and a number of weaker lines. After heating the crystal in an atmosphere of H_2 there appeared another line at 7850 cm^{-1}.

We have no explanation of the spectrum in the infrared. We have tried to fit the spectrum to Fe^{2+} in tetrahedral symmetry. However the multiplicity of the lines and the narrowness seem to be indicative for transitions belonging to some rare earth or uranium group impurity.

ACKNOWLEDGMENT

We are grateful to Dr. E. Fischer of the Weizmann Institute of Science for permission to use the Cary spectrophotometer.

PHYSICAL REVIEW VOLUME 119, NUMBER 5 SEPTEMBER 1, 1960

Critical Fields of Thin Superconducting Films*

WILLIAM B. ITTNER, III
International Business Machines Research Laboratory, Poughkeepsie, New York
(Received April 25, 1960)

The critical fields of thin superconducting films have been calculated on the basis of the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity following a method outlined by Schrieffer. It is shown that it is convenient to use the critical field formula postulated by London where the London penetration depth is replaced by an effective penetration depth which can be specified through the use of the BCS theory. The effective penetration depth, unlike the London penetration depth which, for a given material, varies only with the temperature, is found to vary, in the BCS theory, with both the film thickness and the electronic mean free path of the normal material. This paper attempts to show that the measured critical fields of thin tin films are in general qualitative agreement with the predictions of the BCS theory.

INTRODUCTION

RECENT measurements carried out on thin superconducting tin films[1] have shown that the experimentally measured values of the externally applied longitudinal field required to destroy superconductivity are greater than would be expected on the basis of the London theory of superconductivity. This behavior has, indeed, been anticipated. Lutes,[2] in studying the critical fields of thin relatively impure tin whiskers, concluded that the superconducting penetration depths were considerably greater than the London value. Experiments by Schawlow[3] on several cylindrical tin films likewise indicated that the magnetic penetration depth in a superconductor will be a function, not only of the specimen purity, but of the specimen thickness as well. Likewise, Glover and Tinkham[4] found, in microwave measurements on thin films, that the effective penetration depth was increased appreciably over its value in bulk material.

These experimental results are, in fact, suggested by the various modern theories of superconductivity[5–7] which indicate that the supercurrent density is correctly expressed as an integral of the vector potential and will vary with both the specimen dimensions and the normal state electronic mean free path. As a consequence of this nonlocal aspect of superconductivity, the effective penetration depth (as it appears in the London theory) will be expected to depend on both the specimen dimensions and bulk mean free path. These considerations have been discussed by Tinkham[8] who suggested a practical method of treating the penetration depth to be used in the London theory. The results presented here are in qualitative agreement with the results outlined by Tinkham.

LONDON THEORY

It is instructive to carry out a number of calculations on the basis of the London theory of superconductivity to illustrate the general methods which are involved. Moreover, as the nonlocal theories are rather intractable in a mathematical sense, it will subsequently prove expedient to make use of a modified London equation where the London penetration depth, λ_L, is replaced by an effective penetration depth, λ_ϵ whose magnitude is indicated by the nonlocal theory.

In the London theory[9] the spatial variation of magnetic field, $H(x)$, inside a film of thickness d is

$$\frac{H(x)}{H_0}=\frac{\cosh x/\lambda_L}{\cosh d/2\lambda_L}, \tag{1}$$

* Supported, in part, by the Department of Defense.

[1] The films reported on in this work were fabricated and measured both by Mr. R. G. Blumberg of the IBM Federal Systems Division, Kingston, New York, and by Mr. A. E. Brennemann of the IBM Research Laboratory, Poughkeepsie, New York.

[2] O. S. Lutes, Phys. Rev. **105**, 1451 (1957).

[3] A. L. Schawlow, Phys. Rev. **109**, 1856 (1958).

[4] R. E. Glover and M. Tinkham, Phys. Rev. **104**, 844 (1956); **108**, 243 (1957).

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

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

[7] N. N. Bogoliubov, Nuovo cimento **7**, 794 (1958).

[8] M. Tinkham, Phys. Rev. **110**, 26 (1958).

