that the periodic variations have an amplitude too small for us to see. It would be extremely interesting to see whether flux is quantized in these cylinders by the methods used by Deaver and Fairbank,[4] for such magnetic experiments are not limited in the same way as ours are. Alternatively, vanadium may be a superconductor which does not have an infinite range of order.

In conclusion, I should like to mention one unexplained feature of our measurements which I think should be brought to light. In many samples a considerable amount of structure develops within the individual parabolas as the temperature is reduced so that almost the entire sample is superconducting. This was most clearly shown in a sample of indium. By taking a time exposure, the "noise" in this structure was averaged out and we obtained the picture shown in Fig. 5. Here, in addition to the strong period in $hc/2e$, there appears to be an additional period in $hc/8e$. If higher order correlations than pair correlations existed in this sample, it would show up in just this manner and this may be an indication of such correlations. A considerable amount of further investigation is needed to test the validity of this possibility and we are attempting to extend our measurements right into the pure superconducting state in the belief that here these higher correlations may be stronger.

Discussion 41

A. J. COLEMAN, *Queen's University:* Since I have been thinking about the density matrix for about ten years, Professor Little's remarks interest me very much. You remember that he stated that Yang demonstrated that if there is a large eigenvalue of the second order density matrix, this implies flux quantization. Sasaki has shown that to get a large eigenvalue of the second order density matrix the most favorable type of function is a BCS type of function. In a paper of mine which appears in the July issue of the Reviews of Modern Physics [Rev. Mod. Phys. **35**, 668 (1963)] for a BCS function I give a criterion that there shall be a large eigenvalue. This criterion which assumes only that the wave function is antisymmetric is relatively easy to satisfy. For example, it will be satisfied if the first $2n$ of the h_k, in the usual notation of the BCS theory, are of comparable size, then you can get a large eigenvalue of about $\frac{1}{2}$ n. As you remember Yang requires for long range order an eigenvalue of roughly the order of n.

LITTLE: Could I just mention that in Yang's paper, in the Appendix, he also demonstrates that the BCS state is the only way of obtaining the maximum.

COLEMAN: Not exactly the BCS type, but the BCS type where all h_k's are equal. But with such a function you would not be able to satisfy the Hamiltonian.

MENDELSSOHN: I think Professor Little's crucial experiment on Vanadium can be carried out within the next week in any of 50 laboratories because all that is required is to show that in vanadium you can't have a macroscopic persistent current for any length of time. We have to remember that whereas flux quantization as such wasn't shown until two years ago, flux quantization must exist or you cannot conceive of an ordinary persistent current unless two conditions are satisfied: a grain structure of the states—that implies quantization—and zero fluctuations. Professor Little's experiment really should mean that in vanadium quantization is either nonexistent or that the grain is very small. I imagine nobody has looked for persistent currents in V; they just shouldn't exist.

Consequences of Fluxoid Quantization in the Transitions of Superconducting Films

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I. INTRODUCTION

In this paper we start by recalling what the concept of the *fluxoid*, introduced by London,[1] means, and why the Bohr–Sommerfeld quantum condition applied to pairs requires that it be quantized in units of $hc/2e$. We shall than see how in conjunction with the Ginzburg–Landau[2] theory it leads to an interpretation of the experimental results of Little and Parks[3] on the periodic variation of T_c with flux through a thin-walled cylinder, and of the aperiodic variation

[1] F. London, *Superfluids* (J. Wiley & Sons, Inc., New York, 1960), Vol. 1.

[2] V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. **20**, 1064 (1950); a discussion in English is given by J. Bardeen, in *Handbuch der Physik*, edited by S. Flügge, (Springer-Verlag, Berlin, 1956), Vol. 15, p. 324.

[3] W. A. Little and R. D. Parks, Phys. Rev. Letters **9**, 9 (1962).

of T_c which they also found. Similar considerations lead to some new results on the transitions of thin superconducting films in a perpendicular magnetic field. These results are supported experimentally by results of various workers. The primary result is an expression for the perpendicular critical field, which seems to be in agreement with a large variety of experimental results. Also predicted is an unusual angular dependence of the critical field which has been borne out in several experiments. Finally, we shall note the close correspondence of our result for the critical field and of our model in general with the vortex model for bulk superconductors of the second kind given by Abrikosov.[4] This close connection offers assistance in obtaining a qualitative understanding of the rigorous results of his highly mathematical theory.

