

## Precise Critical-Field Measurements of Superconducting Sn Films in the London Limit

D. H. DOUGLASS, JR.\*

*Department of Physics and Research Laboratory of Electronics,† Massachusetts Institute of Technology, Cambridge, Massachusetts*

AND

R. H. BLUMBERG†

*IBM Command Control Center, Federal Systems Division, Kingston, New York*

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Precise measurements of the critical field of superconducting tin films were made as a function of temperature and thickness. Particular attention was paid to measurements near the transition temperature where the London limit holds. It was found that near the transition temperature the critical field could be expressed in the form  $H_c = (\Delta t)^{1/2} \gamma (1 + \epsilon \Delta t)$ , where  $\gamma$  and  $\epsilon$  are independent of temperature. In terms of the Ginzburg-Landau theory modified to include the lower temperatures, the constant  $\gamma$  determines the penetration depth and the constant  $\epsilon$  is different for different modifications of the field-independent free energy.

Penetration depths determined in this way were found to be a function of thickness. Assuming that the coherence length  $\xi$  in the film is determined by the thickness and using the expression for the thickness dependence of the penetration depth given by Tinkham, one can obtain a bulk coherence length  $\xi_0$  of approximately 2100 Å, as well as a bulk penetration depth of 510 Å, from the data. This way of determining  $\xi_0$  is quite different from the high-frequency method of Pippard, and it is proposed as an independent and alternative method.

Two particular modifications of the Ginzburg-Landau theory are considered. The first, which is the original theory, predicts a value of  $\epsilon = 0.31$ . The second, which is the Gorter-Casimir modification proposed by Bardeen, predicts a value of  $\epsilon = -0.19$ . Experimentally,  $\epsilon$  was determined to have an average value of  $0.14 \pm 0.10$ .

### I. INTRODUCTION

THE various theories of superconductivity are usually worked out explicitly only for two limiting conditions; the Pippard limit (nonlocal limit) in which the coherence length  $\xi$  is much larger than the penetration depth  $\lambda$ , and the London limit (local limit) in which the penetration depth is much larger than the coherence length. If the object of an experiment is to test such theories, then experimental conditions should be chosen, if at all possible, so that either one or the other of these limits is satisfied. Unfortunately, these conditions are sometimes difficult to achieve in the bulk state since the above-mentioned lengths are frequently of the same order of magnitude. One can, however, satisfy the London limit by working very near the transition temperature where the penetration depth is approaching an infinite value. If one works with thin evaporated specimens, which are inherently "dirty," then one would expect that the coherence length would be limited by the size of the specimen. In addition, it is expected that the penetration depth increases as the thickness is made smaller; thus, the London limit is easier to satisfy. By using thin specimens and working near the transition temperature, we have insured that the London limit was satisfied. Thus, it is hoped that a comparison of the data with the theories that are valid in this limit will be meaningful.

\* Now at Department of Physics and Institute for the Study of Metals, University of Chicago.

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In Sec. II we discuss the theory of the critical fields for thin specimens. Section III includes the experimental results, and in Sec. IV we present an analysis of these results.

### II. THEORY

#### A. The Critical Field of Thin Superconducting Films

It has been quite common in the past to apply the London equations to the analysis of critical-field data on thin films. This application is basically incorrect because the London equations are valid only for fields that are much less than the critical field. To describe critical-field phenomenon one must use a nonlinear theory such as the Ginzburg-Landau<sup>1</sup> (GL) theory that was invented explicitly to handle strong magnetic fields and that reduces to the London equations for weak fields.<sup>2</sup> Also, in the light of Gor'kov's<sup>3</sup> derivation of the GL equations from the microscopic theory, the solutions of the GL equations are equivalent to the local nonlinear limit of the microscopic theory.

In the rest of this section we will outline the GL theory, showing how it has been empirically modified to include the lower temperatures. We will then point out that the expression for the critical field of a thin film contains two parameters that can be measured

<sup>1</sup> V. L. Ginzburg and L. D. Landau, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **20**, 1064 (1950).

<sup>2</sup> Physically, the difficulty with the London theory is that it does not consider the field dependence of the penetration depth which is implicitly built into the GL theory. This difficulty becomes most acute for the thin limit.

