

SUPERCONDUCTING CHARACTERISTICS OF RATHER CLEAN THIN FILMS IN STRONG PARALLEL FIELDS

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The Green's functions of a thin film superconductor are calculated to all orders in the ordering parameter in the presence of very strong magnetic fields parallel to the surface, when expansion in powers of the field strength is not appropriate. From these Green's functions the magnitude of the order parameter and the tunneling current are calculated near the critical field.

THE parallel critical fields and tunneling currents for dirty thin superconducting alloy films have recently been shown by Guyon, Meunier, and the author<sup>[1]</sup> to agree very well experimentally with the theory of Maki,<sup>[2]</sup> as extended by us and Baratoff<sup>[3]</sup> to include nonlocal electrodynamics. However, only one experiment was performed on a film of the pure metal because of the lack of a complete theoretical treatment valid for cleaner samples.

Using a quasiclassical method de Gennes and Tinkham<sup>[4]</sup> and Shapoval<sup>[5]</sup> have calculated the value of the critical field expected in all limiting cases of mean free path length and temperature for thin films with diffuse boundary reflection. The present work extends their results by calculating the Green's functions to all orders in the order parameter, from which the magnitude of the order parameter and the tunneling current near the critical field are obtained. In the case of an absolutely clean film at intermediate temperatures the value of the order parameter agrees with that obtained by Ovchennikov,<sup>[6]</sup> which we write more exactly.

1. CLEAN FILMS

We consider first the case of clean films, those having no defects in the bulk, but with diffuse boundary scattering. The anomalous Green's function  $F_{\omega}(\mathbf{r}, \mathbf{r})$  of Gor'kov<sup>[7]</sup> evaluated at one point was found previously to first order in the order parameter  $\Delta(\mathbf{r})$  using the Green's functions  $G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}')$  of the normal metal:<sup>[4, 5]</sup>

$$F_{\omega}^{(1)}(\mathbf{r}, \mathbf{r}) = \int G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}') G_{-\omega}^{(0)}(\mathbf{r}, \mathbf{r}') \Delta(\mathbf{r}') d\mathbf{r}'$$

$$= 2\pi N \left\langle \int_0^{\infty} dt e^{-2|\omega|t + i\varphi(t)} \Delta(t) \right\rangle. \tag{1}$$

The average  $\langle \dots \rangle$  is over all classical electron trajectories at the Fermi surface. The phase

$$\varphi(t) = \varphi(0, t) = 2e \int_0^t \mathbf{A} \cdot \mathbf{v} dt,$$

is twice the line integral of the vector potential  $\mathbf{A}_{\mathbf{y}} = H_z \mathbf{x}$  along the trajectory with velocity  $\mathbf{v}$ .  $N$  is the density of states.  $\omega = (2n + 1) \pi T$ .

The film thickness  $d$  is required to be much less than the bulk coherence length  $\xi_0$ . If in addition

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$d \gg (1 - t_r) \xi_0$ , where  $t_r$  is the reduced temperature  $T/T_c$ , then the critical field will be small,  $eH_c d^2 \ll \omega d/v$ . This case has been treated by expanding in powers of the field strength.<sup>[3]</sup>

What interests us now is the case of strong critical fields,  $eH_c d^2 \gg \omega d/v$ , obtained when  $d \ll (1 - t_r) \xi_0$ . For small  $\omega$ ,  $\omega d/v \ll eH d^2$  and 1, many collisions with the boundaries occur in the relevant time interval  $t \sim 1/\omega$ , so the contribution from the direct trajectory without collisions may be neglected. The portion of a trajectory from one boundary to the other gives no contribution to the phase  $\varphi$ . The important contributions come from the two portions of a trajectory from an end point in the interior to a boundary. Due to the diffuse reflection  $\mathbf{v}(t)$  and  $\mathbf{r}(t)$  are uncorrelated with  $\mathbf{v}(0)$  and  $\mathbf{r}(0)$ , and the averages may be taken separately. The averaged contribution of a trajectory from the interior to a boundary is just.<sup>[4]</sup>

