THIRD CRITICAL FIELD OF A SUPERCONDUCTOR WITH A FILM ON THE SURFACE

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The third critical field is calculated for a system consisting of a thin superconducting film deposited on the plane surface of a bulky superconductor. It is assumed that the film and backing differ only with respect to the electron mean free path. The two extreme cases of thin and thick films are considered. A qualitative explanation is given for the experimental dependence of the third critical field of the superconductor on the mechanical and heat treatment of the surface.

1. INTRODUCTION AND STATEMENT OF THE PROBLEM

THE existence of a third critical field H_{c3} for superconductors was predicted theoretically by Saint-James and de Gennes;[1] later, this effect was discovered experimentally by many investigators. However, the experimentally determined ratio H_{c3}/H_{c2} is rarely close to the theoretical value of 1.69.

Well-annealed alloys of Nb-Ta give $H_{c3}/H_{c2} = 1.71$.[2] Gygax et al.[3] measured this ratio for an indium-lead alloy. It was found that for samples melted under vacuum and annealed for 100 days at room temperature, $H_{c3}/H_{c2} = 4$. After pickling, this ratio became equal to 3, and after a 60-hour tempering at 110°C, it took on the value 1.69 ± 10%.

Thus, the quality of the surface has a great effect on the value of the third critical field. By the word "quality" is meant here the amount of impurities, lattice distortions, dislocations, and other defects in the surface layer of the sample.

In the present work, the problem was to find H_{c3} in the case in which the plane surface of a bulky superconductor is "contaminated" by atoms of a different type, dislocations, and distortions, which decrease the mean free path of the electrons. In other words, the problem was to find H_{c3} for a system consisting of a bulky superconductor on which a surface film is deposited, where the material of this film has the same critical temperature and critical thermodynamic field as in the bulk of the substrate, but in accord with Gor'kov,[4] has a larger value of the parameter κ of the Ginzburg-Landau theory [5] than the substrate.

Let the parameter κ for the bulky material of the substrate be equal to κ_2. A film of thickness d is applied to the plane surface. The material of this film has the parameter κ_1. In what follows, the index 1 always refers to the film and the index 2 to the bulky material of the substrate. The plane boundary of the film with the vacuum coincides with the plane $z = 0$. The film with the substrate occupies the half-space $z > 0$. All the electron characteristics of the materials 1 and 2 are identical, except for the mean free paths of the electron, which are respectively equal to l_1 and l_2. A very large external magnetic field H_0 is applied parallel to the surface of the film, for which the entire system is in the normal state. Let us begin to decrease the field H_0. The problem is to find the field for which the superpenetrability into the surface layer begins to set in. This field we shall denote by H_{c3}.

Inasmuch as we shall limit ourselves to the neighborhood of T_c in what follows, we shall solve the stated problem by the methods of the Ginzburg-Landau theory.[5] Near H_{c3}, the value of Ψ is small; therefore, we can omit the cubic term in Ψ relative to the Ψ term in the Ginzburg-Landau equation for Ψ, thus linearizing the problem.

Furthermore, if we choose (by using the gauge invariance of the equations) such a gauge of the vector potential A that Ψ becomes real, then the set of initial equations takes the form

$$-\frac{\hbar^2}{2m}\frac{d^2\Psi_i}{dz^2}+\frac{2e^2}{mc^2}A^2\Psi_i+a_i\Psi_i=0, \quad (1)$$

$$\frac{d^2A}{dz^2}-\frac{16\pi e^2}{mc^2}\Psi_i^2A=0, \quad i=1,2. \quad (2)$$

As has already been pointed out, the index 1 correspond to the film and the index 2 to the substrate. Here a is the coefficient in the expansion of the free energy in powers of $|\Psi|^2$: $F_s = F_n + a|\Psi|^2 + \frac{1}{2}b|\Psi|^4$, e is the electron charge, m the effective mass of the electron, and c the velocity of light.