<sup>3</sup> L. P. Gor'kov, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **36**, 1918 (1959) [translation: *Soviet Phys.—JETP* **9**, 1364 (1959)].

experimentally, the first of which is the penetration depth  $\lambda$  and is independent of the modifications. The second parameter, a constant that we call  $\epsilon$ , will depend on the modification. Hopefully, measurement of  $\epsilon$  will help to select the correct one.

Ginzburg and Landau, in the derivation of their equations, started with the following free-energy function for the superconducting state in a magnetic field.

$$F_{sH} = F_{s0}(\psi) + \frac{(\nabla \times \mathbf{A})^2}{8\pi} + \frac{\hbar^2}{2m} \left( -i\nabla - \frac{e}{c} \mathbf{A} \right) |\psi|^2, \quad (1)$$

where  $\mathbf{A}$  is the vector potential,  $\psi$  is the order parameter (which was shown by Gor'kov to be proportional to the energy gap), and  $F_{s0}(\psi)$  is the value of the free energy in the absence of a magnetic field. There were, in fact, two independent assumptions in the original GL theory. The first concerns the form of the magnetic contribution to the free energy represented by the last two terms in Eq. (1) and the second pertains to the explicit form of the field-free portion given by the first term.

No exact form that is good for all  $T$  exists, at present, for  $F_{s0}$ . Various phenomenological expressions have been considered, all of which near  $T_c$  must and do reduce to the expression originally given by Ginzburg and Landau.

Bardeen<sup>4</sup> has considered the following  $F_{s0}$ :

$$F_{s0} = F_{n0} + [H_{cb}^2(0)/8\pi] \times \{2t^2[1 - (1 - |\chi|^2)^{1/2}] - |\chi|^2\}, \quad (2)$$

where  $\chi = \psi(T, H)/\psi(0, 0)$  and  $F_{n0}$  is the free energy of the normal state. (He has also considered a completely general  $F_{s0}$ .) This is essentially the free-energy expression of the Gorter-Casimir<sup>5</sup> two-fluid model. We shall call this  $F_{s0}^{(I)}$ .

Ginzburg and Landau expanded  $F_{s0}$  in a power series in  $|\psi|^2$ , which was originally expected to be good only near  $T_c$ :

$$F_{s0} = F_{n0}(T) + \alpha(T) |\psi|^2 + \frac{1}{2} \beta(T) |\psi|^4 + \dots, \quad (3)$$

where  $\alpha$  and  $\beta$  are coefficients depending on the temperature. This expansion was stopped after the  $|\psi|^4$  term.

Ginzburg,<sup>6,7</sup> in an effort to apply (3) to all temperatures, has considered  $\alpha$  and  $\beta$  to be arbitrary functions of temperature. They may be expressed in terms of the temperature dependence of experimentally determined bulk properties such as the critical field  $H_{cb}(T)$ ,

$$\alpha(T) = -[H_{cb}^2(T)/4\pi][1/|\psi(0, T)|^2], \quad (4)$$

$$\beta(T) = [H_{cb}^2(T)/4\pi][1/|\psi(0, T)|^4]. \quad (5)$$

We shall call this  $F_{s0}^{(II)}$ .

<sup>4</sup> J. Bardeen, Phys. Rev. **94**, 554 (1954).

<sup>5</sup> C. J. Gorter and H. B. G. Casimir, Physik Z. **35**, 963 (1934); Z. Tech. Phys. **15**, 539 (1934).

<sup>6</sup> V. L. Ginzburg, J. Exptl. Theor. Phys. (U.S.S.R.) **30**, 595 (1956) [translation: Soviet Phys.—JETP **3**, 621 (1956)].

<sup>7</sup> V. L. Ginzburg, Doklady Akad. Nauk S.S.S.R. **1**, 541 (1956).

It was first pointed out by Marcus<sup>4</sup> that if the coefficients had the following temperature dependence,

$$\alpha(T) = -[H_{cb}^2(0)/4\pi|\psi(0, 0)|^2][(1-t^2)/(1+t^2)], \quad (6)$$

$$\beta(T) = [H_{cb}^2(0)/4\pi|\psi(0, 0)|^4][1/(1+t^2)^2], \quad (7)$$

the theory would give a parabolic critical-field curve and a  $(1-t^2)^{-1/2}$  dependence on temperature for the penetration depth. Clearly Eqs. (6) and (7) are a special case of (4) and (5).