$$\langle e^{i\varphi} \rangle = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} d\psi \exp \left[ ieH \left( \frac{d^2}{4} - x^2 \right) \text{tg } \psi \right]$$

$$= \exp \left[ -eH \left( \frac{d^2}{4} - x^2 \right) \right]. \tag{2}$$

Since the average value of  $\mathbf{x}(t)$  for large  $t$  is independent of time, the average over  $\mathbf{x}(t)$  is carried out independently of the integration over  $t$ , and we obtain  $F^{(1)}$ :

$$F_{\omega}^{(1)}(\mathbf{r}, \mathbf{r}) = \frac{\pi N}{|\omega|} \exp \left( -eH \left( \frac{d^2}{4} - x^2 \right) \right)$$

$$\times \frac{1}{d} \int_{-d/2}^{d/2} d\xi \exp \left[ -eH \left( \frac{d^2}{4} - \xi^2 \right) \right] \Delta(\xi) d\xi.$$

It follows that the order parameter

$$\Delta(x) = \lambda T \sum_{\omega} F_{\omega}(\mathbf{r}, \mathbf{r})$$

has this same exponential dependence on  $\mathbf{x}$ .<sup>[5]</sup>  $F^{(1)}$  and the critical field are expressed in terms of the function  $f$  introduced by Shapoval:<sup>[5]</sup>

$$f = \frac{2}{d} \int_0^{d/2} d\xi \exp \left[ -2eH \left( \frac{d^2}{4} - \xi^2 \right) \right] d\xi,$$

and the value of the order parameter at a boundary, which we write as  $\Delta$  without an argument:

$$F_{\omega}^{(1)}(\mathbf{r}, \mathbf{r}) = \frac{\pi N}{|\omega|} \exp \left[ -eH \left( \frac{d^2}{4} - x^2 \right) \right] f \Delta. \tag{3}$$

The next order term in  $\Delta$  of  $F$  is  $F^{(3)}$ :

$$F_{\omega}^{(3)}(\mathbf{r}, \mathbf{r}) = - \int G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}_1) \Delta(\mathbf{r}_1) G_{-\omega}^{(0)}(\mathbf{r}_2, \mathbf{r}_1) \Delta^*(\mathbf{r}_2) \times G_{\omega}^{(0)}(\mathbf{r}_2, \mathbf{r}_3) \Delta(\mathbf{r}_3) G_{-\omega}^{(0)}(\mathbf{r}, \mathbf{r}_3) d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3. \quad (4)$$

Using the reasoning of de Gennes and Tinkham<sup>[4]</sup> we arrive at the result:

$$F_{\omega}^{(3)}(\mathbf{r}, \mathbf{r}) = -2\pi N \left\langle \int_0^{\infty} dt_1 \int_0^{\infty} dt_2 \int_0^{t_1+t_2} dt_3 \exp[-2|\omega|(t_1+t_3) + i\varphi(0, t_1) + i\varphi(t_1-t_2, t_1-t_2+t_3)] \Delta(t_1) \Delta^*(t_1-t_2) \Delta(t_1-t_2+t_3) \right\rangle. \quad (5)$$

The times concern four successive motions along the trajectory, forward and then backward, finally returning to the initial point. The limit  $t_2 < t_1 + t_3$  arises from the requirement that  $t_4$  be positive. Again the values of  $\mathbf{v}$  and  $\mathbf{r}$  at large times are uncorrelated with their initial values and may be averaged independently of the time integrals. The phase contributions are divided between one for the initial point and one for each  $\Delta(t)$ .

$$F_{\omega}^{(3)}(\mathbf{r}, \mathbf{r}) = - \frac{\pi N}{2|\omega|^3} \exp\left[-eH\left(\frac{d^2}{4} - x^2\right)\right] (f\Delta)^3. \quad (6)$$

