Critical fields of superconductors with magnetic impurities

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The upper critical field \( H_{c2} \), the field \( H_{c3} \) for nucleation of the surface superconductivity, and the thermodynamic \( H_c \) are evaluated within the weak-coupling theory for the isotropic s-wave case and arbitrary transport and pair-breaking scattering. We find that for the standard geometry of a half-space sample in a magnetic field parallel to the surface, the ratio \( R = H_{c3}/H_{c2} \) is within the window \( 1.55 \lesssim R \lesssim 2.34 \), regardless of temperature, magnetic or non-magnetic scattering. While the non-magnetic impurities tend to flatten the \( R(T) \) variation, the magnetic scattering merely shifts the maximum of \( R(T) \) to lower temperatures. Surprisingly, while reducing the transition temperature, magnetic scattering has a milder impact on \( R \) than the non-magnetic scattering. The surface superconductivity is quite robust; in fact, the ratio \( R \approx 1.7 \) even in the gapless state. We used Eilenberger’s energy functional to evaluate the condensation energy \( F_c \), and the thermodynamic critical field \( H_c \) for any temperature and scattering parameters. By comparing \( H_{c2} \) and \( H_c \), we find that unlike the transport scattering, the pair-breaking pushes materials toward type-I behavior. We find a peculiar behavior of \( F_c \) as a function of the pair-breaking scattering parameter at the low-\( T \) transition from gapped to gapless phases, which has recently been associated with the topological transition in the superconducting density of states.

I. INTRODUCTION

The question of the critical fields \( H_{c2} \) and \( H_{c3} \) is practically relevant since it is directly related to the critical temperature \( T_c(H) \) where the superconductivity emerges in the applied magnetic field \( H \). In type-II materials at a fixed \( T_c \), in the increasing field, the vortex phase in the bulk is destroyed at \( H_{c2} \), but the superconductivity may survive in a coherence-length-deep surface layer up to \( H_{c3} \).

In recent decades the interest in limiting fields was further fueled by the significant progress in superconducting resonator cavities used in particle accelerators [1, 2], and even more recently, in the hardware for superconducting circuits–based quantum computing [3, 4]. Of particular interest are effects of a disorder that influences cavi- ties quality factors and superconducting qubits coherence times [1, 5].

The ratio \( R = H_{c3}/H_{c2} \) for the applied field parallel to the surface of isotropic superconducting half-space has been evaluated by Saint-James and DeGennes (SJDG) [6] by solving the linearized Ginzburg-Landau equations for the order parameter \( \Delta \) subject to the boundary condition of vanishing normal gradient, \( \nabla_n \Delta = 0 \), at the sample surface. Their seminal result is \( R \approx 1.695 \). Since then, the surface superconductivity has been observed in many experiments, but the ratio \( R \) varied depending on surface quality, sample anisotropy, set up geometry, scattering, and temperature [7–11]. Theoretically, effects of material anisotropy have been discussed in [12], where it was shown that for sufficiently high anisotropy for some surface orientation, the ratio \( R \) may fall under unity. In other words, surface superconductivity does not exist.

An interesting development came recently, showing that within microscopic BCS theory, \( R(T) \) has a maximum at intermediate temperatures, which, however, disappears with increasing transport scattering [13]. In this contribution, we extend this study to the case when both magnetic and non-magnetic scattering channels are present. The discussion is limited to isotropic material with isotropic Fermi surface and s-wave order parameter. Given that \( H_{c2} \) is enhanced by non-magnetic transport scattering whereas suppressed by magnetic impurities [14], the question of the effect of magnetic impurities on \( H_{c3} \) is not obvious.