In the original theory Ginzburg and Landau expanded  $\alpha$  and  $\beta$  in the neighborhood of  $T_c$ :

$$\alpha(T) \approx (d\alpha/dT)_{T_c}(T - T_c), \quad (8)$$

$$\beta(T) \approx \beta(T_c). \quad (9)$$

Gor'kov's derivation produced the GL equations implicitly in the form of (8) and (9). All of the above-given expressions for  $F_{s0}$  reduce to (8) and (9) near  $T_c$  as they must.

Ginzburg<sup>7</sup> has also considered the expression for the critical field  $H_c$  of a thin film (penetration depth  $\lambda$  much larger than the thickness  $d$ ) in terms of a completely general  $F_{s0}$ . It can be expressed as

$$H_c^2 = 24\eta[\lambda(T, d)/d]^2 H_{cb}^2, \quad (10)$$

where

$$\eta = -[(1/\phi)(\partial f_{s0}/\partial \phi^*)]_{\phi=0}, \quad (11)$$

$$f_{s0} = (4\pi)(F_{s0} - F_{n0})/H_{cb}^2(T), \quad (12)$$

$$\phi = \psi(T, H)/\psi(T, 0), \quad (13)$$

and where  $d$  is the thickness and  $\lambda(T, d)$  is the penetration depth in the limit of zero magnetic field and depends on thickness, as well as temperature. Equation (10) is valid as long as  $d \leq 5^{1/2}\lambda(T, d)$ . Note also that, independently of the form of  $F_{s0}$ ,  $H_c$  varies inversely as  $d$ , since  $F_{s0}$  is independent of thickness. (Experimentally  $T_c$  is observed to change very little with thickness; hence  $F_{s0}$  does not change either.) It may be easily shown that evaluation of  $\eta$  for  $F_{s0}^{(I)}$  and  $F_{s0}^{(II)}$  gives  $\eta = \frac{1}{2}(1+t^2)$  and  $\eta = 1$ , respectively. Thus, for  $F_{s0}^{(I)}$

$$H_c = (24)^{1/2}[\lambda(T, d)/d]H_{cb}(T)[(1+t^2)/2]^{1/2}, \quad (14)$$

and thus, for  $F_{s0}^{(II)}$

$$H_c = (24)^{1/2}[\lambda(T, d)/d]H_{cb}(T). \quad (15)$$

Since we are interested in applying these equations only near the transition temperature, we can express the equations as a function of  $\Delta t = (1-t)$ .

From the data of Muench<sup>8</sup> on bulk Sn one can show that, near  $T_c$ ,

$$H_{cb}(T) = 549.5\Delta t(1 - 0.32\Delta t). \quad (16)$$

It has been determined experimentally<sup>9</sup> for Sn that

<sup>8</sup> N. L. Muench, Phys. Rev. **99**, 1814 (1955).

<sup>9</sup> Erich Erlback, Ph.D. thesis, Columbia University, 1960 (unpublished).

the temperature dependence and the thickness dependence of the penetration depth are factorable, at least for  $d$  between 160 and 400 Å, and for  $\Delta t \ll 1$  it can be expressed as

$$\lambda(T, d) = \lambda(0, d) \{ [0.56 / (\Delta t)^{1/2}] (1 + 0.63 \Delta t) \}. \quad (17)$$

Substitution of Eqs. (16) and (17) in (14) and (15) gives an expression that is valid near  $T_c$  for the critical field of the Sn film,

$$H_c(T) = 1510 [\lambda(0, d) / d] \Delta t (1 + \epsilon \Delta t), \quad (18)$$

in which  $\epsilon$  is a number that is independent of  $d$ , but is dependent on the form of  $F_{s0}$ . In particular,  $\epsilon = -0.19$  and  $\epsilon = +0.31$  for models  $F_{s0}^{(I)}$  and  $F_{s0}^{(II)}$ , respectively.

