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Parallel nucleation field in thin superconducting films

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Nucleation of superconductivity in a thin film situated in a parallel magnetic field is considered. The field dependence of the coherence length is taken into account. In particular, the known result for the nucleation field H near the critical temperature T_c is shown to hold only in the dirty limit. For a finite mean free path H^2 depends linearly on T as in the dirty limit. However, the slope dH^2/dT at T_c may change sign to positive for thin and clean enough films.

The problem of the magnetic field parallel to a thin film, at which superconductivity nucleates, has been considered by many authors starting with Ginzburg and Landau (GL) (Ref. 1) (see Ref. 2, and references therein). For a thin enough film near the critical temperature [the thickness d should not exceed $1.84\xi(T)$ with $\xi(T)$ being the coherence length] the order parameter depends only upon the transverse coordinate (x).[3] The persistent current has a "laminar" pattern; for the field in z direction, current lines are parallel to the y axis. In thicker films vortices start to form. Consequently, the order parameter depends on both x and y, and the formal solution becomes more complex.

In this paper only the laminar case is considered for which the nucleation field near the critical temperature T_c was given by

$$H=\phi_0\sqrt{3}/\pi d\xi\ , \tag{1}$$

where ϕ_0 stands for the flux quantum $hc/2e$.[1-3] It will be shown below that, in fact, this result holds only in the dirty limit.

The approach used in this paper is based upon the quasiclassical formulation of the BCS theory[4] which is useful when the spatial dependence of superconducting parameters is involved. This formulation is convenient in particular near the second-order phase transition, where the equations for the quasiclassical Green's functions can be linearized. The formalism has been further simplified in Ref. 5, where it is shown that the nucleation of superconductivity at a phase boundary can be described by the equation

$$\Pi^2F=k^2F\ , \tag{2}$$

where $F(\mathbf{r},\omega)$ is the Gor'kov Green's function integrated over the energy and averaged over the Fermi surface, ω is the Matsubara frequency, $\mathbf{\Pi}=\nabla+2\pi i\mathbf{A}/\phi_0$, and $\mathbf{A}$ is the vector potential.

It is worth noting that Eq. (2) is analogous to the linearized GL equation. Unlike the latter, however, it is valid at any temperature and field provided the parameter k is taken as to satisfy the self-consistency equation

$$\frac{\Delta}{2\pi T}\ln\frac{T_c}{T}=\sum_{\omega>0}\left(\frac{\Delta}{\hbar\omega}-F\right), \tag{3}$$

which relates the pair potential Δ to F. It is shown in Ref. 5 that this requirement yields

$$\frac{\hbar}{2\pi T}\ln\frac{T_c}{T}=\sum_{\omega>0}\left(\frac{1}{\omega}-\frac{2\tau S}{\beta-S}\right). \tag{4}$$

Here $\tau=l/v$, l is the mean free path for nonmagnetic scatterers, v is the Fermi velocity, and $\beta=1+2\omega\tau$. The quantity $S(k,H,\omega,l)$ is given by

$$S=\sum_{m,j=0}^{\infty}\frac{(-q^2)^j}{j!(2m+2j+1)}\left(\frac{(m+j)!}{m!}\right)^2\times\left(\frac{l}{\beta}\right)^{2m+2j}\prod_{i=1}^{m}[k^2+(2i-1)q^2]\ , \tag{5}$$

$$q^2=2\pi H/\phi_0\ .$$

Hence, Eqs. (4) and (5) determine implicitly $k^2(H,T,l)$. It is worth emphasizing that k^2 comes out of the microscopic theory being field dependent.

The cumbersome series (5) covers all known cases where the coherence length, $\xi=(-k^2)^{-1/2}$, was calculated. For $H=0$ and $T>T_c$, only the term $j=0$ contributes; Eqs. (5) and (4) then recover the "pair penetration depth" into the normal phase.[5] At H_{c2}, where $k^2=-q^2$, only the term $m=0$ remains:

$$S(H_{c2})=\sum_{j=0}^{\infty}(-1)^j\frac{j!}{2j+1}\left(\frac{lq}{\beta}\right)^{2j}=\frac{2\beta}{lq}\int_0^{\infty}e^{-\omega^2}\tan^{-1}(\omega lq/\beta)d\omega\ . \tag{6}$$

This case has been studied in detail by Helfand and Werthamer, and by Eilenberger.[6] The dirty limit $H_{c2}(T)$ is obtained if one keeps only the terms $j=0,1$ and substitutes the truncated $S(H_{c2})$ in Eq. (4).