[9] H. London, Proc. Roy. Soc. (London) **A152**, 650 (1935).

where H_0 is the tangential field strength at the two surfaces of the film, λ_L is the London penetration depth, and x varies from $-d/2$ to $+d/2$. It has been shown that the critical magnetic field to be expected of a particular film can be calculated in terms of the free energy-difference per unit volume between the superconducting and the normal state of the film,[10] i.e.,

$$\frac{F_n-F_s}{v}=\frac{H_c^2}{8\pi}=-\frac{H_0\Delta M}{2}, \tag{2}$$

where H_c is the critical field of the bulk material, H_0 is the field at the surface of the film, and M is the magnetization of the film. ΔM may be written as

$$\Delta M=\frac{1}{4\pi d}\left[\int_{-d/2}^{+d/2}H(x)dx-H_0d\right]. \tag{3}$$

Substituting Eq. (1) into Eq. (3) and integrating yields a value of H_0 for which the field at the surface of the film becomes critical, i.e., a field H_0 equal to the critical field of the film, H_c^f, where

$$H_c^f=H_c[1-(2\lambda_L/d)\tanh d/2\lambda_L]^{-\frac{1}{2}}. \tag{4}$$

This is, of course, the result obtained by London in a somewhat different manner.[11] The important point to be emphasized here is that the critical field of a thin film can be related to the spatial variation of the magnetic field within the film. Consequently, the calculation of the critical magnetic field of a film can be reduced to the calculation of the spatial variation of the magnetic field within the film. Once the field variation, $H(x)/H_0$, is known, Eqs. (2) and (3) can be used to calculate H_c^f/H_c, the ratio of the critical field of the film to the critical field of the bulk material.

It will become apparent in the subsequent discussion that the form of the spatial variation of the magnetic field which is specified by the various nonlocal theories is quite similar to the form given by Eq. (1). In the nonlocal theories, on the other hand, the magnitude is quite different from that indicated by the local London theory. In general, however, the field variation specified by the nonlocal theories can be approximated quite accurately both in form and in magnitude by the London equation if an appropriate effective penetration depth, λ_ϵ, is substituted for the London penetration depth, λ_L. It will be found convenient in much that follows to compare this effective penetration depth of the nonlocal theories, λ_ϵ, (where λ_ϵ will be found to vary both with film thickness and with specimen purity) with the penetration depth of the London theory, λ_L. If, moreover, the forms of the spatial variations of magnetic field do not differ appreciably among the various theories, then, in all cases, the critical field of the film will be related to the effective penetration depth in the manner indicated by Eq. (4) where λ_L is simply replaced by the quantity λ_ϵ.

NONLOCAL THEORIES

J. R. Schrieffer[12] has calculated the variation of magnetic field within a thin film of thickness d as

$$\frac{H(x)}{H_0}=\frac{4}{d}\sum_{n=0}^{n=\infty}\frac{k_n\sin k_nx}{k_n^2+K(k_n)}, \tag{5}$$

where $k_n=(2n+1)\pi/d$, and $K(k)$ is obtained from the transform into wave vector space of the current density-vector potential relationship, i.e.,

$$K(k)=\frac{-4\pi}{c}\frac{\mathbf{j}(k)}{\mathbf{A}(k)}.$$

Schrieffer's equation is obtained under the assumption that the electron scattering at the surface of the specimen is specular. For the case where the film thickness is allowed to become infinitely large, the summation in Eq. (5) may be transformed into an integration and the variation of magnetic field may then be used to obtain the penetration depth for an infinitely thick specimen as