II. THE PROBLEM OF \( H_{c2} \) AND \( H_{c3} \)

Consider an isotropic material with both magnetic and non-magnetic scatterers; \( \tau_m \) and \( \tau \) are the corresponding average scattering times. The problem of the second order phase transition at \( H_{c2} \) and \( H_{c3} \) is addressed on the basis of Eilenberger’s quasiclassical version of Gor’kov’s equations for normal and anomalous Green’s functions \( G \) and \( f \). At the 2nd order phase transition, \( g = 1 \) and we are left with a linear equation for \( f \) [15, 16]:

\[
(2\omega^+ + \mathbf{v} \cdot \mathbf{\Pi}) f = 2\Delta/h + \langle f \rangle /\tau^-, \tag{1}
\]

\[
\omega^+ = \omega + \frac{1}{2\tau^+}, \quad \frac{1}{\tau^\pm} = \frac{1}{\tau} \pm \frac{1}{\tau_m}. \tag{2}
\]

Here, \( \mathbf{v} \) is the Fermi velocity, \( \mathbf{\Pi} = \nabla + 2\pi i \mathbf{A}/\phi_0 \) with the vector potential \( \mathbf{A} \) and the flux quantum \( \phi_0 \), \( \Delta(r) \) is the order parameter; Matsubara frequencies are defined by \( \omega = \pi T(2n + 1) \) with an integer \( n \); in the following (except some final results) we set \( h = 1 \); (…) stand for averages over the Fermi surface. Solutions \( f \) of Eq. (1) along with \( \Delta \) should satisfy the self-consistency equation:

\[
\frac{\Delta}{2\pi T} \ln \frac{T_{c0}}{T} = \sum_{\omega > 0} \left( \frac{\Delta}{\omega} - \langle f \rangle \right), \tag{3}
\]

where \( T_{c0} \) is the critical temperature in the absence of pair-breaking scattering.
Helfand and Werthamer [17] had shown that at the 2nd order phase transition at $H_{c2}$, the order parameter satisfies a linear equation

$$\Pi^2 \Delta = k^2 \Delta. \tag{4}$$

It was realized later that this equation holds at any 2nd order phase transition from normal to superconducting state away of $H_{c2}$, e.g. in proximity systems or at $H_{c3}$, provided $k^2 = -1/\xi^2$ satisfies the self-consistency equation of the theory [18, 19]. It turned out that the coherence length so evaluated depends not only on temperature and scattering but also on the magnetic field (except in the dirty limit or near $T_c$). In this sense, Eq. (4) in fact differs from the linearized Ginzburg-Landau equation that forms the basis for SJDG prediction of the surface superconductivity at $H_{c3}$ [6]. It is worth noting that if $\xi$ would have been $H$ independent, the ratio $H_{c3}/H_{c2}$ would have been constant equal 1.695 at all temperatures. As was shown in [13], this is not so (except for the dirty limit).

Thus, the order parameters at both $H_{c2}(T)$ and $H_{c3}(T)$ satisfy the same Eq. (4). The difference, however, comes from boundary conditions: $\Delta(\sigma)$ should be finite everywhere for $H_{c2}$, whereas $\nabla \Delta(\sigma) = 0$ at the sample surface for $H_{c3}$ ($\nabla \Delta$ is the gradient of $\Delta$ along the normal to the sample surface).

At any 2nd order phase transition, $\Delta \to 0$ and one can deal with linear Eq. (1). Repeating the derivation of Ref. [18], one finds (see the outline in Appendix A):

$$\langle f \rangle = \frac{2\tau - S}{2\omega + \tau - S}, \tag{5}$$

where $S$ is given by a series

$$S = \sum_{j,m=0}^{\infty} \frac{(-q^2)^j}{j!(2m + 2j + 1)} \left(\frac{m + j}{m!}\right)^2 \left(\frac{\ell^+}{\beta^+}\right)^{2m+2j} \times \prod_{i=1}^{m} [k^2 + (2i - 1)q^2], \quad q^2 = \frac{2\pi H}{\phi_0}, \tag{6}$$

where

$$\ell^+ = \nu \tau^+; \quad \beta^+ = 1 + 2\omega \tau^+. \tag{7}$$

This sum can be transformed to an integral, which is more amenable for the numerical work [19]:

$$S = \sqrt{\frac{\pi}{u}} \left[ \frac{1}{\sqrt{u}} \int_0^{1/\sqrt{u}} \left(\frac{1 + \eta^2}{(1 - \eta^2)^{\sigma/2 + 1}} \frac{\text{erfc} \frac{\eta}{\sqrt{u}} - \cos(\pi \sigma) \text{erfc} \frac{1}{\eta \sqrt{u}}}{\eta \sqrt{u}} \right) \right]. \tag{8}$$

