Magnetic-Field Dependence of Radio-Frequency Flux Penetration in Type-II Superconductors*

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The magnetic-field dependence of the penetration of radio-frequency fields has been measured in type-II superconductors. We discuss information obtained concerning the mixed state, the superconducting surface sheath, and superconducting cylindrical thin films. Some of the results, such as structure near H_{c2}, are interpreted in terms of vortex motion and effects of the sheath. Results pertaining to the sheath are compared with the calculations of Fink and Kessinger and of Maki. Other results in the mixed state are related to surface impedance calculations of Maki and of Caroli and Maki, based on the microscopic theory. Penetration depth measurements in the films agree better with a naive model than with Pincus's theory.

I. INTRODUCTION

THE equilibrium properties of type-II superconductors[1] in steady magnetic fields, ranging from zero to just above a value that completely destroys the superconductivity, are mostly well accounted for by the Ginzburg-Landau theory[2] or its microscopic equivalent.[3] In the mixed state—when the field lies between the first and second critical fields, H_{c1} and H_{c2}, and the Meissner effect is no longer observed—flux enters in amounts determined by the fluxoid quantization condition.[4] Each fluxoid has associated with it a vortex in the superconducting electron fluid, at the center of which the superconducting order parameter is zero. Between H_{c2} and the third critical field $H_{c3}(\theta)$, a superconducting layer[5] remains at those parts of the surface of the superconductor that are inclined at an angle θ with respect to the local magnetic field, while the interior behaves as though it were normal.

The original Ginzburg-Landau equations were time-independent, being derived initially from the thermodynamic theory of second-order phase transitions,[6] and thus described only equilibrium phenomena. Much progress towards formulating the time-dependent version of the theory has recently been made.[7] Meanwhile, there have been attempts to describe non-equilibrium phenomena in terms of modifications of the classical hydrodynamic theory of vortex motion in a perfect fluid.[8]

We shall be concerned here with the effects of alternating currents, produced by radio-frequency fields, on the mixed state—which in some cases can be understood in terms of the vortex motion that the currents produce. A steady electric current passed through an array of stationary vortices in a type-II superconductor produces a force which in general is perpendicular to both the vortices and the current.[9] A drift of the vortices, or "flux flow,"[10] may thus be brought about—provided that the force is sufficient to overcome the pinning effects of potential wells caused by inhomogenities in the system.[11] (A weak force can produce vortex hopping or "flux creep" rather than smooth flow.[12]) The pinning effects may be negligible when an alternating current of sufficiently high frequency is applied, since small-amplitude motion can occur within the pinning potential well or at those parts of the vortex which are not close to a pinning center.[13]

The effect of a weak oscillating magnetic field on a type-II superconductor in the mixed state depends on the relative orientations of the oscillating induced currents, the steady magnetic field, and the surfaces of the sample. Except when the steady magnetic field is perpendicular to the surface, the interpretation of the results is complicated by the superconducting

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[1] Elementary survey up to December 1965: E. A. Lynton and W. L. McLean, Advan. Electron. Electron Phys. **23**, 1 (1967); more detailed account: P. G. de Gennes, *Superconductivity of Metals and Alloys* (W. A. Benjamin, Inc., New York, 1966).
[2] V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. **20**, 1064 (1950). See also Ref. 1.
[3] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **36**, 1918 (1959) [English transl.: Soviet Phys.—JETP **9**, 1364 (1959)]. See also N. R. Werthamer in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, Inc., New York, to be published).
[4] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].
[5] D. Saint-James and P. G. de Gennes, Phys. Letters **7**, 306 (1963); D. Saint-James, *ibid.* **16**, 218 (1965).
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[10] Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. **139**, A1163 (1965).
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[12] P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
[13] J. I. Gittleman and B. Rosenblum, Phys. Rev. Letters **16**, 734 (1966).

surface layer. When the field is parallel to the surface, the order parameter is nonzero throughout a layer of depth of the order of the coherence length. If the field is tilted, vortices pierce the layer, but it still continues to have a shielding effect until the field is normal to the surface.[14] There are thus a number of different configurations to consider.

The description just given has been in the spirit of the hydrodynamic models. The major difficulty in these models is in adequately dealing with the core of the vortex, where the order parameter is either zero or so small that the superconductivity is gapless. This is where differences arise between the two most highly developed vortex models, those of Bardeen and Stephen[8] and of Nozières and Vinen.[8] For instance, the predicted field dependences of the Hall angle near H_{c2} are different. An attempted comparison with the present results is discussed in Sec. III A.

A microscopic theory of the response of the mixed state to oscillating fields has been given by Caroli and Maki.[15] One of the possible modes of oscillation corresponds to oscillating flux flow, but differs from flux flow in that it does not correspond to a merely translational oscillation of the pattern of the order parameter. In the configuration where the oscillating currents are parallel to the vortices, the results reduce in the appropriate limit to those obtained by Maki,[16] neglecting any changes in the order parameter produced by the oscillating field. Although the experiments here were not originally designed to test these microscopic theories, some comparisons with them are made in Sec. III A, and remarks about their applicability to realizable experiments are given in Sec. IV.

Structure near to H_{c2} appears not to be due to the predicted vortex waves,[17] which were one of the reasons stimulating the present experiments. Instead, it is attributable partly to the frequently-observed peak effect[18] and partly to the change in vortex length as the field passes through H_{c2}. The latter, probably the same as an effect observed in the critical current measurements of Swartz and Hart,[19] is discussed in Sec. III B.