In a similar way, one should retain in series (5) only the lowest powers of $l\,(m+j=0,1)$ when treating the dirty situation in an arbitrary field (not necessarily equal to 0 or H_{c2}). One derives readily first corrections to the dirty-limit theory by keeping an extra term $(m+j=0,1,2)$:

$$S=1+\frac{l^2k^2}{3\beta^2}+\frac{l^4}{5\beta^4}(k^4+q^4)\ . \tag{7}$$

In order to obtain k^2 for the "moderately dirty" situation one substitutes (7) in Eq. (4) and solves the latter with respect to k^2 for a fixed H, T, and l. In the dirty limit, k^2 so obtained is H independent: The field dependence enters only the last term in Eq. (7), which is neglected in the dirty case. Already the first correction to this limit introduces the H dependence of k.

Let us consider now the problem of the nucleation field $\mathbf{H}=H\hat{\mathbf{z}}$ parallel to a thin film situated in the yz plane. The phenomenon is described by Eq. (2), where $F=F(x,\omega)$. One can take the middle plane of the film as $x=0$ and choose the gauge $A_x=A_z=0$ and $A_y=Hx$. The operator $\mathbf{\Pi}=\{\partial/\partial x, 2\pi i Hx/\phi_0, 0\}$, and Eq. (2) for $F(x)$ reads

$$F''-q^4x^2F=k^2F\,, \tag{8}$$

with q defined in Eq. (5). Introducing q^{-1} as a unit length we obtain Eq. (8) in the dimensionless form

$$F''+(\eta-t^2)F=0,\quad \eta=-k^2/q^2,\quad t=qx\,, \tag{9}$$

which is further reduced to the confluent hypergeometric equation with substitutions $F=u(v)\exp(-v/2)$, $v=t^2$. The general solution in the notation of Ref. 7 can then be written as a sum of even and odd functions:

$$F=e^{-t^2/2}\left[C_1M\left(\frac{1-\eta}{4},\frac{1}{2},t^2\right)+C_2tM\left(\frac{3-\eta}{4},\frac{3}{2},t^2\right)\right]. \tag{10}$$

In the dirty limit the current density is proportional to $\mathrm{Im}F^*\Pi_yF=q^2xF^2(x)$ for a real F. The total current in the film must vanish, i.e., F should be either even or odd in x. On the other hand, in a small field $F(x)$ should be close to the constant value which would be found in the absence of the field. Thus the odd possibility is excluded. Essentially the same argument can be used in the general case to show that within the gauge $A=Hx$, $F(x)$ must be an even function and, therefore, $C_2=0$. The boundary condition $F'(\pm d/2)=0$ now yields

$$(1-\eta)M\left(\frac{5-\eta}{4},\frac{3}{2},t_0^2\right)=M\left(\frac{1-\eta}{4},\frac{1}{2},t_0^2\right),\quad t_0=qd/2\,. \tag{11}$$

This equation gives the nucleation field as a function of d at any temperature.

Near T_c one expects $q^2\propto H\propto|T_c-T|^{1/2}$, while $k^2\propto|T_c-T|$. Then both $\eta=-k^2/q^2$ and t_0^2 behave as $|T_c-T|^{1/2}$. Kummer's series[7] for the functions M in Eq. (11) converge rapidly and one can truncate them at the first nontrivial terms:

$$(1-\eta)\left(1+\frac{5-\eta}{6}t_0^2\right)=1+\frac{1-\eta}{2}t_0^2\,. \tag{12}$$

