ory due to Eliashberg.[10] The time dependence of the theory for a uniform superconductor has already been verified in some detail.[5,6] In addition we have a verification of (6), namely that tunneling measures the Green's function at the tunneling surface.

We wish to thank P. W. Anderson for valuable discussions and an essential suggestion, D. E. Thomas for design of equipment which made the derivative measurement possible, L. Kopf for junction fabrication, and Miss D. R. Margel for the electron-diffraction measurement.

Note added in proof.—It has been brought to our attention that P. D. de Gennes and D. Saint-James [Phys. Letters 4, 151 (1963)] have solved a theoretical model similar to that considered here (but for energies less than Δ) and, indeed, oscillations are quite apparent in their results. We have not succeeded in making a sufficiently thick, clean Ag film to observe the oscillations at these low energies.

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EFFECT OF ELECTRON-SPIN PARAMAGNETISM ON THE CRITICAL FIELD OF THIN Al FILMS*

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We report measurements of the critical field parallel to the plane of ~100-Å Al films, through the field region where spin paramagnetism limits the critical field. The question of electron-spin polarization and superconductivity is of great importance because of the absence of a general explanation of the measurements of the Knight shift (the shift in the nuclear resonance frequency due to the additional field produced by the net electron polarization). The Knight-shift measurements[1] in superconducting tin, mercury, and aluminum yield values at 0°K that are about $\frac{2}{3}$ of the normal-state values. On the basis of the BCS theory where there is complete spin pairing at 0°K, one would expect zero Knight shift at 0°K if only spin polarization effects were important. Various explanations advanced to explain the Knight-shift measurements, including some which do not rely only on spin polarization, have been discussed by Schrieffer.[1] The present measurements yield information about the spin polarization, through its effect on the critical field, and thus complement the Knight-shift measurements.

The effects of electron paramagnetism on the upper field of superconductors were treated by Chandrasekhar[2] and Clogston[3] for the limiting case of a superconductor showing complete field penetration, no Meissner effect, at 0°K. The Clogston[3] calculation, which compares the paramagnetic energy of the normal metal in a field to the gap energy of a superconductor with no Meissner effect, gives the upper limit of the critical field. We have made a simple extension of the Clogston argument to a film at 0°K that does show diamagnetism. From this calculation and our data we can estimate a value for the upper paramagnetic limited critical field for the "no Meissner effect" case.

In the discussions to follow we discuss our data in terms of the simple two-fluid temperature dependence[4]; this temperature dependence is used for convenience and because better theoretical estimates such as those of de Gennes and Tinkham[5] and Maki[6] do not, for our purposes, differ too significantly from this depen-

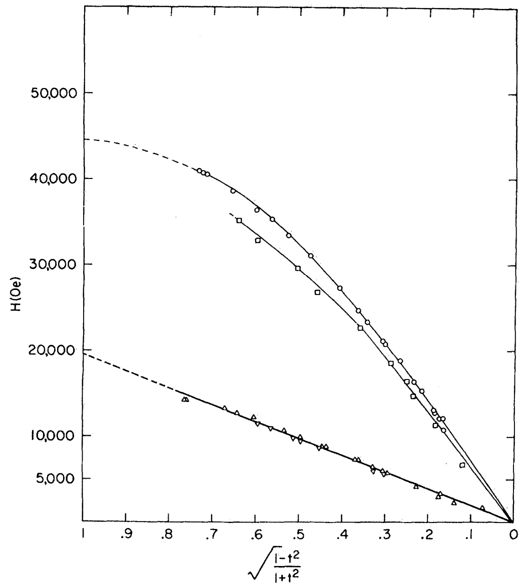

FIG. 1. Critical field versus the two-fluid temperature dependence. This dependence is given by $[(1-t^2)/(1+t^2)]^{1/2}$. ○: ~100-Å film, $T_c=2.23$°K, $H_c{}^*(0)=44\,500$ Oe, $H_c(0)=77\,000$ Oe; □: ~100-Å film, $T_c=1.99$°K, $H_c{}^*(0)=43\,000$ Oe, $H_c(0)=64\,000$ Oe; ∇: 250-Å film, $T_c=1.76$°K data taken in superconducting magnet; Δ: 250-Å film, $T_c=1.76$°K data taken in Varian magnet. $H_c{}^*(0) \sim H_c(0) \sim 19\,500$ Oe.

dence.