At fields above H_{c2}, the surface superconducting layer shields the interior from flux variations. The measured flux penetration is compared with the solutions of the Ginzburg-Landau equations of Fink and Kessinger[20] and of Maki[21] in Sec. III C. A detailed study of the screening effects of the sheath leads to conclusions which differ from some of those drawn by Alais and Simon[22] from their skin-effect experiments, as is discussed in Sec. IV. For instance, we find that the sheath *can* produce a shielding effect. A reason, in keeping with the present results, is suggested for the apparent existence in their experiments of vortices above H_{c2}.

The experiments on the shielding effect of the surface sheath led to similar experiments on cylindrical thin films. Comparisons have been made in Sec. III D with a naive model and with a thorough calculation by Pincus.[23]

Hysterisis effects have been reported in many experiments in the mixed state.[24] In Sec. III E we discuss briefly another mechanism that was suggested by an extensive study of these effects.[25]

II. EXPERIMENTAL METHOD

The penetration of radio-frequency flux was measured by the active method of Schawlow and Devlin.[26] The self-inductance of a coil wound around the sample governs the frequency of a Colpitts oscillator.[27] Changes in the flux penetration into the sample cause a change in the self-inductance and can be found from the consequent frequency changes. The change in the penetration depth (or inductive skin depth) $\delta\lambda$, is proportional to the change in the frequency of the oscillator, δf. The penetration depth at a field H, $\lambda(H)$, is found from the oscillator frequency at H, $f(H)$, by using the relation

$$\lambda(H)-\lambda(0)=[\lambda(H_{c3})-\lambda(0)]\times[f(0)-f(H)]/[f(0)-f(H_{c3})], \quad (1)$$

where $\lambda(0)$ is taken to be 4.4×10^{-6} cm for niobium.[28] For the most part, $\lambda(0)$ was negligible in comparison with $\lambda(H)$—except at the lowest fields. As is well known in this type of experiment,[28] absolute determinations of $\lambda(H)-\lambda(0)$ are limited in accuracy by the difficulty of estimating the geometrical factor $d\lambda/df$. Within the experimental error, the value of the normal-state resistivity deduced from the geometrical factor (estimated using the long solenoid

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[15] C. Caroli and K. Maki, Phys. Rev. **159**, 306 (1967).
[16] K. Maki, Phys. Rev. **141**, 331 (1966).
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[19] P. S. Swartz and H. R. Hart, Jr., Phys. Rev. **137**, A818 (1965).
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[21] K. Maki, Physics **1**, 21 (1964).
[22] P. Alais and Y. Simon, Phys. Rev. **158**, 426 (1967).
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[24] C. P. Bean, Phys. Rev. Letters **8**, 250 (1962); M. Cardona, J. Gittleman, and B. Rosenblum, Phys. Letters **17**, 92 (1965); D. G. Schweitzer, M. Garber, and B. Bertman, Phys. Rev. **159**, 296 (1967); D. G. Schweitzer and M. Garber, *ibid.* **160**, 348 (1967).
[25] D. E. Carlson and W. L. McLean, Bull. Am. Phys. Soc. **11**, 224 (1966).
[26] A. L. Schawlow and G. E. Devlin, Phys. Rev. **113**, 120 (1959).
[27] See, for instance, A. J. Cote and J. B. Oakes, *Linear Vacuum-Tube and Transistor Circuits* (McGraw-Hill Book Co., New York, 1961).
[28] B. W. Maxfield and W. L. McLean, Phys. Rev. **139**, A1515 (1965).

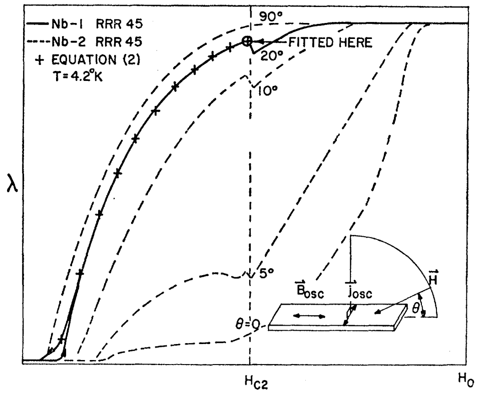

FIG. 1. The penetration depth in niobium, found from the change in the oscillator frequency using Eq. (1), plotted against the magnitude of the external field. The solid curves are for one sample, Nb-1. The dashed curves have been sketched on the same diagram but were obtained on a different sample, Nb-2, of approximately but not exactly the same residual resistance ratio and linear dimensions as Nb-1. They show (a) the dependence of λ on θ, and (b) the typical structure found in fields near to H_{c2}. The crosses were calculated using the classical skin effect theory and the assumption that the flux-flow resistivity was proportional to the flux-density in the plate [see Eq. (2)].

approximation and the measured radii of coil and core) and $f(0)-f(H_{c3})$ agreed with the room-temperature resistivity divided by the residual resistance ratio.