II. THE FLUXOID CONCEPT

The flux Φ through a closed loop can be written in the following ways:

$$\Phi = \iint \mathbf{H}\cdot d\mathbf{S} = \iint \operatorname{curl} \mathbf{A}\cdot d\mathbf{S} = \oint \mathbf{A}\cdot d\mathbf{s}\,, \quad (1)$$

using the fact that $\mathbf{H} = \operatorname{curl} \mathbf{A}$ and a theorem of vector integration. The flux*oid* of London is a closely related quantity Φ' which can be expressed as

$$\Phi' = \iint (\mathbf{H} + c \operatorname{curl} \Lambda\mathbf{J}_s)\cdot d\mathbf{S} = \oint (\mathbf{A} + c\Lambda\mathbf{J}_s)\cdot d\mathbf{s}\,, \quad (2)$$

where the London parameter Λ is defined by

$$\Lambda = m/n_s e^2 = 4\pi\lambda^2/c^2\,. \quad (3)$$

In this, λ is the superconducting penetration depth, and n_s is the "number of superconducting electrons" of mass m and charge e. Note that the fluxoid of a loop differs from the flux by a line integral of the supercurrent about the loop. Thus, if the loop is taken in the interior of a bulk superconductor where the current is zero, the fluxoid equals the flux. Recalling the London equations

$$\mathbf{H} = -c \operatorname{curl} \Lambda\mathbf{J}_s\,,$$
$$\mathbf{E} = \frac{\partial}{\partial t}(\Lambda\mathbf{J}_s)\,, \quad (4)$$

we see that for a loop enclosing purely superconducting material the integrand in (2) vanishes everywhere inside the loop, and the fluxoid would have to be zero. However, if there is a physical hole in the material about which the loop is taken, or if there is a normal spot (where the London equations, of course, do not hold), then the deduction that $\Phi' = 0$ does not hold. Rather, one can only show that the fluxoid has the same value for any loop in the superconductor enclosing the same nonsuperconducting spot and that this value will remain constant in time even if time-varying fields are applied. These properties suggest that the fluxoid might well be quantized. We now note that the Bohr–Sommerfeld quantum condition applied to the condensed superconducting pairs provides such a quantized value.

The Bohr–Sommerfeld quantum condition requires that $\oint \mathbf{p}\cdot d\mathbf{s} = nh$ around the orbit of a particle. In the presence of a vector potential $\mathbf{A}$, the canonical momentum $\mathbf{p}$ is given by $(m^*\mathbf{v} + e^*\mathbf{A}/c)$, where m^* and e^* are the effective mass and charge of the quantized charge carriers. Thus, we have

$$nh = \oint \mathbf{p}\cdot d\mathbf{s} = \oint \left(m^*\mathbf{v} + \frac{e^*\mathbf{A}}{c}\right)\cdot d\mathbf{s}$$
$$= \left(\frac{e^*}{c}\right)\oint (\mathbf{A} + c\Lambda\mathbf{J}_s)\cdot d\mathbf{s} = \left(\frac{e^*}{c}\right)\Phi'\,, \quad (5)$$

and we obtain the quantized values of the fluxoid

$$\Phi' = n(hc/e^*) = n(hc/2e) = n\varphi_0\,, \quad (6)$$

where $\varphi_0 = hc/2e$ is the fluxoid quantum for pairs.

Since it is now generally accepted that pairs are instrumental in producing the superconducting state, we have set $e^* = 2e$ without any detailed discussion, although the step may not be considered to be obvious. In fact, one might well object that it is misleading to treat the superconducting electrons in such strict analogy to a Bose condensation of pairs, since only those electrons near the Fermi surface are strongly bound into pairs, those well inside the Fermi surface being negligibly affected by the superconducting transition. However, the presence of the inner electrons raises the Fermi velocity of the condensed electrons at the Fermi surface, so that in fact the kinetic energy of a current-carrying system of given drift velocity is the same whether or not the inner electrons are paired. A simple analogy is to a spherical fishbowl full of water. When one moves the bowl, the fluid water moves as a rigid body because it is confined within a rigid skin. Similarly, when the electron sphere is displaced in momentum space by an accelerating field, all electrons are accelerated equally and they are kept from relaxing back to a no-current regime by the superconducting "skin" at the Fermi surface. Thus one may think of the entire assembly of electrons as participating in the super-

[4] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].

current flow, not simply the small number of electrons at the Fermi surface which are strongly bound in the superconducting state. The quantum velocity imposed by fluxoid quantization on the pairs is thus imposed on the entire electron assembly. This justifies the use of our shortcut method at the absolute zero,

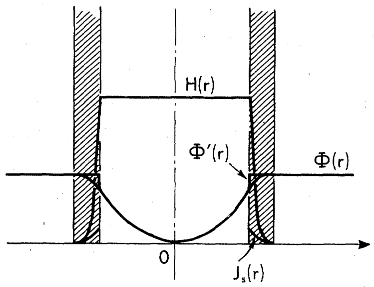


FIG. 1. Diagram showing flux Φ and fluxoid Φ' associated with circular path of radius r in case of thick-walled cylinder containing trapped flux.

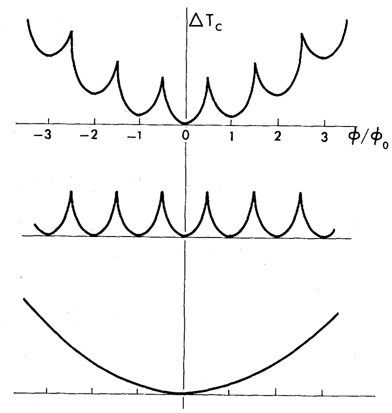
at least. At finite temperatures, the situation is complicated by the presence of quasipartical excitations which can and do quickly relax toward a noncurrent carrying state. This reduces the current from that obtained with all electrons participating rigidly, and this reduction is described by the reduced superfluid density ρ_s or n_s in the two-fluid description.[5] Thus, as the temperature is increased from $T = 0$ to $T = T_c$, n_s falls continuously from a value approximating the number of conduction electrons to zero at the transition. If the mean free path is short, even at $T = 0$, n_s is reduced below the number of conduction electrons by a factor of order (l/ξ_0), corresponding to the increased penetration depth under these circumstances. Since n_s does not enter in the quantum condition, all the considerations discussed in this paragraph are irrelevant to the quantum periodicity. They are concerned only in the quantitative size of the effects observed.

