Transition of Type-I Superconducting Thin Films in a Perpendicular Magnetic Field: A Microwave Study

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Microwave resistance measurements which were undertaken to seek direct evidence for the existence of the mixed state in thin type-I films are described. The measurements were for the most part confined to indium films. The observed change in the microwave resistance of the films as a function of magnetic field applied perpendicular to the surface is, in detail, similar to reported surface resistance measurements on bulk type-II superconductors. This includes the intrinsic hysteresis observed below H_{c2} and the existence of a superconducting surface layer at the edge of the film which presists to H_{c3}.

I. INTRODUCTION

ACCORDING to Tinkham,[1] if a film of a type-I superconductor is sufficiently thin it will undergo a second-order phase transition to the normal state when the magnetic field applied at right angles to its surface is equal to $H_{\perp}=\sqrt{2}\kappa H_c=H_{c2}$. For fields less than H_{c2} the magnetic flux enters the film as an array of quantized vortices rather than being confined to macroscopic normal regions as in the ordinary intermediate state. Such an array is analogous with the mixed state in type-II materials. Unlike the case of type-II superconductors κ may be much smaller than $1/\sqrt{2}$ so that $H_{\perp}$ may be much smaller than H_c. Chang *et al.*[2] have investigated the temperature dependence of $H_{\perp}$ and κ for type-I films. Cody and Miller[3] have shown that the relationships between $H_{\perp}$ and $H_{||}$ (the critical field parallel to the film) are such that the identification of $H_{\perp}$ with H_{c2} is indeed valid. In fact, such an identification permitted the extraction of known parameters of bulk Pb from their data.

Earlier microwave studies of White and Tinkham[4] provided information relative to the behavior of thin films in a parallel field and to the angular dependence of the critical field. However, a critical examination of the perpendicular field properties was not made at that time. The present microwave measurements were undertaken to seek additional and more direct evidence that a mixed state does indeed exist in type-I films. The behavior of the microwave surface resistance of type-II superconductors has been extensively studied[5–7] and the results correlated with the flux–flow losses observed under dc conditions.[7–9] If the microwave losses in type-I films were analogous to those observed in type-II materials, then it would seem reasonable that the parallel is complete and all the properties of the mixed state, including flux flow and flux pinning, are applicable to thin films. The results of these experiments do make a strong case of the existence of a mixed state in type-I films and, in fact, reveal the existence of a superconducting surface layer which persists to a magnetic field corresponding to H_{c3} as in bulk type-II material.

II. EXPERIMENT

The measurements were made on a series of indium films of different thicknesses. Indium was chosen because of its low intrinsic κ. It was chosen rather than tin because its thin films had lower residual resistivities than those of tin and because its properties seemed less sensitive to strain. It was chosen rather than aluminum because of its more convenient temperature range.

The films were made by evaporation in a vacuum of about 5×10^{-6} mm of Hg (more or less depending on the evaporation rate) on to glass substrates which were in thermal contact with a copper base held at liquid N_2 temperature. After evaporation, the films were photoetched in the form shown in Fig. 1. The triangular portions of the films served as tabs to which leads could be attached for measuring dc resistance. Only the narrow portion was exposed to the microwave electric field. Photoetching was necessary because of the extreme sensitivity of the microwaves to any penumbra resulting from evaporation through masks. The photoresist which covered the unetched portion of the film was not removed. It served as protection against damage to the film during mounting and as insulation from the cavity to permit monitoring the electrical resistance during the experiment. Esti-

[1] M. Tinkham, Phys. Rev. **129**, 2413 (1963); Rev. Mod. Phys. **36**, 268 (1964).
[2] G. K. Chang, T. Kinsel, and B. Serin, Phys. Letters **5**, 11 (1963).
[3] G. D. Cody and R. E. Miller, Phys. Rev. Letters **16**, 697 (1966).
[4] R. H. White and M. Tinkham, Phys. Rev. **136**, A203 (1964).
[5] M. Cardona, G. Fischer, and B. Rosenblum, Phys. Rev. Letters **12**, 101 (1964); M. Cardona and B. Rosenblum, Phys. Letters **8**, 308 (1964).
[6] A. Rothwarf, J. I. Gittleman, and B. Rosenblum, Phys. Rev. **155**, 151 (1967) and (to be published).
[7] M. Cardona, J. Gittleman, and B. Rosenblum, Phys. Letters **17**, 92 (1965).
[8] Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev **139**, A1163 (1965).
[9] J. Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).

mates of the impurity mean free path from residual resistance ratios and boundary scattering formulas were much greater than 10^4 Å.