Here,

$$u = \left(\frac{q \ell^+}{\beta^+}\right)^2 = \frac{h}{(P^+ + t(2n + 1))^2}, \tag{9}$$

where the reduced field $h$, temperature $t$, and the scattering parameter $P^\pm$ are introduced:

$$h = H \frac{\hbar^2 v^2}{2\pi \phi_0 T_{c0}}, \quad t = \frac{T}{T_{c0}}, \quad P^\pm = \frac{h}{2\pi T_{c0} \tau^\pm}. \tag{10}$$

(h is written explicitly to stress that $h$ and $P^\pm$ are dimensionless).

The parameter $\sigma$ is defined as

$$\sigma = \frac{1}{2} \left(\frac{k^2}{q^2} - 1\right) = -\frac{1}{2} \left(\frac{1}{q^2 \xi^2} + 1\right). \tag{11}$$

At $H_{c2}$, $\sigma = -1$ and

$$S(u) = \sqrt{\frac{\pi}{u}} \int_0^1 \frac{d\eta}{1 + \eta^2} \left[ \text{erfc} \frac{\eta}{\sqrt{u}} + \text{erfc} \frac{1}{\eta \sqrt{u}} \right]. \tag{12}$$

Near $T_c$, the order parameter satisfies linearized GL equation $-\xi^2 \Pi^2 \Delta = \Delta$, and at $H_{c3}$ SJDG obtained $\xi^2 q^2 = 1.695$ [6].

Therefore, at $H_{c3}$ we get

$$\sigma = -\frac{1}{2} \left(1 - \frac{1}{1.695} + 1\right) \approx -0.795. \tag{13}$$

Thus, to calculate $H_{c3}(t, P, P_m)$ with the help of Eqs (3) and (5) one has to use $S$ of Eq. (8) with $\sigma = -0.795$.

For numerical work we recast the self-consistency equation to dimensionless form:

$$-\ln t = \sum_{n=0}^{\infty} \left[ \frac{1}{n + 1/2} - \frac{2tS}{2(n + 1/2) + P^+ - SP^+} \right]. \tag{14}$$

The calculated ratio $R = H_{c3}/H_{c2}$ as function of the reduced temperature $T/T_{c0}$ for a few values of scattering parameters $P$ and $P_m$ is shown in the upper panel of Fig. 1: the lower panel shows the same results plotted vs $T/T_c$. One can see that the maximum of $R$ shifts to lower $T$ with increasing transport scattering $P$. Effects of magnetic scattering are mostly in suppressing the actual critical temperature $T_c$ and larger values of $R$ at low temperatures as compared to purely transport scattering.

It was shown in Ref. [13] that in the absence of magnetic impurities and a strong transport scattering, the ratio $R(T)$ flattens and the $T$ dependence disappears in the dirty limit in which $R(T) \approx 1.7$ at all $T$s. This is due to the disappearing field dependence of the coherence length $\xi$ [19] in this limit. It is thus instructive to see that the magnetic scattering does not change this qualitatively, see the upper panel of Fig. 2.

Hence, magnetic impurities do not change drastically the behavior of $H_{c3}$ relative to $H_{c2}$. We find that within the isotropic s-wave theory, for the standard geometry of a half-space sample in a field parallel to the surface, the ratio $R = H_{c3}/H_{c2}$ is within the window $1.55 \lesssim R \lesssim 2.34$, regardless of temperature, magnetic or non-magnetic scattering.