A simple illustration of the distinction between flux and fluxoid is given in Fig. 1, which represents the situation when flux is trapped in a thick-walled cylinder as in the experiments of Doll and Näbauer[6] and of Deaver and Fairbank.[7] If the cylinder is long, the field H is uniform over a cross section far from an end, as is shown in the figure. This results in a flux $\Phi(r)$ which rises quadratically with r until the wall is reached. In the wall, $H(r)$ falls off to zero in a penetration depth because of the screening supercurrent $J_s(r)$, which excludes the field from the interior of the superconductor. Thus, in the wall $\Phi(r)$ rises more slowly and then levels off to a constant value, the trapped flux. However, since the fluxoid contains a term in J_s, which drops as $\Phi(r)$ rises, the fluxoid $\Phi'(r)$ is constant through the thickness of the wall. Now, if $\Phi(r)$ did not approach a quantum value, it would be necessary for there to be a "body current" J_s as well as the indicated surface current in order for the fluxoid to have a proper quantized value. Since such a current would greatly increase the kinetic energy of the system, only quantum flux values are expected under the usual experimental conditions. This is in agreement with the findings of Doll and Näbauer and of Deaver and Fairbank.

III. THE LITTLE–PARKS EXPERIMENT

Special interest is attached to the transition into the superconducting state, since the behavior there determines the value of the fluxoid quantum number which will be maintained as one cools further below T_c. The effect upon the transition of flux through a thin-walled cylinder has been studied by Little and Parks.[3] They found a variation of resistance within the width of the resistive transition which can be interpreted in terms of a variation of T_c with flux of the sort depicted in Fig. 2. Evidently this variation

FIG. 2. Schematic diagram of results of Little-Parks experiment on the variation of T_c with flux through a thin-walled cylinder. Upper curve shows experimental result; lower curves show its decomposition into periodic and aperiodic parabolic parts.



can be decomposed into two components as indicated. One component is periodic in flux with period φ_0; the other is a pure quadratic, aperiodic background. Little and Parks concentrated on the periodic effect, and produced a theory, based on the Bardeen–Cooper–Schrieffer (BCS) theory,[8] which accounted for the existence of a periodic quadratic variation, but due to an error it underestimated the size of the

[5] J. Bardeen and J. R. Schrieffer, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1961), Vol. 3, p. 263.
[6] R. Doll and M. Näbauer, Phys. Rev. Letters **7**, 51 (1961).
[7] B. S. Deaver, Jr., and W. M. Fairbank, Phys. Rev. Letters **7**, 43 (1961).

[8] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

effect, and it failed to account for a mean-free-path effect which they demonstrated subsequently. We now give a simple theory, based on the Ginzburg–Landau[2] (GL) theory, which treats the consequences of fluxoid quantization in these experiments.

We start by recalling that GL assume a difference in free energy density between superconducting and normal states in the absence of fields and currents given by

$$f(\omega,T) = -a(T)\omega + \tfrac{1}{2}\, b(T)\omega^2 \,. \tag{7}$$

In this, ω is an order parameter equal to $n_s/n_s(T = 0)$ or $\lambda_e^2(0)/\lambda^2$, which takes on a value such as to minimize f and which goes to unity at the absolute zero and to zero at T_c. By requiring the theory to reproduce the experimental temperature dependences of the penetration depth $\lambda_e\,(T)$ and of the bulk critical field $H_{cb}\,(T)$, the two coefficients $a(T)$ and $b(T)$ may be shown to have the values

$$a(T) = \frac{\lambda_e^2(T)H_{cb}^2(T)}{\lambda_e^2(0)4\pi}\,, \quad b(T) = \frac{\lambda_e^4(T)H_{cb}^2(T)}{\lambda_e^4(0)4\pi}\,. \tag{8}$$