Keeping only terms of the order $|T_c-T|^{1/2}$, i.e., neglecting ηt_0^2, one obtains $t_0^2=3\eta$ or

$$q^4=(2\pi H/\phi_0)^2=-12k^2/d^2\,. \tag{13}$$

[The result is the same if one keeps terms ηt_0^2 and t_0^4 of the order $|T_c-T|$. Numerical solution of Eq. (11) shows that Eq. (13) holds within less than 3% accuracy up to $t_0=1$ and $\eta=\frac{1}{3}$.] Thus, the assumption $q^4\propto k^2\propto|T_c-T|$ is, in fact, justified. Equation (13) coincides with Eq. (1) obtained within the GL method, if one replaces $-k^2$ in (13) with ξ^{-2}, where the coherence length ξ is understood as $(\phi_0/2\pi H_{c2})^{1/2}$ [the minus sign in Eq. (13) is due to the choice of the sign in Eq. (2)]. Note that, in general, Eq. (13) does not solve the problem. The parameter k^2 itself depends upon H (or q) so that Eq. (13) gives H only implicitly.

To evaluate $k^2(H,T)$ in the GL domain one notes that as $T\to T_c$, both k^2 and q^2 go to zero. Then one can retain only the terms with $m+j=0,1,2$ in the series (5). Formally, the procedure is the same as that used to derive Eq. (7). Note, however, that in the moderately dirty case parameters lk and lq are small due to a small l, whereas in the GL domain they are small because of k and q. Consequently, Eq. (7) holds in the moderately dirty case for any T, while in the GL domain it is valid for any l.

Let us assume once again that $q^4\propto k^2\propto|T_c-T|$; the assumption will be justified by the result. Then one can neglect k^4 with respect to q^4 in the last term of Eq. (7). Substitute now this short version of Eq. (7) in Eq. (4) and, after simple algebra, obtain near T_c,

$$\frac{\hbar(T_c-T)}{2\pi T_c^2}=-\frac{l^2k^2}{6\tau}\sum_{\omega>0}\frac{1}{\omega^2\beta}-\frac{l^4q^4}{10\tau}\sum_{\omega>0}\frac{1}{\omega^2\beta^3}\,, \tag{14}$$

where T is replaced with T_c everywhere except the term $\ln(T_c/T)=(T_c-T)/T_c$.

At $q=0$, Eq. (14) yields the known result for the GL coherence length:

$$k^2(0,T)=-\xi^{-2}=-\frac{3\tau\hbar(T_c-T)}{\pi T_c^2l^2}\left(\sum\frac{1}{\omega^2\beta}\right)^{-1}. \tag{15}$$

With the choice of the sign of k^2 in Eq. (2), $k^2(0,T)<0$ in the superconducting phase, while it is positive in the normal one. It should be stressed that both Eqs. (14) and (15) are valid below T_c as well as above it.

In the field one obtains the following from Eqs. (14) and (15):

$$k^2(H,T)=k^2(0,T)-3l^2q^4\gamma(\lambda)/5\,,$$
$$\gamma(\lambda)=\frac{\lambda^2\sum_{n=0}^{\infty}(2n+1)^{-2}(2n+1+\lambda)^{-3}}{\sum_{n=0}^{\infty}(2n+1)^{-2}(2n+1+\lambda)^{-1}}\,, \tag{16}$$

where the impurity parameter $\lambda=\hbar v/2\pi T_cl$. It is seen that in the limit $l\to0$, k^2 is field independent. The first correction to the dirty limit ($\lambda\gg1,\gamma\approx1$) yields

$$k^2(H,T)=k^2(0,T)-3l^2q^4/5\,. \tag{17}$$

In the clean case ($\lambda\ll1$), $\gamma(\lambda)=0.955\lambda^2$, and[8]

$$k^2(H,T)=k^2(0,T)-0.573\left(\frac{\hbar v}{2\pi T_c}\right)^2q^4\,. \tag{18}$$

To find the nucleation field in the GL domain one substitutes $k^2(H,T)$ of Eq. (16) in (13) and solves for H,

$$\left(\frac{2\pi H}{\phi_0}\right)^2=-\frac{12k^2(0,T)}{d^2-7.2\gamma(\lambda)l^2}\,. \tag{19}$$

This coincides with Eq. (1) only in the dirty limit. For any nonvanishing l the slope of $H^2(T)$ at T_c is larger than that predicted in Eq. (1).