Since the experimental curves of  $H_c(T)$  near  $T_c$  do, indeed, have the form of Eq. (18), we can determine for each film  $\lambda(0, d)$  and the coefficient  $\epsilon$ , to test the validity of various forms of  $F_{s0}$ .

### B. Variation of Penetration Depth with Thickness

Since we will be able to determine experimentally the variation of the penetration depth with thickness, we now consider what can be said in a theoretical way. The simplest considerations follow that of Tinkham.<sup>10</sup> He suggested, after considering the Pippard equations,<sup>11</sup> that one could incorporate the nonlocal effects of the theory in the penetration depth without too great an error by letting the penetration depth be a function of the coherence length  $\xi$  in the following way:

$$\lambda = \lambda_B (\xi_0 / \xi)^{1/2},$$

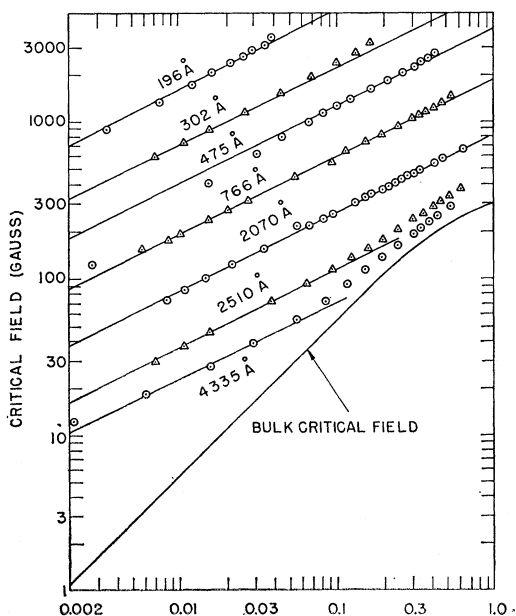


FIG. 1. Critical field of Sn vs temperature for various thicknesses.

<sup>10</sup> M. Tinkham, Phys. Rev. **110**, 26 (1958).

<sup>11</sup> A. B. Pippard, Proc. Roy. Soc. (London) **216**, 547 (1953).

where  $\lambda_B$  is the bulk penetration depth and  $\xi_0$  is the bulk coherence length. Since evaporated films are inherently quite "dirty," the most obvious assumption concerning  $\xi$  is that for the very thin limit it is equal to the thickness  $d$ . Since for the thick films  $\xi$  must approach  $\xi_0$ , a reasonable variation for all  $d$  might be

$$1/\xi = (1/\xi_0) + (1/d).$$

The penetration depth will now be

$$\lambda = \lambda_B (1 + \xi_0/d)^{1/2}, \quad (19)$$

which is the formula given by Tinkham.

There have been several discussions of the mean free path variation of the penetration-depth limit. Gor'kov<sup>12</sup> has shown that the penetration depth in the GL theory is a function of the mean free time between collisions, and Douglass<sup>13</sup> has extended it in the manner of Tinkham. The treatment that we shall use, and which is the most precise, is that of Miller.<sup>14</sup> Miller has calculated from the Bardeen-Cooper-Schrieffer theory the penetration depth as a function of  $\xi_0/l$  and has presented the results in the form of useful tables. We consider below how to relate the mean free path  $l$  to the thickness. All of these calculations give the same functional form at high ratios of  $\xi_0/l$ —namely,  $\lambda \propto \lambda_L (\xi_0/l)^{1/2}$ . The resulting functional form of the critical field is  $H_c \propto (\lambda_L/d) (\xi_0/l)^{1/2}$  which becomes  $(\lambda_L^2 \xi_0/d^3)^{1/2}$  if the mean-free path is limited by the thickness; this result was previously obtained.<sup>13,15</sup>

### III. EXPERIMENTAL RESULTS

Critical-field measurements were made on 24 Sn films as a function of temperature. Thicknesses ranged from 196 to 4330 Å. These films were deposited by vacuum deposition at a controlled rate of 100 Å/sec onto a soft soda-glass substrate held at a temperature of 77°K. The substrates were initially baked at 670°K for 10 h. The pressure during evaporation was less than  $10^{-6}$  mm Hg. In order to eliminate penumbra effects the edges of the film were trimmed with a diamond-tooled ruling engine and the thickness was measured by optical interferometry to an accuracy of  $\pm 40$  Å. The width of the film was measured by a traveling microscope. X-ray diffraction methods showed a strong [100] orientation with the  $c$  axis located at random in the plane of the film. Electron microscopy shows that the grain size is nearly constant at 1000 Å.