The frequencies usually used were between 1 and 5 MHz with the amplitude of the applied oscillating field between a few hundreths and several oersted. The effects of field-dependent losses on the oscillator (arising from the magnetoresistance of the coil or the surface resistance of the sample) were reduced by having a relatively large spacing between the windings of the coil and the sample. A frequency-to-voltage converter was used to display on an x-y recorder the difference-frequency between the Colpitts oscillator output and a fixed frequency standard. The x axis was driven by a voltage proportional to the strong steady magnetic field. The noise level was lower than the systematic errors, which were estimated to be less than 2%. The frequency meter was calibrated using a Hewlett-Packard frequency meter reading to about 10 Hz.

The magnetization of the specimens was measured by the method of Fietz,[29] which employs two series-opposed coils of the same number of turns. One of them contains the sample and when the external field is changed, a net emf is induced which is proportional to the rate of change of the magnetization of the sample. This emf can be amplified by an integrating circuit, which gives an output voltage proportional to the magnetization. Drift in the output voltage, owing to the internal drift of the amplifier and stray thermal emf's, was found to cause the output voltage to vary by as much as several millivolts in two minutes. The effect of the drift on the measurement of the surface sheath was minimized by sweeping small hysteresis loops from which the over-all magnetization curve could be determined.

Five niobium samples of varying purity and several indium-bismuth alloys were used in the experiments. The alloys were prepared from 99.999% pure indium and 99.999% pure bismuth. The surfaces of the alloys were left untreated since chemical polishing and electropolishing can alter the chemical composition near the surface.[30] Most of the samples were in the form of thin plates (typically $0.02\times5\times30$ mm^3), while the rest were circular cylinders (e.g., 5 mm in diameter and 30 mm long).

III. EXPERIMENTAL RESULTS AND THEIR ANALYSIS

A. Flux Penetration in Mixed State

Figure 1 shows how the penetration depth [obtained from the measured frequency using Eq. (1)] in a niobium sample typically depends on the magnitude and the orientation of the magnetic field. In this case, the steady field was always perpendicular to the oscillating currents on the large faces of the sample. The surface sheath had only a small effect when the strong field was inclined by more than a few degrees to the large faces of the plate samples. The results obtained with the oscillating currents on the large faces perpendicular to the vortices were easily interpreted in terms of flux flow,[10] as can be seen from the results for the specimen Nb-1 ($\theta=20°$) shown by the solid curve. The crosses on the graph were calculated from the classical skin effect theory assuming the bulk resistivity to be given by $\rho=\rho_n B/H_{c2}$, where B is the magnetic induction and ρ_n the resistivity in the normal state. This expression for ρ has been found from many dc measurements of the resistivity in the mixed state of different type-II superconductors at temperatures much less than the transition temperature and when there is no pinning.[10] We thus find for the penetration depth,

$$\lambda=\mathrm{Re}\left[\int_0^\infty \frac{b(z)\,dz}{b(0)}\right]=\left(\frac{\rho}{8\pi\omega}\right)^{1/2}=\tfrac{1}{2}(B/H_{c2})^{1/2}\delta_c\,, \quad (2)$$

where $b(z)$ is the complex oscillating magnetic induction at a depth z below the surface, and $\delta_c=(\rho_n/2\pi\omega)^{1/2}$ is the classical skin depth in the normal state. The induction was found from the external field using a correction (only important near H_{c1}) for the demagnetization effects. Similar agreement with the above expression for λ was obtained when the surface was plated with normal metal or when the steady field was perpendicular to the large faces, thus destroying the surface sheath. In our purest niobium specimen it was necessary to allow for the modifica-

[29] H. Fietz, Rev. Sci. Instr. **36**, 1621 (1965).

[30] D. E. Carlson and H. J. Blech, Bull. Am. Phys. Soc. **12**, 38 (1967).

tion of the skin effect by the Hall effect, as is discussed later in this section.

The pinning of vortices in the niobium samples appeared to become important as the amplitude of the oscillating magnetic field was reduced to small values. (The penetration depth obtained in the way described above was amplitude-independent at the highest levels of oscillation.) There was a close correspondence between these results and direct current measurements of ρ at current densities comparable with the amplitudes of the oscillating currents. The indium-bismuth alloys showed similar pinning effects—but even at the largest amplitudes of the oscillating fields. These were again corroborated by dc resistivity measurements. Since the pinning effects were so pronounced, most of the comparison of the results with theories has been restricted to the niobium samples—except in one case where the large pinning effects were useful in preventing flux flow (see later in this section).

The purest of our niobium specimens had a Hall angle at H_{c2} of about 36° (residual resistance ratio ~3000). In the normal state of pure metals in high magnetic fields, the Hall effect leads to propagating modes of electromagnetic waves—either helicon or Alfven waves.[31] The skin effect in such a case can be dealt with by the theory of Chambers and Jones,[32] which is sufficiently accurate for the present purpose. The inductive skin depth is then given by