The following observations are evident from the data. (1) The thick film (~250 Å, $T_c \sim 1.75$°K) obeys the two-fluid dependence. This is to be expected since its 0°K critical field of about 19 000 Oe is well below the Clogston value of about 35 000 Oe and the critical field is thickness and mean-free-path limited. Agreement with the two-fluid model is indicated in Fig. 1, by a linear dependence of $H_c{}^*$ on $[(1-t^2)/(1+t^2)]^{1/2}$. (2) At the low temperatures, where T/T_c is near 0.5, the critical fields of the 100-Å films deviate significantly from a linear dependence on the plot in Fig. 1, indicating that the two-fluid temperature dependence is not obeyed in this region.

In Fig. 2(a), $[H_c{}^*(t)/H_c{}^*(0)]^2$ vs T/T_c is plotted. $H_c{}^*(t)$ is the measured[7] critical field at temperature T. $H_c{}^*(0)$ is the "measured field" at 0°K, obtained from Fig. 1 by extrapolating to 0°K the data at the lowest temperatures where T/T_c was about 0.5. For the 100-Å films this plot illustrates the large deviation from the two-fluid dependence at all temperatures, when $H_c{}^*(0)$ is used in the denominator of the above ratio. The data are also far above the other dependences.[5,6] Note that the 250-Å film val-

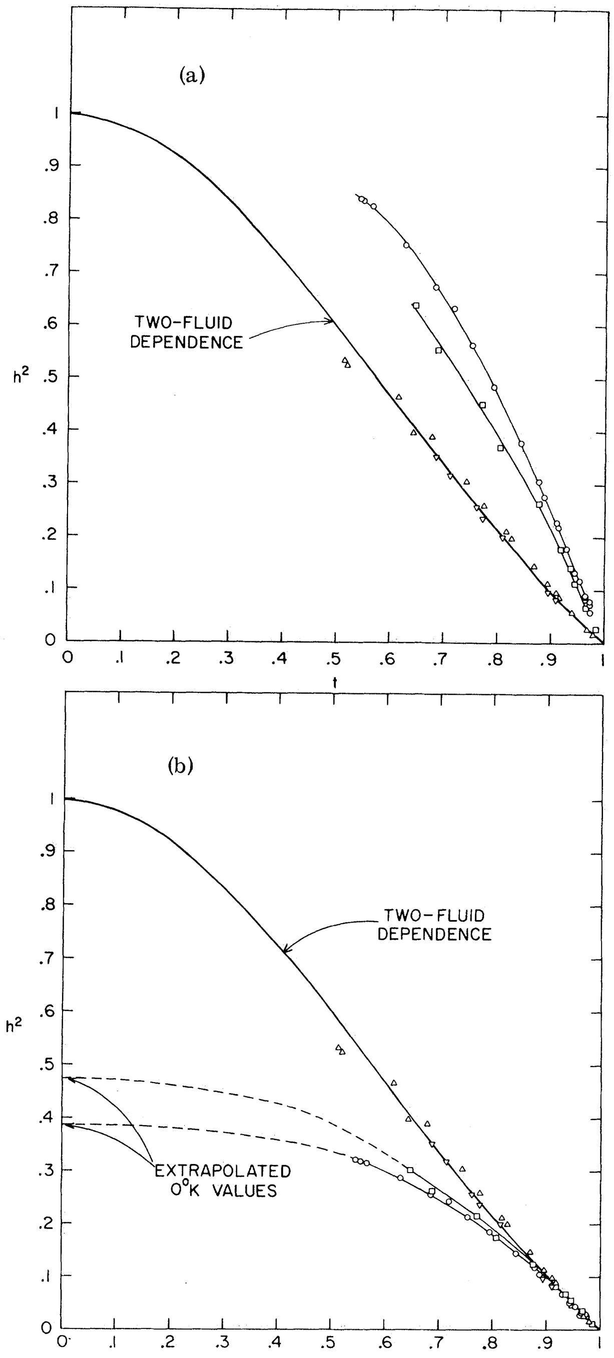

FIG. 2. (a) h^2 vs t where $h \equiv H_c{}^*(t)/H_c{}^*(0)$, t is the reduced temperature, and $H_c{}^*(t)$ is the measured field. (b) h^2 vs t where $h=H_c{}^*(t)/H_c(0)$, $H_c(0)$ is the critical field at 0°K in the absence of paramagnetism, t is the reduced temperature, and $H_c{}^*(t)$ is the measured field. ○: ~100-Å film, $T_c=2.23$°K, $H_c{}^*(0)=44\,500$ Oe, $H_c(0)=77\,000$ Oe; □: ~100-Å film, $T_c=1.99$°K, $H_c{}^*(0)=43\,000$ Oe, $H_c(0)=64\,000$ Oe; ∇: 250-Å film, $T_c=1.76$°K data taken in superconducting magnet; Δ: 250-Å film, $T_c=1.76$°K data taken in Varian magnet. $H_c{}^*(0) \sim H_c(0) \sim 19\,500$ Oe.

ues fall close to the two-fluid dependence.