The critical fields were determined by measuring the resistance of the film as a function of field. The transition was arbitrarily defined as one-half of the normal-state resistance and was found to be independent of the

<sup>12</sup> L. P. Gor'kov, J. Exptl. Theoret. Phys. (U.S.S.R.) **37**, 1407 (1959) [translation: Soviet Phys.—JETP **10**, 998 (1959)].

<sup>13</sup> D. H. Douglass, Jr., Phys. Rev. **124**, 735 (1961).

<sup>14</sup> Piotr Miller, Phys. Rev. **113**, 1209 (1959).

<sup>15</sup> Similar results were presented by R. A. Ferrell and A. J. Glick and by A. Toxen to the American Physical Society, New York, January 24–27, 1962.

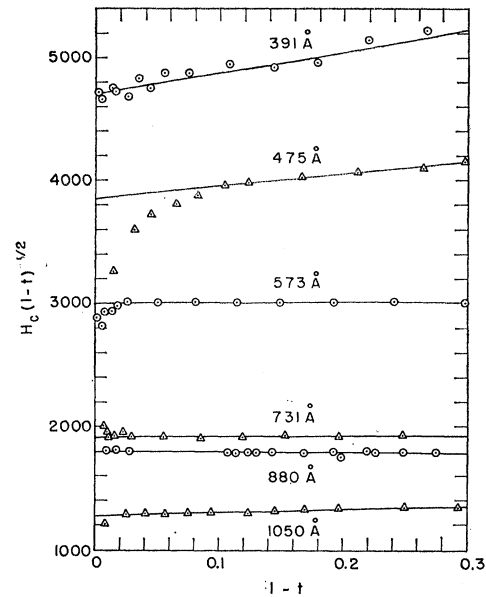
TABLE I. Tabulations of thicknesses, resistivities, penetration depths, and values of  $\epsilon$  for the 24 films investigated.

Film No.	Thickness (angstroms)	Resistivity at 4.2°K ( $\mu\Omega\text{-cm}$ )	$\lambda(0,d)$ (angstroms)	$\epsilon$
Sn 136-3	196	2.99	1880	...
136-4	197	3.23	2070	...
195-1	297	1.89	1410	...
195-2	302	1.97	1410	...
193-3	388	1.44	1240	+0.33
193-4	391	1.51	1230	+0.34
93-1	421	1.51	1230	...
93-2	398	1.40	1160	...
128-2	475	1.58	1220	+0.23
133-2	573	1.33	1150	+0.09
94-1	766	0.925	960	+0.02
94-2	731	0.915	920	+0.02
199-3	880	1.51	1040	+0.02
199-4	900	1.54	1050	+0.05
121-2	1050	0.920	890	+0.18
105-1	1070	0.685	870	...
105-2	1030	0.628	820	...
198-3	1570	1.12	850	...
198-4	1590	1.39	860	...
120-1	2070	0.599	728	...
120-2	2060	0.614	721	...
98-2	2510	0.402	660	...
95-1	4330	0.547	630	...
95-2	4330	0.533	645	...

measuring current. A sixth-order solenoid with a variation of less than 0.1% over the film was designed to apply the magnetic field; calibration was with a nuclear magnetic-resonance probe. The earth's and stray magnetic fields were reduced by two orders of magnitude by a  $\mu$ -metal shield surrounding the test station. Temperatures were determined by vapor pressure measurements, with the 1958T scale.

Critical-field measurements are shown in Fig. 1 for representative films (data on some films have been omitted for clarity). Here  $\log H_c(T)$  has been plotted vs  $\log(1-t)$ . The solid lines are the best straight lines through the data with slope 1/2. (It is seen that for some the fit is not too good.) The bulk critical-field curve taken from Muench is shown; as a check, one film of 50 000 Å was made and found to have essentially the same critical-field curve.