$$\lambda=\tfrac{1}{2}[(1+u^2)^{1/4}\cos(\theta/2)]\delta_c,$$

where θ is the Hall angle, $u=\tan\theta=AH/\rho$, and A is

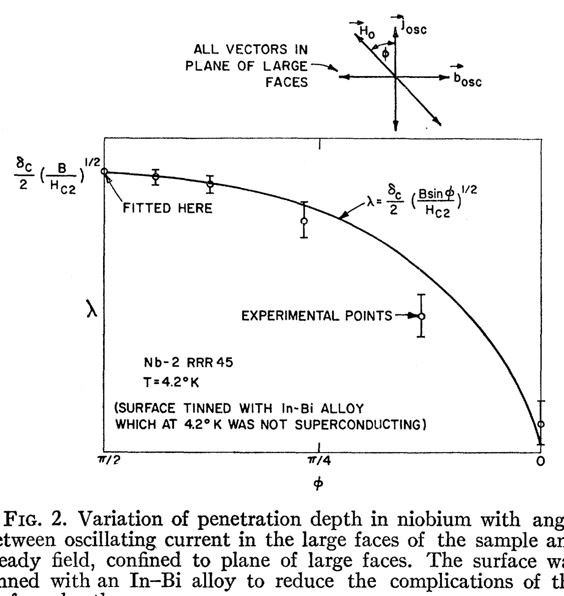

FIG. 2. Variation of penetration depth in niobium with angle between oscillating current in the large faces of the sample and steady field, confined to plane of large faces. The surface was tinned with an In–Bi alloy to reduce the complications of the surface sheath.

[31] See, for instance, S. J. Buchsbaum, in *Plasma Effects in Solids*, edited by J. Bok (Academic Press Inc., New York, 1965).

[32] R. G. Chambers and B. K. Jones, Proc. Roy. Soc. (London) **A270**, 417 (1962).

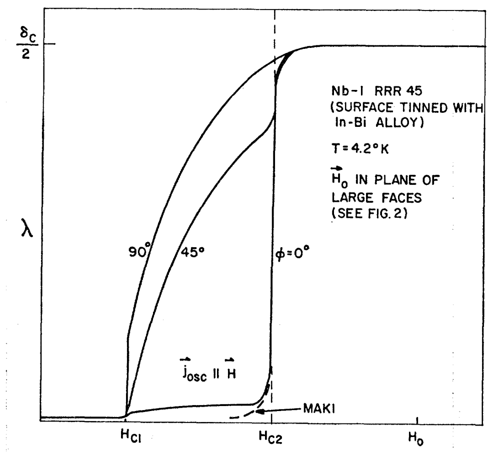

FIG. 3. Penetration depth in niobium, tinned with In–Bi, versus external field. ϕ is the angle between the oscillating currents in the large faces of the plate and the steady field. The dashed curve is predicted by the theories of Maki and of Caroli and Maki. The poor fit is due to vortex motion near the edges of the plate where flux emerges.

the Hall coefficient at field H. An attempt was made to use this equation to determine the tangent of the Hall angle in the mixed state. However, it was not possible from the results to distinguish between the predictions of the hydrodynamic models of Bardeen and Stephen[8] and of Nozières and Vinen.[8] The main obstacle was the small step near H_{c2}, the origin of which is discussed later.

As a simple check on possible systematic errors due to boundary effects, the penetration depth has been measured with different angles between the steady field (confined to the plane of the large faces of the sample) and the oscillating currents in the large faces. The power dissipation, and therefore the flux-flow resistivity, should be proportional to the cosine of the angle between the current and electric field, and thus to the sine of the angle ϕ between the current and steady magnetic field.[8] (It should be noted that the pinning forces become important when $\phi\sim 0$.) The results are shown in Fig. 2. Complications due to the superconducting surface sheath were reduced by tinning the surface of the niobium plate with an indium-bismuth alloy that was normal at 4.2°K.

A case of particular interest is that of the current parallel to the vortices ($\phi=0$), when no motion should be produced. The calculation by Maki[16] of the surface impedance Z very close to H_{c2}, gives

$$\lambda=\mathrm{Re}(-iZ/4\pi\omega)=[\hbar\beta(2\kappa_2^2-1)/4e(H_{c2}-H)]^{1/2}, \quad (3)$$

where $\beta=1.16$ for a triangular lattice of vortices, κ_2 is the Ginzburg-Landau parameter defined in terms of the slope of the magnetization curve at H_{c2}, and e the electronic charge (in cgs, emu). A discrepancy between the experimental results and this expression (see Fig. 3) is to be expected, since although the

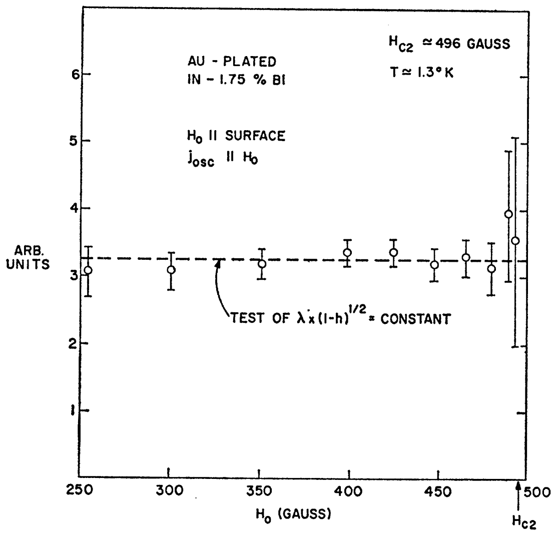

FIG. 4. Test of the field dependence of the penetration depth in In–1.75% Bi predicted by Maki and by Caroli and Maki. The oscillating currents in the large faces are parallel to the vortices. Strong pinning evidently stops the vortex motion that prevents such a test in the niobium samples.

current flow is parallel to the vortices on the large faces of the specimen, it is not so on the edges. The discrepancy was much smaller in gold-plated In–1.75% Bi, as can be seen in Fig. 4. Here we have plotted against field the quantity $\lambda[(H_{c2}-H)/H_{c2}]^{1/2}$ which, according to Eq. (3), should be independent of field. The reason for the discrepancy being less here than in the niobium specimens is probably that the severe pinning effects (found at the other orientations) prevented the vortices from moving at the edges of the specimen perpendicular to the steady field.