The microwave cavity and the relative orientation of the specimen are shown in Fig. 2. To ensure linearity, the coupling of the cavity to the system was slightly less than critical and the specimen was weakly coupled to the cavity by being placed near a node of the electric field. The klystron was stabilized to the resonant frequency of the cavity which was about 23 GHz. The change in the microwave power reflected from the cavity as a function of applied field was proportional to the change in the power absorbed by the specimen. The change in the power absorbtion is in turn proportional to the microwave resistivity of the film.

The magnetic field was provided by a small pair of superconducting Helmholtz coils which could be rotated to provide fields both perpendicular and parallel to the specimen. The microwave electric field was parallel to the film strip. Two appropriately oriented Hall probes (Siemens type RHY17) were used to measure the magnetic field.

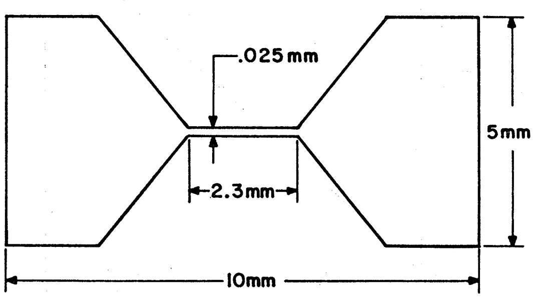

FIG. 1. Specimen shape and dimensions.

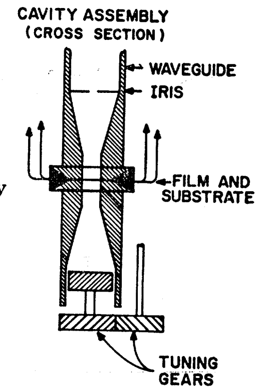

FIG. 2. Microwave cavity assembly (cross section).

III. RESULTS

Figure 3 gives a typical low-temperature result for the perpendicular field configuration. It is a tracing of the change in power reflected from the sample cavity as a function of magnetic field as obtained on an X-Y recorder. The general features of these data are:

1. As the magnetic field is increased, $P-Po$ increases in a curve which is concave upward until the magnetic field designated $H_\perp$ is attained. At this point there is a break in the curve after which $P-Po$ continues to increase reaching a limiting value at the field designated H_n.

2. For $H_\perp<H<H_n$, $P-Po$ is completely reversible.

3. For $0<H<H_\perp$, a hysteresis is observed. There are two distinct curves, one for increasing field and one for decreasing field. If on the increasing curve the field is suddenly decreased, then $P-Po$ changes along a connecting curve until the decreasing curve is reached. The connecting curves are completely reversible with respect to increasing and decreasing fields.

4. The dc resistance of the specimens is restored in the neighborhood of H_n.

The curves obtained at all temperatures were similar to the one shown. The effect of increasing temperature was to: (1) decrease P_n-Po, (2) decrease $H_\perp$ and H_n, (3) cause the hysteresis to be less distinct, and (4) cause the break at $H_\perp$ to be less distinct.