The denominator in Eq. (19) changes sign at a critical thickness d_c such that

$$d_c^2=7.2\gamma(\lambda)l^2 . \qquad (20)$$

The value of d_c increases from $2.68l$ for $\lambda \gg 1$ up to the clean limit $2.62\hbar v/2\pi T_c$.[9] For $d > d_c$, the curve $H^2(T)$ is situated in the domain $T < T_c$, where $k^2(0,T)$ is negative. For $d < d_c$ there are no solutions of Eq. (19) for $T < T_c$; the curve $H(T)$ starts at T_c and bends to higher temperatures: $H \propto (T-T_c)^{1/2}$ as shown in Fig. 1.

The prediction just made, for thin enough and sufficiently clean films, implies that the phase boundary for these films might have a shape sketched by the dashed line in Fig. 1. In particular, it would mean that the critical temperature, $T_c(H)$, of a film can be enhanced by applying a parallel magnetic field (which should not be too large). Then, some temperature, T^*, should exist at which the curve $H(T)$ turns over to the usual behavior and $H'(T^*)=\infty$ (see Fig. 1). In the domain (T_c,T^*) function $H(T)$ is double valued. At a constant T in this domain the superconductivity being absent in zero field, should occur at a field H_1 and disappear again at some $H_2 > H_1$. This type of behavior of the phase boundary $H(T)$ does not violate any basic physical requirement. In fact, some magnetic structures show "field-induced superconductivity."[10] It would be surprising, however, to have the field-induced superconducting phase in a plane film made of a BCS kind of metal.

Further study is needed to estimate a possible critical temperature enhancement, T^*-T_c. Within the method used the difficulty of this calculation lies primarily in evaluation of the function S represented by the asymptotic series (5) out of the immediate vicinity of the point $T=T_c$, $H=0(k=q=0)$. A mere addition of terms in the truncated series S makes the problem analytically untractable and an outcome absolutely unreliable. One may try

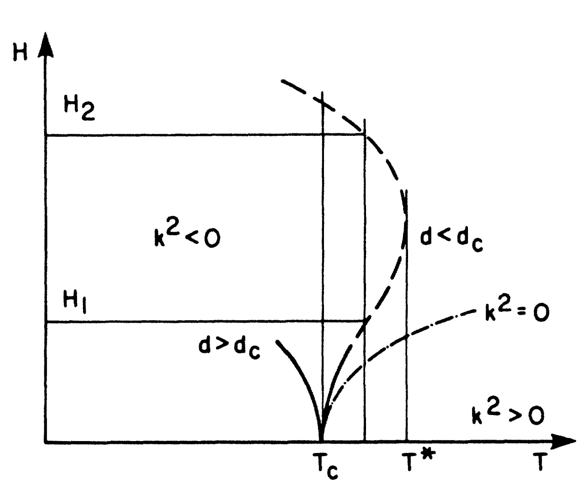

FIG. 1. The phase diagram of a thin film. The nucleation field $H(T)$ for $d < d_c$ is sketched by the dashed line. The curve $k^2=0$ separates domains of positive and negative $k^2(H,T)$.

to obtain an integral representation of S similar to that given in Eq. (6) for $S(H_{c2})$. Progress in this direction has been made and will be reported elsewhere. It should be noted, however, that as far as the slope dH^2/dT at zero field T_c is concerned, the truncation used above is certainly justified. In this respect, the condition $d < d_c$ for the anomalous phase boundary to occur holds irrespective of however important and intriguing the behavior of the whole curve $H(T)$ could be.

Formally, one can draw a boundary in the H,T plane which cannot be crossed by the nucleation curve $H(T)$. To this end, integrate Eq. (8) over the film and take the condition $F'(\pm d/2)=0$ into account to obtain $q^4=-k^2\langle F\rangle/\langle x^2F\rangle$, where $\langle\,\rangle$ stands for integrals over x. Hence, the whole curve $H(T)$ must be situated in the domain $k^2(H,T)<0$ of the H,T plane.

