Discussion 48

MEISSNER: (Discussing first two papers). We have looked at the complete transition in pulsed currents for a small strip over a ground plane and it turned out that, even at currents about 4 or 5 times the current which restores half the normal resistance, we find that the thin film still is partially superconducting. Now a remark to the last paper (Mitescu's). I think one has to be very careful. You will always get some field lines breaking through the film. I do not believe that you really can have your field lines truly all parallel to the film but that some will break through and you will have the effects which Tinkham observed and described as flux inclusions. It's not quite clear that these are necessarily always resistanceless if you get this breakthrough in an alternating field.

C. D. MITESCU, *California Institute of Technology:* The fact that the persistent currents which have been observed in stationary d.c. conditions are comparable to our critical currents makes the presence of ordinary, normal resistance rather unlikely. I don't think that flux leakage could account for the observed very sharp exponential dependence on the current.

MEIKLEJOHN: I might make a remark on one of Meissner's remarks. It depends on just how sharp your transition is as to whether or not this resistance at which you make your measurement is important. We did not find any difference in our temperature dependence if we went down to a tenth of this voltage sensitivity which would be a different resistance level.

BEAN: I'd like to ask Dr. Mitescu if he has checked for the absence of static hysteresis in the magnetic properties of his ring when he has less than complete penetration. The reason I ask this is, if I understand the theory, there is no hysteresis at low frequencies because the Ginzburg–Landau theory does not have strong inertial terms. If one does have hysteresis due to the irreversible or partially irreversible motion of flux lines as Professor Meissner mentioned, then one would generate harmonics in the sense that I showed in my talk on the first day.

MITESCU: There is no hysteresis in going above the critical transition or coming down through it. One point which might argue against the flux line motion is that, if I remember rightly, in Dr. Bean's paper the dependence in his model was quadratic in the amplitude. I think one would expect some kind of power law in the case of flux line motion, unless it is a very strange kind of motion.

DOUGLASS: I have a question to Dr. Meiklejohn. In regard to the exponents on Δt, I noticed some of your points were very close to the transition temperature, a millidegree or thereabouts. These points strongly influence the slope of these curves and any small error in the transition temperature could appreciably change the value of the exponent. I'd like to know what the width of the transitions were.

MEIKLEJOHN: It is true that the actual magnitude of the critical temperature can influence these slopes. However, we were as high as 50 mdeg in our measurements and the points fartherest away from the critical temperature were not greatly influenced by changing slightly the critical temperature. The widths of the transition varied from about 4 mdeg to about 16 mdeg in the various films that were measured.

Temperature Dependence of the Critical Fields of Thin Superconducting Films

ARNOLD M. TOXEN

International Business Machines Corporation, Thomas J. Watson Research Center, Yorktown Heights, New York

I. INTRODUCTION

To interpret and understand critical magnetic field measurements on superconducting films, a theory is needed which includes both strong-field effects and nonlocal effects–strong field, to describe phenomena occurring at the critical field; nonlocal, to adequately describe thickness and mean free path effects.

In a previous paper[1] (hereafter referred to as I), a simple strong-field, nonlocal theoretical model was presented which relates the critical magnetic fields of superconducting films to the kernel of the current–vector–potential relationship for any theory of superconductivity. In I, the theoretical model was worked out in detail for the Pippard nonlocal kernel with specular boundary conditions in the superconductive state. Comparison of the theoretical model to critical field data for pure indium films indicated that the theoretical model predicted quite well the thickness dependence of critical field at temperatures near the critical temperature. In a second paper[2] (hereafter referred to as II), the theoretical model was compared to critical field measurements made on dilute alloy films of indium containing up to 5 at.% tin. In II, it was shown that the theory using the Pippard kernel predicted quite well the variation of critical field with mean free path at temperatures near T_c.

In this paper, we review the theoretical model and

[1] A. M. Toxen, Phys. Rev. **127**, 382 (1962).