In Fig. 2(b), $[H_c*(t)/H_c(0)]^2$ vs T/T_c is plotted, where $H_c(0)$ is the critical field at 0°K in the absence of paramagnetism. $H_c(0)$ is obtained by extrapolating to 0°K the linear region near T_c of the curve of $H_c*(t)$ vs $[(1-t^2)/(1+t^2)]^{1/2}$ in Fig. 1. This linear region near T_c indicates agreement with the two-fluid temperature dependence and was also assumed to indicate negligible paramagnetic limiting near T_c.[8] Hence the extrapolation of this linear part to 0°K is assumed to yield $H_c(0)$, the critical field in the absence of paramagnetism. If there were still paramagnetic limiting near T_c our $H_c(0)$ values would be too small. In the plot in Fig. 2(b) it is evident that the 100-Å film data agree with the two-fluid dependence at the higher temperatures near T_c and then fall far below this dependence at the lower temperatures where T/T_c is near 0.5. The large deviations from the two-fluid and the other models[5,6] at the lower temperatures are taken as evidence for the effect of electron paramagnetism on the upper critical field at these temperatures. Paramagnetic effects should be expected in this region since the measured fields are of the order of the paramagnetic-limited critical field estimated from Clogston's formula[9] that $H_p(0) = 18\,400 T_c$ Oe. We emphasize again that in the 250-Å film where the effects of paramagnetism are unimportant, agreement is obtained with the two-fluid dependence.

To find a value for $H_p(0)$ from our measurements we refer first to Fig. 3. This figure qualitatively shows the energy balance between the paramagnetic normal metal and the superconducting film in a field and shows the relationship of "measured field" $H_c*(0)$ to the other fields $H_c(0)$ and $H_p(0)$. In the argument to follow we try to give some idea of how the measured field can be related to the upper paramagnetic field. We follow Clogston's argument except that we add an energy term due to the diamagnetism of the film. For simplicity we assume a superconducting film susceptibility, χ_d, that is constant with field. We have then at 0°K

$$0.5\chi_p H_c*(0)^2 = H_{cB}(0)^2/8\pi - 0.5\chi_d H_c*(0)^2. \quad (1)$$

We emphasize that the above equation assumes complete antiparallel pairing and constant gap parameter at 0°K until the transition field $H_c*(0)$. Hence this is the case of a first-order phase transition. χ_p is the paramagnetic spin susceptibility of the normal metal and H_{cB} is the thermodynamic critical field.

Following Clogston we define $H_p(0)$ by

$$0.5\chi_p H_p(0)^2 = H_{cB}(0)^2/8\pi = 0.5N(0)\epsilon(0)^2. \quad (2)$$

This is the case for a film with thickness approaching zero. For this case

$$H_p(0) = \epsilon(0)[N(0)/\chi_p]^{1/2}.$$

We also have that

$$0.5\chi_d H_c(0)^2 = H_{cB}(0)^2/8\pi = 0.5N(0)\epsilon(0)^2. \quad (3)$$

$H_c(0)$, as mentioned previously, is the critical field at 0°K in the absence of paramagnetism. By combining Eqs. (2) and (3) with (1), we get

$$H_c*(0)^2 = H_p(0)^2[1 - H_c*(0)^2/H_c(0)^2];$$

thus

$$H_c*(0) = H_p(0) H_c(0)[H_c(0)^2 + H_p(0)^2]^{-1/2}. \quad (4)$$

By using this formula we can solve for H_p by using the extrapolated $H_c*(0)$ and $H_c(0)$ values of 44 500 and 77 000 Oe, respectively, for the 2.23° film.[10] We obtain for this film that $H_p \sim 54\,500$ Oe which is ~35% above Clogston's result that $H_p(0) = 18\,400 T_c$ Oe. Professor M. Tinkham has indicated to us that the Ginzburg-Landau theory can be used in a simple form to obtain an expression for $H_p(0)$. In this case one says that $\chi_d = \chi_d(H=0)(1 - H_c*^2/H_c^2)$ and the diamagnetic energy is given by $\int \chi_d H dH$. The diamagnetic energy contribution is then calculated to

