Limiting Critical Field in Thin Superconductors*

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The Ginzburg-Landau theory has been formulated in a proper form to include the magnetization effect due to penetration of the magnetic field within the superconductor. Modified Ginzburg-Landau equations were obtained and solved for the case of thin films and filaments *in vacuo* or imbedded in normal material. The resulting expressions describe the behavior of the critical field of thin films and filaments as functions of thickness and temperature. They reduce to Clogston's critical field in the limit of vanishing thickness. They also provide a simple criterion for distinguishing between different types of the superconducting-normal transitions.

I. INTRODUCTION

In this paper the Ginzburg-Landau[1] (GL) phenomenological theory of superconductivity is reformulated in proper thermodynamic integral form so as to include the magnetization energy due to penetration of the external magnetic field inside the superconductor. The requirement that the thermodynamic potential of a system in equilibrium be a minimum leads to a nonlinear pair of GL equations which include the magnetization effect of the external magnetic field. This pair of equations is then solved, approximately, for cases of thin superconducting filaments and laminas *in vacuo* or imbedded in a normal material. The error involved in this approximation is indicated.

The existence of superconducting regions in hard superconductors in the presence of very high fields has led to a renewed theoretical interest[2,3] in the maximum obtainable critical field. BCS (Bardeen-Cooper-Schrieffer) theory, while explaining the nature of the superconducting phenomenon, treats superconductors of infinite extent and cannot be easily applied to solution of the boundary-value problems. As the dimensions of the superconducting body decrease, the importance of the surface energy terms increase, until even the order of the superconducting-normal transition in a magnetic field is changed from that of a bulk body.[1,4]

Derivation of the behavior of the critical field of a superconducting body as function of its dimensions involves the solution of a boundary value problem which is handled best within a phenomenological theory. But, so far, the derivation of such behavior has been hampered by the divergence of the corresponding theoretical expressions in the limit of vanishing thickness. The best developed phenomenological theories of superconductivity of London and London[5] and of Ginzburg-Landau[1] both seem to predict an infinite critical field for vanishingly thin superconductors. Physically, this is due to the penetration of the field within the superconductor with corresponding lowering of the diamagnetic energy term. Yet, once the penetration of the external field becomes appreciable, we have to consider the difference in free energies between the superconducting and the normal states due to magnetization of the superconducting body.[6,7] Neither the Londons nor GL have considered this magnetization term, which finally prevents the critical field from growing indefinitely as the characteristic dimension of a specimen approaches zero.

In the following, this magnetization term is included in a systematic manner in the GL theory. The modified GL equations are solved for thin films and filaments. These solutions are then used to bridge the gap between the limiting expressions for the critical field of thin specimens as obtained by general thermodynamic arguments[7,8] and those given by the London and GL theories.[2,3] The magnetostriction of a superconducting body has been neglected as insignificant.

II. THERMODYNAMIC RELATIONS

It has been shown previously[9] that during an isothermal process in equilibrium in a magnetic field the following thermodynamic potential must have a minimum:

$$\phi(T,H)=E-TS-\frac{1}{4\pi}\int_V \mathbf{B}\cdot\mathbf{H}_0 dv, \tag{1}$$

where E is the internal energy of the body, S the entropy, $\mathbf{B}$ the actual magnetic induction, and $\mathbf{H}_0$ the magnetic field intensity in absence of the body. The integral on the right-hand side of (1) is taken over the entire space.

Consider now a superconducting body. Denoting by subscripts s and n superconducting and normal states, and by subscripts h and 0 the presence or absence of an external magnetic field, we may write the GL expres-

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[9] V. L. Ginzburg, Zh. Eksperim. i Teor. Fiz. **34**, 113 (1958) [translation: Soviet Phys.—JETP **7**, 78 (1958)].

sions[3,9] for the $E-TS$ term as

$$E-TS=\int_V F_{sh}dv, \tag{2}$$

where

$$F_{s0}=F_{n0}+\alpha|\psi|^2+\tfrac{1}{2}\beta|\psi|^4$$

and

$$F_{sh}=F_{s0}+\frac{1}{4m}\left|-i\hbar\nabla\psi-\frac{2e}{c}\psi\mathbf{A}\right|^2+\frac{\mathbf{B}\cdot\mathbf{H}}{8\pi}.$$