A comparison of the above results for a field *perpendicular* to the film with the microwave surface resistance measurements[5,7] of type-II materials in which the magnetic field is *parallel* to the specimen and *perpendicular* to the microwave electric field reveals a striking similarity. The reversibility for $H_\perp<H<H_n$ is very similar to that observed in the type-II materials for $H_{c2}<H<H_{c3}$. Also, the hysteresis observed for fields below $H_\perp$ bear a marked resemblance to the intrinsic hysteresis observed in the type-II materials below H_{c2}.[7] The only real difference rests in the detailed shape of the curves. However, the shapes should not necessarily be the same since the surface resistance was measured in the earlier case and the film resistance was

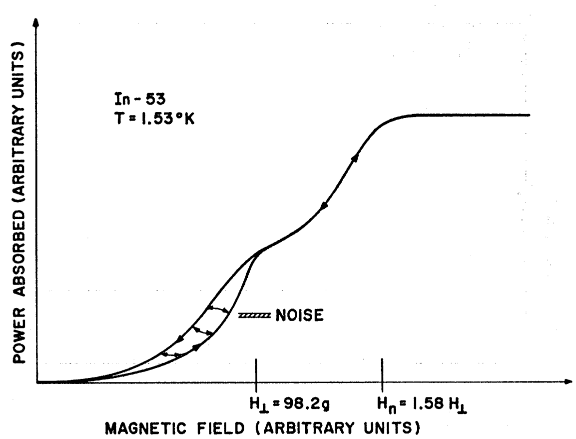

FIG. 3. Microwave power absorbed by specimen versus magnetic field applied perpendicular to the film.

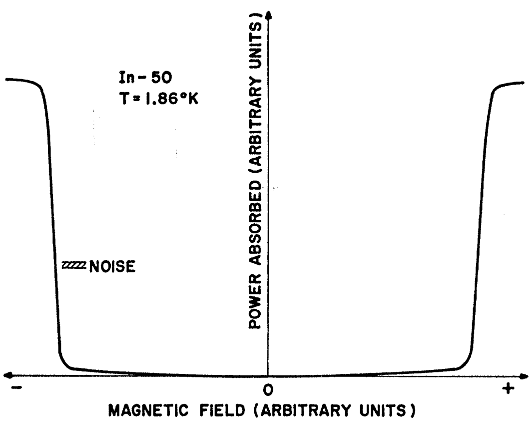

FIG. 4. Microwave power absorbed by specimen versus magnetic fields applied parallel to the film.

measured in the present case. Considerations of the shape of the curve will be made later. The similarity is sufficient to allow the tentative identification of $H_\perp$ with H_{c2} and H_n with H_{c3}. For fields between $H_\perp$ and H_n, the film strip is considered to be normal except for thin surface layers along the edges. It is then an objective of the remainder of this paper to examine the consistency of our data, subject to these identifications, with current theories and other experiments.

Before doing this the measurements in the parallel field configuration will be considered. Figure 4 gives a typical low-temperature result. It is a tracing of the power reflected from the sample cavity as a function of magnetic field. This curve is nearly identical with resistance-versus-field curves measured at dc. At temperature approaching T_c, the critical field $H_{||}$ is equally well defined. However, $P_n - P_0$ becomes smaller and the curve becomes bowed upward as expected. For thicknesses greater than 1500 Å, supercooling can be observed suggesting the onset of a first-order transition. As has been observed and predicted by many workers,[10–12]

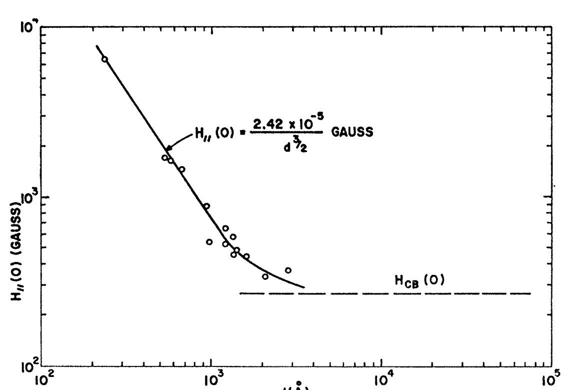

FIG. 5. Parallel critical field ($T=0$°K) versus film thickness.

