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CRITICAL FIELDS OF THIN SUPERCONDUCTING FILMS. III. THE CASE $L \sim l$. COMPARISON WITH EXPERIMENT

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The dependence of the critical field strength is investigated in detail for a case which is intermediate with respect to those previously studied, viz. when the film thickness is of the order of the electron volume mean free path. Comparison of the results with experiment yields the BCS correlation parameter and the volume mean free path. The results are also compared with published experimental data on critical fields of thin films of tin, indium, and mercury.

CRITICAL magnetic fields of thin films for diffuse and specular reflection of electrons from a film surface for various impurity concentrations have been obtained in the work of one of the authors[1] (quoted below as I and II; see also the paper by de Gennes and Tinkham[2]). However, previously obtained results refer only to the various possible and very numerous limiting cases which are usually not realized under experimental conditions. The only exception is the case of Maki[3] ($l \ll L$, l is the mean free path and L the film thickness) which occurs for not too thick unannealed films when the linearized Ginzburg-Landau equation is applicable in its original (for $1 - T/T_c \ll 1$) or modified form (at arbitrary temperatures).

In present-day techniques of preparing thin films the volume mean free path is usually comparable with the film thickness; in other words, we are dealing with a case which is intermediate between those considered by de Gennes and Tinkham and Maki. The expression for the critical field applicable in this instance (assuming diffuse reflection of the electrons from the surface) has been obtained in the Appendix. The equation for the critical fields is of the form

$$\left(\frac{eH}{c\hbar}\right)^2 = \frac{128}{\pi}\frac{T_c}{\hbar v L^3} f_1\left(\frac{L}{l}\right) F\left(\frac{T}{T_c}\right) \approx \frac{7.35}{\xi_0 L^3} f_1\left(\frac{L}{l}\right) F\left(\frac{T}{T_c}\right) \quad (1a)$$

or

$$\left(\frac{eH}{c\hbar}\right)^2 = \frac{72}{\pi}\frac{T_c}{\hbar v l L^2} f_2\left(\frac{l}{L}\right) F\left(\frac{T}{T_c}\right) \approx \frac{4,14}{\xi_0 l L^2} f_2\left(\frac{l}{L}\right) F\left(\frac{T}{T_c}\right). \quad (1b)$$

The functions $f_1(x)$, $f_2(x)$, and $F(t)$ are defined in the Appendix [(A.9), (A.14), and (A.13) respectively] and their plots are shown in Figs. 1–3. The limiting values of these functions are as follows:

$$f_1(x) = \begin{cases} 1 + {}^8/_{15}x, & x \ll 1 \\ {}^{81}/_{128} + {}^9/_{16}x, & x \gg 1 \end{cases}, \quad (2)$$

$$f_2(x) = \begin{cases} 1 + {}^8/_9 x, & x \ll 1 \\ {}^{128}/_{135} + {}^{16}/_9 x, & x \gg 1 \end{cases}, \quad (3)$$

$$F(t) = \begin{cases} \pi^2/8\gamma, & t = 0 \\ 1 - t, & 1 - t \ll 1 \end{cases}. \quad (4)$$

It is seen from the graphs that the functions $f_1(x)$ and $f_2(x)$ differ little from linear functions; they can be approximated with good accuracy by the straight lines:

$$f_1(x) \approx 1 + 0.52x, \quad (5)$$

$$f_2(x) \approx 0.9 + 1.78x. \quad (6)$$

One can readily verify that in the limiting cases $l \ll L$ and $l \gg L$ Eqs. (1a) and (1b) are equivalent to expressions (3.15) and (3.22) of I. For this it is sufficient to utilize the corresponding limiting values (2) and (3) of the functions $f_1(x)$ and $f_2(x)$.

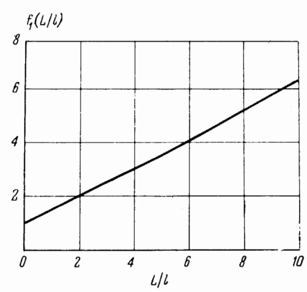

FIG. 1. Plot of the function $f_1(x)$.