The expansion for F_{s0} in terms of the order parameter $|\psi|^2$ was obtained from general considerations of the second-order transitions. The F_{sh} term describes the effect of penetration of the magnetic field into the superconducting body. Employing (2), Eq. (1) may be rewritten as:

$$\phi_s(T,H)=\int_V\left(F_{n0}+\alpha|\psi|^2+\frac{\beta}{2}|\psi|^4+\frac{1}{4m}\left|-i\hbar\nabla\psi-\frac{2e}{c}\psi\mathbf{A}\right|^2+\frac{B^2}{8\pi\mu}-\frac{\mathbf{B}\cdot\mathbf{H}_0}{4\pi}\right)dv. \tag{3}$$

Since in equilibrium ϕ_s has a minimum, (3) may be varied with respect to ψ^* and $\mathbf{A}$ to obtain the equilibrium values of ψ and $\mathbf{A}$. By varying (3) with respect to ψ^*, leaving the boundary conditions free, we get

$$\delta\phi_s/\delta\psi^*=0$$

provided

$$\nabla^2\psi=k^2(-\psi+|\psi|^2\psi+\psi A^2)+2ik\mathbf{A}\cdot\nabla\psi \tag{4a}$$

and

$$\mathbf{n}\cdot(\nabla\psi-ik\mathbf{A}\psi)=0 \quad \text{on} \quad S, \tag{4b}$$

where S is a surface bounding the superconductor and $\mathbf{n}$ is a unit, vector, normal to S. In (4) we have introduced the GL parameter $k=k(T)$, which depends on the properties of the material in bulk, and have performed the GL normalization as modified by Bardeen.[1,10]

We obtain an equation for $\mathbf{A}$ by varying (3) with respect to $\mathbf{A}$ subject to the condition that $\nabla\cdot\mathbf{A}=0$ and subject to the constraint of fixed boundary conditions ($\mathbf{n}\times\delta\mathbf{A}=0$ on S):

$$\frac{\delta\phi_s}{\delta A}=0 \quad \text{if} \quad \nabla^2\mathbf{A}=\frac{i\mu}{2k}(\psi^*\nabla\psi-\psi\nabla\psi^*)+\mu|\psi|^2\mathbf{A} \quad \text{for} \quad \mathbf{B}=\nabla\times\mathbf{A}, \tag{5}$$

where again the GL normalization has been performed.

In the following, we shall consider the critical field of infinite superconducting laminas and circular filaments. Laminas or filaments may be imbedded in normal material or *in vacuo*. In either case we are dealing with shapes which have a demagnetizing factor equal to zero. Therefore, we may write (3) as

$$\frac{\phi_s-\phi_n}{V}=\frac{\mu_n-\mu_s}{8\pi}H_0^2+\frac{1}{V}\int_V\left[\alpha|\psi|^2+\frac{\beta}{2}|\psi|^4+\frac{1}{4m}\left|-i\hbar\nabla\psi-\frac{2e}{c}\psi\mathbf{A}\right|^2+\frac{\mu_s}{8\pi}(\mathbf{H}_0-\mathbf{H})^2\right]dv, \tag{6}$$

where the integrals now are taken only over the volume of the body. We consider thicknesses small enough to exclude metastable states. Then at the transition point, as H_0 is raised to the critical value H_c, when transition takes place at constant temperature the left-hand side of (6) reduces to zero.

We proceed now to evaluate integrals on the right-hand side (6).

III. THIN FILM

Consider now the one-dimensional problem of a thin superconducting film, extending from $-d\leq z\leq +d$, and infinite in x and y directions. Let a uniform magnetic field due to external sources (H_0) be directed along the y axis. The vector potential $\mathbf{A}$ and the superconducting current $\mathbf{j}$ will then have only x components and be functions of z only. Considering the z components of (5), we conclude that ψ must necessarily be real. Thus, we may write (4) and (5) in this case as

$$A''=\mu_s\psi^2A, \qquad A'(\pm d)=B_0, \tag{7a}$$

$$\psi''=k^2(-\psi+\psi^3+\psi A^2), \quad \psi'(\pm d)=0. \tag{7b}$$

Except for a μ_s factor in Eq. (7) for A, this set of second-order, third-degree equations is the same as that given in reference 1. This set is most commonly solved by setting $k=0$. An approximate solution for $\mu_s=1$, $k\neq 0$ was given by GL,[1] but their method of solution did not permit the error involved to be estimated. To circumvent this limitation of the GL method, we resort rather to a series method of solution.