The theory of Caroli and Maki[15] also leads to Eq. (3) for the case where the oscillating currents flow parallel to the vortices, since, in this case, its most general results in the limit $\hbar\omega \ll kT$ reduce to those of Maki.[16] The results in Fig. 4 thus confirm one aspect of the theory of Caroli and Maki. In the case where the oscillating currents are perpendicular to the vortices, this theory predicts a greater penetration depth than in the other configuration. The experiments, on the other hand, show a much greater anisotropy, as can be seen from Fig. 3. The discrepancies are discussed later in Sec. IV.

B. Structure in Penetration Depth near H_{c2}

As the magnetic field is varied near H_{c2}, two different types of orientation-dependent features can be seen in the resulting variation of the penetration depth. We shall refer to these as the dip and the step. (See Fig. 1, $\theta=5°$.)

The dip appears to be due to the frequently observed "peak effect,"[19] which in the simplest case shows up in the flux-flow resistivity as a sharp decrease just before the (increasing) magnetic field reaches H_{c2}—at which value the resistivity finally rises quickly to its normal-state value. An explanation by Anderson and Kim[12] is that the vortices near H_{c2} interact so strongly that when one of them becomes pinned, the motions of many are hindered, thus reducing the amount of flux flow and, consequently, the flux-flow resistivity. This explanation is substantiated by the dependence of the dip on oscillating field amplitude and on the relative orientations of the induced currents and of the vortices. There seems to be no evidence that the dip is connected with wave propagation along the vortices.

The other feature—the step—occurs at H_{c2} for all orientations, except when the steady field is either parallel or perpendicular to the large surfaces of the sample (see Fig. 1). As the steady field is increased through H_{c2} the penetration depth falls abruptly by an amount that depends on the orientation and can be as much as one-tenth of its normal-state value. This effect appears to arise from the change in the effective length of a vortex as the field passes through H_{c2}, and consequently an increase in the effectiveness of surface pinning in preventing the vortex from moving. In the mixed state near H_{c2}, the oscillating currents penetrate to a depth approximately equal to $\frac{1}{2}\delta_c$, the penetration depth in the normal state. The approximate length of a vortex subject to the effect of the oscillating currents is $\delta_c/2\sin\alpha$, where α is the angle between the surface and the vortex. The driving force on the vortex in thus $(J\phi_0)\delta_c/2\sin\alpha$, where ϕ_0 is the flux quantum. At fields above H_{c2}, magnetic flux still penetrates the superconducting surface layer—of thickness $\sim\xi$, the coherence length. The experiments of Giaver[33] on induced flux flow in adjacent parallel films suggest that the current in the normal region below the surface sheath produces no driving force on the flux piercing the sheath. The driving force above H_{c2} would then be reduced to $J\phi_0\xi/\sin\alpha$, thus making the effect of surface pinning more effective. This explanation is also supported by the following observations: the disappearance of the step when the surface was coated with a normal metal (in order to reduce the sheath effect); a decrease in the step when the amplitude of the oscillating field was decreased until the pinning in the mixed state became important. It is probably also the reason for the step seen by Swartz and Hart[19] in the critical surface current which first produces a measurable resistance.

C. Penetration Depth above H_{c2}

The variation of the penetration depth with field above H_{c2}—found from the measured frequency shifts by the procedure described in Sec. II—can be seen from Fig. 1. Qualitatively, the results reflect the gradual reduction in the efficiency of the screening

[33] I. Giaver, Phys. Rev. Letters **15**, 825 (1965).

of the superconducting surface sheath as the field is raised—owing to the field dependence of the inhomogeneous superconducting order parameter in the sheath. We begin by assuming that the order parameter is unaffected by the small oscillating magnetic field—mainly because the amplitude of the oscillating induced current density was much less than the steady shielding current density. We defer considering whether the assumption is in fact true until the discussion in Sec. IV.

The simplest model describing the screening effects of the surface sheath is a plane superconducting layer, of constant thickness $\sim$ the coherence length ξ and uniform order parameter, superimposed on a semi-infinite normal region. In the superconducting layer it is assumed that London's equations hold, while in the normal region Ohm's law applies. The penetration depth is given by

$$\lambda=\mathrm{Re}\int_0^\infty \frac{b(z)dz}{b(0)}=\mathrm{Re}\left(\frac{[(q+k)+(q-k)e^{-2qd}]}{\{[(q+k)-(q-k)e^{-2qd}]q\}}\right), \tag{4}$$

where $q^2=4\pi n_se^2/m$, n_s is the density of superelectrons in the layer, $k=(1+i)/\delta_c$, δ_c is the classical skin depth of the normal material, and d is the thickness of the superconducting layer. Except immeasurably close to H_{c3}, the normal-electron current in the superconducting layer can be neglected at the frequencies used here.