On the dirty side (with a strong transport scattering), the maximum of $R(T)$ moves to $T \approx 0$ as is seen in the lower panel of Fig. 2. Effects of pair breaking here are not drastic, even for the gapless situation ($0.128 < P_m < 0.14$, [16]) we still have $R \approx 1.7$. 
III. TYPE OF SUPERCONDUCTIVITY AND MAGNETIC IMPURITIES

The basic Eqs. (1) and (3) can be obtained minimizing the energy functional as is done in the original Eilenberger’s paper [15] for exclusively transport scattering:

\[ \Omega = N(0) \left[ \Delta^2 \ln \frac{T}{T_c} + 2\pi T \sum_{\omega > 0} \left( \frac{\Delta^2}{\omega} - \langle I \rangle \right) \right] , \] (15)

\[ I = 2\Delta f + 2\omega(g - 1) + \frac{f(f)}{2\tau^-} + \frac{g(g) - 1}{2\tau^+} . \] (16)

The function \( g \) in (16) is an abbreviation for \( \sqrt{1 - f^2} \). The free energy difference between superconducting and normal states is obtained by substituting solutions of Eqs. (1) in \( \Omega \). In particular, taking account of the self-consistency equation, we obtain for the condensation energy density \( F_c = F_n - F_s \):

\[ \frac{F_c}{2\pi TN(0)} = \sum_{\omega > 0} \left( \Delta f + 2\omega(g - 1) + \frac{f(f)}{2\tau^-} + \frac{g(g) - 1}{2\tau^+} \right) . \] (17)

This expression reduces to the known BCS result for isotropic s-wave cases with or without magnetic impurities [20]. For uniform zero-field state, the averaging brackets can be omitted, and the scattering part is \( -f^2/\tau_m \). Introducing dimensionless order parameter \( \delta = \Delta/2\pi T_c \) one has:

\[ \frac{F_c}{4\pi^2 T_c^2 N(0)} = \sum_{n=0}^{\infty} \left[ \delta f + t(2n + 1)(g - 1) - P_m f^2 \right] . \] (18)
The thermodynamic critical field follows

\[ H_c = \sqrt{8\pi F_c} = \left\{ 32\pi N(0)T^2_{\text{c1}} \right\}^{1/2} U(t), \]  

(19)

where

\[ U(t) = \left\{ t \sum_{n=0}^{\infty} \left[ \delta + t(2n + 1)(g - 1) - P_m f^2 \right] \right\}^{1/2}. \]  

(20)

Hence, we have the dimensionless thermodynamic field:

\[ h_c = \frac{H_c h^2 v^2}{2\pi \phi_0 T_{\text{c1}}} = \frac{h^2 v^2}{\phi_0 T_{\text{c1}}} \sqrt{8\pi N(0)} U(t). \]  

(21)

The pre-factor by \( U \), which is a characteristics of the clean material, can be expressed in terms of GL parameter \( \kappa_0 \) for the clean limit [21]:

\[ \kappa_0 = \frac{3\phi_0 T_{\text{c1}}}{h^2 v^2 \sqrt{7\zeta(3)\pi N(0)}}. \]  

(22)

Thus, we obtain

\[ h_c(t, P_m) = \frac{3}{\kappa_0 \sqrt{8}} \sqrt{7\zeta(3)} U(t, P_m). \]  

(23)

To evaluate the condensation energy (18) one first has to find \( f \) and \( \delta \) for given \( t \) and \( P_m \). For the uniform zero-field state these are solutions of the Eilenberger equation for \( f \) and of the self-consistency equation. In our notation this system of two equations for \( f(t, P_m) \) and \( \delta(t, P_m) \) reads:

\[ \sqrt{1 - f^2 (\delta - P_m f)} - t(n + 1/2)f = 0, \]  

(24)

\[ -\delta \ln t = \sum_{n=0}^{\infty} \left( \frac{\delta}{n + 1/2} - tf \right). \]  

(25)

The system can be solved numerically with the help of Wolfram Mathematica or MATLAB. Evaluation of \( h_c(t, P_m) \) is then straightforward.