[2] A. M. Toxen and M. J. Burns, Phys. Rev. **130**, 1808 (1963).

present new theoretical and experimental results relating to the temperature dependence of critical field. In particular, we show that although the theoretical model using the Pippard kernel describes the observed temperature variation of critical field approximately, there exist systematic discrepancies between experiment and the predictions of the theoretical model. We also show that preliminary calculations indicate that the discrepancies are at least partly resolved if the theoretical calculations are carried out with the BCS kernel rather than the Pippard kernel.

II. THEORETICAL MODEL

For films thin enough so that the order parameter ψ_0 can be considered constant over the thickness of the film, Eqs. (61) and (62) of Ginzburg–Landau[3] give the following expressions for the film critical field:

$$(h_c/H_c)^2 = \psi_0^2(2 - \psi_0^2)/[1 - (1/\eta)\tanh\eta]\,, \quad (1)$$

and

$$(h_c/H_c)^2 = [4\psi_0^2(\psi_0^2 - 1)\cosh^2\eta]/[1 - (1/2\eta)\sinh 2\eta]\,, \quad (2)$$

where $\eta \equiv \psi_0\, a/\delta_0$. The quantity a is the film half-thickness, h_c is the film critical field, H_c is the bulk critical field, and δ_0 is the weak field penetration depth. For $h_c/H_c > 1$, Eqs. (1) and (2) can be solved simultaneously to yield h_c/H_c as a function of δ_0/a, i.e.,

$$h_c/H_c = F(\delta_0/a)\,, \quad (3)$$

where $F(x)$ is a function which is plotted in Fig. 1 of I. From (66) of Ref. 3, we obtain a relationship between the film susceptibility in a weak magnetic field and the weak field penetration depth.

$$\kappa/\kappa_0 = 1 - (\delta_0/a)\tanh(a/\delta_0)\,, \quad (4)$$

where κ is the film susceptibility in a weak magnetic field and κ_0 is the weak field bulk susceptibility. From (3) and (4) we can obtain a relationship between the film critical field and weak field susceptibility which is of the form

$$h_c/H_c = G(\kappa/\kappa_0)\,, \quad (5)$$

where G is a function which can be numerically evaluated and is plotted in Fig. 2 of I.

The weak field susceptibility can in turn be related to the appropriate nonlocal parameters by means of a calculation due to Schrieffer.[4] Schrieffer, for the case of specular reflection in the superconducting state, obtained the following expression for the weak field susceptibility of a film:

$$(\kappa/\kappa_0)_{\text{spec}} = 1 - (2/a^2)\sum_{n=0}^{\infty}[k_n^2 + K(k_n)]^{-1}\,, \quad (6)$$

where $k_n = (2n+1)\pi/2a$ and $K(k)$ is the kernel obtained from the relationship between the supercurrent density and the vector potential in wave-vector space. If one takes the Fourier transform of the expression for the supercurrent density in the gauge $\nabla\cdot\mathbf{A} = 0$, then one obtains a relationship of the form

$$(-4\pi/c)\mathbf{j}(k) \equiv K(k)\mathbf{A}(k)\,, \quad (7)$$

where $\mathbf{j}$ and $\mathbf{A}$ are the current density and vector potential, respectively. Relation (7) then defines the kernel $K(k)$. For the Pippard and BCS theories $K(k)$ can be expressed as[5]

$$K(k) = \frac{3}{4\pi\xi_0\lambda_L^2(T)}\int_0^{\infty}\int_0^{\pi}\int_0^{2\pi}\sin^3\theta\sin^2\phi e^{-R/l} \times e^{ikR\cos\theta}J(R,T)d\phi d\theta dR\,, \quad (8)$$

where ξ_0 is the coherence distance in pure material, λ_L is the London penetration depth and l is the electronic mean free path in the normal state. For the Pippard theory,[6] the expression for $J(R,T)$ is