The principal defect of the layer model is the assumption of a uniform superelectron density. A better description can be given in terms of the Ginzburg-Landau theory, where the superelectron density is described by $|\psi(z)|^2$, $\psi(z)$ being the order parameter at a depth z below the surface. A generalization of Eq. (4) can be obtained by integrating the Ginzburg-Landau equation $\partial^2A/\partial z^2=4\pi e^2|\psi(z)|^2A/m$, where A is the vector potential of the oscillating field. In this way it can be shown that, near to H_{c3}, λ is given by expanding the exponentials in Eq. (4) and replacing $q^2d=4\pi e^2n_sd/m$ by

$$4\pi e^2\int_0^\infty|\psi(z)|^2dz/m.$$

It is convenient[20] to define the "sheath thickness"

$$\zeta=\int_0^\infty|\psi(z)|^2dz/|\psi(0)|^2,$$

and the reduced order parameter $F(z)=\psi(z)/\psi_0$, where ψ_0 is the order parameter in the absence of a magnetic field. Both ζ and $F(0)$ have been obtained explicitly by Fink and Kessinger[20] from numerical solutions to the Ginzburg-Landau equations. Near H_{c3}, ζ is approximately equal to the coherence length ξ, but as the field is reduced below H_{c3}, it changes significantly. $|F(0)|^2$ is found to vary almost linearly with field between H_{c2} and H_{c3}. A similar field dependence follows from Maki's[21] extension of the microscopic approach of Gor'kov[3] to the Ginzburg-Landau theory. By combining Maki's expression for the gap parameter at the surface,[34]

$$|\Delta(0)|^2=[0.59eckT(H_{c3}-H)]/[\sqrt{2}\sigma\psi_2(\kappa_2^2-0.156)],$$

with the relation between the order parameter and the gap parameter,

$$|\psi(0)|^2=[m\sigma\psi_2|\Delta(0)|^2]/[\pi e^2hckT],$$

we find that

$$|F(0)|^2=[\sqrt{2}\kappa_1/(\kappa_2^2-0.156)][(H_{c3}-H)/H_{c3}],$$

where $\kappa_1=\xi/\lambda_0$, λ_0 is the penetration depth in weak fields, and κ_2 is related to the slope of the magnetization curve at H_{c2} by $(4\pi dM/dH)^{-1}=1.18(2\kappa_2^2-1)$. Not only is Maki's result restricted to fields near H_{c3}, but the whole approach is valid only in alloys with electronic mean free paths in the normal state

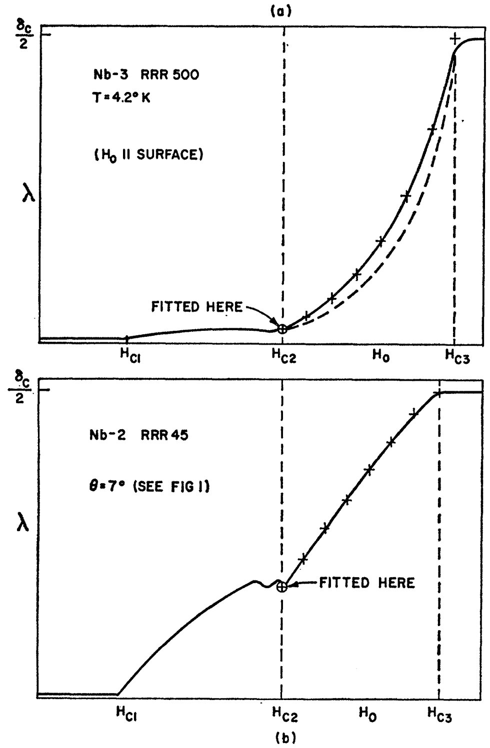

FIG. 5. Penetration depth in niobium versus field, between H_{c2} and H_{c3}. Solid curves are the experimental results. (a) The dashed curve is calculated from the simple layer model, assuming its thickness to be constant and that the uniform superelectron density varies linearly with field. The crosses take into account the thickness variation with field as calculated by Fink and Kessinger. The steady field was parallel to the large faces of the plate and perpendicular to the oscillating currents. (b) A similar comparison but with the field inclined at 7° to the surface. At greater inclinations there was evidently vortex motion in the sheath.

[34] Orsay Group on Superconductivity, in *Quantum Fluids*, edited by D. F. Brewer (North-Holland Publishing Company, Amsterdam 1966).

much less than the coherence length. This may be the reason why the coefficient of $(H_{c3}-H)$ in the expression for $|F(0)|^2$ is greater by a factor of 1.3 than the value of $(-d|F(0)|^2/dH)$ near to H_{c3} obtained from the numerical results of Fink and Kessinger.

In Fig. 5(a) we compare the measured λ at different fields with values obtained from Eq. (4). The dashed curve was obtained by assuming $\zeta=\xi$, and $|F(0)|^2$ to be proportional to $(H_{c3}-H)$. The constant of proportionality was fixed by fitting the theoretical expression to the experimental curve at the one point $H=H_{c2}$. The crosses in Fig. 5(a) were obtained in the same way, except that the values of ζ estimated by Fink and Kessinger were used instead of assuming $\zeta=\xi$.

This procedure for analyzing the experimental results also works well even when the steady field is inclined up to about 7° with the surface, as can be seen from Fig. 5(b). At larger inclinations the results were strongly amplitude-dependent and could not be fitted by Eq. (4). Apparently vortices in the sheath were almost completely pinned at small inclinations, but at larger inclinations it appeared as though flux flow was occurring. At the smaller inclinations the results were independent of amplitude.