$$\lambda_\infty=\frac{2}{\pi}\int_0^\infty\frac{dk}{k^2+K(k)}. \tag{6}$$

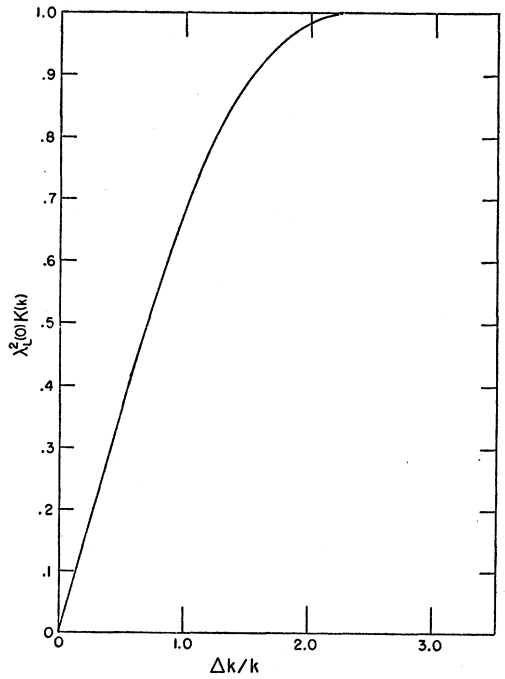

FIG. 1. The BCS kernel, $K(k)$, plotted as a function of $\Delta k/k$, where $\Delta k=\pi/\xi_0$. $K(k)$ has been normalized to unity in the limit as k approaches zero by multiplying by $1/\lambda_L^2(0)$.

[10] P. M. Marcus, Phys. Rev. 88, 373 (1952).

[11] F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), Vol. 1, see p. 130.

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

In the London theory, the supercurrent-vector potential relationship leads to $K(k)=1/\lambda_L^2$, where λ_L is the London penetration depth. $K(k)$ is thus a simple constant independent of k. Substitution of this value of $K(k)$ into Eq. (5) yields the Fourier expansion of an equation of the form of (1) in the interval from $x=0$ to $x=d$.

In the BCS theory, the kernel $K(k)$ is a function not only of the temperature, but of the mean free path of the normal electrons and the superconducting coherence length, ξ_0. $K(k)$ can be expressed in a direct analytical fashion only under certain simplifying assumptions. For the case where $kl\gg1$ (corresponding to a large mean free path in the normal state, i.e., a pure material) the BCS theory gives, at the absolute zero of temperature,

$$\lim_{k\to0}K(k)=1/\lambda_L^2(0),$$

and

$$\lim_{k\to\infty}K(k)=\frac{3}{4}\frac{\pi}{\xi_0}\frac{1}{\lambda_L^2(0)}\frac{1}{k}\left(1-\frac{16}{\pi^3k\xi_0}\ln(\pi k\xi_0)\right).$$

Letting $\Delta k=\pi/\xi_0$, the BCS expression for $K(k)$ in the limit as $kl\gg1$ can be rewritten as

$$\lim_{k\to0}K(k)=1/\lambda_L^2(0),$$

and

$$\lim_{k\to\infty}K(k)=\frac{3}{4}\frac{1}{\lambda_L^2(0)}\frac{\Delta k}{k}\left(1-\frac{16}{\pi4}\frac{\Delta k}{k}\ln\frac{\pi^2k}{\Delta k}\right).$$

Intermediate values of $K(k)$ can be found by graphic interpolation between the limits given above. A plot of $K(k)$ as a function of $\Delta k/k$ is shown in Fig. 1. A variation of $K(k)$ with $1/k$ similar to that shown in Fig. 1 is also given by Pippard's[5,12] nonlocal theory of superconductivity.

In the instance where $kl\ll1$ (corresponding, for example, to an impure material) it is not possible to obtain a simple analytical expression for $K(k)$. Miller[13] has calculated $K(k)$ in the limit as T approaches zero for the cases where $\xi_0>l$ and $\xi_0<l$. The Miller formulas can be used to obtain the asymptotic value of $K(k)$ in the limit as $\Delta k/k$ goes to infinity since the condition $kl<1$ is always satisfied. For the sake of simplicity we have assumed that the shape of the function $K(k)$ is that given by Fig. 1, and that it is only the asymptotic value which is changed by variations of mean free path or specimen purity. The detailed variation of $K(k)$ with k, l, and ξ_0 is thus appropriately left as a challenge to the theoretician.

While it is possible to calculate the asymptotic values of $K(k)$ using the formulae developed by Miller, we have chosen to determine them empirically from experimental data for two reasons. First there exist some uncertainties in the exact values to be used for $\lambda_L(0)$ and ξ_0. These quantities must, in fact, be inferred indirectly from various directly measured quantities and the literature is replete with estimated values for both quantities. Secondly, the Schrieffer equations are derived on the basis of a specular reflection of electrons from the surface of the film. Most measurements on bulk material yield data which numerically is in better agreement with those formula derived on the assumption that the surface scattering is diffuse. For example, Miller's calculations of the penetration depth in bulk materials as a function of the material mean free path are in reasonably good agreement with the values measured by Pippard in experiments with microwaves.[5] In general, the difference between the results obtained by assuming either a specular or diffuse surface scattering appears as numerical constant and the mechanism of surface scattering does not generally alter the qualitative features of the results. Thus it seems reasonable to determine the asymptotic values of $K(k)$ by an empirical method which accords with previous experiment.