The boundary conditions of the problem are

$$H|_{z=0}=H_0, \quad \left.\frac{d\Psi_1}{dz}\right|_{z=0}=0, \quad \Psi_2|_{z=\infty}=0. \quad (3)$$

Moreover, we must also state the conditions of joining together the functions Ψ_1 and Ψ_2 on the boundary separating materials 1 and 2. For this purpose, we use for $z = d$ the boundary conditions for Ψ and $d\Psi/dz$ obtained by Zaĭtsev:[6]

$$\frac{1}{\sqrt{\chi_1}}\Psi_1=\frac{1}{\sqrt{\chi_2}}\Psi_2, \quad \sqrt{\chi_1}\frac{d\Psi_1}{dz}=\sqrt{\chi_2}\frac{d\Psi_2}{dz}.$$

Here χ is the dimensionless function of the mean free path of the electrons, introduced in the work of Gor'kov.[4] It is equal to unity for $l \to \infty$ and tends to zero as $l \to 0$. The constants of the Ginzburg-Landau theory—κ for the alloy and κ_0 for the pure metal (here the alloy differs from the pure metal only by the short free path of the electrons)—are connected by the relation

$$\varkappa=\varkappa_0/\chi. \quad (4)$$

In the zeroth approximation, $\Psi = 0$ and Eq. (2) is solved with account of the boundary conditions (3):

$$A=H_0(z-z_0), \quad (5)$$

where z_0 is the constant of integration.

We simplify Eq. (1) by transformation to dimensionless units. As was done by Abrikosov in [7], we choose the quantity $(\hbar c/2eH_0)^{1/2}$ for the unit of length:

$$\xi = \sqrt{2eH_0/\hbar c}\, z.$$

We now turn our attention to the fact that this unit of length depends only on the magnetic field H_0 but not on the material of the sample. We transfer the origin of the coordinates to the point $z = z_0$. Now Eq. (1) takes the particularly simple form:

$$d^2\Psi_i / d\xi^2 + (\beta_i - \xi^2)\Psi_i = 0, \qquad i = 1, 2, \tag{6}$$

where

$$\beta_i = \frac{m|a|c}{e\hbar H_0} = \frac{\sqrt{2}\,\varkappa_i H_{cm}}{H_0} = \frac{H_{c2}(i)}{H_0}. \tag{7}$$

Here H_{cm} is the critical thermodynamic field, $H_{c2}(i)$ is the second critical field of the ith material. The boundary conditions and the joining conditions are

$$\left.\frac{d\Psi_1}{d\xi}\right|_{\xi=-\eta} = 0, \quad \Psi_2|_{\xi\to\infty} = 0, \quad \sqrt{k}\,\Psi_1|_{\xi=s-\eta} = \Psi_2|_{\xi=s-\eta},$$
$$\frac{1}{\sqrt{k}}\left.\frac{d\Psi_1}{d\xi}\right|_{\xi=s-\eta} = \left.\frac{d\Psi_2}{d\xi}\right|_{\xi=s-\eta}. \tag{8}$$

Here $\eta = (2eH_0/\hbar c)^{1/2} z_0$, $s = (2eH_0/hc)^{1/2} d$, i.e., η and s are equal to z_0 and d, but expressed in dimensionless units; furthermore, we have introduced the notation $k = \kappa_1/\kappa_2$.

The general solution of Eq. (6) will be:

$$\Psi_1 = e^{-\xi^2/2}\left[C_1\Phi\left(\frac{1-\beta_1}{4}, \frac{1}{2}; \xi^2\right) + C_2\xi\Phi\left(\frac{3-\beta_1}{4}, \frac{3}{2}; \xi^2\right)\right], \tag{9}$$

$$\Psi_2 = e^{-\xi^2/2}\left[C_3\Phi\left(\frac{1-\beta_2}{4}, \frac{1}{2}; \xi^2\right) + C_4\xi\Phi\left(\frac{3-\beta_2}{4}, \frac{3}{2}; \xi^2\right)\right]. \tag{10}$$

Here Φ is the confluent hypergeometric function, and $C_1, \ldots, C_4$ are arbitrary constants. The four conditions (8) permit us to eliminate these constants and establish the functional relation between the parameters β_i and η. The critical field for the appearance of surface superconductivity H_{c3} will correspond (see [7]) to the maximum value of the external field for which there still exists a nontrivial solution of Eq. (6). As in [7], this field is determined by ascertaining whether the function $\beta_i(\eta)$ has a minimum, since $\beta_i = H_{c2}(i)/H_0$. Simultaneously, the equilibrium value of η_0 at which β_i reaches a minimum is determined. Getting ahead of ourselves, we can state that η_0 is the expression, in dimensionless form, for the distance between the boundary with the vacuum and the place where the density of the superconducting electrons in the superconducting surface layer is a maximum.