As a test of Eq. (18), the data were replotted in terms of  $H_c(1-t)^{-1/2}$  vs  $(1-t)$  and some of the results are shown in Fig. 2. The data should, when plotted, result in a straight line if Eq. (18) is obeyed. Any error in the transition temperature will cause these curves to deviate from a straight line for very small  $(1-t)$ ; the deviations are largest for the 475 Å film. We therefore neglected the region very near  $T_c$  and drew the best straight line through the remaining points. According to Eq. (18), the intercept gives us  $\lambda(0,d)$  and the slope determines the constant  $\epsilon$ . The data on all 24 films are tabulated in Table I. Values of  $\lambda(0,d)$  were found for all films; but  $\epsilon$  for only nine films was obtainable for the following reasons: For the smaller thicknesses only a narrow temperature range could be scanned because of a 3300-G limitation of the solenoid. For the thicker specimens,


 FIG. 2. Ratio of critical field to  $(1-t)^{1/2}$  vs temperature for various thicknesses.

the small region of validity of the London limit restricted the measurements to a very narrow temperature range.

The resistivities of the films were measured at 273, 77, and 4.2°K, in order to determine  $l$  for the films. These values are then used, with Miller's tables, to calculate  $\lambda$ . These data were analyzed on the basis of the expression by Fuchs<sup>16</sup> for the resistance of a thin film with diffuse scattering assumed,

$$\frac{\rho}{\rho_\infty} = \frac{4l'}{3d} \frac{1}{[\ln(l'/d) + 0.4228]}, \quad \frac{d}{l'} \ll 1 \quad (20)$$

$$\frac{\rho}{\rho_\infty} = 1 + \frac{3l'}{8d} \frac{d}{l'}, \quad \frac{d}{l'} \gg 1, \quad (21)$$

where  $\rho$  and  $d$  are the resistivity and the thickness of the film, and  $\rho_\infty$  and  $l'$  are the resistivity and the mean free path of the "bulk" state. The data at 273°K were plotted as  $\rho$  vs  $d^{-1}$ , and application of Eq. (21) to the slope and intercept of the straight line through the data gave  $l'$  and  $\rho_\infty$ . The data at 77°K were analyzed in the same way. Equation (20) was applied to the data at 4.2°K, where  $(\rho d)^{-1}$  was plotted vs  $\log d$ . Slope and intercept again gave  $\rho_\infty$  and  $l'$ . These data are summarized in Table II, together with other measurements on Sn.<sup>17-19</sup> It is seen that the product  $\rho_\infty l'$  is essentially independent of temperature or thickness, as one would expect from the free-electron model. Assuming this, we

<sup>16</sup> K. Fuchs, Proc. Cambridge Phil. Soc. **34**, 100 (1938).

<sup>17</sup> E. R. Andrews, Proc. Phys. Soc. (London) **A62**, 86 (1949).

<sup>18</sup> R. G. Chambers, Proc. Roy. Soc. (London) **A215**, 481 (1952).

<sup>19</sup> J. E. Kunzler and C. A. Renton, Phys. Rev. **108**, 1297 (1957).

TABLE II. Tabulation of  $l'$ ,  $\rho_\infty$ , and  $\rho_\infty l'$  as determined by various investigators.

$T$ (°K)	$l'$ (Å)	$\rho_\infty$ (ohm-cm)	$\rho_\infty l'$ (ohm-cm <sup>2</sup> )
This work	(Thin films)		
273	160 ± 30	(11.4 ± 0.3) × 10 <sup>-6</sup>	(1.9 ± 0.4) × 10 <sup>-11</sup>
77	670 ± 130	(2.7 ± 0.1) × 11 <sup>-6</sup>	(1.8 ± 0.4) × 10 <sup>-11</sup>
4.2	6300 ± 1500	(0.26 ± 0.02) × 10 <sup>-6</sup>	(1.5 ± 0.3) × 10 <sup>-11</sup>
Andrews	(Thick foils) <sup>a</sup>		
291	170	11.8 × 10 <sup>-6</sup>	2.0 × 10 <sup>-11</sup>
3.8	950 000	2.1 × 10 <sup>-9</sup>	2.0 × 10 <sup>-11</sup>
Chambers	(Bulk) <sup>b</sup>		
4.2	...	...	(1.05 ± 0.10) × 10 <sup>-11</sup>
Kunzler	(Bulk single crystal) <sup>c</sup>		
4.2	4,500,000	5.2 × 10 <sup>-10</sup>	2.3 × 10 <sup>-11</sup>

<sup>a</sup> See reference 17.<sup>b</sup> See reference 18.<sup>c</sup> See reference 19.

take the effective mean free path  $l$  to be given by  $l = \rho_\infty l' / \rho$ .