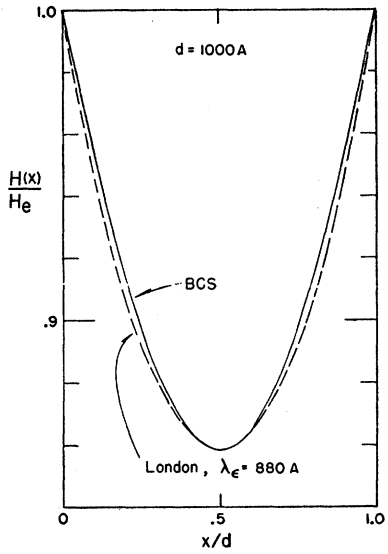

FIG. 2. Variation of the magnitude of magnetic field through the cross section of a 1000 A thick film. The curve calculated from the BCS theory is compared to the curve calculated from the London equation (1) where an effective penetration depth of 880 A is used in the London equation.

In particular, the asymptotic value of $K(k)$ was chosen so that the value of the effective penetration depth for an infinitely thick film [given by Eq. (6)] corresponds to the value of the effective penetration depth calculated by Miller for different mean free paths in a situation where the surface scattering is diffuse. This asymptotic value for $K(k)$ can then be used along with the data contained in Fig. 1 to calculate the field variation through the film as given by Eq. (5). The method, in essence, corresponds to normalizing the BCS kernel $K(k)$ to ensure that the effective pene-

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

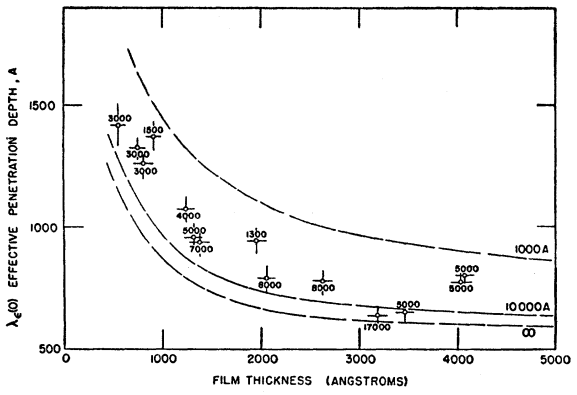

FIG. 3. The variation of effective penetration depth, $\lambda_\epsilon(0)$, as a function of film thickness for a number of bulk material mean free paths. The figures in parenthesis beside each data point indicate the bulk mean free path calculated from the measured low-temperature resistivity of the various films.

tration depth for an infinitely thick film corresponds to the value obtained by Miller for bulk material.

The variation of magnetic field through a film (whose thickness is chosen typically as 1000 A) calculated by the above method is shown in Fig. 2. Figure 2 also contains a plot of the field variation given by the London theory [Eq. (1)] where the effective penetration depth in the London theory has been set equal to 880 A to normalize the two curves at the center of the film. The two curves differ in magnitude by less than $\frac{1}{2}\%$. In view of this correspondence it is possible to use an effective penetration depth (here 880 A) in the London expression for the critical field [as represented by Eq. (4)] and to circumvent the necessity of graphically integrating the BCS curve for $H(x)$ required to obtain the critical magnetic field through Eqs. (2) and (3). Since the form of the magnetic field variation predicted by the BCS theory is not ostensibly different from that of the London theory, the error involved in this procedure is negligible. It is now possible to define an effective penetration depth, λ_ϵ, as being that penetration depth which gives a magnetic field at the center of the film which agrees in magnitude with the field calculated from the BCS theory.