$$J_P(R,T) = \exp(-R/\xi_0)\,. \quad (9)$$

For the BCS theory, as modified by Mattis and Bardeen[7] to include impurity scattering,

$$J_{BCS}(R,T) = \frac{2\lambda_L^2(T)\epsilon_0^2}{\pi\epsilon_0(0)\lambda_L^2(0)}\int_0^{\infty}\left\{\frac{1-2f(\epsilon_0)}{\epsilon_0} - \frac{1-2f(E)}{E}\right\}\frac{\sin 2\alpha\epsilon}{\epsilon}d\epsilon\,, \quad (10)$$

where ϵ_0 is the temperature-dependent energy gap, α is equal to $R/\hbar v_F$, v_F is the Fermi velocity, $f(x)$ is the Fermi function $(\exp[x/kT] + 1)^{-1}$, and $E = (\epsilon_0^2 + \epsilon^2)^{\frac{1}{2}}$. The function defined by (10) is in fact quite similar to that of (9). For

$$\int_0^{\infty}J_{BCS}(R,T)dR = \xi_0 \equiv \hbar v_F/\pi\epsilon_0(0)\,. \quad (11)$$

In this case ξ_0 is a temperature-independent parameter corresponding to Pippard's coherence length. In addition, J_{BCS} is a slowly varying function of tem-

[3] V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. **20**, 1064 (1950).

[4] J. R. Schrieffer, Phys. Rev. **106**, 47 (1957).

[5] P. B. Miller, Phys. Rev. **113**, 1209 (1959).

[6] A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953).

[7] D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

perature and is not too different from the exponential of (9). (See for example Fig. 5 of Ref. 8.)

Let us now consider how the critical fields are calculated. Depending on which theory of superconductivity one wishes to use, one picks a particular kernel $K(k)$. From (6), the weak field susceptibility

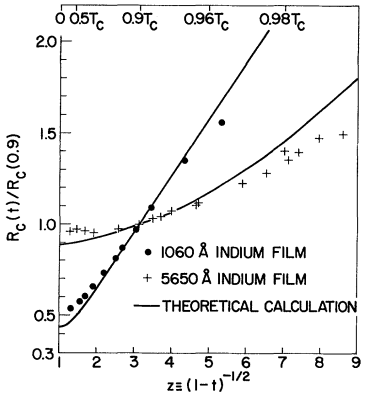

FIG. 1. The temperature variation of the ratio of film critical field to bulk critical field, $R_c \equiv h_c/H_c$. The dots and pluses represent critical field data for indium films of thicknesses 1060 and 5650Å, respectively. The values for $R_c(t)$ have been normalized to the measured value at a reduced temperature of $t = 0.9$ and are plotted as a function of the temperature parameter $Z = (1-t)^{-\frac{1}{2}}$. The corresponding curves are the results calculated from the theoretical model of Eqs. (5), (6), (8), and (9) with values for the nonlocal parameters discussed in Sec. III of the text.

is then calculated. The results of (6) are then substituted into (5) which relates film critical field to weak field susceptibility. Thus, the critical field ratio h_c/H_c can be calculated in a straightforward manner, once $K(k)$ is known. If one wishes to calculate critical fields assuming random surface scattering in the superconductive state, one cannot use (6) which was derived on the basis of specular boundary conditions. Instead, κ/κ_0 is calculated by solving an integral equation as related by Schrieffer.[4] One then substitutes this value of κ/κ_0 into (5) to obtain h_c/H_c.

