Critical fields of mesoscopic superconductors

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Recent measurements have shown oscillations in the upper critical field of simply connected mesoscopic superconductors. A quantitative theory of these effects is given here on the basis of a Ginzburg-Landau description. For small fields, the \( H - T \) phase boundary exhibits a cusp where the screening currents change sign for the first time thus defining a lower critical field \( H_{c1} \). In the limit where many flux quanta are threading the sample, nucleation occurs at the boundary and the upper critical field becomes identical with the surface critical field \( H_{c3} \).

I. INTRODUCTION

As is well known, superconductivity only exists at sufficiently low temperatures \( T \) and small external magnetic fields \( H \). The resulting \( H - T \) boundary for the normal to superconducting transition is determined by the Ginzburg parameter \( \kappa = \lambda/\xi \). For type II superconductors with \( \kappa > 1/\sqrt{2} \) one obtains a lower (\( H_{c1} \)) and an upper (\( H_{c2} \)) critical field which - for bulk samples - are universal functions of temperature \( \xi \). However, it was realized long ago by Saint-James and de Gennes \( \xi \) that in the presence of a surface, these results are changed considerably. Regarding \( H_{c1} \), there is a surface barrier for the entrance of the first flux quantum. Thus the field up to which the sample stays in the Meissner phase may be much larger than the thermodynamic \( H_{c1} \). In the case of the upper critical field, superconductivity in a bounded sample persists even in the range \( H_{c2} < H < H_{c3} = 1.69H_{c2} \) provided the external field is parallel to the surface. In this regime only a thin sheet at the sample boundary of the order of the zero field coherence length \( \lambda \) is superconducting. Quite generally, in samples whose size is of the order of the \( T = 0 \) coherence length, one expects that the \( H - T \) phase boundary will strongly depend on the detailed form of the sample, reflecting the possible eigenmodes for the complex superconducting order parameter \( \psi(\mathbf{r}) \).

Experimentally this was recently studied by Moshchalkov et al., who investigated the temperature dependence of the upper critical field of small mesoscopic aluminium samples with typical sizes of only a few coherence lengths. By a careful solution of the boundary value problem for the linearized GL-equation near \( T_{c1} \), we are able to quantitatively describe the observed structure in the upper critical field of a small disc. In the limit where many flux quanta are threading the sample, the upper critical field is in fact a surface critical field, reproducing the standard \( H_{c3} \) value of a semi-infinite geometry. Moreover we determine a generalized lower critical field for mesoscopic discs and rings. An interesting point is that the eigenvalue spectrum which determines the suppression of the critical temperature \( T_{c}(H) \) as a function of magnetic field, is rather different from the case of electron levels in a quantum dot, because of the different boundary conditions.

II. CRITICAL FIELDS OF SUPERCONDUCTING DISCS AND RINGS

Let us consider a small disc with radius \( R \) and thickness \( d \) in an external magnetic field \( \mathbf{H} = H\mathbf{e}_z \), which is perpendicular to the sample surface at \( z = \pm d/2 \). Near the normal to superconducting transition the change in the free energy with respect to the normal state can be expressed in terms of a GL-functional of the complex superconducting order parameter \( \psi(\mathbf{r}) \)

\[
F[\psi] = F_n + \int_V \left\{ \frac{\mu^2}{2} \left| (\nabla - \frac{2ie}{mc} \mathbf{A})\psi(\mathbf{r}) \right|^2 + a|\psi(\mathbf{r})|^2 + \frac{B^2}{8\pi} |\psi(\mathbf{r})|^4 + \frac{\mu^2}{2} |\psi(\mathbf{r})|^4 \right\} d^3r.
\]

Here \( \mathbf{B} = \nabla \times \mathbf{A} \) is the magnetic field in the sample with volume \( V \), \( a = a'(T - T_{c})/T_{c} \) and \( b \) are the standard GL-coefficients and \( \mu \) the effective electron mass. In principle there are also surface contributions to the free energy functional \( \frac{\mu^2}{2} |\psi(\mathbf{r})|^4 \) which may be important for mesoscopic samples with a large surface to volume ratio. In our treatment below such contributions are neglected, which is justified only a posteriori. In the vicinity of the transition the order parameter and the screening currents are small. To lowest order we may therefore neglect the