Figure 3 contains the variation of effective penetration depth $\lambda_\epsilon(0)$, calculated as a function of film thickness for a number of bulk material mean free paths. These calculations were made assuming a value of 2.56×10^{-5} cm for ξ_0 and adjusting the ratio of $\xi_0/\lambda_L(0)$ so that the bulk penetration depths calculated from Eq. (6) correspond to the Miller values. The general feature of the results shown in Fig. 3 may be summarized by the statement that the penetration depth is relatively independent of both mean free path and film thickness as long as both are greater than the superconducting coherence length ξ_0. When either the bulk mean free path or the specimen dimensions becomes smaller than the superconducting coherence length the effective penetration depth increases significantly.

EXPERIMENTAL

In order to compare experimentally measured values of the critical magnetic field with those derived theoretically, it is important, if meaningful results are to be obtained, that two experimental conditions be fulfilled. First, it is necessary that the films correspond geometrically to the mathematical model, i.e., that, in so far as is possible, the films correspond to a uniformly thin layer of material bounded by two plain surfaces. Examination of electron microscope surface replicas of various thin films is sufficient to indicate that what may appear to the naked eye as a thin film is frequently a thin agglomeration of particles or crystals which bears no geometrical resemblance to the mathematical model of a film. It is possible, however, to produce a reasonably uniform and flat film by a number of methods. In particular, the tin films reported on here were deposited by relatively rapid vacuum evaporation on either quartz or glass substrates held at liquid nitrogen temperature. Such films, where in the depositing atoms have, after giving up their latent heat of condensation, a limited mobility, are found to consist of extremely small grains or crystals whose dimensions are small compared to the film thickness. The films are thus considerably smoother than films deposited, for example, on warm substrates where, characteristically, the surfaces are rough and the films are composed of large gains. In order to eliminate effects due to a varying film thickness at the edges of the film, the lines used in the critical field measurements were mechanically cut from a larger area of film with a diamond tip mounted in a ruling engine.

The second requirement, if the experimental results are to be meaningful, is that the true thermodynamic transition between states be determined. The most convenient method of observing the transition in a thin film is to measure the change in resistance as the film is driven normal. It is difficult to be certain that a measurement of the transition made in this manner really corresponds to the actual thermodynamic transition of the material and not to the transition of some filament or substructure within the film.[14] While it is not possible to be completely certain that the true thermodynamic transition was actually observed, the following facts suggest that this was actually the case for the films reported here.

1. The transitions were relatively sharp and generally took place over a field interval amounting to 1 or 2% of the critical field with no measurable hysteresis between the superconducting to normal and the normal to superconducting transition.

2. The transitions were surprisingly reproducible (to

[14] W. B. Ittner and J. F. Marchand, Phys. Rev. **114**, 1268 (1959).

within several percent) among a series of four lines all laid down in a single evaporation. Moreover transitions measured after successive edge cuts (corresponding to different widths of a single line) were essentially identical.

3. The transitions as determined by resistance measurements were essentially independent of measuring current over a current range of several orders of magnitude.[15]

The measurements of critical field were carried out with conventional cryogenic apparatus. The greatest possible source of error in the critical field-temperature determinations lies in the determination of the critical temperature T_c. Near the critical temperature the low critical fields become comparable to the stray magnetic fields which are present and the geometry of the specimen is such that fields perpendicular to the plane of the film are tremendously magnified near the specimen edges. It is virtually impossible to prevent the specimen from entering an intermediate state where the transitions are greatly influenced by the measuring current. In short, it is quite difficult to determine the critical temperature to closer than about 5 or 10 millidegrees. Since the penetration depth varies most rapidly with temperature near T_c, uncertainties in T_c lead to large uncertainties in the temperature variation of the penetration depth. For this reason the data has only been used to look at the variation of penetration depth with temperature at temperatures such that $T/T_c<0.9$.

Following the measurements of critical field, the thickness of the film, d, was measured by silvering the entire sample and using a standard interferometer technique to measure the distance between the film surface and the substrate. The film thickness was also calculated by a method which involves measuring the temperature variation of the resistance, separating the residual from the temperature dependent resistance, and assuming the temperature dependent resistivity to be equal to the bulk temperature dependent resistivity. (Some credence is lent to this assumption by the fact that the temperature dependence of the resistivity in these films was essentially identical to that of bulk material.) Unfortunately, while there were some notable exceptions, the optically measured thickness frequently differed from the calculated film thickness by as much as 10% and it must be concluded that the thickness was, in most instances, not known with great accuracy.