The function $k^2(H,T)$ has been studied in Ref. 5; we describe briefly its relevant features. First, $k^2=-2\pi H_{c2}/\phi_0$ at the bulk upper critical field $H_{c2}(T)$. Further, $k^2>0$ in zero field for $T>T_c$; Eq. (2) in this region describes an exponential attenuation of superconductivity in the normal phase. Being a continuous function of H and T, k^2 must turn zero at a curve which starts at the zero field T_c. As H increases, this curve bends to higher temperatures for any $l\neq 0$; it is shown by the dash-dot line in Fig. 1. Hence, the nucleation field in a film with $d<d_c$ is situated above the boundary at which k^2 changes sign. In other words, the coherence length ξ defined by $\xi^2=-k^{-2}(H,T)$ at the phase boundary $H(T)$ is real, even if a part of the curve $H(T)$ lies in the domain $T>T_c$.

The effect described is a direct consequence of the H dependence of the coherence length. To demonstrate this qualitatively let us first observe that Eq. (2) is similar to the Schrödinger equation for a particle in uniform magnetic field. In zero field we have $-F''=\varepsilon_0 F$ with $\varepsilon_0=-k^2(0,T)$ being the "energy eigenvalue." Under the condition $F'(\pm d/2)=0$, the solution is $F=F_0=$const and $k^2(0,T)=0$; this happens at $T=T_c$. When a small field is applied, the coherence length becomes shorter: $\xi^2(H,T)=\xi^2(0,T)-\alpha^2H^2$ with a positive constant α^2 (the first correction due to the field must be even in H). This translates into $-k^2(H,T)=-k^2(0,T)+L^2q^4$ with some positive material-dependent constant L^2. Substitute this in Eq. (8) and rearrange terms to obtain

$$-F''+q^4(x^2-L^2)F=-k^2(0,T)F .$$

In a small field the "potential," $V=q^4(x^2-L^2)$, may be treated as a perturbation so that the new "energy" $\varepsilon=\varepsilon_0+\langle V\rangle$. This yields

$$-k^2(0,T)=-k^2(0,T_c)+F_0^2dq^4(d^2-12L^2)/12 .$$

Now, normalize the unperturbed "wave function," $F_0^2d=1$, denote $d_c^2=12L^2$, and recall that $k^2(0,T_c)=0$ to obtain the main result given in Eq. (19).

One might wonder why the result (1) derived from the GL equations (the latter are obtained from the microscopic theory for *any* impurity concentration) turns out to hold only in the dirty limit? To answer the question one recalls that the GL equations are derived from the microscopic theory not under the sole condition $|T_c-T|\ll T_c$. An additional restriction is imposed upon the magnetic field which must be small. As Ref. 11 indicates, the field is not supposed to exceed the order of magnitude of H_{c2}. The latter is proportional to $(T_c - T)$ and therefore it is much smaller than the nucleation field in a thin film proportional to $|T_c - T|^{1/2}$. Corrections to the GL equations when the field is no longer small in the sense indicated, are hard to obtain using the Gorkov's equations as a starting point. The quasiclassical Eilenberger's method is more suitable for these corrections to be made. One obtains as a result the field dependence of the coherence length.

Reservations might be expressed as to validity of the quasiclassical formalism in films for which the thickness is on the order of the mean free path or of the zero-temperature coherence length (in the clean case). The very presence of sharp film surfaces might be a problem for the quasiclassical method which is valid, strictly speaking, only for spacial variations slow with respect to k_F^{-1}. In this context the question arises whether or not the boundary condition $F'(\pm d/2) = 0$ used above is the right one. To estimate how sensitive the effect described might be to boundary conditions, perhaps the most severe one, $F(\pm d/2) = 0$, was tested. Although the expression for d_c came out different and more restrictive than Eq. (20), there is still a "window" of l's and d's where the effect should occur.

In any event, experimental verification (or otherwise) of the effect described is desirable, although it might prove to be difficult. The point is that the requirements of "thin and clean enough" are usually hard to meet in the same film. This might be the reason why the effect has not been already observed. The existing data on the parallel nucleation field in thin films confirm the $|T_c - T|^{1/2}$ temperature dependence. However, to the author's knowledge, the slope dH^2/dT at T_c for some reason was neither monitored carefully nor was it compared with other characteristics of the same film (unlike the case of the perpendicular field). Thus, both the commonly accepted equation (1) and equation (19) obtained in this work, are still subject to experimental scrutiny.

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