The generalization to all orders in  $\Delta$  is thus evident:

$$F_{\omega}(\mathbf{r}, \mathbf{r}) = \frac{\pi N f \Delta}{[\omega^2 + (f\Delta)^2]^{3/2}} \exp\left[-eH\left(\frac{d^2}{4} - x^2\right)\right]. \quad (7)$$

The Green's function  $G_{\omega}(\mathbf{r}, \mathbf{r})$  is obtained in the same manner. The normal state Green's function  $G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r})$  is independent of the field in this approximation.

$$G_{\omega}^{(2)}(\mathbf{r}, \mathbf{r}) = - \int G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}_1) \Delta(\mathbf{r}_1) G_{-\omega}^{(0)}(\mathbf{r}_2, \mathbf{r}_1) \Delta^*(\mathbf{r}_2) G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2 = 2i\pi N \text{sign } \omega \int_0^{\infty} dt_1 \int_{-\infty}^0 dt_2 \exp[-2|\omega|(t_1-t_2) + i\varphi(t_1) - i\varphi(t_2)] \Delta(t_1) \Delta^*(t_2) = i \frac{\pi N \text{sign } \omega}{2\omega^2} (f\Delta)^2. \quad (8)$$

To all orders in  $\Delta$ :

$$G_{\omega}(\mathbf{r}, \mathbf{r}) = \pi N \omega / i [\omega^2 + (f\Delta)^2]^{3/2}. \quad (9)$$

We note that the density of states  $N(\omega) = -\text{sign } \omega \pi^{-1} \text{Im } G_{1\omega}$  is independent of the initial direction of the electron's velocity, in contrast to the case of a clean film with specular boundary reflection investigated by Ovchennikov,<sup>[8]</sup> and that there is an energy gap in the spectrum of  $2f\Delta$ . Further we observe that  $N(\omega)$  is independent of position, in contrast to the dirty case.<sup>[9]</sup> Due to the independence of  $G_{\omega}(\mathbf{r}, \mathbf{r})$  on position the screening current vanishes in this approximation. Therefore the phase transition to the normal state will be of second order.

The magnitude of  $\Delta$  is obtained from  $F_{\omega}(\mathbf{r}, \mathbf{r})$ :

$$\Delta = \lambda T \sum_{\omega} \frac{f\Delta}{2[\omega^2 + (f\Delta)^2]^{3/2}}$$

The effective interaction strength  $\lambda$  is determined from  $T_c$  and may be eliminated:

$$\ln t_r = 2\pi T \sum_{\omega > 0} \left( \frac{f}{[\omega^2 + (f\Delta)^2]^{3/2}} - \frac{1}{\omega} \right). \quad (10)$$

As stated earlier, the expression for  $F$  (7) applies only for small  $\omega$ . From the work of Shapoval<sup>[5]</sup> to logarithmic

mic accuracy  $f$  is replaced by 1 when  $\omega > eHd$ . If  $eHd^2 > 1$  then in the region  $v/d < \omega < eHd$   $f$  is replaced by 0. The critical field  $H_c$ , as shown by Shapoval,<sup>[5]</sup> follows from the equation:

$$\ln t_r = 2\pi T \sum_{\omega > 0} \frac{f_c - 1}{\omega}, \quad f_c = f(H_c). \quad (11)$$

To obtain the magnitude of  $\Delta$  near the critical field we expand (10) in powers of  $\Delta^2$  and subtract (11):

$$\pi T \sum_{\omega > 0} \frac{f - f_c}{\omega} = \pi T \sum_{\omega > 0} \frac{f^3 \Delta^2}{2\omega^3}. \quad (12)$$

The sum over  $\omega$  on the left side of (12) is the same as on the right side of (11) if  $eHd^2 < 1$ , so we obtain

$$\frac{7\zeta(3)}{8} \frac{f^3 \Delta^2}{(\pi T)^2} = (f - f_c) (f_c - 1)^{-1} \ln t_r = \frac{f - f_c}{f} \left[ \ln 4\pi e H d \xi_0 - \frac{7}{3} \right]. \quad (13)$$

When  $eHd^2 \ll 1$  the result of Ovchennikov<sup>[6]</sup> (and additionally the coefficient of the argument of the logarithm) is obtained by substituting  $f = 1 - 1/3 eHd^2$ . When  $eHd^2 \gg 1$  the second equality of (13) still holds to logarithmic accuracy, and the limiting form  $f = 1/eHd^2$  may be substituted.

