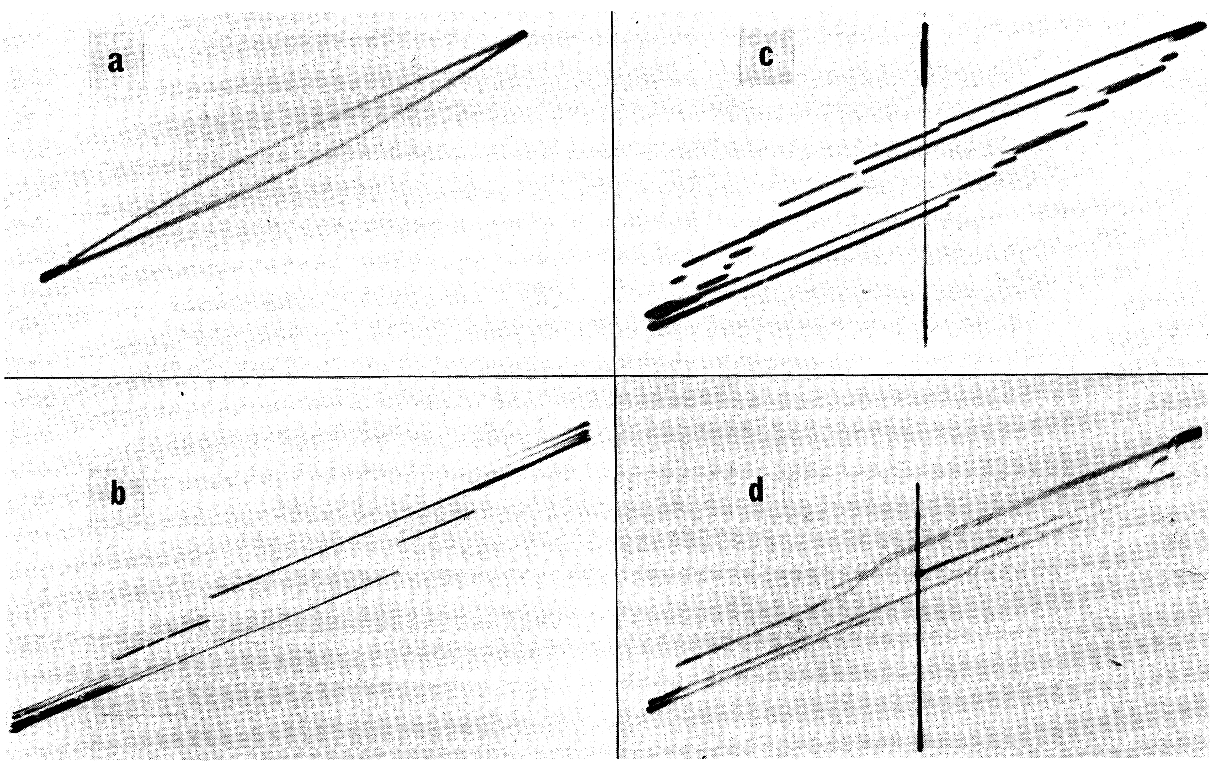

FIG. 1. Oscillograms taken at (a) T = 7.62°K, peak value of H_P = 15.2 Oe, frequency 18 cps; (b) 7.12°K, 30.4 Oe, 18 cps; (c) 4.2°K, 22.8 Oe, 25 cps, covering 1.75 cycles; (d) 4.2°K, 23.2 Oe, 0.048 cps, covering 1.97 cycles, virgin curve. Film thickness 1050 Å.

certain critical value, it suddenly drops within less than 10^{-4} sec to a lower figure. It then follows H_p with the "superconducting" slope until it has again reached a critical value, when another step follows. At zero field and between 7.7° and 6.7°K these steps are large and reproduce themselves in consecutive