RESULTS

The experimentally determined critical fields, $H_c'(T)$, were used, along with bulk values of $H_c(T)$,[16] to determine the ratio $\lambda_\epsilon(T)/d$ as it is given by Eq. (4). It is then possible to examine the ratio $\lambda_\epsilon(T)/\lambda_\epsilon(0)$ by an appropriate choice of $\lambda_\epsilon(0)/d$; and, by using an experimentally determined value for d, to determine $\lambda_\epsilon(0)$.

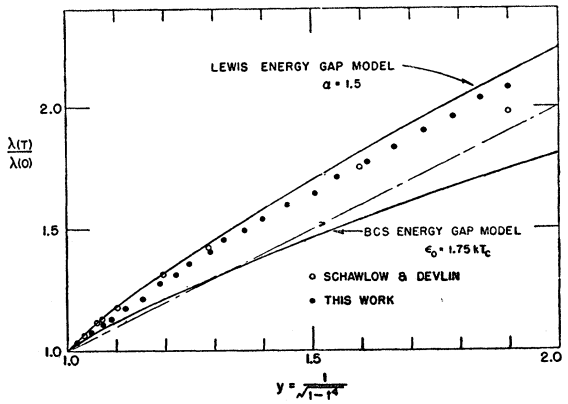

FIG. 4. The temperature variation of the penetration depth as a function of the parameter, y. The solid curves correspond to the temperature variations predicted by the Lewis energy gap model and the BCS theory, while the dotted line corresponds to the variation used in the Gorter-Casimir two fluid model of superconductivity.

In order to choose an appropriate value for $\lambda_\epsilon(0)/d$, it is desirable to choose some sort of model for the variation of λ_ϵ with temperature and to extrapolate measurements of $\lambda_\epsilon(T)$ made at finite temperatures to $\lambda_\epsilon(0)$ at the zero of temperature. Lewis[17] has calculated the temperature variation of the penetration depth which would result if the superconducting state were separated from the normal state by a temperature independent energy gap. While the Lewis model differs appreciably from the BCS model (in which the energy gap varies with temperature and disappears at the critical temperature) the former model does predict a temperature variation of the electronic specific heat which is in rough agreement with theory and does predict a variation of penetration depth with temperature which is in good agreement with the measurements made by Schawlow and Devlin[18] on bulk tin. Since our measurements of the variation of the penetration depth with temperature are quite similar to those found by Schawlow and Devlin we have chosen to use the Lewis model to determine the ratio $\lambda_\epsilon(0)/d$. This has been done by reducing a series of values of $\lambda_\epsilon(T)/d$ to $\lambda_\epsilon(0)/d$ and averaging the results to obtain the final value.

Figure 4 contains a plot of the quantity $\lambda_\epsilon(T)/\lambda_\epsilon(0)$ as a function of the parameter $y=(1-t^4)^{-\frac{1}{2}}$. The variation shown in Fig. 4 was typical of the results obtained on a number of films ranging in thickness from about 500 to 5000 angstroms. Within the experimental errors, it was impossible to distinguish any change in the variation of $\lambda_\epsilon(T)/\lambda_\epsilon(0)$ with y as a function of the film

[15] This is true except for measurements very close to the critical temperature where the field produced by the measuring current becomes comparable to the critical field of the specimen. Near T_c the shape of the transition may be quite sensitive to the magnitude of the measuring current.

[16] J. M. Lock, A. B. Pippard, and D. Shoenberg, Proc. Cambridge Phil. Soc. **47**, 811 (1951).

[17] H. W. Lewis, Phys. Rev. **102**, 1508 (1956).

[18] A. L. Schawlow and G. E. Devlin, Phys. Rev. **113**, 120 (1959).

thickness. The temperature variation of penetration depth is essentially that found by Schawlow and Devlin whose data is also shown in Fig. 4.