Using the condition (8) and the asymptotic expansion of the confluent hypergeometric function for large ξ, we eliminate all four coefficients C and obtain the following equation:

$$\Big[\sqrt{k}\,F_\alpha(\xi_1)\Psi_\gamma'(\xi_1) - \sqrt{k}\,\xi_1 F_\alpha(\xi_1)\Psi_\gamma(\xi_1) - \frac{1}{\sqrt{k}}F_\alpha'(\xi_1)\Psi_\gamma(\xi_1) \tag{11}$$
$$+ \frac{1}{\sqrt{k}}\xi_1 F_\alpha(\xi_1)\Psi_\gamma(\xi_1)\Big][\xi_0^2\Phi_\alpha(\xi_0) - \Phi_\alpha(\xi_0) - \xi_0\Phi_\alpha'(\xi_0)]$$
$$= \Big[\frac{1}{\sqrt{k}}\Phi_\alpha(\xi_1)\Psi_\gamma(\xi_1) + \frac{1}{\sqrt{k}}\xi_1\Phi_\alpha'(\xi_1)\Psi_\gamma(\xi_1) - \frac{1}{\sqrt{k}}\xi_1^2\Phi_\alpha(\xi_1)\Psi_\gamma(\xi_1)$$
$$- \sqrt{k}\,\xi_1\Phi_\alpha(\xi_1)\Psi_\gamma'(\xi_1) + \sqrt{k}\,\xi_1^2\Phi_\alpha(\xi_1)\Psi_\gamma(\xi_1)\Big][F_\alpha'(\xi_0) - \xi_0 F_\alpha(\xi_0)].$$

The prime denotes the derivative of the corresponding function with respect to ξ at the points ξ_0 and ξ_1. Here we use the following notation:

$$\alpha = \frac{1-\beta_1}{4}, \quad \gamma = \frac{1-\beta_2}{4}, \quad F_\alpha(\xi) = \Phi\left(\frac{1-\beta_1}{4}, \frac{1}{2}; \xi^2\right),$$
$$F_\gamma(\xi) = \Phi\left(\frac{1-\beta_2}{4}, \frac{1}{2}; \xi^2\right), \quad \Phi_\alpha(\xi) = \Phi\left(\frac{3-\beta_1}{4}, \frac{3}{2}; \xi^2\right),$$
$$\Phi_\gamma(\xi) = \Phi\left(\frac{3-\beta_2}{4}, \frac{3}{2}; \xi^2\right), \quad \xi_0 = -\eta, \quad \xi_1 = s - \eta,$$
$$\Psi_\gamma = -\frac{1}{\Gamma(2\gamma+1)}F_\gamma(\xi) + \psi_\gamma\xi\Phi_\gamma(\xi),$$
$$\psi_\gamma = 2\sqrt{\pi}\,e^{-p\gamma}\frac{\gamma}{\Gamma^2(\gamma+1)}, \quad p = 2\ln 2. \tag{12}$$

Equation (11) will be fundamental for all the subsequent exposition. In it is also included in implicit form the connection between β_i and η which was mentioned above.

Our task reduces now to obtaining this connection in explicit form. In the general case (it suffices to consider (11)), this is not feasible. However, there is a possibility of completely solving this problem by the method of successive approximations for two limiting cases $s \ll 1$ and $s \gg 1$. The analysis of these cases gives a qualitative answer also for the intermediate case. For both limiting cases, it is assumed that η is a small parameter. As will be seen in what follows, this assumption is correct.