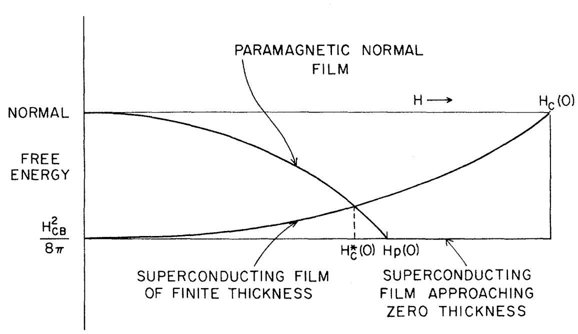

FIG. 3. Gibbs free energy of a film at 0°K versus magnetic field. $H_c(0)$ is the critical field in the absence of paramagnetism at 0°K, $H_c*(0)$ is the "measured field" at 0°K, and $H_p(0)$ is the paramagnetic-limited critical field at 0°K.

be $(H_{cB}{}^2/8\pi)(H_c{}^*/H_c)^2[2-(H_c{}^*/H_c)^2]$, and an analysis similar to the one we have given above yields that $H_c{}^*(0)=H_p(0)[1-H_c{}^*(0)^2/H_c(0)^2]$. In our formula $[1-H_c{}^*(0)^2/H_c(0)^2]$ entered as a square root. Tinkham indicates that his formula and the data give $H_p(0)\sim 67\,000$ Oe, and points out that this field is consistent, within experimental errors, with the Knight-shift measurements and a χ_{ps} in the superconducting state that is about $\frac{2}{3}$ as large as the normal state χ_p. This use of the Ginzburg-Landau theory is an improvement over the more elementary analysis above. Dr. Arthur Paskin has mentioned to us that a further improvement can be made on using the Ginzburg-Landau expression for χ_d, by also adding a term of the form $\int\chi_{PS}(H)HdH$ to the right-hand side of Eq. (1) to describe the loss in energy of the superconducting state due to the paramagnetism of the normal electrons which are created as the field is increased. We also mention that while the Ginzburg-Landau theory is quantitatively correct near T_c, small numerical factors may enter into extensions to 0°K.[11]

An important point to remember in considering the data is the effect of surface scattering in these disordered films.[12] Ferrell[13] first suggested that spin reversal might occur by boundary scattering, through spin-orbit coupling, in the case of a system composed of small particles.[14] This then leads to spin paramagnetism in the superconducting state and would then explain the Knight-shift results. It is also easily seen that finite spin paramagnetism in the superconducting state would add a term $0.5\chi_{ps}H_c{}^*(0)^2$ to the right-hand side of Eq. (1). This would then raise the estimate of $H_p(0)$. For instance an $H_p(0)$, determined from Eq. (4), 35% above Clogston's result, might be explained by a χ_{ps} of near one-half the normal χ_p.

In concluding, we emphasize that the data together with the simple models described in this paper lead to $H_p(0)$ values that are from 35% higher than the Clogston value up to about 1.7 times the Clogston value, depending on the model. Hence, although the details must rely on a better theory, it is probably safe to say that there is a significant paramagnetic susceptibility in the superconducting state of these Al films. It is also clear that experimental values of χ_p in normal Al are needed to relate the present results to the pairing in the superconducting state.

We thank Mr. George Hrabak for expert assistance in the construction of the apparatus and A. Ingraham for the preparation of the thin-film samples. We are greatly indebted to Professor M. H. Cohen for enlightening discussions and suggestions on the interpretation of these results and their relation to the Knight-shift measurements. We also thank Dr. Arthur Paskin for advice and discussions on this work, Professor Kazumi Maki for discussions on his work, and Professor M. Tinkham for pointing out improvements in the manuscript and allowing us to include his analysis in this paper.

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[2]B. S. Chandrasekhar, Appl. Phys. Letters 1, 7 (1962).

[3]A. M. Clogston, Phys. Rev. Letters 9, 266 (1962).

[4]The Gorter-Casimir two-fluid dependence is obtained from the dependence of $H_c(t)$ on the penetration depth and the bulk critical field, and is given by $[(1-t^2)/(1+t^2)]^{1/2}$. This formula is discussed in Ref. 5. D. H. Douglass and R. H. Blumberg [Phys. Rev. 127, 2038 (1964)] also discuss another dependence in the Ginzburg-Landau free-energy expression. The agreement of the 250-Å film data with $[(1-t^2)/(1+t^2)]^{1/2}$ excludes this other expression.

[5]P. G. de Gennes and M. Tinkham, Physics 1, 107 (1964).

[6]K. Maki, Progr. Theoret. Phys. (Kyoto) 31, 731 (1964).

[7]The $H_c{}^*$ values were obtained by taking the half-way point for the ac resistance-versus-field transition at a given temperature. T_c was defined by the half-way point of the temperature transition in zero field. The T_c transition was normally about 0.1°K wide and the field transition was normally about 7500 Oe wide.