$H_{||}$ was found to be very nearly proportional to $[(1-t^2)/(1+t^2)]^{1/2}$, where t is the reduced temperature, T/T_c, over the temperature range of measurement $(0.5<t<1)$. For the thinner films, $H_{||}$ was found to be proportional to $d^{-3/2}$ in agreement with Toxen.[13] Figure 5 is a plot of $H_{||}(0)$ versus d as determined from the microwave data. If one assumes[14] that

$$H_{||} = (24)^{1/2}(\lambda/d)H_c$$

and

$$\lambda = \lambda_L[\xi_0/d]^{1/2},$$

one obtains equally good agreement with either the suggested values of Toxen[13] [$\xi_0 = 2600$ Å, $\lambda_L(0) = 350$ Å] or those of Burton[15] [$\xi_0 = 3400$ Å, $\lambda_L(0) = 275$ Å]. $H_{||}(0)$ is the value of $H_{||}$ at absolute zero obtained by extrapolation; d is the film thickness; λ is the weak-field penetration depth; λ_L is the London penetration depth; and ξ_0 is the Pippard coherence distance.

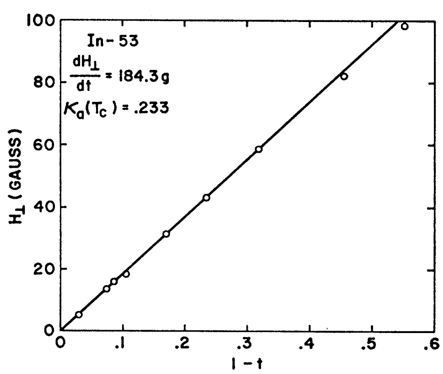

FIG. 6. Perpendicular critical field versus $(1-t)$.

A. Analysis of the Critical Field Data

The first and simplest point to check is the ratio of $H_n/H_\perp$. For some twenty specimens, including one specimen of lead, $H_n/H_\perp$ was found to have an average value of 1.6 independent of temperature, the smallest measured value being 1.5 and the largest 1.7. This is in excellent agreement with the expected[16] value for H_{c3}/H_{c2}.

The second point to note is that, where it is possible to check, $H_\perp$ as determined from the microwave data corresponded to the field for which the magnetic moment vanished.[17] The magnetic-moment measurements were not sensitive enough to detect superconductivity in a strip about ξ wide at the specimen edge.

Since we assume that $H_\perp = \sqrt{2}\kappa(t)H_c(t)$, the value of

[10] D. H. Douglass and R. H. Blumberg, Phys. Rev. **127**, 2038 (1962).
[11] A. M. Toxen, Rev. Mod. Phys. **36**, 308 (1964).
[12] P. G. de Gennes and M. Tinkham, Physics **1**, 107 (1964).
[13] A. M. Toxen, Phys. Rev. **127**, 382 (1962).
[14] V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. **20**, 1064 (1950).
[15] R. Burton, Cryogenics **6**, 257 (1966).
[16] D. Saint-James and P. G. de Gennes, Phys. Letters **7**, 206 (1963).
[17] G. D. Cody and R. E. Miller (unpublished data).

TABLE I. Summary of the critical field data.

Sample	$R_4/(R_{300}-R_4)$	d[a] (Å)	T_c (°K)	$\lambda(0)$[b] (Å)	$\kappa_a(T_c)$[c]	$\kappa_b(T_c)$[c]	$H_n/H_\perp$
In-50	0.067	1460	3.45	780	0.348	0.366	1.64
In-51	0.038	1203	3.41	580	0.22	0.20	1.50
In-52	0.055	575	3.42	683	0.27	0.28	1.70
In-53	0.036	927	3.41	605	0.23	0.22	1.58
In-53	0.041	1366	3.41	595	0.228	0.213	1.51

[a] Obtained from $H_\parallel/H_\perp$ and Refs. 16 and 18.
[b] $\lambda(0)$ is obtained from curves similar to Fig. 7.
[c] κ_a is obtained from Eq. (1) and κ_b from Eq. (3).