III. RESULTS

Now let us consider the temperature dependence of critical field. In Fig. 1 are plotted experimental data for two of the pure indium films studied, a thick film and a thin film. In Fig. 2, data are plotted for an alloy film of indium containing 4.6 at. % tin. Along with the experimental data in Figs. 1 and 2 are plotted the corresponding results calculated from the theoretical model presented previously, using the Pippard kernel, specular boundary conditions, and the values for the nonlocal parameters previously used in I and II, i.e., $\xi_0 = 2600$ Å and $\xi_0\lambda_L^2 = 1.62 \times 10^9(\text{Å})^3$ at 0.9 T_c. In carrying out the calculations, ξ_0 was taken to be temperature-independent (as suggested by the BCS theory) and the temperature dependence of λ_L was determined from the calculations of Mühlschlegel.[9] For the alloy film, ξ was calculated from the film resistivity to be 990 Å. In each figure we have plotted, not the critical ratio $R_c \equiv h_c/H_c$ itself, but this ratio normalized to its value at a reduced temperature of $t = 0.9$. This temperature was picked because it was the temperature at which the earlier thickness and composition results were presented. Also, we have plotted the critical field ratio, not versus temperature, but versus the parameter $Z \equiv (1-t)^{-\frac{1}{2}}$. This has the advantage of expanding the temperature scale near T_c which is where R_c varies most rapidly. In addition, because the penetration depth is proportional to Z near T_c, the plots come out to be nearly linear. As Figs. 1 and 2 indicate, the theoretical model predicts fairly well the

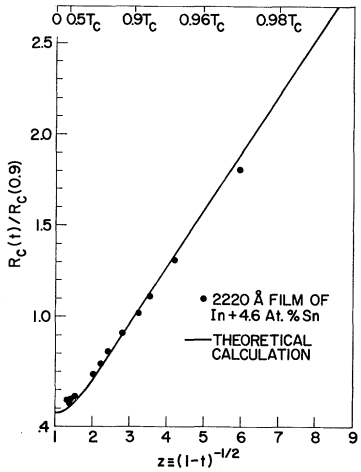

FIG. 2. The temperature variation of the ratio of film critical field to bulk critical field, $R_c \equiv h_c/H_c$. The dots represent critical field data for an alloy film of indium +4.6 at. % tin, of thickness 2220 Å. The values for $R_c(t)$ have been normalized to the value measured at a reduced temperature of $t = 0.9$ and are plotted as a function of the temperature parameter $Z = (1-t)^{-\frac{1}{2}}$. The corresponding curve is the result calculated from the theoretical model of Eqs. (5), (6), (8), and (9) with values for the nonlocal parameters discussed in Sec. III of the text.

observed temperature variation of critical field, particularly for the thin and "impure" films. For these, the maximum discrepancy is ~10%. For thicker films, the discrepancies are somewhat larger. The theoretical model also bears out the over-all quali-

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

[9] B. Mühlschlegel, Z. Physik **155**, 313 (1959).

tative feature of the data—that the temperature variation of critical field is greatest for the thinnest or most impure films and decreases with increasing thickness and/or purity.

Although the discrepancies between the experimental results and the theoretical model are not large they are consistent. For each film the observed variation of R_c with temperature is somewhat less than that predicted by the theoretical model. Preliminary calculations indicate that at least part of the discrepancy can be resolved by using the BCS nonlocal kernel instead of the Pippard kernel. Comparison of $J_P(R,T)$ to $J_{BCS}(R,T)$, relations (9) and (10), indicates that the Pippard and BCS kernels will lead to slightly different temperature dependences of critical field. Calculations with the BCS kernel, however, are

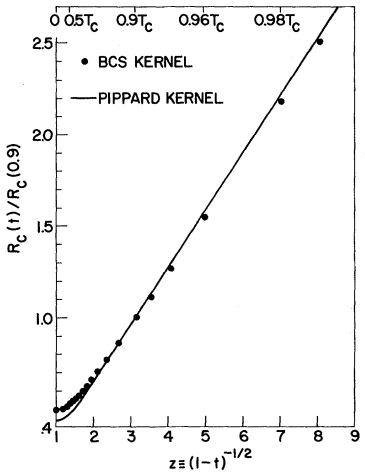

FIG. 3. The temperature dependence of the critical field ratio $R_c \equiv h_c/H_c$ in the thin film limit. The dots are values calculated from the theoretical model of the text using the BCS kernel [Eqs. (12), (13), (5), and (6)]. The solid curve was calculated using the Pippard kernel [Eqs. (5), (6), (8), (9)]. In both cases, the values for $R_c(t)$ are normalized to the value at $t = 0.9$, and are calculated with values for the nonlocal parameters discussed in Sec. III of the text.

quite difficult, a fact which motivated the use of the Pippard kernel in the earlier calculations. In certain limits, however, the BCS kernel simplifies and we can easily carry out the necessary calculations. More complete calculations are now underway and will be reported at a later date.