#### IV. ANALYSIS OF DATA

In Fig. 3 values of  $\lambda^2(0, d)$  are plotted vs  $d^{-1}$ . The extrapolation of the data to  $d^{-1} = 0$ , which should correspond to the bulk state, gave a value of intercept corresponding to 510 ± 50 Å. This is to be compared with the currently accepted value of 510 Å for the bulk penetration depth.<sup>19</sup> The data were normalized to this value, and are presented by the circles in Fig. 3. The data are compared with the solid lines representing the predictions of Miller and Tinkham [Eq. (19)] for various  $\xi_0$ , where  $l$  is arbitrarily taken to be equal to the thickness in the application of Miller's calculations.

Curve *a* is the best curve that passes through the points for large  $d^{-1}$  computed from Miller's table with  $\xi_0 = 2500$  Å, and curve *b* is the best curve through the smaller values of  $d^{-1}$  computed from Miller's table with

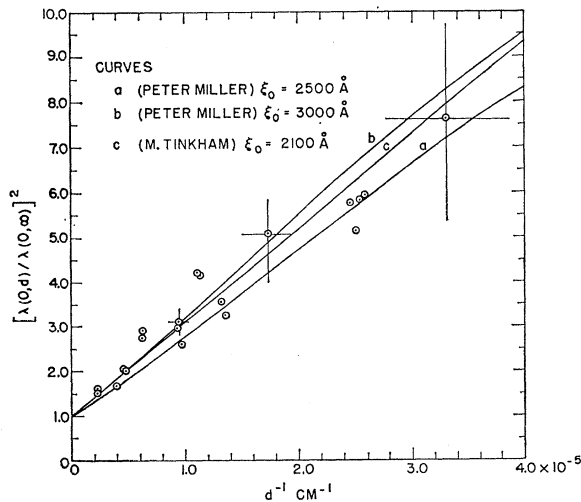


FIG. 3. Penetration depth vs thickness.

$\xi_0 = 3000$  Å. Curve *c* is the best fit of the Tinkham expression [Eq. (19)], which is a straight line on this plot, with  $\xi_0 = 2100$  Å. These values are to be compared with the currently accepted value<sup>20</sup> of 2300 Å as determined from anomalous skin-effect measurements.

Although the above-given agreement is good, the analysis is somewhat dissatisfying because of the non-exact arguments leading to Eq. (19) and the questionable assumption that the mean free path equals  $d$ .

A less-questionable analysis would be to use Miller's table, together with measured values of the mean free path  $l$  which can be obtained from the electrical resistivity.

According to the free-electron model, the mean free path is inversely proportional to the normal-state resistivity. In the free-electron model, the expression for the  $\rho l$  product can be put into the form

$$\rho l = h/e^2 (3/8\pi N^2)^{2/3}, \quad (22)$$

where  $N$  is the number of electrons per unit volume. Since  $N$  is not expected to vary much with temperature or thickness,  $\rho l$  should be nearly the same and independent of these variables. Table II shows that although  $\rho$  and  $l$  separately vary over many orders of magnitude, the product remains the same within a factor of 2. However, this work apparently detects a slight temperature variation of  $\rho l$ .