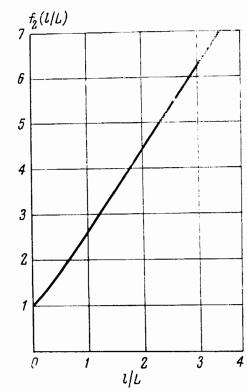

FIG. 2. Plot of the function $f_2(x)$.

Strictly speaking, expressions (1a) and (1b) are applicable if the following conditions

$$\min\{\xi_0^2, l^2\} \lesssim \xi_0 L / (1 - T/T_c), \quad (7)$$

$$L^2 p_0 \gg \min\{\xi_0, l\}, \quad (8)$$

$$(eH/c\hbar)L^2 \ll 1. \quad (9)$$

are fulfilled. The fulfillment of conditions (7) and (8) is in practice uniquely related to the temperature de-

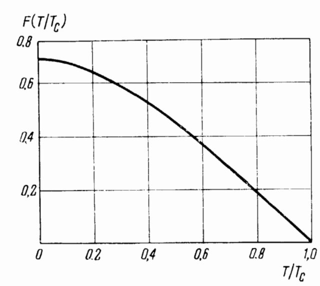

FIG. 3. Plot of the function F(t).

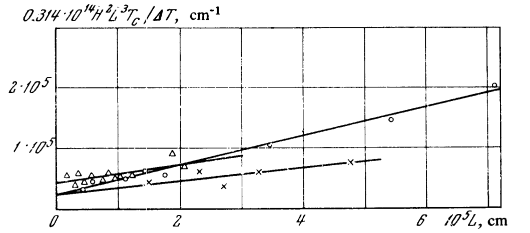

FIG. 4. Experimental results on the critical fields of thin films of tin treated with the use of the function $f_1(x)$: X – data of [7], O – data of [5], Δ – data of [6].

pendence of the critical field: when $H^2(T)$ is proportional to $F(T/T_c)$ (at least in the narrow temperature range near T_c where $F = \Delta T/T_c$) then if condition (9) is fulfilled relations (1a) and (1b) are valid, whereas in the opposite case they are not valid. The restriction (9) is not so essential, since the case when $eHL^2/c\hbar \gg 1$ corresponds (for the same temperature dependence of the critical field) to the region where the Ginzburg-Landau theory is applicable. This case has been studied in detail in Abrikosov's papers.[4]

A comparison of expressions (1a) and (1b) obtained by us with experimental data provides information about the value of the BCS parameter ξ_0 and the volume mean free path l if it is assumed that the volume mean free path is (in a given series of experiments) the same for films of various thicknesses. By plotting the dependence of $H^2L^3/F(T/T_c)$ on L and of $H^2L^2/F(T/T_c)$ on 1/L from experimental data we obtain curves which should correspond to the functions f_1 (Fig. 1) and f_2 (Fig. 2), i.e., they should approximately be straight lines. From the slope of these straight lines and the values of these functions for zero argument one can determine ξ_0 and l.

We have thus investigated the published experimental data on the critical magnetic fields of thin films of tin, indium, and mercury. The data of Zavaritskiĭ,[5] Blumberg,[6] and of one of the authors[7] on the critical fields of thin films of tin processed by the indicated method are presented in Figs. 4 and 5; the data of Appleyard[8] for thin films of mercury are presented in Figs. 6 and 7. As can be seen from these figures, the spread of the experimental results is very considerable; this is apparently connected with errors in determining the film thickness (the thickness appears in the formula as a square and cube). The large spread is also possibly due to differences in the method of preparation of films of various thicknesses—a small difference in the technique and in the temperature of deposition and annealing can noticeably affect the mean free path of the electrons.