[8]K. Maki [Physics 1, 127 (1964)] has shown that the effect of electron paramagnetism in type-II superconductors becomes negligible near T_c. The linear region near T_c, in Fig. 1, in the 100-Å films probably indicates that this is true in the thin-film case. The behavior of films near T_c has been investigated, for example, by D. H. Douglass, Jr., and R. H. Blumberg, Phys. Rev. 127, 2038 (1964).

[9]Clogston obtained this value by setting

$$H_p=\epsilon(0)[N(0)/\chi_p]^{1/2}=\epsilon(0)[N(0)/2N(0)\mu_B{}^2]^{1/2}=\epsilon(0)/\sqrt{2}\mu_B.$$

[10]It should be mentioned that errors may be involved in the extrapolations to obtain $H_c{}^*(0)$ and $H_c(0)$. The slope near T_c in Fig. 1 is, of course, very sensitive to the value of T_c and we estimate that there may be an error of about 5% in the extrapolation to 0°K to ob-

tain $H_c(0)$.

[11]First-principle attempts at describing the effects of paramagnetism have been made by K. Maki [Physics 1, 127 (1964)], who has investigated the effect of paramagnetism on H_{c2} for type-II superconductors at all temperatures, for the case of a small gap parameter and weak paramagnetic limiting. For this case Maki gets

$$H_{c2}(0)^* = H_{c2}(0)H_p(0)[2H_{c2}(0)^2 + H_p(0)^2]^{-1/2}.$$

Because of the restrictions of small gap parameter and weak paramagnetic limiting, as mentioned above, it is not clear whether the experimental results can be described by this analysis.

[12]One must keep in mind that the Al films we have used were very disordered. In the 100-Å films the mean free path was estimated to be about 20 Å.

[13]R. A. Ferrell, Phys. Rev. Letters 3, 262 (1959).

[14]K. Maki has indicated that he and others have now considered the problem of the critical field in the presence of a strong Pauli effect as well as spin-orbit scattering.

FLUX PINNING AND FLUX-FLOW RESISTIVITY IN MAGNETICALLY COUPLED SUPERCONDUCTING FILMS

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It has recently been shown that when two superconducting films are placed sufficiently close together they become magnetically coupled[1]; i.e., if a dc current is passed along one film such that it enters a resistive yet superconducting state, a dc current and a dc voltage may be induced along the other film. This effect apparently takes place whether the films are in the mixed state[1] or in the intermediate state.[2] In this Letter, I wish to report some further observations on such a system; and, in particular, effects related to the concepts of flux pinning and flux-flow resistivity in type-II superconductors.

The appearance of a voltage in a type-II superconductor has been associated with the motion of quantized flux vortices (fluxons) perpendicular to the current direction.[3] A very simple criterion for the motion of fluxons has been established by Kim, Hempstead, and Strnad[4] who consider the average forces per unit length acting on one fluxon,

$$\eta v_L = (\varphi_0/c)J - F_p. \tag{1}$$

$(\varphi_0/c)J$ is the Lorentz force acting on the fluxon, where φ_0 is the flux quantum, c the velocity of light, and J the current density. F_p is the so-called pinning force. The pinning force is generally associated with lattice defects which, in effect, form energy barriers and tend to trap the fluxons. When the Lorentz force exceeds the pinning force, the fluxons start to move through the lattice with a velocity v_L. This motion is thought of as a viscous flow, and the fluxons will be subjected to a third force ηv_L where η is the viscosity coefficient. The observed electric field E along a type-II superconductor is taken to be proportional to v_L:

$$E = \frac{n\varphi_0 v_L}{c} = \frac{Bv_L}{c}, \tag{2}$$

where n is the fluxon density. By combining Eqs. (1) and (2), the flux-flow resistivity ρ_f is obtained:

$$\rho_f = dE/dJ = \varphi_0 B/c^2\eta. \tag{3}$$

Unfortunately, these simple formulas are not directly applicable to thin films and, in particular, not when the applied magnetic field is zero. (In most experiments dealing with type-II superconductors, the applied field is much larger than the self-field from the transport current.) In this Letter, I am mainly concerned with how the coupling between two films affects the flux pinning and the flux-flow resistivity. Thus, I shall rely upon analogous equations, even though they may not be valid in detail.

The samples were prepared by vacuum-depositing a film of tin onto a microscope glass slide, then insulating it with a thin layer of silicon oxide,[5] and finally depositing a film of tin on the top. The bottom Sn film is referred to as the primary; the top Sn film, which is narrower than the primary, is referred to as