The tunneling current from a superconducting film through a barrier into a normal metal is the same as from a bulk type one superconductor with gap  $f\Delta$ . If  $f\Delta \ll \pi T$  the density of states may be expanded in powers of  $\Delta/\pi T$  and the differential conductivity obtained:<sup>[10]</sup>

$$d(V) = \frac{1}{C} \frac{dI}{dV} = 1 + \frac{1}{8} \left( \frac{f\Delta}{\pi T} \right)^2 \text{Re } \psi_3 \left( \frac{1}{2} - \frac{ieV}{2\pi T} \right), \quad (14)$$

where  $C$  is the normal state conductivity,  $V$ —the potential difference, and  $\psi_n$ —the  $(n-1)$ st derivative of the digamma function. Thus the slope  $S$  at the critical field, which is usually measured,<sup>[11]</sup> is

$$S = - \frac{2f'}{f^2} \ln(1.22eHd\xi_0), \quad (15)$$

where  $f' = df/dH$  and the limiting forms of  $-f'/f^2$  are  $ed^2/3$  where  $eHd^2 \ll 1$  and  $ed^2$  when  $eHd^2 \gg 1$ .

## 2. FILMS WITH IMPURITIES OR DEFECTS

Scattering from inhomogeneities in the film interior may be included using the techniques developed by Abrikosov and Gor'kov.<sup>[11]</sup> As we noted earlier<sup>[3]</sup> their original derivation also applies in the presence of fields and spatial variations of  $\Delta(\mathbf{r})$  if isotropic scattering is assumed. Their method replaces  $\omega$  and  $\Delta$  in the Green's functions by  $\bar{\omega}$  and  $\bar{\Delta}$ , where

$$\bar{\omega} = \omega + \frac{1}{2\tau} \frac{i}{\pi N} G_{\omega}(\mathbf{r}, \mathbf{r}), \quad \bar{\Delta}(x) = \Delta(x) + \frac{1}{2\tau} \frac{1}{\pi N} F_{\omega}(\mathbf{r}, \mathbf{r}); \quad (16)$$

and  $v\tau = l$ —the mean free path.

The expansions in powers of the field strength used earlier<sup>[3]</sup> apply when

$$eH_e d^2 \ll \frac{d}{v} \left( \omega + \frac{1}{2\tau} \right),$$

obtained when  $(1 - t_r)\xi^2 \ll d\xi_0$  and  $\xi_0 \xi$ , where  $\xi^{-1} = \xi_0^{-1} + l^{-1}$ . We consider thin films with thickness  $d < \sqrt{\xi_0 \xi}$ .

Now we are interested in the case of stronger critical fields

$$eHcd^2 \gg \frac{d}{v} \left( \omega + \frac{1}{2\tau} \right),$$

obtained for cleaner films at low temperatures, when  $(1 - t_r)\xi^2 \gg d\xi_0$ . From (7) and (8) we obtain F and G.

$$\bar{\omega} = \omega + \frac{1}{2\tau} \frac{\bar{\omega}}{[\bar{\omega}^2 + (f\bar{\Delta})^2]^{1/2}},$$

$$\bar{\Delta}(x) = \Delta(x) + \frac{1}{2\tau} \frac{f\bar{\Delta}}{[\bar{\omega}^2 + (f\bar{\Delta})^2]^{1/2}} \exp \left[ -eH \left( \frac{d^2}{4} - x^2 \right) \right]. \quad (17)$$