In Figs. 4 and 6 the quantity $(eH/c\hbar)^2L^3/7.35F(T/T_c)$ is plotted as a function of L[1)] so that the obtained dependence should correspond to $\xi_0^{-1}f_1(L/l)$. If it is approximated by a straight line, then the ordinate for L = 0 is according to (1a) and (6) equal to the reciprocal of the BCS correlation parameter ξ_0^{-1} and the slope is equal to $0.55/\xi_0 l$. Analogously, the dependence of

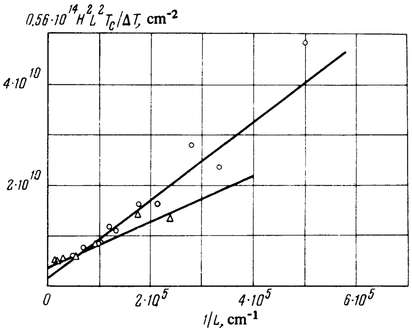

FIG. 5. Experimental results on the critical fields of thin films of tin treated with the use of the function $f_2(x)$: Δ – data of [5], O – data of [6].

$(eH/c\hbar)^2L^2/4.14F(T/T_c)$ on 1/L[2)] corresponding to $(l\xi_0)^{-1}f_2(l/L)$ is given on Figs. 5 and 7. If it is approximated by a straight line then the value of the ordinate for zero argument is equal [in accordance with (1b) and (6)] to $0.9(l\xi_0)^{-1}$ and the slope corresponds to $1.78\xi_0^{-1}$.

We note the clear discrepancy between the observed results and the Ginzburg-Landau theory: this theory would lead in Figs. 4 and 6 to a straight line passing through the origin, and in Figs. 5 and 7 to a horizontal straight line.

The values of ξ_0 and l which we obtained as a result of such a treatment are tabulated. Since the values of ξ_0 and l for tin determined by two methods [with the use of the functions $f_1(x)$ and $f_2(x)$] do not differ appreciably (~10 percent), we present their average values. The considerable discrepancy between the values of ξ_0 obtained from the data of Zavaritskiĭ[5] and Blumberg[6] can be explained by differences in the method of preparation of the samples and in the measurement of their thickness: a systematic error of the order of 15–20% in determining the thickness will lead to precisely such a discrepancy in the values of ξ_0. We note that the data of[7] treated in the same way led to approximately the same value of ξ_0 as the results of Zavaritskiĭ. The same method of determining the film thickness was used in these papers.

The two methods of processing the experimental data described above lead in the case of mercury to somewhat different values of ξ_0 and l (based on the data of Appleyard[8]); both results are therefore presented in the Table. The table also includes values of ξ_0 and l for indium, obtained from the data of Toxen.[9]

For comparison the table presents values of ξ_0 from Lynton's book[10] calculated according to the formula

$$\xi_0 = 0.18\hbar v / T_c, \tag{10}$$

where the velocity v on the Fermi surface was obtained from data on the anomalous skin effect, as well as values of ξ_0 which we obtained by recalculating the data of Faber[11] on the critical supercooling field in accordance with the formula

$$\xi_0^2 = \frac{24\gamma^2}{7\pi^2\zeta(3)} \frac{c\hbar}{eH_{c1}} \frac{\Delta T}{T_c} \approx 0.918 \frac{c\hbar}{eH_{c1}} \frac{\Delta T}{T_c}. \tag{11}$$

1) Since in the absolute system $(e/c\hbar)^2 = 2.31 \times 10^{14}$, measuring the field in Oersteds and the film thickness in centimeters, we plot along the ordinate axis the quantity $0.3^{14} \times 10^{14} H^2 L^3 T_c/\Delta T$ [cm^{-1}].

2) If the field is measured in Oersteds and the sample thickness in centimeters, then one plots on the ordinate axis 0.56×10^{14} $H^2 L^2 T_c/\Delta T$ [cm^{-2}].