It should be noted that in spite of the apparent agreement in Fig. 5 between experiment and theory, a large discrepancy is found between the values of $|F(0)|^2$ obtained by Fink and Kessinger and the values found from the experimental results (without fitting at H_{c2}) by using the values of ζ calculated by Fink and Kessinger in the generalized form of Eq. (4). In both cases $|F(0)|^2$ varies linearly with $(H_{c3}-H)$ but the slope from the theory (assuming $\kappa=\kappa_1=\kappa_2=1.0$) is 3.5 times the slope found from the combination of experiment and theory. The disagreement may be partly due to the difficulty of estimating the normal-state skin depth used in the scaling between frequency and penetration depth, and partly due to surface inhomogeneities lowering the local value of the order parameter. It must also be remembered that our formulation in terms of the "sheath thickness" ζ is valid only when $\lambda \gg \xi$. However, similar discrepancies between theory and experiment have been observed in low κ superconductors by Carlson and Bleck,[30] Rothwarf *et al.*,[35] and by Deutscher.[36]

When the steady field was confined to the plane of the large faces of the plate specimens, it was found that the flux penetration varied appreciably with the angle between the oscillating currents and steady field. But when the small-area faces over which the oscillating currents also flow become normal, a change in shielding occurs which is difficult to allow for in estimating quantitatively the effects due to the large faces.

D. Penetration Depth in Cylindrical Thin Films

The similarity between the surface sheath and a cylindrical thin film prompted us to measure the penetration depths of such films. Figure 6 shows the field dependence of the results for an In–2.0% Bi film of thickness 375 Å at 1.3°K. Measurements on bulk In–2.0% Bi between H_{c2} and H_{c3} had suggested that the electron density n_s in the uniform-layer model varied in proportion to $(H_{c3}-H)$. If this were also true for the film, one would expect $\lambda(H_{c3}-H)/H_{c3}$ to be independent of H, the external field, where H_{c3} is the upper critical field at which the transition to the normal state occurs. [When the film thickness d is much less than the London penetration depth $\lambda_L=(m/4\pi n_s e^2)^{1/2}$, the measured penetration depth $\lambda \sim \lambda_L^2/d \propto 1/n_s$.] On the other hand, Pincus[23] has predicted that it is $\lambda(H_{c3}^2-H^2)/H_{c3}^2$ that should be independent of field. This result is based on a solution to the Ginzburg-Landau equations, assuming that although the steady field can penetrate through the film, the oscillating field varies so rapidly that its fluxoid remains constant. As can be seen from the figure, there is better agreement with the naive model than with the theory. "Pulling" of the oscillator, owing to the large induced electric field in the film, made it impossible to analyze the results meaningfully between 700 G and $H_{c3}\sim$800 G.

Again, the comparison in Fig. 6 avoids using the geometrical factor in which there is a large experimental error—as mentioned in Sec. II.

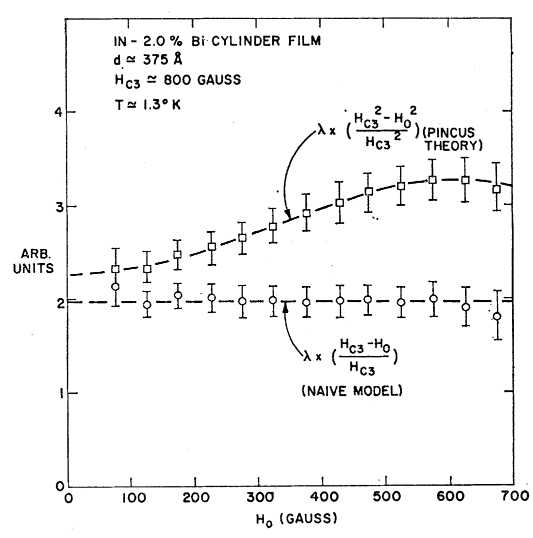

FIG. 6. A test of the field dependence of the penetration depth in a cylindrical thin film of In–2.0% Bi predicted by (i) Pincus's theory (ii) a simple model based on the observed field dependence of the order parameter in the sheath.

[35] A. Rothwarf, J. I. Gittleman, and B. Rosenblum, Phys. Rev. **155**, 370 (1967).
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E. Hysteresis in the Penetration Depth

Large hysteresis effects were often observed in the penetration depth measurements. Various mechanisms have been suggested by others as possible causes of hysteresis in measurements of the microwave surface resistance and of the magnetization: a difference between the external field and the field below the vortex-free surface layer owing to an induced current in that layer which reverses sign when the rate of change of external field is reversed[37]; an interaction of the vortices with the gradient of the steady magnetic field near the surface[38]; with the gradient of the order parameter in defect-free specimens[39]; with pinning centers[40]; or with their images in the surface boundary.[41]

A detailed examination of the hysteresis loops in thin cylindrical films between H_{c2} and H_{c3} has suggested yet another mechanism. This involves a current-driving force similar to the one which sets up the screening currents in a cylinder of a type-I superconductor when it is cooled through its transition temperature in the presence of a weak axial field. In that case, the Meissner-effect screening currents flow in a direction opposite to the direction in which a current produced solely by electromagnetic induction would flow. However, there is another mechanism that has been suggested by Park,[42] which also explains the observed results: the slight inhomogeneity of the order parameter, caused by the combination of the induced and the diamagnetic screening currents, is different in increasing and decreasing fields. The hysteresis effects were too sensitive to structure—metallurgical and surface conditions—to make it worthwhile analyzing them in any more detail.