In the thin limit, i.e., $kl \gg 1$, $k\xi_0 \gg 1$, the BCS kernel becomes[5]

$$K_{BCS}(k) \simeq [3\pi/4\xi_0\lambda_L^2 k]F(t)\,, \tag{12}$$

where

$$F(t) \equiv \frac{\lambda_L^2(t)}{\lambda_L^2(0)}\,\frac{\epsilon_0(t)}{\epsilon_0(0)}\tanh\frac{\epsilon_0}{2kT}\,. \tag{13}$$

Since the quantity in square brackets in Eq. (12) is just the Pippard kernel $K_P(k)$ in the thin limit,

$$\therefore K_{BCS}(k)/K_p(k) = F(t)\,. \tag{14}$$

In Fig. 3 we compare the results of the Pippard and BCS kernels in the thin limit. The solid line is $R_c(t)/R_c(0.9)$ calculated using the Pippard kernel; the dots, using the BCS kernel. It is clear that the BCS results show a striking similarity to the measured critical field ratio for the thin indium film, and the alloy film as well.

In the impure limit, we can obtain simple expressions for $K_{BCS}(k)$ only at T_c and 0°K. At $T = 0$°K for $kl \ll 1$, $l/\xi_0 \ll 1$,

$$K_{BCS}(k) \simeq l/\xi_0\lambda_L^2(0)\,, \tag{15}$$

a result identical to $K_P(k)$ at $T = 0$°K. At $T = T_c$, $kl \ll 1$, $l/\xi_0 \ll 1$,

$$K_{BCS}(k) \simeq 1.35\, l/\xi_0\lambda_L^2(T)\,, \tag{16}$$

which differs from the Pippard results only by the factor of 1.35. Since $F(T_c)/F(0) = 1.33$ in the thin limit compared to 1.35 which we just obtained in the impure limit, the implication is that the temperature dependence of critical field ratio in the impure limit, as calculated from the BCS kernel, is nearly identical to that in the thin limit. The experimental results bear out this conjecture, as a comparison of Figs. 1 and 2 indicates.

Thus, to summarize our results, comparison of the nonlocal, nonlinear theoretical model to measurements of the temperature variation of critical field of indium and indium–tin films yields fairly good agreement when the Pippard nonlocal kernel is used. However, consistent discrepancies are noted. Recalculation of the theoretical results using the BCS nonlocal kernel in the thin and impure limits indicates the small discrepancies between experiment and theory previously observed can be partly (or perhaps completely) resolved by use of the BCS kernel in the theoretical calculation.

Discussion 49

P. V. MASON, *California Institute of Technology:* Am I right in believing in all cases you assume specular reflection?

TOXEN: You are right. In all of the calculations I have assumed specular reflection. However, I would like to point out that the results with diffuse reflection are quite similar and one really wouldn't expect to find any striking differences.

MASON: In some work I have done, I have found a rather considerable difference. Although I admit that the difference here was that the film had a field applied on only one side. But in this case there was a rather sizeable qualitative difference between diffuse and specular reflection.

TOXEN: Well, if you go back and look in Schrieffer's paper on the calculation of the susceptibility in the superconducting state, and if you look at his curves, you'll find that those for the specular and diffuse reflection are nearly the same. However, here we are not considering the one-sided case, but rather the case in which the field is the same on either side of the film. I have also carried out some calculations for the diffuse scattering case and verify Schrieffer's results.