Now, if there are supercurrents present, they contribute a kinetic energy density

$$T = \tfrac{1}{2}\, n_s m v_s^2 = \tfrac{1}{2}\, \Lambda J_s^2 \tag{9}$$

in the superconducting state, which makes it less energetically desirable. As a result, there is a downward shift in the temperature T_c at which a nonzero order parameter can first be sustained. Because of the need to maintain fluxoid quantization everywhere, the supercurrent is forced to have the value

$$J_s(r) = \frac{n\varphi_0 - \Phi(r)}{2\pi r c \Lambda} \tag{10}$$

because of cylindrical symmetry. Since we are working right at the transition, where $n_s \rightarrow 0$ and $\lambda \rightarrow \infty$, supercurrent screening effects are negligible, and $\Phi(r)$ is simply $\pi r^2 H$, where H is the applied field. If we neglect the thickness of the cylinder wall we now see that when H is such that the flux $\pi R^2 H$ through the cylinder has a quantum value, then J_s is zero. In this case, there is no supercurrent and associated kinetic energy, and the transition should occur at exactly the same temperature as in the absence of the field. As we depart from a quantum value of flux, however, J_s will increase linearly with the discrepancy, leading to a quadratic variation of the kinetic energy and hence of the transition temperature. The maximum periodic depression of the transition temperature will occur when the flux is midway between two quantum values, since at that point the least current able to restore fluxoid quantization has its maximum value. Setting $n\varphi_0 - \Phi(R) = n\varphi_0 - \pi R^2 H = \frac{1}{2}\,\varphi_0$, and solving for the shifted transition temperature, we find

$$\left(\frac{\Delta T_c}{T_c}\right)_{\max} = \frac{\varphi_0^2}{32\pi^2 R^2 \lambda_e^2(0) H_{cb}^2(0)}\,, \tag{11}$$

where $\lambda_e(0)$ is the equilibrium or weak-field value of the penetration depth at $T = 0$ for the film. Details of this calculation have been given elsewhere.[9] Since $1/\lambda_e^2(0) \approx (1/\lambda_L^2)(l/\xi_0)$ for the short mean free paths found in these dirty film samples, this formula predicts that the magnitude of the variation of T_c should be proportional to the mean free path l. Such a variation has, in fact, been observed by Little and Parks. In a typical case, the order of magnitude of ΔT_c is predicted to be 10^{-2} to 10^{-3} °K, again in agreement with the results of Little and Parks.

The remaining question is the origin of the aperiodic quadratic background. Since this has been discussed in detail previously,[9] we shall simply summarize our results. The point is that, for cylinder walls of non-negligible thickness, $J_s(r)$ as given by (10) will be zero only for one value of r for any given H. The kinetic energy is least if this value of r is chosen to fall halfway through the thickness of the wall. In this event, $J_s(r)$ flows in opposite directions inside and outside of this radius R of null current. The strength of this current is proportional to $d\Phi/dr$, i.e., to H. Hence, the kinetic energy and depression of T_c at the quantum values of H increase as H^2 with no periodic return to zero. The coefficient of H^2 in this aperiodic term is down from that in the periodic term by a factor of $d^2/3R^2$, where d is the wall thickness and R is the mean radius of the cylinder. Incidentally, this aperiodic term is exactly the same as would be found for a plane film in a parallel field. Another source of an aperiodic quadratic effect is any misalignment of the field with respect to the cylinder axis. The coefficient of H^2 due to this mechanism should be down by a factor of $4 \sin^2\theta$ from the periodic term, where θ is the angle of misalignment. It seems probable that the observed quadratic effect may be explained by a combination of these two effects.

IV. PLANE FILM IN NORMAL MAGNETIC FIELD

Another, and more novel, application of these ideas is to the problem of determining the critical field of a thin superconducting film in a perpendicular magnetic field. In contrast, the parallel field geome-

[9] M. Tinkham. Phys. Rev. **129**, 2413 (1963), referred to in text as T.

try has been understood for many years, the GL theory leading to a parallel transition field $H_{T\|}$ given by

$$H_{T\|} = 2(\sqrt{6})(\lambda/d)H_{cb} \qquad (12)$$

provided that $d < (\sqrt{5})\lambda$, in which case the transition is of second order. Thus, for a thin film, $H_{T\|}$ can be much greater than the thermodynamic bulk critical field H_{cb}. The situation in normal field has been much less clear because of the awkwardness of the geometry. It has been known that the transition field $H_{T\perp}$ was of the same order of magnitude as H_{cb} and that it did not seem to depend strongly on the thickness or area of the film. Also, from the thermal conductivity measurements of Morris[10] and other data, it seemed experimentally to be a second-order transition, even for films thick enough to show a first-order transition in parallel field. Assuming a second-order transition, the GL theory should be applicable near the transition, even for temperatures well below T_c. These considerations led me to consider a simple model of the transition region which has been presented in detail in T (Ref. 9).

The model assumes that very near $H_{T\perp}$ the field penetrates uniformly, essentially undiminished by the small screening currents left as the order parameter goes to zero. The currents are assumed to form small circular vortices, each surrounding a point at which the order parameter goes to zero, allowing a nonzero fluxoid quantum number n. The size of each vortex is such that the flux through its perimeter is n quantum units. Thus as H increases, the vortices shrink closer together, additional ones forming at the edges of the sample. The spatial variation of ω within each vortex is chosen to minimize the sum of the kinetic energy due to the circulating current, the energy associated with the gradient of the order parameter in the GL theory, and the ordinary condensation energy terms given in (7). The conclusion reached is that the maximum field under which some superconductivity can be maintained is probably independent of the fluxoid quantum number n. This independence has been brought out particularly clearly by Miller, Kington, and Quinn.[11] However, the configuration with $n = 1$ seems to be favored on energetic grounds for any given field near $H_{T\perp}$. The transition field found in this way is