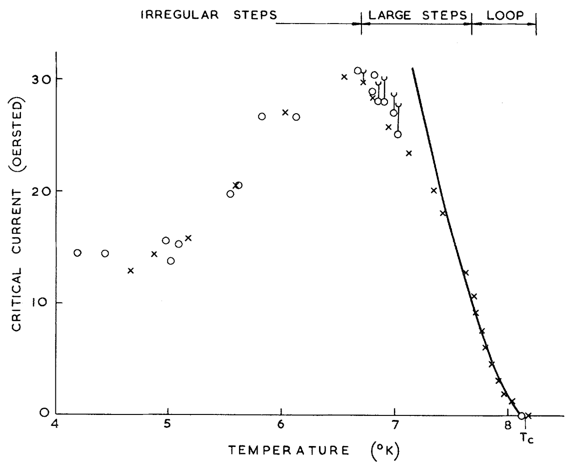

FIG. 2. Critical current as function of temperature. The curve represents $H_{Ic} = 31.1(T_c - T)^{\frac{3}{2}}$. Film thickness 1050 Å.

cycles. As the temperature is lowered there is a tendency for the pattern to become asymmetrical and to change its shape. Finally, below 6.7°K the steps become smaller and more numerous. Now the pattern alters from one cycle to the next [Figs. 1(c) and 1(d)]. In this region the onset of the critical current becomes spurious and ill defined.

The dependence of H_I on temperature is shown in Fig. 2 and on the superimposed magnetic field H_s in Fig. 3. In both cases there is an initial rise of H_I followed by a decrease to zero.

The thermodynamical critical field H_c of niobium at 4.2°K is 1560 Oe, therefore, the ratio $h_u = H_u/H_c$ of the two films is between 19 and 12. This high value cannot be due to the ordinary thin film effect[2–4] which leads to a high ratio only if the film thickness d is small compared with the penetration depth. In this case $h_u \propto d^{-\frac{3}{2}}$.[4] We must therefore assume that our films behave like superconductors of the second kind[5–7] with a high value of the parameter $\kappa = h_u/\sqrt{2}$.

[2] H. London, Proc. Roy. Soc. (London) **A152**, 650 (1935); E. T. S. Appleyard, J. R. Bristow, H. London, and A. D. Misener, *ibid.* **A175**, 540 (1939).
[3] V. L. Ginzburg, Dokl. Akad. Nauk SSSR **118**, 464 (1958) [English transl.: Soviet Phys.—Dokl. **3**, 102 (1958)].
[4] W. B. Ittner, III, Phys. Rev. **119**, 1591 (1960).
[5] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].
[6] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **36**, 1918 (1959) [English transl.: Soviet Phys.—JETP **9**, 1364 (1959)].
[7] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **37**, 1407 (1959) [English transl.: Soviet Phys.—JETP **10**, 998 (1960)].

Such a high κ can be due to impurities and strains in the film. It is decreased by heat treatment (see Table I). The behavior of the critical current density in the region above the maximum is in accordance with observations on other hard superconductors which have been treated theoretically by Gorter,[8] Kim,[9] and Anderson.[10]

However, at fields below Abrikosov's[5] lower critical field H_L, the superconductor should revert to classical behavior and carry currents up to the value $H_I = H_L$. H_L can be related to H_U by a semiempirical relation of Goodman.[11] This leads to $H_L = 240$ Oe and 375 Oe for the 440-Å and 1050-Å films, respectively, vastly in excess of the observed values of H_{Ic}.

The dependence on temperature is in close agreement with the observations on tin of Mercereau and Crane,[1] who found a maximum of similar shape, and proportionality of H_{Ic} with $(T_c - T)^{\frac{3}{2}}$ in the neighborhood of the critical temperature T_c. Our measurements near T_c can also be fitted to a $(T - T_c)^{\frac{3}{2}}$ law. This means that in this region the penetration depth becomes larger than the film thickness.[12,13]

At somewhat lower temperatures, where the H_{Ic} vs T curve becomes linear, H_{Ic} may possibly be identified with H_L as discussed above, though the observed H_{Ic} is only about $\frac{1}{4}$ of what one would expect.

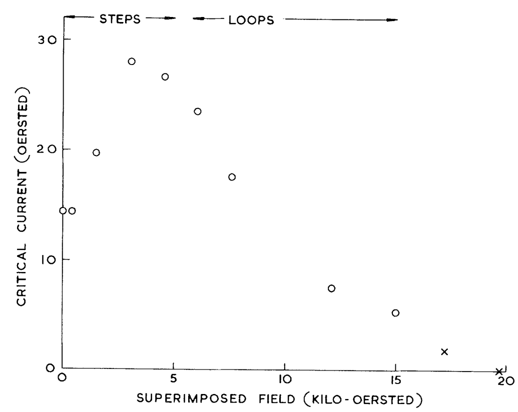

FIG. 3. Critical current as function of a superimposed magnetic field at 4.2°K. Film thickness 1050 Å.