LYNTON: With regard to the thickness dependence of the critical field in the thin field limit, I believe that you use the customary procedure of saying that in the limit of a very short mean free path the coherence length is essentially equal to the mean free path. There is reason to believe as a number of people have pointed out (de Gennes, Goodman, and others) that the coherence length should in fact be taken as the square root of the bulk coherence length times the mean free path. Would that affect your results in any sense?

TOXEN: Actually I didn't do that. Following the spirit of Schrieffer's calculations the coherence length is that of the bulk material and the coherence length which I used was not taken to depend on thickness. Actually I took ξ_0 to be the same for all of the films. The effective coherence distance ξ was then calculated by determining the intrinsic mean free path. That is, I measured the film resistivity in the normal state, measured the film thickness, calculated the intrinsic mean free path, assuming boundary scattering, and then calculated the effective coherence distances in the straightforward manner.

LYNTON: In what straightforward manner?

TOXEN: Well the reciprocal of the effective coherence distance was the sum of the reciprocals of ξ_0 and the mean free path.

LYNTON: I would like to hear Dr. de Gennes on whether this indeed is the correct way of calculating ξ.

DE GENNES: For phenomena involving the thickness of a transition layer of a superconductor of the first kind, the correct coherence length is $(\xi_0 l)^{\frac{1}{2}}$, while if we compute the current at a point in terms of the vector potential at other points the formula which we must use is $1/\xi = 1/\xi_0 + 1/l$. The reason is as follows: (1) For the transition layer problem, we compute the order parameter $\Delta(\mathbf{r})$ at point $\mathbf{r}$ in terms of $\Delta(\mathbf{r}')$ at surrounding points. The corresponding kernel involves essentially the correlation function:

$$\langle \delta[\mathbf{r} - \mathbf{r}(0)]\delta(\mathbf{r}' - \mathbf{r}(t))\rangle$$

where $\mathbf{r}(0)$ and $\mathbf{r}(t)$ are successive *positions* of one electron, in the normal state, and the average is taken on electrons at the Fermi energy. The relevant time t is of order $\hbar/k_B T_c$. In a "dirty" alloy the correlation function is ruled by a diffusion process, and the range of the kernel is $\sim(Dt)^{\frac{1}{2}} \sim (\xi_0 l)^{\frac{1}{2}}$ (where $D = \frac{1}{3}v_F l$ is the diffusion coefficient). (2) For the calculation of the current $\mathbf{j}(\mathbf{r})$ in terms of the potential $\mathbf{A}(\mathbf{r}')$, what comes in the kernel is a correlation function between *velocities:* this correlation is essentially destroyed by one collision, and the range of the kernel in the "dirty" limit is the transport mean free path l.

Microwave Nonlinearities in Thin Superconducting Films

M. D. SHERRILL and K. ROSE

General Electric Research Laboratory, Schenectady, New York

Nethercot[1] has shown that superconducting tin films thicker than 2000 Å can produce second harmonics of 10 kMc/sec microwaves when they are placed in a cavity and properly biased with a magnetic field.

In the present experiment a much thinner tin film is placed directly across an X-band waveguide so that the waveguide opening is completely covered. The film is not biased. In a certain power range the film transmission is found to be highly nonlinear and third harmonics of the fundamental frequency are generated. No detectable second harmonic is generated.

If the film's behavior is to be substantially different in the superconducting and normal states the normal resistance per square should not be too small compared with the guide impedance, which is about 500 Ω. To control the agglomeration of the tin and thereby obtain films with resistances per square of 10 to 110 Ω at helium temperature, the substrates were prepared for the tin evaporation by first evaporating upon them a very thin nonconducting gold layer. The substrates were of fused or crystal quartz and had four gold patches evaporated on them to provide dc connections to the film. The substrate was at room temperature during the tin evaporation. After the tin evaporation the films were removed

[1] A. H. Nethercot, Jr., Phys. Rev. Letters **7**, 226 (1961).