IV. DISCUSSION

The flux penetration measurements in the mixed state have been interpretable mainly in terms of the bulk parameters of the medium—the flux-flow resistivity and the Hall coefficient. Like the microwave surface resistance measurements of Rosenblum and Cardona,[43] the present results agree well with the flux-flow model. On the other hand, apart from the case where the induced currents flow parallel to the vortices, the microscopic theory of Caroli and Maki[15] does not appear to apply—in spite of the response of the system being linear. Since the high-frequency theory[15] does not go over into the flux-flow theory as the frequency is reduced to zero, it thus appears that there is some, as yet unstated, criterion for the high-frequency theory to apply. In terms of the hydrodynamic picture, this might be supposed to be that the amplitude of the oscillations of the vortices be *much less* than their equilibrium separation, which near H_{c2} is approximately the coherence length. But the amplitude of the oscillations in the present experiments on the pure niobium near H_{c2} was at the most about 10^{-8} cm, while the coherence length was about 10^{-6} cm. Unless *much less* should mean many orders of magnitude less, it appears that the simple-minded criterion is not correct. Perhaps this should be expected, since the modes of oscillation of the theory do not correspond to a simple translational oscillation of the order parameter pattern.

The expressions calculated by Caroli and Maki[15] and by Maki[16] for the surface impedance in the mixed state, with the steady field parallel to the surface, do not take into account the strong screening effects of the vortex-free layer at the surface. It was for this reason that comparisons with the theory were made only for samples that had been coated with normal metal in order to try to destroy the order parameter at the surface. This, of course, is no guarantee that the vortices near the surface will behave as supposed in the theory. On the other hand, it seems likely that the field dependence of the order parameter near the surface in unplated samples will have as big an effect on the surface impendance as the variation of the bulk conductivity of the interior.

Another experimental difficulty in testing the theory was mentioned near the end of Sec. III A: the large effect due to currents on the smaller faces where vortex motion can occur, while on the large faces the oscillating currents are parallel to the vortices. It might be possible to devise an arrangement at microwave frequencies in which this difficulty was eliminated. A few that have been considered by the authors introduce other systematic errors that also seem difficult to eliminate. Clearly, any test of the anisotropy predicted by the theory can only be made if such effects are eliminated.

It was mentioned in Sec. III C that the data had been analyzed assuming that the order parameter was unchanged by the presence of the rf field. The theoretical work of Caroli and Maki[15] shows that this should not be the case in the mixed state, and that it is unlikely to be true whenever the order parameter has a spatial variation—as is the case near the surface in fields between H_{c2} and H_{c3}. It would thus be interesting to compare the present results with a basic theory dealing with this field region, but again it would be useful to have a clear

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criterion for its applicability. As mentioned in Sec. III C, there are practical difficulties in testing the dependence on the angle between the oscillating currents and the steady field, owing to the normal regions at the surface where the flux emerges.

As we have already mentioned in the Introduction, it is possible to reinterpret some of the results of the skin effect measurements of Alais and Simon[22] in the light of the "step" in Sec. III B and the inclination and amplitude dependences discussed in Sec. III C. The screening or shielding by the surface sheath may not be appreciable if the surface is rough, or if the field is not aligned parallel to it and the flux pierces the sheath in the form of vortices. Provided the amplitude of the oscillating field is sufficiently large to overcome pinning, the oscillating flux can easily move the vortices along the sheath—thus enabling flux to pass from above the sheath to below, in the manner found by Boato *et al.*[44] If, however, the pinning is strong or the surface smooth, and the field accurately aligned parallel to it, small amplitude oscillating fields do not cause any flux flow, and large shielding effects can be observed. It seems that if vortices exist above H_{c2}, they must be confined to the sheath. However, if they move, they can give rise to the appearance of vortex motion similar to that observed below H_{c2}.

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Anomalous Thermal Conductivity of Superconductors due to Impurity Spin Ordering

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It is shown that spontaneous and also a magnetic-field-induced ordering of the paramagnetic impurity spins in superconductors gives rise to an anomalous temperature dependence of the electronic thermal conductivity.

ONE of the most important aspects of a study of superconductors containing paramagnetic impurities is the problem of the coexistence of superconductivity and magnetic ordering. There exists now some experimental and theoretical evidence for the coexistence of superconductivity and impurity spin ordering.[1–3] It is the purpose of this paper to show that the electronic thermal conductivity should strongly reflect the coexistence of superconductivity and magnetic ordering, possibly as strongly as the specific heat, and to stimulate experimental studies bearing upon the theoretical predictions presented here. The anomalous dependence of the superconducting transition temperature on the concentration of paramagnetic impurities,[1,2] which has been explained as resulting from spontaneous ordering of the impurity spins,[4] directly reflects the coexistence of superconductivity and impurity spin ordering only at temperatures close to the superconducting transition temperature. However, a study of the electronic thermal conductivity and the specific heat,[5,6] for example, should clearly reflect the coexistence of superconductivity and magnetic ordering at all temperatures below the superconducting transition temperature.

If the paramagnetic impurity spins order, then the theory by Gor'kov and Rusinov,[3] using a static *s-d* exchange coupling between the conduction electrons and the paramagnetic impurities, needs to be extended by using a dynamic *s-d* exchange coupling. The dynamics of the *s-d* exchange coupling become very important if the superconducting transition temperature

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