$\kappa(T_c)$ can be determined from the $H_\perp$ data. Thus, taking $H_c(t)=H_c(0)[1-t^2]$,

$$\kappa(T_c)=\frac{1}{2\sqrt{2}H_c(0)}\left|\frac{dH_\perp}{dt}\right|_{t=1}. \qquad (1)$$

κ determined in this way is designated as $\kappa_a(T_c)$. de Gennes and Saint-James[16] and Burger *et al.*[18] have given $H_\parallel/H_\perp=f(H_\parallel\pi d^2/2\phi_0)$, where ϕ_0 is the flux quantum. Both the parallel and perpendicular transitions are assumed to be of the second order. f is such that if d is sufficiently large, $f\rightarrow1.7$ (a limit not obtainable in indium because of the onset of a first-order parallel transition). If d is sufficiently small, $f\rightarrow3/(H_\parallel\pi d^2/2\phi_0)$. The latter limit is obtainable directly from the Ginzburg-Landau theory.[14] If this relationship is used to compute the film thickness, the value obtained for a given sample is constant over a wide range of temperatures. The value of d obtained in this way varied only 20% from the value obtained from room-temperature and helium-temperature dc resistance measurements.

From the Ginzburg-Landau theory[14]

$$H_\parallel=\sqrt{24}[\lambda(0)H_c(0)/d][1\quad t]^{1/2},$$

where $\lambda(t)$ is the weak-field penetration depth and is a function of d. Thus the value of $\lambda(0)$ can be computed. Also, from Ginzburg-Landau

$$\kappa(t)=2\sqrt{2}\pi\lambda^2(t)H_c(t)/\phi_0, \qquad (2)$$

so that putting $\lambda^2=\frac{1}{4}[\lambda^2(0)](1-t)^{-1}$ and

$$H_c=2H_c(0)(1-t),$$

$$\kappa(T_c)=\sqrt{2}\pi\lambda^2(0)H_c(0)/\phi_0. \qquad (3)$$

$\kappa(T_c)$ computed from this relationship is designated as $\kappa_b(T_c)$. For all computation $H_c(0)$ was taken as 280 G.

Figure 6 is a plot of $H_\perp$ as a function of $(1-t)$ and Fig. 7 is a plot of $H_\parallel$ as a function of

$$\left[\frac{1\quad t^2}{1+t^2}\right]^{1/2}\left[\lim_{t\rightarrow1}\left(\frac{1-t^2}{1+t^2}\right)^{1/2}=[1-t]^{1/2}\right].$$

The scatter indicated for this sample is typical of all the samples. Table I is a summary of the analysis of the critical field data. It is clear that the results are consistent with the identification of $H_\perp$ with H_{c2} and H_n with H_{c3}.

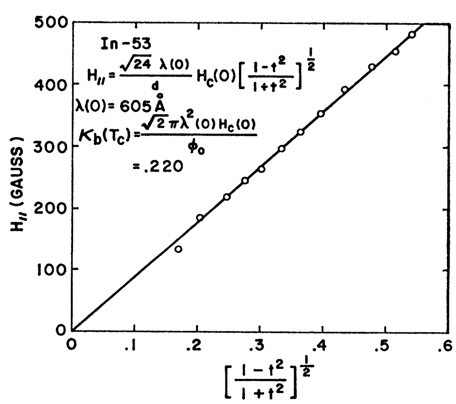

FIG. 7. Parallel critical field versus $[(1-t^2)/(1+t^2)]^{1/2}$.

[18] J. P. Burger, G. Deutscher, E. Guyon, and A. Martinet, Phys. Rev. **137**, A853 (1965).

B. The Dependence of Reflected Microwave Power on Magnetic Field

It is desirable to subject the microwave-power-versus-perpendicular-field data to an analysis analogous with the treatment of the surface resistance data.[6] To do this it is necessary to impose certain reasonable assumptions and construct a model from which the power absorbed by the specimen can be computed. It was pointed out earlier that the specimen was weakly coupled to the cavity. Therefore, the first assumption is that the specimen sees a high-impedance, essentially constant-current, microwave source. Then the power absorbed by the specimen is proportional to its microwave resistance or $P\propto \mathrm{Re}(Y)/|Y|^2$, where Y is the admittance of the film strip.