Letting $\psi(z)=\psi_0+\varphi(z)$, $\psi(0)=\psi_0$, and using the series method, we get

$$A=\frac{B_0\sinh(\alpha z)}{\alpha\cosh(\alpha d)}+O(z^5d^2), \tag{8a}$$

$$\varphi(z)=\frac{\psi_0(\psi_0^2-1)}{3\psi_0^2-1}[\cosh(pz)-1]+\frac{2\psi_0k^2B_0^2}{q^4\cosh^2(\alpha d)}\times[\cosh(qz)-1]-\frac{\psi_0k^2B_0^2z^2}{q^2\cosh^2(\alpha d)}+O(z^6d^2),$$

$$\alpha^2=\mu_s\psi_0^2, \quad p^2=k^2(3\psi_0^2-1), \quad q^2=p^2+4\alpha^2. \tag{8b}$$

The determinantal equation for ψ_0 is given by

$$\psi_0^2-1=\frac{2B_0^2}{q^2\cosh^2(\alpha d)}\left[1-\frac{\sinh(qd)}{qd}\right]\frac{pd}{\sinh(pd)}+O(d^6). \tag{9}$$

[10] J. Bardeen, Rev. Mod. Phys. 34, 667 (1962).

All quantities in the Eqs. (8) and (9) are normalized. Equation (8) is similar to Eq. (58) of reference 1, but does not reduce to it for $\mu_s=1$. Equation (9) reduces to Eq. (59) of reference 1 only for $\mu_s=1$, $k=0$.

With the aid of the above solutions for φ and A, integrals in Eq. (6) may now be evaluated:

$$\frac{H_c^2}{H_{cb}^2}=\frac{\psi_0^2(2-\psi_0^2)+O(d^6)}{\mu_n-\mu_s+\mu_s\{1-[\tanh(\mu_s^{1/2}\psi_0 d/\delta)]/\mu_s^{1/2}\psi_0 d/\delta\}}, \tag{10}$$

where H_{cb} is the critical field for a bulk sample and δ is the small field penetration depth obtained from its London value and the data of reference 11. For $k=0$, $\mu_s=\mu_n=1$, the above equation reduces to the corresponding GL expression for H_c which diverges as the thickness of the film approaches zero. Knowledge of the bulk parameters of the materials as a function of temperature permits us, within the indicated error, to solve Eqs. (9) and (10) to obtain the critical field of thin films as a function of thickness and temperature.

We may also notice that at any temperature $\lim_{d\to 0}\psi_0^2=1$. Therefore,

$$\lim_{d\to 0}\frac{H_c^2}{H_{cb}^2}=\frac{1}{4\pi(\chi_n-\chi_s)}. \tag{11}$$

This result is independent of the temperature region of validity of the GL theory, and may be obtained directly from (6).

IV. THIN CYLINDRICAL FILAMENT

Consider now a thin infinitely long superconducting cylinder with its center on the z axis and its generating line parallel to the z axis. In this case, the vector potential $\mathbf{A}$ and superconducting current $\mathbf{j}$ will have only φ components and will be functions of r only. Considering again r components of Eq. (5) we agaln conclude that ψ may be chosen real. For real ψ, Eqs. (4) and (5) in cylindrical coordinates become

$$\frac{d^2A}{dr^2}+\frac{1}{r}\frac{dA}{dr}-\frac{1}{r^2}A-\mu_s\psi^2A=0,$$

$$\frac{1}{r}\frac{d}{dr}(rA)\Big|_{r=r_0}=B_0, \tag{12a}$$

$$\frac{d^2\psi}{dr^2}+\frac{1}{r}\frac{d\psi}{dr}+k^2\psi-k^2\psi^3-k^2\psi A^2=0,\quad \frac{d\psi}{dr}\Big|_{r=r_0}=0. \tag{12b}$$