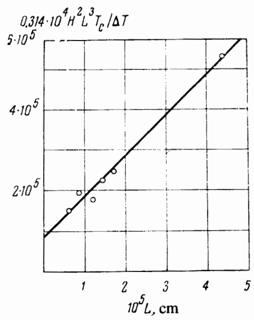

FIG. 6

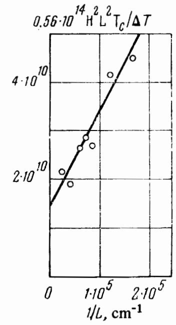

FIG 7.

FIG. 6. Experimental results on the critical fields of mercury films [8] treated with the use of the function $f_1(x)$.

FIG. 7. Experimental results on the critical fields of mercury films [8] treated with the aid of the function $f_2(x)$.

$l \cdot 10^5$, cm	$\xi_0 \cdot 10^5$, cm	Method of determination	Experimental data
		Tin	
0.6	4,0	From the critical magnetic fields of the films	[5]
1.1	3,8		[6]
2,0	2,3		[7]
—	2,3	From the anomalous skin effect	[10]
—	4,2	From the supercooling field	[11]
		Mercury*	
0,5	1,2	From the critical magnetic fields of the films	[8]
0,7	0,9		
		Indium	
0,8	3,4	From the critical magnetic fields of the films	[9]
—	4,4	From the anomalous skin effect	[10]
—	5,2	From the supercooling field	[11]

*Upper line—data obtained with the use of $f_1(L/l)$, and lower line—with the use of $f_2(l/L)$.

The considerable discrepancy between the values of ξ_0 for tin obtained from the data of [10] and [11] is apparently connected with the strong anisotropy of tin.

In conclusion, it should be noted that in complex investigations of thin films it is extremely desirable to measure the residual resistivity of the investigated films in order to be able to check the volume mean free path. The accuracy in the determination of the thickness of the samples should be sufficiently high, and the method of preparing films of various thicknesses must be absolutely identical, in order to insure that the volume mean free path be independent of the sample thickness.

APPENDIX

The problem of finding the critical field reduces, as is well known, to finding the maximum field for which there exists a nontrivial solution of the integral equation

$$\frac{1}{v|\lambda|}\Delta^*(z_1) = \int_{-L/2}^{+L/2} K(z_1, z_2)\Delta^*(z_2)\,dz_2, \quad (A.1)$$

where the kernel of the integral equation can be represented in the form

$$K(z_1, z_2) = 4\pi T \sum_{\omega>0} \Phi(\omega; z_1, z_2). \quad (A.2)$$

The functions $\Phi(\omega; z_1, z_2)$ for pure films have been obtained for diffuse and specular reflection laws in I and II. Analogous functions for contaminated films $\Phi_\tau(\omega; z_1, z_2)$ can be found, as was shown in I, from the integral equation[3)]

$$\Phi_\tau(\omega; z_1, z_2) = \Phi(\omega + 1/2\tau; z_1, z_2)$$
$$+ \frac{1}{\tau}\int_{-L/2}^{+L/2} \Phi\left(\omega + \frac{1}{2\tau}; z_1, z'\right)\Phi_\tau(\omega; z', z_2)\,dz'. \quad (A.3)$$

If we expand the function $\Phi(\omega; z_1, z_2)$ which is symmetrical with respect to z_1 and z_2 in the eigenfunctions:

$$\Phi(\omega; z_1, z_2) = \sum_n \frac{\varphi_n(\omega, z_1)\varphi_n(\omega, z_2)}{\lambda_n(\omega)}, \quad (A.4)$$

then

$$\Phi_\tau(\omega; z_1, z_2) = \sum_n \frac{\varphi_n(\omega + 1/2\tau, z_1)\varphi_n(\omega + 1/2\tau, z_2)}{\mu_n(\omega)}, \quad (A.5)$$

where

$$\frac{1}{\mu_n(\omega)} = \frac{\tau}{\tau\lambda_n(\omega + 1/2\tau) - 1}. \quad (A.6)$$