Results for \( h_c \) and \( h_{\text{c2}} \) are shown in the left panel of Fig. 3 for the clean case. For the parameter \( \kappa = 0.5 \), we have a type-I behavior, while the type-II is realized for \( \kappa = 1 \) and 2, as should be since the boundary value is \( \kappa_0 = 1/\sqrt{2} \approx 0.7 \). Effect of the pair-breaking scattering is non-trivial; to see this we show the case of exclusively pair-breaking scattering, in which for both \( \kappa = 0.5 \) and \( \kappa = 1 \) the material behaves as type-I since \( h_c > h_{\text{c2}} \).

One may say that the pair-breaking scattering pushes materials toward type-I, the conclusion we arrived at in [22] in a different manner.

IV. SUMMARY

We have studied the effects of transport and pair-breaking scattering on the upper critical field \( H_{\text{c2}} \), the thermodynamic critical field \( H_c \), and the nucleation field \( H_{\text{c3}} \) of surface superconductivity for the field parallel to the plane surface of the half-space isotropic sample. We did not touch on questions of surface roughness, surface curvature, inhomogeneous distribution of impurities [5], material anisotropy [12], etc.

Whereas \( H_{\text{c2}} \) is suppressed by the pair-breaking scattering, \( H_{\text{c3}} \) is found to be suppressed as well so that the ratio \( R = H_{\text{c3}}/H_{\text{c2}} \) is within the window 1.55 \( \lesssim R \lesssim 2.34 \) regardless of temperature, magnetic or non-magnetic scattering. We find that the magnetic impurities do not change qualitatively the behavior of the ratio \( R \) with changing temperature and transport scattering; \( R(T) \) is equal to SJDG value 1.695 at \( T_c \) but increases on cooling.
goes through a maximum at intermediate temperatures and then drops to a $P$ dependent value at $T = 0$ [13]. The addition of magnetic impurities does not change this qualitative behavior, the suppression of the critical temperature notwithstanding.

The thermodynamic critical field $H_c$, along with the condensation energy, is also suppressed by the pair-breaking scattering, but depending on material parameters and temperature the speed of this suppression could be larger or smaller than that of $H_{c2}$. On the other hand, the value of $H_c$ relative to $H_{c2}$ is crucial for the type of emerging superconductivity: type-I for $H_c > H_{c2}$, whereas type-II for $H_c < H_{c2}$. A possibility of changing the type of material superconductivity by changing the concentration of magnetic impurities has also been discussed in [22].

The summary of our results for the critical fields at a low temperature is given in Fig. 4.

Using our routine of calculating the condensation energy, we looked at the phase transition between gapped and gapless superconductivity, the question recently attracted the community’s attention. We found that the third derivative of the free energy with respect to the pair-breaking parameter has a singular discontinuous jump at $T = 0$ predicted in [23, 24]. The transition, however, broadens at finite $T$, see Appendix B.

We mention yet another example of the 2nd order phase transition that can be treated within the same formal scheme as $H_{c3}$. This is the problem of nucleation of superconductivity in thin films in parallel applied field [19]. The boundary condition $\nabla_n \Delta(r) = 0$ should now be obeyed at both film surfaces and the emerging state, being physically similar to that of surface superconductivity of SJJDG, can even be nucleated at $T > T_c$ at a non-zero magnetic field. The $T_c(H)$ enhancement in this geometry had been observed [25, 26], but a careful investigation of this intriguing possibility is still to be done.

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Appendix A: The sum $S$ in the presence of magnetic impurities

The solution $f$ of Eq. (1) can be written as

$$f = (2\omega^+ + v\Pi)^{-1}(F/\tau^- + 2\Delta)$$

$$= \int_0^\infty d\rho e^{-\rho(2\omega^+ + v\Pi)}(F/\tau^- + 2\Delta). \quad (A1)$$

Taking the Fermi surface average we get

$$F = \frac{1}{\tau} \int_0^\infty d\rho e^{-2\omega^+ \rho \langle e^{-\rho\Pi} \rangle} (F + 2\Delta \tau^-). \quad (A2)$$

The term $\langle \ldots \rangle$ does not contain the scattering parameters, hence it is the same as that calculated in [18] for the clean case:

$$\langle e^{-\rho\Pi} \tilde{F} \rangle = \sum_{m,j} \frac{(-q^2)^j}{(m!)^2 j!} \frac{(2\mu)!!}{(2\mu + 1)!!} \left( \frac{\rho v}{2} \right)^{2\mu} (\Pi^+)^m (\Pi^-)^m \tilde{F}. \quad (A3)$$