According to the constant energy gap model, the electronic specific heat would be expected to vary as $\exp(-\alpha T_c/T)$ where for tin, α is experimentally[19] close to 1.5. The temperature variation of the penetration depth in the tin films reported here actually fits best a constant energy gap model in which α is equal to 1.7, although the experimental errors preclude our attaching any real significance to this fact. The temperature variation of the penetration depth is, on the other hand, in rather poor agreement with the predictions of the BCS theory as is evident from an examination of Fig. 4. The BCS variation of the penetration depth which appears in Fig. 4 is that predicted for a bulk material in which there is a single energy gap approaching $3.5Tk_c$ at the absolute zero of temperature. While the BCS theory gives a somewhat different variation of the penetration depth with temperature for a thin film,[13] the difference is not enough to account for the discrepancy shown in Fig. 4. Moreover, the data of Schawlow and Devlin obtained by measurements on bulk material is also in poor agreement with the BCS theory. Recent measurements[20] of the ultrasonic attenuation in bulk superconductors have indicated that the energy gap in tin may take on different values in different crystal orientations and this suggests that it is probably not correct to use a single value for the energy gap as it appears in the theory. Just how real crystalline anisotropies in the energy gap will modify the temperature variation of the penetration depth is not yet clear but it is reasonable to assume that an effect will be evident.

J. M. Lock[21] has pointed out that a variation of the superconducting penetration depth with magnetic field would be expected to make itself evident in the penetration depths inferred from critical field measurements on thin films. The available evidence,[22] however, indicates that in bulk materials the critical field is able to change the penetration depth by only a few percent. Conceivably the effect could be greater in thin films, but at present there is no real evidence for or against this hypothesis. At the moment, therefore, the temperature variation of the penetration depth is not understood and further work on this problem is obviously needed.

The variation of $\lambda_\epsilon(0)$ with film thickness is shown for a variety of films in Fig. 3. The experimental uncertainties indicated in the figure arise almost entirely from uncertainties in the measured film thicknesses. It is seen that in a qualitative manner, the effective penetration depths do increase for the thinner films. The effective mean free paths (indicated in parenthesis) of the bulk material of which the various films are composed have been estimated by using the measured low temperature resistivity, Fuchs' formula,[23] and Chamber's value[24] of the ratio of σ/l for tin. Several of the films were found to have, inadvertently, somewhat higher resistivities and, consequently, somewhat lower mean free paths than the majority of the films studies. The impurer films exhibited a somewhat higher effective penetration depth in accord with theoretical expectations. While the data shown in Fig. 3 is qualitatively in good agreement with the theoretical predictions, the quantitative agreement is occasionally poor. This is perhaps not too surprising. It is known, for example, that T_c and H_c for bulk tin depend to some extent on the purity of the material. Likewise it might be suspected that ξ_0 is a function of the specimen purity. In calculating the theoretical values of $\lambda_\epsilon(0)$ we have taken the $H_c(t)$ curve for pure bulk tin and a constant value for ξ_0. It is possible, of course, to carry out the theoretical development under a variety of assumptions regarding the variation of $K(k)$, ξ_0, and $H_c(t)$ with the mean free path. There is at the moment, however, insufficient evidence to indicate the most profitable extension of the present development. It would, of course, ultimately be desirable to carry out the thin film calculations under the assumptions of a diffuse surface scattering mechanism. At the moment, it is sufficient to say that the experimental data overwhelmingly supports a nonlocal picture and indicates that the penetration depths in thin films is considerably higher than is predicted by the London picture.

ACKNOWLEDGMENTS

The author is deeply indebted to R. G. Blumberg for bringing this problem to his attention and for making available the thin film data on which this analysis is based. In addition the author would like to thank Dr. A. M. Toxen and Dr. J. C. Swihart for many stimulating and fruitful discussions.

[19] W. S. Corak, B. B. Goodman, C. B. Satterthwaite, and A. Wexler, Phys. Rev. **102**, 656 (1956); **102**, 662 (1956).
[20] R. W. Morse, T. Olsen, and J. D. Gavenda, Phys. Rev. Letters **3**, 1, 15 (1959).
[21] J. M. Lock (private communication).
[22] See, for example, M. Spiewak, Phys. Rev. **113**, 1479 (1959).
[23] K. Fuchs, Proc. Cambridge Phil. Soc. **34**, 100 (1938).
[24] R. G. Chambers, Proc. Roy. Soc. (London) **A203**, 378 (1950).