$$H_{T\perp}(T) = \frac{4\pi\lambda_e^2(T)H_{cb}^2(T)}{\varphi_0} = \sqrt{2}\,\kappa(T)H_{cb}(T)\,, \qquad (13)$$

where κ is the GL parameter $\sqrt{2}(e^*/\hbar c)\lambda_e^2(T)H_{cb}(T)$. Thus the normal critical field is found to be given by the same expression as was found by Abrikosov for negative surface energy superconductors, which he showed also break up into a similar periodic array of vortices with $n = 1$. If one inserts the usual Gorter–Casimir temperature dependences for H_{cb} and λ_e, Eq. (13) becomes

$$H_{T\perp}(t) = \frac{4\pi\lambda_e^2(0)H_{cb}^2(0)}{\varphi_0}\,\frac{(1-t^2)}{(1+t^2)}\,, \qquad (14)$$

where $t = T/T_c$. This temperature dependence differs from that of a bulk sample by a factor of $(1 + t^2)^{-1}$. A final conclusion from the model calculation is that the normal component of the magnetic field contributes linearly to the free energy, rather than quadratically as does the parallel component. This leads one to expect an angular dependence of the critical field given by

$$(H_T \sin\theta/H_{T\perp}) + (H_T \cos\theta/H_{T\|})^2 = 1\,. \qquad (15)$$

The experimental picture that emerges from magnetization measurements such as those of Chang, Kinsel, and Serin[12] (see Fig. 3) is that, as the normal

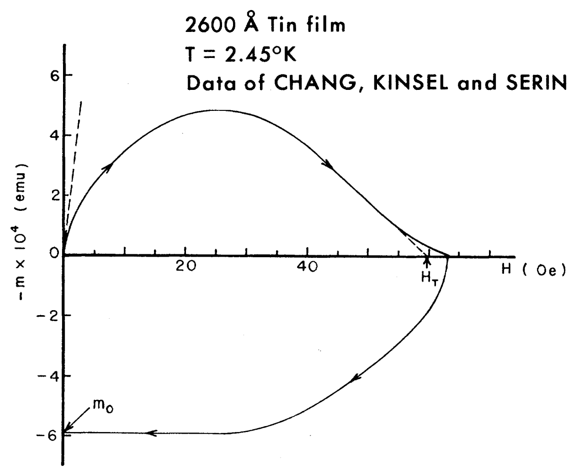


FIG. 3. Magnetization data of Chang, Kinsel, and Serin (Ref. 12) on a tin film in perpendicular field. The transition field $H_{T\perp}$ is defined by the indicated extrapolation. The remanent moment after saturation is indicated by m_0.

field is increased from zero, induced supercurrents circulate around the periphery of the film to screen the interior. This produces a large diamagnetic moment linear in H corresponding to flux being excluded from a volume of order R^3 rather than R^2d. At a field of the order of a few Gauss, the energy of

[10] D. E. Morris and M. Tinkham, Phys. Rev. Letters **6**, 600 (1962); also, Ph. D. thesis of D. E. Morris, University of California, Berkeley, 1962 (unpublished).

[11] P. B. Miller, B. W. Kington, and D. J. Quinn, Rev. Mod. Phys. **36**, 70 (1964).

[12] G. K. Chang, T. Kinsel, and B. Serin, Phys. Letters **5**, 11 (1963).

these currents becomes too great to be sustained, and the flux begins to enter in an irreversible way. From this point on, trapped flux remains if the applied field is reduced to zero. Also, the increase in magnetization is no longer linear in H. In still higher fields, the magnetization rolls over and decreases toward zero as the superconducting order parameter is driven down toward zero. The final decrease to zero magnetization appears to be linear in $(H\text{-}H_{T\perp})$, just as in the Abrikosov type II superconductors, and as predicted by our simple model. As this final transition to the normal state is approached, the field distribution is essentially uniform over the sample, and effects of sample size and macroscopic inhomogeneity seem to be unimportant. It is in this regime that our simplified theory is expected to be useful.

The point of view taken in interpreting the above results is that until there is flux penetration, we can view the film as a whole as having fluxoid quantum number zero. After flux penetrates, there will be normal spots where flux goes through. These may form either a macroscopic intermediate state pattern or microscopic units containing only one or a few fluxoid units. Some of the currents are probably best thought of as macroscopic currents circulating around the entire sample. Finally, near the transition, the field is uniform, and one expects a uniform array of vortices, each containing a single fluxoid unit. The measurements of Miller *et al.*,[11] and of Chang *et al.*,[12] show that the slope of the magnetic moment near $H_{T\perp}$ is roughly 100 times as large as that given by this simple picture. Although this discrepancy seems large, one must recall that macroscopic currents circulating over the entire surface of the film will give a moment larger by a factor of order (radius of film/radius of vortex) than currents of the same density circulating in small vortices. Since this ratio of radii is typically of order 10^4, even a relatively small circulating current over the entire film can completely overshadow the moment due to the vortices.