Again the low values of H_{Ic} near and below the maximum cannot be explained. Perhaps an answer to the problem may be found on the lines of the following speculations.

[8] C. J. Gorter, Phys. Letters **2**, 26 (1962).
[9] Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. Letters **9**, 306 (1962).
[10] P. W. Anderson, Phys. Rev. Letters **9**, 309 (1962).
[11] B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962).
[12] D. H. Douglass, Jr., Phys. Rev. **124**, 735 (1961).
[13] J. E. Mercereau and T. K. Hunt, Phys. Rev. Letters **8**, 243 (1962).

In the region in question the film behaves like a classical superconductor with a penetration depth smaller than or, at best, comparable with the thickness of the film. Under these conditions a frozen-in radial magnetic field may become concentrated into certain small portions of the film and become much larger than the axial magnetic fields. Under the influence of the axial fields the radial field lines may cut through the film in an axial direction. There are some indications in the irregular hysteresis loops associated with the region under discussion [Figs. 1(c) and 1(d)] which point to a tendency of the trapped flux to escape when H_p passes through zero. However, experiments in which precautions were taken to compensate the horizontal component of the earth's magnetic field and in which the virgin branch of the hysteresis loop was studied [Fig. 1(d)] gave critical currents which were only about 30% above the values observed after the first breakdown of H_I.

We must therefore look for further causes for a concentration of the applied magnetic fields. With the geometry of short rings used by Mercereau,[1] such concentration can be expected to occur at the edges of the ring and may possibly account for his results. Perhaps in our geometry a similar field concentration is started off in pinholes or other defects we have noticed in our films.

Free Energy of Composite Wires in the Superconducting State

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PHENOMENOLOGY OF COMPOSITE STRUCTURES

Qualitative Effects

Widespread interest[1–7] has been aroused in the phenomena associated with superconductivity in composite superconducting normal metal films. The authors, in a recent note,[8] reported use of a new method (suggested by R. Garwin) to investigate superconductivity for filamentary specimens. The present work extends this technique and reports some qualitatively new observations which seem of fundamental interest.

Previous workers had already noted one primary effect, the depression of critical temperature, critical current, and critical field for a superconductor in contact with normal metal. The effect is a strong function of thickness of either element,[5] increasing with decreasing thickness of superconductor and increasing thickness of normal metal. Indeed, the superconductor has an apparent critical size, ~100 Å, below which it will not support superconductivity in contact with a normal metal. The depression in T_c is also found to be strongly dependent upon the characteristics of the normal metal,[7] being much larger for a paramagnetic element as compared to nonparamagnetic.

A second primary effect, the appearance of superconducting order in the normal metal has also been discussed in the authors' preliminary report.[8] In that report the effect was detected in Al well above its critical temperature by flux exclusion, while in an earlier article Smith *et al.*[4] report electron tunneling into Ag, demonstrating a superconducting energy gap when in contact with Pb.

Semiquantitative Theory of Composite Specimens

Although de Gennes[9] has suggested that the GL theory may not work for very thin films, it is of interest to examine this theory and, if possible, to apply

[1] E. F. Burton, J. O. Wilhelm, and A. D. Misener, Trans. Roy. Soc. Canada, Sec. III **28**, 65 (1934).
[2] A. D. Misener and J. O. Wilhelm, Trans. Roy. Soc. Canada, Sec. III **29**, 5 (1935).
[3] H. Meissner, Phys. Rev. **109**, 686 (1958), Report, Office of Naval Research, Contract NONR 248 (49), 1959; in *Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications, Ltd., London, 1962).
[4] P. S. Smith, S. Shapiro, J. Miles, and J. Nicol, Phys. Rev. Letters **6**, 686 (1961); J. L. Miles and P. S. Smith, J. Appl. Phys. **34**, 2109 (1963).
[5] P. Hilsch, Z. Physik **167**, 511 (1962). P. Hilsch, R. Hilsch, and G. V. Minnigerode, in *Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications, Ltd., London, 1962).
[6] A. C. Rose-Innes and B. Serin, Phys. Rev. Letters **7**, 278 (1961).
[7] W. Simmons and D. H. Douglas, Jr., Phys. Rev. Letters **9**, 153 (1962).
[8] D. P. Seraphim, F. M. d'Heurle, and W. R. Heller, Appl. Phys. Letters **1**, 93 (1962).
[9] P. G. de Gennes and E. Guyon (to be published).