Here $\tilde{F} = F + 2\Delta \tau^-$, $\mu = m + j$, and $\Pi^\pm = \Pi_{xx} \pm i\Pi_{xy}$. After integrating over $\rho$, one obtains from Eq. (A2)

$$F = \frac{1}{2\omega^+ \tau^-} \sum_{m,j} \frac{(-q^2)^j}{j!(2\mu + 1)} \left( \frac{\mu!}{m!} \right)^2 \left( \frac{\ell^+}{\beta^+} \right)^{2\mu} (\Pi^+)^m (\Pi^-)^m \tilde{F}$$

$$\ell^+ = v\tau^+, \quad \beta^+ = 1 + 2\omega^+ \tau^+. \quad (A4)$$

One can check that if no magnetic impurities are involved, this reduces to Eq. (12) of [18]. Using commutation properties of operators $\Pi^\pm$ in uniform field, one manipulates

$$(\Pi^+)^m (\Pi^-)^m \tilde{F} = \tilde{F} \prod_{i=1}^{m} [k^2 + (2i - 1)q^2] \quad (A5)$$

and obtains:

$$F = \Delta \frac{2\tau^- S}{2\omega^+ \tau^- - S}, \quad (A6)$$

$${S} = \sum_{m,j} \frac{(-q^2)^j}{j!(2\mu + 1)} \left( \frac{\mu!}{m!} \right)^2 \left( \frac{\ell^+}{\beta^+} \right)^{2\mu} \prod_{i=1}^{m} [k^2 + (2i - 1)q^2]$$

Appendix B: Condensation energy vs pair-breaking parameter $P_m$

Recently the character of the quantum phase transition between gapped and gapless superconductivity at $T = 0$ [24] when the pair-breaking scattering parameter $P_m$ varies through the value $e^{-\pi/4-\gamma/2} = 0.128$ (see e.g. [16]). Remarkably, it turned out that the superconducting density of states as function of energy $\omega$ and of the order parameter $\Delta(P_m)$ undergoes a topological transition at $P_m = 0.128$. In Ehrenfest classification, the transition
can be considered as of the 2.5-order, at which the third derivative of the free energy $\partial^3 F_c/\partial P_m^3$ is singular.

Although this question is out of main subject of this paper, we utilize here the functional (15), (16), and the condensation energy $F_c(T, P_m)$ of Eq. (17) derived above and valid for any $T$ and any scattering parameters $P, P_m$.

One should mention that solving a coupled system of Eqs. (24) and (25) is not a trivial task. The lower the temperature, the more Matsubara summations are required. Initially, calculations were conducted with the help of Wolfram Mathematica, however, it could not handle the lowest temperatures. Final calculations were performed within MATLAB that still required at least 100,000 summations to obtain the reported results (interested readers are welcome to contact the authors for further technical details).

As can be seen in Fig. 5, temperature affects the behavior of the third derivative of the condensation energy dramatically when compared to the exact result at $T = 0$ obtained using the Eq. (71) from Maki [20]. Our calculations confirm the existence of a very sharp discontinuity of $\partial^3 F_c(T, P_m)/\partial P_m^3$ at $T \rightarrow 0$ at $P_m = 0.128$ (or at $\zeta = \hbar/\Delta \tau_m = (2\pi T_{co}/\Delta)P_m = 1$ in notations of Ref. [23]). However, we were unable to confirm the claim that the discontinuity is preserved at non-zero temperatures [25]; our calculation shows that the singularity in $F_c''''(P_m)$ broadens with increasing $T$. In fact, at finite temperatures, the singularity disappears while its trace moves to lower scattering rates $P_m$. As expected, this confirms that the critical magnetic scattering rate for a transition to the gapless state decreases starting from $P_m = 0.128$ at $T = 0$ to lower values at higher temperatures [27].

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