In spite of this, it is probably a quite valid assumption that the mean free path is inversely proportional to the resistivity. Moreover, the resistivity measures the "effective" mean free path from impurities and boundary scattering. The mean free path entering the superconducting penetration depth was derived by Miller only for impurity scattering. Since one would expect the electrons "not to care" what it was that scattered them, be it impurities or surface roughness,<sup>21</sup> then one could apply the calculations directly to other kinds of scattering by introducing an effective mean free path that determines the value of the resistivity is the same as that determining the value of the penetration depth. Taking the value  $\rho l = 1.5 \times 10^{-3} \Omega\text{-cm}^2$ , which was determined at 4.2°K on these films, we calculated  $l^{-1}$  for each film. The values of  $\lambda^2(0, d)$  were plotted vs  $l^{-1}$  and  $\xi_0$  was obtained by fitting the "best" curve through the data points with the use of Miller's table. Values of  $\xi_0$  obtained were approximately 50% higher than the values obtained by the first method. This is related to the fact that the mean free path is approximately 50% larger than the thicknesses. If  $\xi_0 = 2300$  Å is accepted as "true" value, then one is forced to the conclusion that for these particular "dirty" films the coherence

<sup>20</sup> C. J. Gorter, *Progress in Low Temperature Physics* (North-Holland Publishing Company, 1961), p. 243.

<sup>21</sup> L. Lynton and D. McLacklan (to be published) have shown that the mean free path from boundary scattering is equivalent to the mean free path from impurity scattering in changing the transition temperature of a superconductor.

length in film is limited by the thickness, not the mean free path. As further evidence for this we noticed in the plot  $\lambda^2$  vs  $l^{-1}$  that the scatter of the data was noticeably greater than in the  $\lambda^2$  vs  $d^{-1}$  plot. Since there is considerable anisotropy in the resistive properties of Sn films,<sup>22</sup> slightly different preferred orientations, which are quite possible, would affect the mean free path and not the thickness. This conclusion, however, is quite tenuous and should await more substantial evidence.

It would appear that the simple expression [Eq. (19)] is adequate enough to determine both the bulk penetration depth and the coherence length of tin to moderate accuracy. The fair agreement between values determined by this method and the more exact, and in some ways more difficult, microwave method, would suggest that one could use this method to determine both the bulk penetration depth and the bulk coherence length without serious error.

In Fig. 4, the coefficient  $\epsilon$ , as determined by Eq. (18), has been plotted as a function of  $d$ . Although neither model considered earlier predicts any variation of  $\epsilon$  with  $d$ , there is a suggestion of a variation from the data.

Systematic errors are not ruled out. If there is any variation of  $\epsilon$  with thickness, the values at small  $d$  agree quite well with model  $F_{s0}^{(II)}$  ( $\epsilon = +0.31$ ). Then  $\epsilon$  appears to drop to zero and then to increase after that; in no case is  $\epsilon$  negative as predicted by model  $F_{s0}^{(I)}$ . Although the data favor  $F_{s0}^{(II)}$  over  $F_{s0}^{(I)}$ , neither fit very well, and a more suitable  $F_{s0}$  must be sought.

## V. SUMMARY

Superconducting tin films were prepared and measurements performed under strictly controlled con-

<sup>22</sup> R. H. Blumberg and D. P. Seraphim, J. Appl. Phys. (to be published).

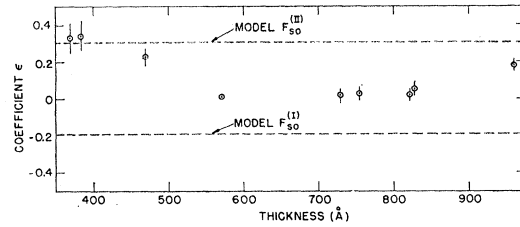


FIG. 4.  $\epsilon$  vs thickness.

ditions. Critical-field measurements made as a function of thickness were restricted to the immediate vicinity of the transition temperature where the penetration depth is much larger than the coherence length.

The penetration depth was determined as a function of thickness. Extrapolation of the data to  $d = \infty$  gave a value of the bulk penetration depth which is equal to 510 Å. Assuming that the coherence length in the film equals the thickness and using Tinkham's expression, we obtained a value for the bulk coherence length of approximately 2100 Å. We suggest that this method be used as an alternate to measure these two constants in superconductors which have not been determined in any other way.

Finally, by examining the critical-field curves at lower temperatures, it is possible, in principle, to distinguish between various forms of the free energy in zero field. Measurements on these films favor Ginzburg's expression for the free energy.

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