9 Dedicated to Prof. W. Götze on the occasion of his 60th birthday.
quartic term in $F[\psi]$ and replace the magnetic field by the external one. The most probable configuration of the order parameter which follows from the mean field equation $\delta F[\psi]/\delta \psi^* = 0$ is then determined by the eigenvalue problem

$$-\frac{\hbar^2}{4\mu} \left( \nabla - \frac{2ie}{\hbar c} \mathbf{A} \right)^2 \psi = -a\psi$$  \hspace{1cm} (2)$$

for a particle with charge $2e$ in an external magnetic field ($e < 0$). Assuming that the sample is embedded in an insulating medium, the relevant boundary condition is that of vanishing current normal to the sample surface $\partial \psi/V$. In covariant form the corresponding Neumann boundary condition is

$$\mathbf{n} \cdot \left( \nabla - \frac{2ie}{\hbar c} \mathbf{A} \right) \psi \bigg|_{\partial \mathcal{V}} = 0,$$  \hspace{1cm} (3)$$

where $\mathbf{n}$ is a unit vector normal to the sample surface. In order to determine the $H - T$ phase boundary, we must find the lowest eigenvalue $E_0(H)$ associated with a nonzero order parameter $\psi(\mathbf{r}) \neq 0$. From $E_0(H)$ the transition from the normal to the superconducting state is determined by

$$-a = \alpha' T_c - T_c(H) \left/ T_c \right. = E_0(H).$$  \hspace{1cm} (4)$$

Here $T_c$ is the (mean field) transition temperature of the infinite system with zero field. Since $E_0(H) \geq E_0(0)$ quite generally, the transition temperature at finite field is always smaller or equal than at $H = 0$. In order to treat the case of discs or rings, it is convenient to introduce cylindrical coordinates $(\rho, \phi, z)$. In the appropriate gauge $\mathbf{A} = H \rho \mathbf{e}_\phi/2$, the solution of the Schrödinger equation (2) can then be written as

$$\psi = \mathcal{R}(\rho) e^{im\phi} e^{ikz}. \hspace{1cm} (5)$$

Here $m \in \mathbb{Z}$ is the angular momentum quantum number and $k_\rho = \nu \pi/d$ with $\nu \in \mathbb{N}_0$ the discrete wavevector for motion in the $z$-direction. Since the lowest eigenvalue has always $\nu = 0$, we will omit the $z$-dependence and the associated quantum number $\nu$ in the following. Introducing a dimensionless variable $\zeta = \rho^2/2l_H^2$ with $l_H = (hc/2e|H|)^{1/2}$ the magnetic length for charge $2e$, the differential equation for $\mathcal{R}(\zeta)$ can be reduced to Kummer’s confluent hypergeometric equation (8)

$$\frac{\partial^2 w}{\partial \zeta^2} + (|m| + 1 - \zeta) \frac{\partial w}{\partial \zeta} - \alpha w = 0 \hspace{1cm} (6)$$

by the substitution $\mathcal{R}(\zeta) = e^{-\frac{\zeta}{2}} \zeta^{|m|/2} w(\zeta)$. The dimensionless parameter $\alpha$ is directly related to the effective energy $E(H)$ by

$$\alpha = \frac{-E(H)}{\hbar \omega_c} + \frac{1}{2}(|m| + m + 1) \hspace{1cm} (7)$$

with $\omega_c = |H|/\mu c$ the standard cyclotron frequency. Using $-E_0(H) = a$ and $a' = \hbar^2/4\mu \xi^2(0)$ with $\xi(0)$ the zero temperature GL-coherence length, the maximum value of $\alpha$ - which always has $m \leq 0$ - determines the magnetic field shift of the transition temperature by

$$\left( \frac{T_c - T_c(H)}{T_c} \right) = \left[ \frac{1}{2} - \alpha_{\max}(H) \right] \frac{4\hat{\Phi}}{\Phi_0} \hspace{1cm} (8)$$

with $\hat{\Phi} = \pi \xi^2(0)\Phi$ and $\Phi_0 = hc/2e|\Phi|$ the superconducting flux quantum. In an infinite sample the ground state is the lowest Landau level with $E_{0}^{\infty} = \hbar \omega_c/2$, i.e. $\alpha^{\infty} = 0$. The phase boundary is then given by

$$\left( \frac{T_c - T_c(H)}{T_c} \right) = 2 \frac{\hat{\Phi}}{\Phi_0}, \hspace{1cm} (9)$$