Since we expect A and ψ to be finite at the origin we exclude the possibility of modified Bessel functions of the second kind appearing in the solutions for A and ψ. Therefore, the series method is again applicable. Solving the system (12) we get

$$\psi(r)=\psi_0+\varphi(r),\quad \psi(0)=\psi_0,$$

$$A=\frac{B_0I_1(\alpha r)}{\alpha I_0(\alpha r_0)}+O(r^5r_0^2), \tag{13a}$$

$$\varphi(r)=\frac{\psi_0(\psi_0^2-1)}{3\psi_0^2-1}[I_0(pr)-1]+\frac{\psi_0k^2B_0^2}{q^4I_0^2(\alpha r_0)}\times[I_0(qr)-1]+\frac{\psi_0k^2B_0^2r^2}{4q^2I_0^2(\alpha r_0)}+O(r^6r_0^2), \tag{13b}$$

with the determinantal equation for ψ_0 as

$$\psi_0^2-1=\frac{B_0^2}{q^2I_0^2(\alpha r_0)}\left[1-2\frac{I_1(qr_0)}{qr_0}\right]\frac{pr_0}{2I_1(pr_0)}+O(r_0^6), \tag{14}$$

where the definition of α, q, and p is the same as above and all quantities are GL normalized. As before, this solution immediately enables us to calculate the integrals in (6), yielding

$$\frac{H_c^2}{H_{cb}^2}=\frac{\psi_0^2(2-\psi_0^2)+O(r_0^6)}{\mu_n-\mu_s+\mu_s[1-2I_1(\alpha r_0/\delta)/(\alpha r_0/\delta)I_0(\alpha r_0/\delta)]}. \tag{15}$$

Knowledge of the bulk critical field and the small field penetration depth (δ), permits simultaneous solution of the (14) and (15). This solution expresses the critical field of the filament function of temperature and radius.

Again, since $\lim_{r_0\to 0}\psi_0^2=1$, we obtain for any temperature.

$$\lim_{r_0\to 0}\frac{H_c^2}{H_{cb}^2}=\frac{1}{4\pi(\chi_n-\chi_s)}. \tag{16}$$

V. DISCUSSION

The description of the fields outside a singly connected bulk superconducting body is considerably simplified if we assign to the body the equivalent external values $\mu_s=0$, $\chi_s=-1/4\pi$. However, these equivalent values do not reflect the actual relation between B and H inside the superconductor. Relations, formally similar to our Eqs. (11) and (16), but using equivalent χ_s, have been given in the literature previously [see Eq. (2.15), reference 10]. As is clear from Sec. II, our χ_s equals the normal susceptibility, modified by the removal of some of the electrons into a superconducting paired state. Our χ_s never reduces to $-1/4\pi$ inside a bulk superconducting body.

If we assume: (a) that the change in susceptibility between the normal and superconducting states is due to free electrons only; (b) use the BCS expression for superconducting susceptibility[12] ($\chi_s=0$ at $T=0$); (c)

[11] D. H. Douglass, Jr., Phys. Rev. **124**, 735 (1961).

[12] K. Yosida, Phys. Rev. **110**, 769 (1958); A. A. Abrikosov and L. P. Gorkov, Zh. Eksperim. i Teor. Fiz. **39**, 480 (1960) [translation: Soviet Phys.—JETP **12**, 337 (1961)].

take the Pauli spin susceptibility for the susceptibility of the normal state, $\chi_P = 2\mu_B^2 N(0)$, and eliminate the density of states at Fermi level $N(0)$ by use of the BCS[13] expression $H_{cb}(T=0) = 1.75[4\pi N(0)]^{1/2} kT_c$; then (11) for films and (16) for filaments in the limit as r_0, d approach zero and at zero temperature, *both yield Clogston's limiting relation*,[7]

$$H_{c0} = H_{cb}(4\pi\chi_P)^{-1/2} = 1.75k(\sqrt{2}\mu_B)^{-1}T_c = 18\,400T_c,$$

where H_{c0} is the critical magnetic field in gauss at 0°K and T_c is the critical temperature in zero field. *The above theory does not substantiate the limiting expression*[8] $H_{c0} = 26\,000T_c$.

Equations (11) and (16) show that Clogston's limiting relation follows rigorously from the GL theory even when the surface energy term is taken into account.