Since the vortices considered in the theory are too small to be seen by any optical technique, the theory must be tested largely through its prediction of the critical field value, a property which is unfortunately insensitive to the unit fluxoid aspect of the model. Nonetheless, it is satisfying to note that the theory does give a good account of a large variety of data. Some of this was cited in T. Subsequently better data have been obtained by various workers in attempts to check the theory more closely. Only the new data will be discussed here. These data confirm that $H_{T\perp}$ is essentially independent of the film thickness, except as the thickness affects the penetration depth, so long as the film is thin ($d < \xi_0$). They also confirm that the observed data fit our formula (13) with reasonable values of λ_e, and that the angular dependence of (15) is supported in several cases.

First, let us consider the magnetization data of Chang, Kinsel, and Serin[12] shown in Fig. 3. The high initial slope of $M(H)$ is shown, together with the linear approach of M to zero at $H_{T\perp}$ (apart from a slight tailing assumed due to inhomogeneity). The slope of this approach is much greater than that expected from the vortices alone, and the area under the $M(H)$ curve is orders of magnitude larger than the $H_{cb}^2/8\pi$ expected if the process were reversible. All this, together with the trapped flux, points up the importance of irreversible effects of the sort emphasized by Bean[13] in his model of hard superconductors. Thus, only the transition field itself can be compared with our theory. This is done in Fig. 4, which shows data for a stack of similar films as well as for a single film. The transition field is taken as the extrapolated end point of the linear part of the $M(H)$ curve, as indicated in Fig. 3. The data are plotted so as to facilitate comparison of the measured temperature dependence with that predicted by this model and with that of bulk tin, which is well approximated by $(1 - t^2)$. It is obvious that the $(1 - t^2)/(1 + t^2)$ dependence gives much the better fit to the data. In addition to this excellent fit for the temperature dependence, the absolute value of the penetration depth at $T = 0$ determined from the fit (namely, 580 Å) is very reasonable.

Second, we consider the data of Mercereau and Crane.[14] In their experiment they measure both the penetration depth and the perpendicular critical field as functions of temperature by measuring a supercurrent screening effect. Assuming the thermodynamic critical field is that of bulk tin, they are then able to test Eq. (13) with no free parameters. Their results show that the observed transition field points overlap completely with the transition fields computed from the measured penetration depth using Eq. (13).

A third type of experiment determining a critical field is the microwave absorption measurements of White.[15] Some typical data on a thin indium film are shown in Fig. 5. Since the field-free energy gap is about an order of magnitude greater than the

[13] C. P. Bean, Phys. Rev. Letters 8, 250 (1962).
[14] J. E. Mercereau and L. T. Crane, Phys. Rev. Letters 11, 107 (1963).
[15] R. H. White, Ph. D. thesis, University of California, Berkeley, 1964 (unpublished).

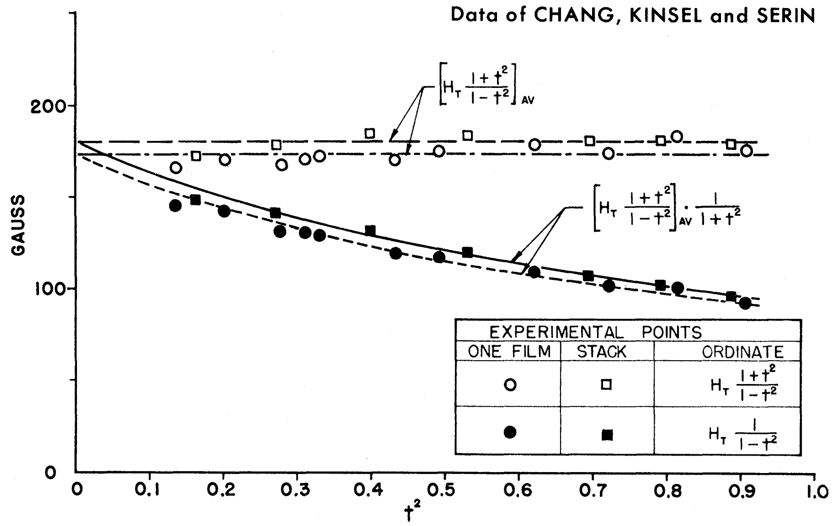


FIG. 4. Transition field data of Chang, Kinsel, and Serin. The constancy of the ordinate in the upper sets of points confirms the fit to the temperature dependence given in Eq. (14).

energy of the microwave photons used, the absorption is attributed to "normal" electrons (quasiparticles) thermally excited into the continuum above the gap. Unlike the dc resistance of the film, which is completely shorted out if $\omega > 0$ the microwave resistance measured here varies smoothly with the order parameter, and it can be used to trace the course of the transition to the normal state. The extrapolated end point of the linear rise in absorption toward the normal value is taken as the critical field. Data are shown for cases in which the static magnetic field makes angles with the plane of the film ranging from 0° to 90°. In all cases, the transition to the normal absorption is continuous, as is consistent with our assumption of a second-order phase transition. Note the extreme sensitivity of the results to angle near the parallel orientation. We infer that the parallel position can be determined within less than $\frac{1}{4}$°. This rapid variation near $\theta = 0$ is as expected from our theoretical conclusion that there is an energetic term linear in the normal component of H as opposed to the more usual quadratic dependence. The critical fields for this film are plotted as a function of angle in Fig. 6, together with the theoretical angular dependence and two alternate dependences. The dashed curve arises from a formula like (15) but with both components entering quadratically; the chain line curve results if both enter linearly as suggested by Morris.[10] The agreement with the theoretical dependence is remarkably good. Similarly good