which is equivalent to the standard relation $H = H_{c2}(T) = \Phi_0/2\pi \xi^2(T)$. For the finite system, the spectrum of eigenvalues follows from the Neumann boundary condition (3) at the inner ($R_i$) and outer ($R$) radius of the ring. The general solution of (8) is a linear combination of Kummer functions (6). In the case of a disc geometry only

$$w_1(\zeta) = \Phi(\alpha, |m| + 1, \zeta) = 1 F_1(\alpha, |m| + 1, \zeta) \hspace{1cm} \hspace{1cm} (10)$$

is allowed since the second linear independent solution diverges at the origin. Using standard recursion relations for $1 F_1$, it is straightforward to show that the boundary condition at $\rho = R$, which simply reads $dR/d\rho = 0$, since $\mathbf{A} \cdot \mathbf{n} = 0$, leads to

$$\left( |m| + 1 - \alpha \right) \Phi(\alpha - 1, |m| + 1, \zeta_R) \hspace{1cm} - \Phi(\alpha, |m| + 1, \zeta_R) + \alpha \Phi(\alpha + 1, |m| + 1, \zeta_R) = 0 \hspace{1cm} (11)$$

with $\zeta_R = R^2/2l_H^2$. For each given $m$ equation (11) determines a discrete series of eigenvalues $\alpha_{nm}(H)$, $n \in \mathbb{N}_0$, which are decreasing with increasing $n$. They obey $\alpha_{nm} \leq 1/2$ and are continuous in $\zeta_R = \Phi/\Phi_0$ which is just the external flux $\Phi = \pi R^2 H$ through the area of the disc in units of the flux quantum. Analytical results for the spectrum can be obtained in the low field limit $\Phi \to 0$. In this limit it is straightforward to treat the general case of a ring with $\sigma = R_i/R \leq 1$. Standard second order perturbation theory in the magnetic field then leads to a shift in the transition temperature which is given by

$$\left( \frac{T_c - T_c(H)}{T_c} \right) = \frac{1}{2} \left( 1 + \sigma^2 \right) \frac{\hat{\Phi} \Phi}{\Phi_0} \hspace{1cm} + \ldots. \hspace{1cm} (12)$$

The corrections to this result are of order $\Phi^4$, since the ground state energy is even in $\Phi$. For a very thin ring with $\sigma \to 1^-$ this agrees with the low field limit of the Little-Parks result (8)

$$\left( \frac{T_c - T_c(H)}{T_c} \right) = \frac{\xi^2(0)}{R^2} \min_{m \in \mathbb{Z}} \left| \frac{m - \Phi}{\Phi_0} \right|^2, \hspace{1cm} (13)$$
as expected. For general magnetic fields the phase boundary can only be obtained numerically. To this end we have directly solved the transcendental equation (11) which allows us to determine the spectrum without any discretization error. The energy levels are thus obtained with arbitrary accuracy, in contrast to previous work by Saint-James [10] or by Nakamura and Thomas [11] who consider Dirichlet boundary conditions. The results are shown in Fig. 1, where the dimensionless eigenvalues 

\[
\frac{\hbar^2}{\mu R^2} \tilde{E}_{nm}
\]

for \(n = 0\) and \(m = 2, 1, 0, -1, ..., -10\) are plotted as functions of \(\Phi/\Phi_0\).

Evidently the lowest eigenvalue exhibits an oscillatory behaviour with cusps at values \(\Phi^{(j)}\), \(j = 1, 2, ...\) where the magnetic quantum number of the lowest eigenstate jumps by one unit. The dimensionless distances between successive cusps

\[
\Delta_j = \frac{\Phi^{(j)} - \Phi^{(j-1)}}{\Phi_0} \quad (\Phi^{(0)} = 0)
\]

are given in Table 1 with an accuracy corresponding to the last given digit.

| \(\Phi^{(j)}\) | \(\Phi_0\) | \(\Delta_j\) |
|---------|--------|---------|
| 1.923765 | \(\Phi_0\) | 1.923765 |
| 3.392344 | \(\Phi_0\) | 1.468579 |
| 4.747920 | \(\Phi_0\) | 1.355676 |
| 6.045882 | \(\Phi_0\) | 1.297902 |
| 7.30680 | \(\Phi_0\) | 1.26092 |
| 8.54233 | \(\Phi_0\) | 1.23552 |
| 9.75843 | \(\Phi_0\) | 1.21612 |
| 10.9591 | \(\Phi_0\) | 1.20072 |
| 12.1477 | \(\Phi_0\) | 1.1886 |
| 13.3255 | \(\Phi_0\) | 1.1778 |