As has been shown in I, for $eHL^2 \ll 1$ the solution of Eq. (A.1) corresponding for given T to maximum H (or for given H to maximum T) does not depend on the z coordinate, so that only the functions $\varphi_0(\omega, z)$ which are almost independent of z are in this instance important in the kernel $K(z_1, z_2)$ of (A.2). Then, taking into account (A.5) and (A.6), the equation for the critical field takes on the following form:

$$\frac{1}{v|\lambda|} = 4\pi T \sum_{\omega>0} \frac{\tau}{\tau\lambda_0(\omega + 1/2\tau) - 1}. \quad (A.7)$$

The maximum eigenvalue of $\lambda_0(\omega)$ can be found by integrating the general expression for $\Phi(\omega; z_1, z_2)$[4)] obtained in I [see (A.1)] over z_1 and z_2 with the condition $eHL^2 \ll 1$:

$$\frac{1}{\lambda_0(\omega)} = \frac{1}{2\omega} - \frac{(eH)^2 L^3 v}{32\omega^2} f_1^{-1}\left(\frac{\omega L}{v}\right), \quad (A.8)$$

where

$$f_1^{-1}(x) = 4x^2 \int_{-1/2}^{+1/2}\int d\xi_1\, d\xi_2 \int_1^\infty \frac{d\mu}{\mu}(\mu^2 - 1)(\xi_1^2 - \xi_2^2)^2 \exp[-2x\mu|\xi_1 - \xi_2|]$$
$$+ 8x\int_{-1/2}^{+1/2} d\xi \int_1^\infty \frac{d\mu}{\mu}\left(\mu - \frac{1}{\mu}\right)\left(\frac{1}{4} - \xi^2\right)^2 \exp\left[-2x\mu\left(\xi + \frac{1}{2}\right)\right]. \quad (A.9)$$

These expressions are not difficult to integrate; as a result one obtains a very cumbersome formula which we do not present here. The limiting values of $f_1(x)$ have been presented above in (2); the graph of the function is shown in Fig. 1.

[3)] Here as in I and II we consider only the case of isotropic scattering on the impurities. As was shown by Maki [3] and by de Gennes and Tinkham, [2] for very contaminated films the critical fields depend on the transport times and the mean free path.

[4)] We consider here the more realistic case of diffuse reflection. Specular reflection has been studied by Thompson and Baratoff. [12,13] It appears useful to compare the experimental data also with their results. [12,13] However, in view of the large spread of the experimental points, it has so far been impossible to differentiate between diffuse and specular reflection on the basis of experiments on the critical fields of thin films.

Substituting (A.8) in (A.7) and taking into account the fact that for

$$(eH)^2L^3l \ll 1 \qquad (A.10)$$

one can neglect the second term of (A.8) (if $\omega \gtrsim 1/\tau$), we obtain the equation

$$\frac{1}{\nu|\lambda|} = 4\pi TL \sum_{\omega>0}\left[2\omega + \frac{(eH)^2L^3v}{16} f_1^{-1}\left(\frac{L}{l}\right)\right]^{-1}. \qquad (A.11)$$

Summing over ω with account of the cut-off at the Debye frequencies, we find an equation for the critical field:

$$\ln\frac{T}{T_c} = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{(eH)^2L^3v}{64\pi T} f_1^{-1}\left(\frac{L}{l}\right)\right). \qquad (A.12)$$

From (A.12) one can obtain H as an implicit function of the temperature. If one introduces the function $F(T/T_c)$ defined by the expression

$$\ln t = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{2}{\pi^2 t} F(t)\right), \qquad (A.13)$$

then one obtains for the critical field the formula (1a). The graph of the function $F(t)$ is shown in Fig. 3 and its limiting values are given in (4).

In place of $f_1(x)$ one can introduce the function

$$f_2(x) = \frac{16}{9x} f_1\left(\frac{1}{x}\right). \qquad (A.14)$$

The critical field is then given by expression (1b).

The limits of applicability of (1a) and (1b) [see (7) and (8)] follow from (A.10) and from the condition of applicability of the quasi-classical approximation (see I and II) respectively.

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