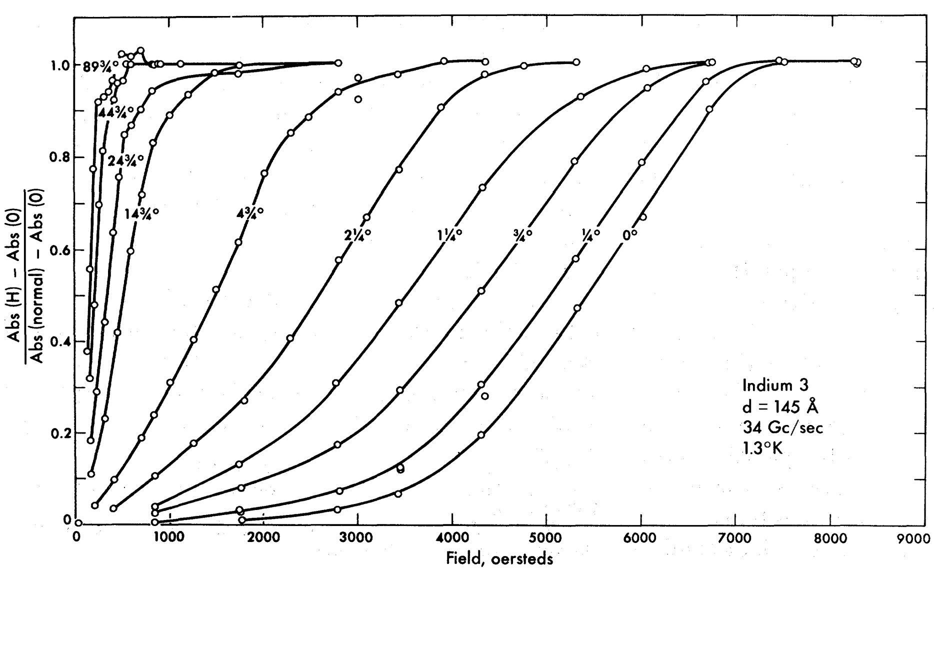


FIG. 5. Microwave absorption in a thin indium film as a function of angle and strength of static applied field. Absorption is normalized to unity in the normal state, and the zero is taken at the level of field-free absorption. Data is that of White (Ref. 15).

agreement has been obtained with a thin tin film, but a lead film has been found to fit the doubly quadratic curve better. Since the data of Morris on a lead film quoted in T agree with the theoretical curve, the source of the discrepancy mentioned is not clear.

Finally, we mention an experiment being pursued by Parks[16] which *does* give some evidence for the size of a single circular vortex. The setup is similar to that in the Little–Parks experiment discussed above, except that he measures shifts in the resistive transition of a narrow strip film in a normal field rather than the resistance of a cylindrical film in an axial field. In this experiment, Parks finds a dip in the generally rising curve of resistance vs applied field at a field such that circular vortices containing one flux quantum approximately fit in the width of the film strip. This behavior has been verified with strips of several widths. Even without a detailed analysis, this effect qualitatively supports the existence of circular vortices of unit flux. The detailed interpretation is, however, still in doubt.

V. CONCLUDING REMARKS

We have seen that by using the concept of fluxoid quantization to determine currents in the framework of the GL theory we have been able to give a good account of the periodic and aperiodic variations of T_c with flux through a thin-walled cylinder. We have also been able to derive a formula for the transition field of a plane film with the field at normal incidence, and an interpolation formula for the transition field at intermediate angles. All of these predictions have received considerable experimental support. The model used in treating the normal field case involves an array of circular vortices each containing a single quantum of flux. As such, it forms a highly simplified version of the Abrikosov theory[4] of type II superconductors, and it leads to the same expression for the critical field as does that rather abstruse theory. This is of pedagogical value in understanding Abrikosov's analysis. In particular, we see that the limiting scale of subdivision in the negative surface energy superconductors can be considered to be set by Plank's constant through the flux quantum, a point not brought out explicitly in his paper.

Despite the close similarity of the two cases, we must remember that there is not a complete equivalence. Since Abrikosov is treating bulk material of infinite extent, the irreversibility associated with the large demagnetizing coefficient of the film geometry is avoided. Also, films should show the vortex behavior even when the material in bulk form would not have negative surface energy behavior. The point is that the film must subdivide on a scale of the order of the film thickness before reasonably small demagnetizing coefficients for superconducting domains can be reached. But if the film thickness is less than a coherence length, this implies a microscopic "mixed state" rather than a macroscopic intermediate state. Thus, the film geometry encourages the material to go into a mixed state even though the material may have a value of $\kappa < 1/\sqrt{2}$, in which case the transition field is less than the bulk critical field, and the second type behavior would not normally be expected.