We may also expand (9) and (10), and also (14) and (15), for small r_0/δ and d/δ, keeping in the expansion terms up to a second order only, obtaining

$$\frac{(H_c^2)_{\text{filament}}}{H_{cb}^2} = \frac{1}{\mu_n - \mu_s + \mu_s^2 r_0^2/8\delta^2} \tag{17}$$

and

$$\frac{(H_c^2)_{\text{film}}}{H_{cb}^2} = \frac{1}{\mu_n - \mu_s + \mu_s^2 d^2/3\delta^2}. \tag{18}$$

Setting $\mu_n = \mu_s = 1$, we obtain the divergent expressions for the critical field of films and filaments previously discussed by Hauser and Helfand.[3] The arguments of Hauser and Helfand bearing on the relative stability of the filamentary versus laminar structure still apply here. Though as r_0 and d simultaneously approach zero, films and filaments go to the limit of the same critical field, Eqs. (17) and (18) show that for r_0 equal to d, different from zero, the critical field of a filament is larger than that of the film:

$$(H_c^2)_{\text{filament}} - (H_c^2)_{\text{film}} = \frac{5\mu_s^2 d^2 (H_c^2)_{\text{film}}}{24(\mu_n - \mu_s)\delta^2 + 3\mu_s^2 d^2} > 0,$$

making the filamentary structure more stable. The laminar structure as the critical field of a film is reached will not go directly into the normal state but break into a filamentary one. Previous discussions[3] of the structure of the intermediate state involved consideration of the critical field of films and filaments *in vacuo*. Due to formulation of the problem in the proper integral form it is possible to discuss the intermediate state directly. The above results apply to a filament or a film imbedded in the normal material, so long as the field at the surface of the film or filament may be considered uniform. In this case, the external field should be interpreted as the magnetic field that would exist at the surface of the film or filament if the entire body were in the normal state.

To obtain the temperature dependence of the critical field of filamentary structure let us: (a) approximate in the (17) the Yosida's susceptibility[12] by a parabola $\chi_s/\chi_n = t^2$; (b) use the BCS[13] expression $\delta^2 = \delta_0^2(2-t)/2(1-t)$; and (c) employ $H_{cb} = H_0(1-t^2)$, where t is the reduced temperature, to get

$$H_c = \frac{a(1+t)(2-3t+t^2)^{1/2}}{[1+bt(1-t)]^{1/2}},$$
$$a = \frac{2\delta_0 H_0}{(\mu_s^2 r_0^2 + 32\pi\delta_0^2\chi_n)^{1/2}}, \quad b = \frac{16\pi\delta_0^2\chi_n}{\mu_s^2 r_0^2 + 32\pi\delta_0^2\chi_n}. \tag{19}$$

If we pass a parabola $H_p = \text{const}(1-t^2)$ through the points $t=1$ and $t=t_1>\frac{1}{2}$ on the critical field curve of Eq. (19), we find that for any t less than t_1, H_p is larger than H_c.

On the other hand, for a number of alloys[14] (Nb_3Sn, Mo—Re, etc.), H_c follows a straight line over a wide range of temperatures and H_c (alloy) is larger than H_p. Such a straight line is best approximated by $\text{const}\times H_{cb}(t)/\delta(t)$ or $\text{const}\times H_{cb}(t)/\delta(t)^{1/2}$, which would require the appearance of $1/r^2$ or $1/r$ terms in our solution for $\psi(r)$. These terms were excluded from our solution by the requirement that $\psi(r)$ be finite within the superconducting region, but would enter, provided the point r equals to zero is outside the superconducting region, i.e., normal. Behavior such that H_c is larger than H_p, therefore, requires for its explanation a vortex-type Abrikosov[15] structure.

A quick glance at the critical field versus temperature curve thus permits us to tell whether the critical field is due to superconducting filaments, structureless transition, or flux filaments depending on the critical field values being below, on, or above the parabola fitted through the first few points around T_c.

In summary we have: (a) formulated the problem in the proper integral form, (b) obtained and solved the GL equations, including the magnetization effect, (c) derived the result that the filamentary structure of the intermediate state is more stable than the laminar one, (d) substantiated Clogston's value of the limiting critical field, (e) discussed the behavior of the critical field as function of temperature for different types of filamentary structures of the intermediate state, and (f) obtained a criterion for easy identification of these filamentary states.

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

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