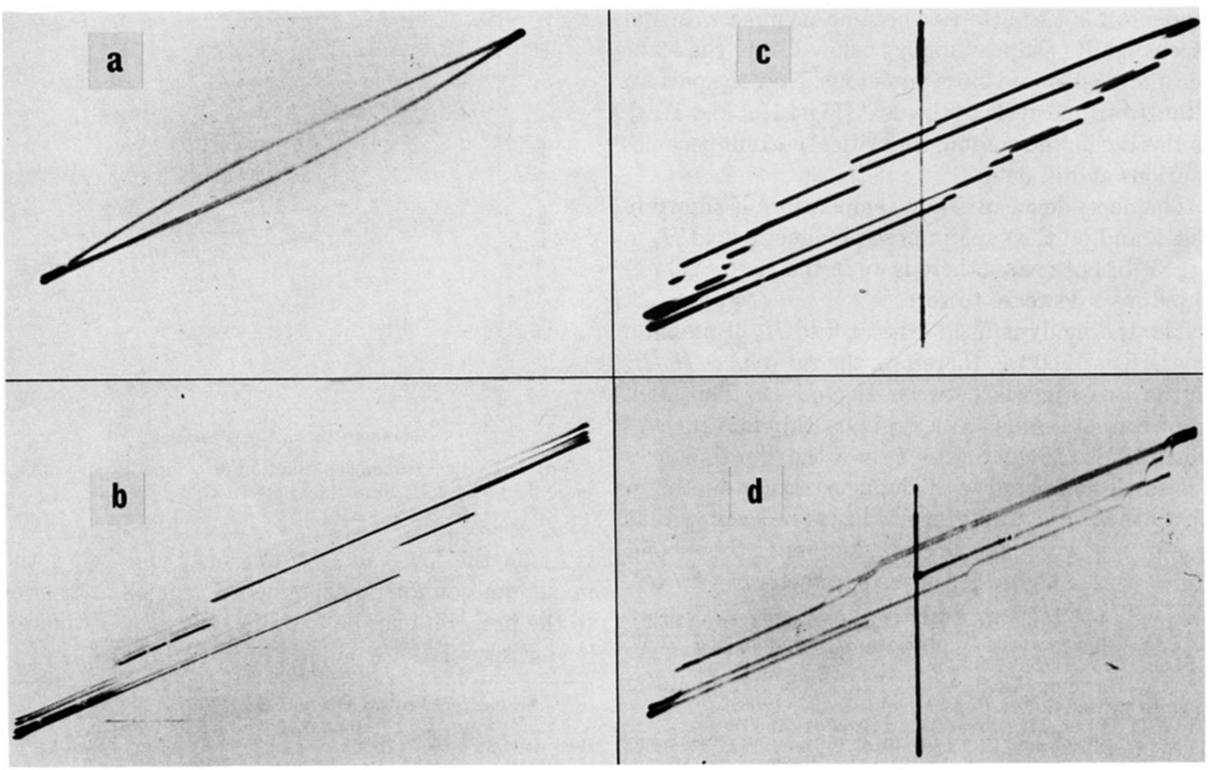

FIG. 1. Oscillograms taken at (a) $T = 7.62°K$, peak value of $H_P = 15.2$ Oe, frequency 18 cps; (b) 7.12°K, 30.4 Oe, 18 cps; (c) 4.2°K, 22.8 Oe, 25 cps, covering 1.75 cycles; (d) 4.2°K, 23.2 Oe, 0.048 cps, covering 1.97 cycles, virgin curve. Film thickness 1050 Å.