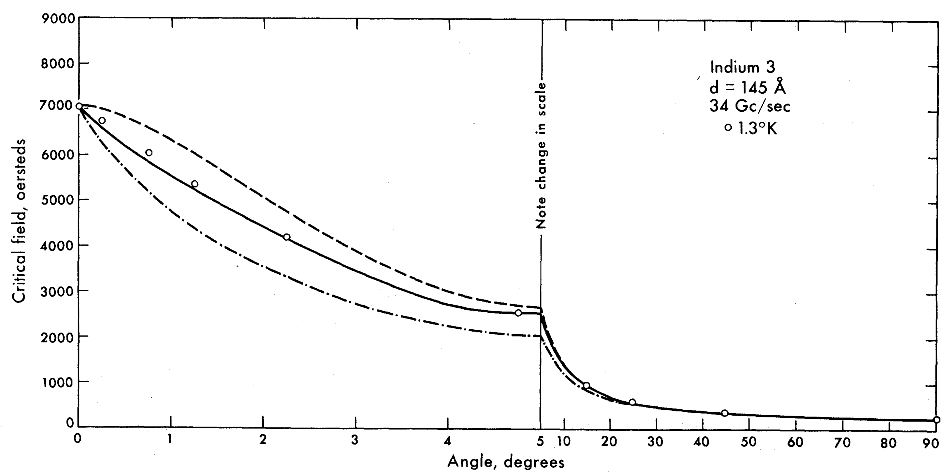


FIG. 6. Angular dependence of critical field found from microwave absorption measurements shown in Fig. 5. Critical field is taken as the extrapolated intersection of the linear part of the curve with the level of normal absorption. Solid curve is plot of Eq. (15) with $H_{T\|}$ and $H_{T\perp}$ fitted at $\theta = 0°$, 90°. Dashed and chain curves are similar, but with both components entering quadratically or linearly, respectively.

[16] R. D. Parks, private communication, to be published.

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Discussion 42

GORTER: The Rutgers group who studied the dependence of H_{c2} as a function of temperature were very happy to find agreement with your theory—the factor of 2 in the temperature dependence of H_{c2}. This essentially means, if we remember the discussion of two days ago, that it agrees with the formula given on the basis of a combination of Abrikosov and Ginzburg's formulas. The Gor'kov theory gave 1.25 as far as I remember, so saying that there's general agreement with theory means certainly not with the theory of Gor'kov.

TINKHAM: On this matter of 2 vs 1.25, I haven't studied these papers carefully. Is there a difference depending on whether the mean free path is important or not? In the model that we're using, of course, the penetration depths, etc., are those of ordinary soft superconductors.

GORTER: If I remember well, Dr. Werthamer was telling us that Shapoval's calculation for the short mean free path case was wrong and for both cases he expected the factor 1.25.

GAULÉ: What is your theoretical or experimental expectation for the size of the small vortices?

TINKHAM: The only case which I've treated is the case in which you have complete penetration so the field is just the applied field. The size of the vortex is just determined by geometry, $\pi r^2 H$ has to be 2×10^{-7} G cm^2. Depending on what system you're talking about, the vortex will be given by that. Specifically, the radius is about a thousand Å in these systems.

SERIN: In reply to Professor Gorter I'd just like to make the point that we are quite aware of the two different temperature dependences. I'd also like to say that we have some data on a very much thicker film where we no longer get or expect any sensible agreement with this theory. The calculated penetration depth is very very small, so it looks as if we're approaching intermediate state behavior. We get a temperature dependence of the Ginzburg–Landau type even in this circumstance. So there may be a difference between a rather pure substance and one with a short mean free path, although we're not at all certain of this.

PIPPARD: I would like to express what Dr. Tinkham has been saying about the Little experiment on the cylinder in terms that perhaps Heinz London would have used in 1935 if he had known about flux quantization. If you have a thin film of a superconductor, you can destroy superconductivity by means of an external applied magnetic field or by means of a current, and London worked out a theory of the field and current required. Now in the cylinder you have in effect a combination of the two: when you apply magnetic field to the outside you have (a) the mean field between the inside and outside which is equivalent to an external applied field, and (b) the difference between internal and external fields which is due to a current. Now both these effects separately produce a quadratic variation of transition temperature with the strength of the effect and the explanation that Dr. Tinkham gave of the slow parabolic variation is of course simply the change of transition temperature of a film in a steady applied field. The oscillatory effects arise from the fact that quantization forces certain values on the current, and this gives the whole explanation if it's right. Now, one can check whether it's right by perhaps a not very simple experiment, which is to split the cylinder so that you get no steady current flowing around but only the external field; then you ought to get simply the parabolic variation, the same as before, without the oscillation superposed.

LITTLE: We had a crazy idea once of making a cylinder in which it was superconducting most the way round and normal on the other part considering that the electrons must always have their wave functions single valued whether they are in the normal state or in the superconducting state and looking for any periodic variation. We didn't see anything. I think this is equivalent to your expectation and that we did see just the general quadratic background.