2. THE CASE $s \ll 1$

In this case, ξ_0 and ξ_1 are small parameters. The functions F_α, Φ_α, Ψ_γ and their derivatives are expanded in power series in ξ. We seek α and γ in the form of power series in ξ_0 and ξ_1: $\alpha = \alpha_0 + \alpha_1 + \alpha_2 + \ldots$, $\gamma = \gamma_0 + \gamma_1 + \gamma_2 \ldots$. The index denotes the order of smallness of the corresponding term. Substituting these series in (11) and separating the terms of equal order of smallness, we can compute α_n and γ_n, where $n = 0, 1, 2, \ldots$ Carrying out the calculations up to third order, we get

$$\gamma = \frac{1}{2\sqrt{\pi}}\eta + \frac{1}{4\pi}(p-4)\eta^2 + f\eta^3 + \frac{1}{2\sqrt{\pi}}\frac{k-1}{k}(s\eta^2 - \eta s^2) + \frac{k^2-1}{6\sqrt{\pi}\,k}s^3 \tag{13}$$

where $f = (1/8)\pi^{-3/2}(2d_2 - C^2 - 12p + (3/2)p^2 + 16) \approx 0.0134$. Here $C \approx 0.577$ is Euler's constant, $d_2 = 1/2(C^2 - \zeta(2)) \approx -0.656$, $\zeta(x)$ is the Riemann zeta function, and $p = 2\ln 2$.

It has been said above that it is necessary to test $\beta_2(\eta)$ for a minimum; but $\gamma = (1 - \beta_2)/4$, therefore, this is equivalent to a test of $\gamma(\eta)$ for a maximum. Up to now, η has been a free parameter. That value of η_0 which corresponds to the minimum of β_2 (maximum of γ) will correspond to the minimum of the free energy of the system.[7] Hence, precisely this value will also be realized under equilibrium conditions.

Limiting ourselves to the linear approximation in s, we find η_0:

$$\frac{\partial\gamma}{\partial\eta} = \frac{1}{2\sqrt{\pi}} + \frac{1}{2\pi}(p-4)\eta_0 + 3f\eta_0^2 + \frac{1}{\sqrt{\pi}}\frac{k-1}{k}s\eta_0 = 0. \tag{14}$$

Solving this quadratic equation relative to η_0 and expressing γ from (13) in terms of β_2, and β_2 in terms of H_0, we find the maximum possible H_0, i.e., H_{c3} as a function of the parameters of the material $k = \varkappa_1/\varkappa_2$ and the thickness of the film s:

$$\eta_0 \approx 0.37 + 1.2\frac{k-1}{k}s, \quad \gamma(\eta_0) = 0.100 + 0.15\frac{k-1}{k}s.$$

Thus, the maximum value is obtained for γ. The minimum value of β_2 corresponding to this value will be, according to (12) and (7), equal to

$$\beta_{2min} = 1 - 4\gamma_{max} = H_{c2}(2) / H_{c3}.$$

We then immediately obtain the desired expression for H_{c3}:

$$\frac{H_{c3}}{H_{c2}(2)} \approx 1.67\left(1 + \frac{k-1}{k}s\right).$$

In ordinary units, this formula has the form

$$\frac{H_{c3}}{H_{c2}(2)} \approx 1.67\left(1 + \frac{\varkappa_1 - \varkappa_2}{\varkappa_1} d\sqrt{\frac{3.4 H_{c2}(2) e}{\hbar c}}\right). \tag{15}$$

This formula can be written in another way. Inasmuch as it follows that

$$\varkappa_2 = 2\sqrt{2}\, e\delta_{02}^2 H_{cm}/\hbar c, \quad H_{c2}(2) = \sqrt{2}\,\varkappa_2 H_{cm}$$

$$H_{c2}(2) = \varkappa_2^2 \hbar c / 2e\delta_{02}^2.$$

Here δ_0 is the penetration depth of the weak magnetic field. Substituting this $H_{c2}(2)$ in (15), we get

$$\frac{H_{c3}}{H_{c2}(2)} \approx 1.67\left(1 + \frac{\varkappa_1 - \varkappa_2}{\varkappa_1}\sqrt{1.7}\,\varkappa_2 \frac{d}{\delta_{02}}\right). \tag{16}$$

From the result obtained, it is seen that as $\varkappa_1 \to \varkappa_2$ or $d \to 0$, Eq. (16) goes over into the formula $H_{c3} = 1.67\, H_{c2}$, which differs but slightly from the formula of Saint-James and de Gennes, $H_c = 1.69\, H_{c2}$. This difference is explained by the fact we have solved Eq. (11) approximately, limiting ourselves to the third approximation. In the fourth approximation H_{c3}/H_{c2} was shown to be equal to 1.68 for the pure surface.