TABLE 1

Experimentally the oscillatory behaviour of the ground state energy is directly reflected in the \(H - T\) phase boundary. For the case of a disc discussed here, this was actually first observed by Buisson et al. [12]. In their experiment, however, the presence of two gold contacts led to a boundary condition which is different from (8) over part of the sample boundary. While the oscillations were still present, a detailed comparison with theory was difficult (for instance the first cusp was observed at \(\Phi \approx 2.5\Phi_0\) compared to \(\Phi = 1.924\Phi_0\) in the pure Neumann case). The more recent experiments of Moshchalkov et al. [5], however, are in very good agreement with the theoretical predictions. This may be seen from a comparison with the measured deviation of the temperature shift \(\Delta T_c = T_c - T_c(H)\) from the average linear behaviour which is shown in Fig. 2.

Here we have used the experimental value \(\xi(0) = 1\mu\), and a disc area which is only 2.7 % smaller than the area of the almost rectangular sample used in the experiment. It is important to note that the periods \(\Delta_j\) decrease monotonically from \(\Delta_1 = 1.924\) to \(\Delta_\infty = 1\) (see Table 1 and below) in contrast to an anomalous first period \(\Delta_1 \approx 1.8\) and constant successive ones \(\Delta_2 \approx \Delta_3 \approx \Delta_4 \approx 1.3\) which were quoted by Moshchalkov et al. [5].

The field at which the ground state changes from \(m = 0\) to \(m = -1\) allows us to extract a lower critical field

\[
H_{c1}^{\text{disc}} = \frac{1.92376 \Phi_0}{\pi R^2}
\]
for a mesoscopic system with size $R$ of order $\xi(0)$. Here $H_{c1}$ is defined via the condition that for $H < H_{c1}$ the sample tries to screen out the applied flux, whereas for $H > H_{c1}$ the free energy is minimized by accepting one flux quantum. It is interesting to compare this with Fetter’s theory of flux penetration in a superconducting disc [13], which is based on calculating the self energy of a vortex. In the limit where the disc radius $R$ is much smaller than the effective thin film penetration depth $\lambda_{2d} = \lambda^2/d$, it turns out, that it is energetically favourable for a vortex to enter if $H > H_{c1}$ with [3]

$$H_{c1} = \frac{\Phi_0}{\pi R^2} \ln \frac{R}{r_c} \quad \lambda_{2d} \gg R \gg r_c. \quad (16)$$

Here $r_c \approx \xi(0)$ is the core radius, which is always assumed to be much smaller than $R$. Obviously for samples whose size is of the order of the coherence length $\xi(0)$, the expression [14] is no longer applicable. In this limit the approximation that the order parameter is constant beyond $r_c$ becomes invalid. As found above the lower critical field is then replaced by our result [15], with a crossover at about $R \approx 7\xi(0)$. Here it is important that for $R \approx \xi(0)$, linearized GL-theory is sufficient to calculate $H_{c1}$, because it is the sample boundary which limits the magnitude of the order parameter instead of the quartic term as usual. Finally consider a ring with inner radius $R_i \gg r_c$. Then the lower critical field is simply determined by the condition that half a flux quantum is applied, i.e.

$$H_{c1}^{\text{ring}} = \frac{1}{2} \frac{\Phi_0}{\pi R^2}. \quad (17)$$

Indeed this follows from the quantization of the fluxoid [2], and is valid irrespective of the thickness of the ring. Comparing (17) with the result (13) for a disc, we find that $H_{c1}$ in the latter case is almost four times larger. Qualitatively this is due to the additional condensation energy in the center of the disc which is required for a vortex to enter.

As a second point let us discuss the behaviour at $\Phi \gg \Phi_0$ where many flux quanta have entered. In this limit the ground state has angular momentum $|m| \gg 1$. The associated eigenfunction is thus concentrated near the disc boundary. It is then obvious that our upper critical field for $\Phi \gg \Phi_0$ is in fact a surface critical field. If this is correct, it should asymptotically approach the value obtained by Saint-James and de Gennes [4] for a surface with a radius of curvature large compared to the coherence length. This can be verified by considering the special values $\Phi_m, m = 1, 2, \ldots$ in Fig. 1, where the tangent to $E_0(|m|)(\Phi)$ goes through the origin (i.e. we are considering successive approximations to the envelope). These values are given in table 2 together with the corresponding values of $\alpha_{\text{max}}$.