Since the film strip is very narrow (2.5×10^{-3} cm) compared with a wavelength, we assume that the microwave electric field is uniform across the width of the sample. By analogy with the situation in type-II materials and in accordance with the calculations of

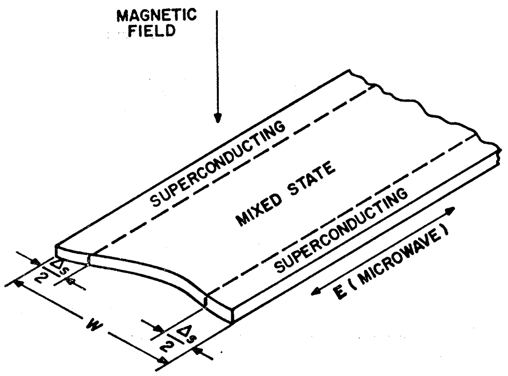

FIG. 8. Assumed structure in specimen under the influence of a perpendicular magnetic field.

Fink and Kessinger,[19] the sample is assumed to be composed of three parallel strips. The center strip is in the mixed state for $H<H_\perp$ and in the normal state for $H>H_\perp$. The two outer strips are assumed to be in the superconducting state for $H<H_n$. The order parameter in the outer strips is assumed to be large and constant for $H<H_\perp$. Thus, the film strip is pictured schematically as shown in Fig. 8.

Since the electric field is presumed to be uniform across the strip, although the total microwave current is constant (by virtue of the high-impedance nature of the source) it cannot be uniform across the specimen. The cores of the vortices are essentially normal and occupy a fraction of F_n of the volume of the sample. Thus, the admittance for the center strip can be written as

$$Y_m=\{[\sigma_nF_n+\sigma_1(1-F_n)]-i\sigma_2(1-F_n)\}(w-\Delta_s),$$

where σ_n is the normal-state conductivity, $\sigma_1-i\sigma_2$ is the complex conductivity of the superconducting areas surrounding the cores, and $(w-\Delta_s)$ is the width. We assume $\omega\tau\ll1$ throughout, where ω is the microwave frequency and τ the normal-state electron scattering time. For each of the outer strips, the admittance is

$$Y_s=-i\sigma_{2s}(\Delta_s/2),$$

where $\Delta_s/2$ is the width of an outer strip and, for simplicity, the rea parts of its conductivity was assumed negligible. Because the electric field is uniform across the width, the total admittance, Y, is the sum of the admittances so that

$$Y=[\sigma_n(F_n)+\sigma_1(1-F_n)](w-\Delta_s)\\ -i[\sigma_2(1-F_n)(w-\Delta_s)+\sigma_{2s}\Delta_s].$$

The power absorbed therefore is

$$P\propto\frac{\sigma_nF_n+\sigma_1(1-F_n)}{(w-\Delta_s)\{[\sigma_nF_n+\sigma_1(1-F_n)]^2+[\sigma_2(1-F_n)+\sigma_{2s}(\Delta_s/(w-\Delta_s))]^2\}}.$$

Then if $w-\Delta_s\simeq w$

$$P=P_n\frac{F_n+(\sigma_1/\sigma_n)(1-F_n)}{[F_n+(\sigma_1/\sigma_n)(1-F_n)]^2+[(\sigma_2/\sigma_n)(1-F_n)+X_s]^2}\qquad(4)$$

where

$$X_s=\frac{\sigma_{2s}}{\sigma_n}\frac{\Delta_s}{w-\Delta_s}$$

and P_n is the power absorbed in the normal state. Furthermore, if $P=P_\perp$ at $H=H_\perp$ and $P=P_0$ at $H=0$,

$$P_\perp=P_n\frac{1}{1+X_s^2}$$

and

$$P_0=P_n\frac{\sigma_1/\sigma_n}{(\sigma_1/\sigma_n)^2+(\sigma_2/\sigma_n+X_s)^2}.$$

Taking the two-fluid model-temperature dependence $\sigma_1/\sigma_n=t^4$ and

$$\frac{\sigma_2}{\sigma_n}=\frac{\rho_n}{\omega\mu_0\lambda^2(0)}(1-t^4),$$

where

ρ_n = normal-state resistivity,

$\omega=2\pi\times$frequency,

$\lambda(0)$ = the weak-field penetration depth,

and

$\mu_0=4\pi\times10^{-9}$ h/cm,

then X_s can be determined from the experimental values of $P_n-P_\perp$ and P_n-P_0. Using the value of $\lambda(0)$ obtained from parallel-field data, the ratio $(P-P_0)/(P_\perp-P_0)$ can be plotted as a function of $H/H_\perp$ as shown in Figs. 9 and 10. Figure 9 is a plot for low-temperature data and the experimental fit is obtained by taking $F_n=H/H_\perp$. Figure 10 is for a high temperature and here $F_n=(H/H_\perp)/(2-H/H_\perp)$.