3. THE CASE $s \gg 1$

In this limiting case, we shall consider $\xi_0 = -\eta$ as the small parameter. This is validated by the fact that in the limit as $s \to \infty$ the equilibrium value of η tends to 0.73. We shall consider the quantity $\xi_1 = s - \eta$ as large ($\xi_1 \gg 1$). We return to our initial and very general equation (11). For the case under consideration, it is convenient to write it differently—in terms of parabolic cylinder functions. If we then use the asymptotic expansion of these functions for large values of the argument, we can reduce the initial equation (11) to the form:

$$\frac{2\alpha}{\Gamma(2\alpha+1)}\psi_\alpha A_\alpha(\xi_0) + \frac{2\alpha}{\Gamma^2(2\alpha+1)}B_\alpha(\xi_0) + \omega = 0, \tag{17}$$

$$\omega = \frac{1}{\sqrt{\pi}} e^{-p\alpha}\xi_1^{1-4\alpha} e^{-\xi_1^2}\frac{k-1}{k+1}\left[\psi_\alpha A_\alpha(\xi_0) - \frac{1}{\Gamma(2\alpha+1)}B_\alpha(\xi_0)\right], \tag{18}$$

where $p = 2 \ln 2$, ψ_α is defined in (12), and the notation

$$A_\alpha(\xi_0) = \xi_0^2\Phi_\alpha(\xi_0) - \Phi_\alpha(\xi_0) - \xi_0\Phi_\alpha'(\xi_0),$$
$$B_\alpha(\xi_0) = F_\alpha'(\xi_0) - \xi_0 F_\alpha(\xi_0).$$

is employed.

We can regard Eq. (17) as an implicit form of writing the functional dependence $\alpha = \alpha(\xi_0, \xi_1)$. Let us find this dependence. It is easy to see that as $\xi_1 \to \infty$ the quantity $\omega \to 0$. Solution of (17) without the ω term is already known to us. It corresponds simply to the case of a pure surface of a semi-infinite superconducting space,[1] and in our methodology, this is the case $k = 1$ of the previous section. Therefore, it is natural to solve (17) by the method of successive approximations.

We seek α in the form

$$\alpha = \varepsilon_0(\xi_1) + [b_1 + \varepsilon_1(\xi_1)]\xi_0 + [b_2 + \varepsilon_2(\xi_1)]\xi_0^2 + \cdots. \tag{19}$$

The numbers $b_1, b_2, \ldots$ are known from previous considerations (see (13)) ($b_1 = -\frac{1}{2}\sqrt{\pi}$, $b_2 = (p-4)/4\pi$), for when $\xi_1 \to 0$ the quantities $\epsilon_0, \epsilon_1, \epsilon_2 \to 0$, and α depend only on ξ_0 according to the already discovered law. We limit ourselves to the terms that are linear in all the ϵ functions. We expand (17) in powers of ξ_0, and substitute the expression (19) in the first two components in (17), and the expression $\alpha = b_1\xi_0 + b_2\xi_0^2$ in the component ω. By comparing the coefficients for each power in the series, we finally obtain

$$\varepsilon_0 = -\frac{1}{\sqrt{\pi}}\frac{k-1}{k+1}\xi_1 e^{-\xi_1^2} \qquad \varepsilon_1 = -\frac{1}{\pi}\frac{k-1}{k+1}\xi_1(2 + C + 2\ln\xi_1)e^{-\xi_1^2},$$

$$\varepsilon_2 = \frac{1}{2\pi\sqrt{\pi}}\frac{k-1}{k+1}\xi_1\left[pC - 8C - C^2 - \frac{p^2}{4} + 4p - 8 + (2p - 4C - 16)\ln\xi_1 - 4\ln^2\xi_1\right]e^{-\xi_1^2}.$$

For the determination of the critical field H_{c3}, as before, it is necessary to find that value of η_0 which corresponds to the maximum value of $\alpha = \epsilon_0 - (b_1 + \epsilon_1)\eta + (b_2 + \epsilon_2)\eta^2$. The latter expression follows from (19), since $\xi_0 = -\eta$.