| $m$ | $\Phi/\Phi_0$ | $\alpha_{\text{max}}$ | $T_c - T_c(H)/T_c$ |
|-----|--------------|-----------------|------------------|
| 1   | 2.44         | 0.28761         | 0.849            |
| 2   | 3.92         | 0.26561         | 0.937            |
| 3   | 5.28         | 0.25514         | 0.979            |
| 4   | 6.56         | 0.24872         | 1.005            |
| 5   | 7.82         | 0.24426         | 1.023            |
| 6   | 9.09         | 0.24091         | 1.036            |
| 7   | 10.28        | 0.24389         | 1.046            |
| 8   | 11.46        | 0.25638         | 1.054            |
| 9   | 12.68        | 0.23449         | 1.062            |
| 10  | 13.86        | 0.23300         | 1.067            |
| 100 | 110.23       | 0.21395         | 1.144            |
| 200 | 214.72       | 0.21130         | 1.154            |
| 1000| 1033.76      | 0.20778         | 1.169            |
| 10000| 10109.33    | 0.20583         | 1.177            |

TABLE 2

It is obvious that $\alpha_{\text{max}}$ converges to a limiting value $0.2058\ldots$. Using (16) the associated transition temperature is then given by

$$\frac{T_c - T_c(H)}{T_c} = 1.177 \frac{\Phi_c}{\Phi_0}. \quad (18)$$

As expected this is completely equivalent to the well known result $H = H_{c3}(T) = 1.695H_{c2}(T)$ for the surface critical field [1]. The coefficient $1 - 2\alpha_{\text{max}}^m < 1$ is in fact just the ratio between the ground state energy in the disc and the energy $\hbar\omega_{2c}/2$ of the lowest Landau level in an infinite sample. Edge states centered near the sample boundary have thus a lower energy than bulk levels. Note that this behaviour is just the opposite of the case with Dirichlet boundary conditions (relevant e.g. for edge states in the Quantum Hall Effect) where edge states are above the corresponding bulk Landau levels [12]. Finally let us discuss the behaviour of the periods $\Delta_j$ of the ground state oscillations for large flux $\Phi \gg \Phi_0$. Due to the factor $\rho^{\text{max}}/\pi^2$ the order parameter for increasing magnetic quantum number $|m| \gg 1$ is more and more concentrated near the sample boundary, but is practically zero in the interior of the sample. The simply connected disc thus effectively behaves like a ring with a normal core of size $R - c_1 l_H$, where $c_1$ is a constant [12]. The periodicity observed in $E_0(H)$ is then simply determined by the condition that one additional flux quantum enters the area of the normal core, i.e.

$$\Delta_j(\Phi) = 1 + 2c_1 \frac{l_H}{R} \rightarrow 1. \quad (19)$$

In fact this field dependance was observed in the experiments by Buisson et al. already for $\Phi/\Phi_0 > 5$ [12]. In the asymptotic limit, which is however only reached for $\Phi/\Phi_0 > 10^3$ (see table 2) the coefficient $c_1$ can be obtained analytically as $2c_1 = \sqrt{0.59} \approx 0.76$ [10].
III. DISCUSSION

Using linearized GL-theory we have calculated the nucleation field of a small superconducting disc with a radius which is of the order of the coherence length $\xi(0)$. The good agreement with the experimentally observed $H - T$ phase boundary suggests that the macroscopic GL-description remains valid in this regime which is not obvious a priori. A surprising feature of our results is that Aharonov-Bohm like oscillations are present even in a simply connected sample. The physical origin of this effect is that already the entrance of a single flux quantum effectively makes the sample a multiply connected one. In the limit $\Phi \gg \Phi_0$ the disc behaves like a thin walled ring, leading to oscillations in $T_c(H)$ which are completely equivalent to the well known Little-Parks experiment. It is interesting to note that these effects depend crucially on the Neumann boundary conditions. In fact the equivalent eigenvalue spectrum with Dirichlet boundary conditions, which was studied by Nakamura and Thomas [11], does not exhibit any oscillations in the ground state energy $E_0(\Phi)$. It is an interesting future problem to investigate similar effects in the fluctuation diamagnetism [14] or extend the calculations above to more complicated geometries. This would allow to study eigenvalue spectra for systems with classical chaotic dynamics [15,16] without the complications due to electron-electron interactions which are unavoidable in non-superconducting mesoscopic systems.

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