The excellent agreement between the model and the experimental data is, to some degree, fortuitous. First of all, arguments can be made for choosing either of the expressions for F_n given above.[6,8,9] However, it is difficult to understand why one expression should be

[19] H. J. Fink, Phys. Rev. Letters **14**, 309 (1965); H. J. Fink and R. D. Kessinger, Phys. Rev. **140**, A1937 (1965).

applicable at low temperatures and one at high temperatures. The difference between the two expressions is well beyond experimental accuracy. On the other hand, simplifying assumptions were made in deriving Eq. (4). The two assumptions which probably are the most important with regard to the functional dependence of P on magnetic field are (1) ignoring the real part of the conductivity of the surface layers, and (2) assuming that X_s is independent of magnetic field. In fact, it is perfectly reasonable for Δ_s, and hence X_s, to change with field as it does in the case considered by Fink and Kessinger.[19]

IV. CONCLUSIONS

The resolution of the field dependence of F_n without the introduction of additional assumptions requires the application of the theory to the specific boundary-value problem presented by the film strip. However, in spite of the fact that such a calculation is lacking, Eq. (4) does seem to describe the microwave properties of the thin films. It perhaps should be pointed out that if the film thickness becomes large compared with the microwave skin depth, Eq. (4) is no longer applicable. Equation (4) was derived assuming that the power absorbed by the specimen was proportional to its net resistance (no skin effect). However, for thick specimens, the power will be proportional to the surface resistance. Analysis of data using Eq. (4) was confined to the thinner specimens.

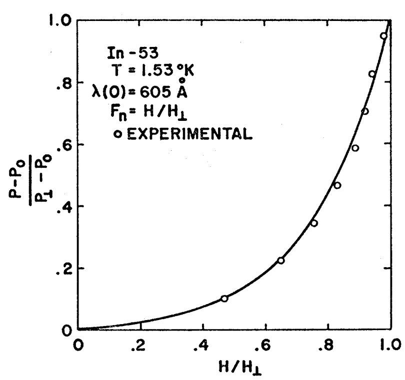

FIG. 9. Low-temperature power absorption versus magnetic field [from Eq. (4)].

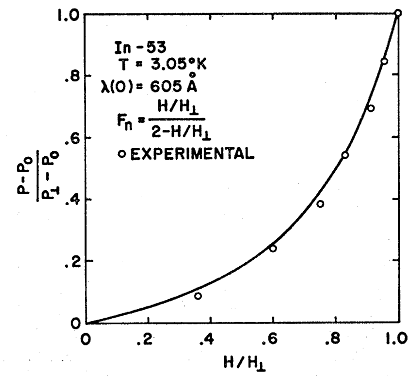

FIG. 10. High-temperature power absorption versus magnetic field [from Eq. (4)].

The results of the parallel and perpendicular critical-field measurements, as well as the field dependence of the microwave absorption strongly support other evidence[2,3,18] that thin films in perpendicular magnetic fields do indeed exhibit a second-order transition to the normal state at $H_{c2}=\sqrt{2}\kappa H_c$. Also, for $H<H_{c2}$, the flux enters the film in quantized vortices forming a mixed state as in type-II materials. In addition, the experiments show that for $H>H_{c2}$ a surface layer at the edge of a film remains superconducting until a field $H_{c3}\simeq 1.6H_{c2}$ is reached. The existence of surface superconductivity makes the identity of flux penetration for perpendicular fields in thin type-I films with the flux penetration in type-II superconductors complete.

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