We seek η_0 and, substituting in the expression for α, we get

$$\alpha_{max} = -\frac{b_1^2}{4b_2}\left(1 + 2\frac{\varepsilon_1}{b_1} - \frac{\varepsilon_2}{b_2}\right) + \varepsilon_0.$$

Substituting all the necessary numerical values and carrying out the arithmetic computations, we finally obtain

$$\frac{H_{c3}}{H_{c2}(1)} = \frac{1}{\beta_{1\,min}} = \frac{1}{1 - 4\alpha_{max}}$$
$$= 1.62\left[1 - \frac{1}{\sqrt{\pi}}\frac{k-1}{k+1}\xi_1(3.43 + 2.41\ln\xi_1 + 1.90\ln^2\xi_1)e^{-\xi_1^2}\right]. \tag{20}$$

From this expression, it is seen that $\xi_1 \to \infty$ as $H_{c3}/H_{c2}(1) \to 1.62$. This value differs from the value 1.67 in (16) simply for the reason that in (16) three orders were taken into account, while here we have limited ourselves to two orders in the expansion of α in powers of ξ_0. However, this is not essential, since we are interested in the correction term.

The term in the parentheses in Eq. (20) is a slowly varying function of ξ_1. Denoting this function by K and assuming it to be constant, we obtain, with logarithmic accuracy,

$$H_{c3}(d) = H_{c3}(\infty)\left[1 - \sqrt{\frac{1,7}{\pi}}\frac{\varkappa_1 - \varkappa_2}{\varkappa_1 + \varkappa_2}\frac{\varkappa_1 d}{\delta_{01}}Ke^{-1,7(\varkappa_1 d/\delta_{01})^2}\right]. \tag{21}$$

In this formula, we have already returned to ordinary units.

4. DISCUSSION OF RESULTS

It has been shown that the decrease in the mean free path of the electrons l in the surface layer of a superconductor (this corresponds to an increase in the parameter $\varkappa$ in this layer) leads to an increase in the third critical field. According to Gor'kov,[4] the gap in the superconducting alloys (when $l \ll \xi_0$) decreases materially over distances $\sim\sqrt{\xi_0 l}$. The boundary conditions of Zaĭtsev[6] are valid in this case only up to the point where the thickness $d > \sqrt{\xi_0 l}$. Consequently, the latter inequality gave the region of applicability of our results.

We now discuss the experimental results of[3].

We assume that vacuum melting produces on the surface of the indium-lead alloy a film of thickness greater than $\sqrt{\xi_0 l}$, which is rich in inhomogeneities, impurities, and other defects that lead to a material decrease in the free path of the electron in the surface layer of the sample. This should lead to an increase in H_{c3}/H_{c2}. Finally, the long annealing equalizes the characteristics of the surface layer and of the whole material, the layer actually disappears, and H_{c3}/H_{c2} becomes equal to 1.7.

It is of interest to note that our calculation also holds in the case $\kappa_1 < \kappa_2$. In this case, one must expect a decrease in H_{c3}/H_{c2} in comparison with 1.7.

We now turn our attention to another aspect of the study of surface superconductivity. It is well known that a stretched wire has a fibrous microstructure, in which the fibers are elongated grains. Williamson and Furdyana[8] have noted recently that the critical field of wires made from type II superconductors is larger than H_{c2}. The authors relate this with the appearance of surface superconductivity inside the wire along the boundaries of the grains that have been stretched in the drawing process. It must be mentioned immediately here that an appreciable difference of H_{c3} from H_{c2} should be expected only if the superconductor borders on a dielectric and not on a metal.[6] Such a situation can arise if, for example, oxides are separated out along the boundaries of the grains. On the other hand, the grain boundaries are enriched by impurities, dislocations, etc., which lead to a decrease in the mean free path of the electron near the surface of the grain. According to our calculation, this increases H_{c3} and consequently also the critical field of the wire.

We now note that a comparison of Eqs. (16) and (21) shows a monotonic variation of H_{c3} with thickness of the film. This means that it is impossible to attribute the high critical field of alloys to the formation in the normal matrix of a filamentary superconducting structure along the dislocation lines. Such filaments will not have an essentially higher critical field as would be the case if they were in a dielectric, thanks to the large transmission coefficient of electrons through the filament-wire boundary.

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