So  $\bar{\Delta}(x)$  has the same spatial dependence as  $\Delta(x)$ . Writing  $u = \bar{\omega}/\bar{\Delta}$  we obtain from (17):

$$\frac{\omega}{\Delta} = u \left( 1 - \frac{1-f}{2\tau\Delta} \frac{1}{\sqrt{u^2 + f^2}} \right). \quad (18)$$

In the case  $eHd^2 \ll 1$  this relation for  $u$  has the same form found by Maki<sup>[2]</sup> to apply to dirty superconductors:

$$\frac{\omega}{\Delta} = u \left( 1 - \zeta \frac{1}{\sqrt{u^2 + 1}} \right), \quad (19)$$

where  $\zeta = 2\tau\alpha/\Delta$  and  $\alpha$  is now  $1/3 eHd^2/(2\tau)^2$  instead of Maki's value  $e^2H^2(d^2/12)(v^2/3)$ .

The density of states is given by

$$-\text{sign } \omega \text{ Im} (Nu / \sqrt{V^2 - u^2}),$$

and thus the gap is

$$\omega_0 = f\Delta \left[ 1 - \left( \frac{1-f}{2\tau f\Delta} \right)^{2/3} \right]^{3/2} \text{ when } \frac{1-f}{2\tau f\Delta} < 1,$$

is less than 1 and vanishes when it is greater.

The critical field and magnitude of  $\Delta$  near  $H_C$  are determined from the analogy to (10):

$$\ln t_r = 2\pi T \sum_{\omega>0} \left( \frac{f}{\Delta\sqrt{u^2 + f^2}} - \frac{1}{\omega} \right). \quad (20)$$

Using (18) the critical field follows from (20) when  $\Delta \rightarrow 0$ .

$$\ln t_r = 2\pi T \sum_{\omega>0} \left( \frac{f}{\omega + (1-f)/2\tau} - \frac{1}{\omega} \right), \quad (21)$$

Expanding (20) in powers of  $\Delta^2$  near the critical field we find its magnitude.

$$\pi T \sum_{\omega>0} (j - f_c) \left\{ \frac{f}{2\tau(\omega + (1-f)/2\tau)^2} + \frac{1}{\omega + (1-f)/2\tau} \right\}$$

$$= \frac{1}{2} \sum_{\omega>0} \frac{\omega}{(\omega + (1-f)/2\tau)^4} f^3 \Delta^2. \quad (22)$$

Similarly expanding the density of states, the differential conductivity is given by:

$$D(V) = 1 + \frac{1}{8} \left( \frac{f\Delta}{\pi T} \right)^2 \text{Re } \psi_3 \left( \frac{1}{2} + \frac{1-f}{4\tau\pi T} - \frac{ieV}{2\pi T} \right). \quad (23)$$

In the case of pure films ( $\tau \rightarrow \infty$ ) these results coincide with (11), (12), and (14). On the other hand, in case  $\xi_0 \gg l \gg \sqrt{\xi_0 d}$  the results are closely analogous to those in the dirty limit. As shown by Shapoval<sup>[5]</sup> the critical field is given by

$$1 - f_c = 1/3 eH_c d^2 = \tau/\tau_u(t_r), \quad (24)$$

where  $\tau_u(t_r)$  is the universal function of reduced temperature which occurs in many expressions evaluated in the dirty limit.

$$\ln t_r = \psi(1/2) - \psi(1/2 + \rho/2), \quad 2\pi T\rho = \tau_u(t_r)^{-1}.$$

Therefore  $\Delta^2$  and  $S$  may be expressed in terms of the same universal functions of temperature used in the dirty limit. In particular:

$$\Delta^2 = 4\pi^2 T^2 \rho \psi_2 / f_1, \quad (25)$$

$$S = \frac{\rho \psi_2}{H_c} \left( -\frac{\psi_3}{2f_1} \right), \quad (26)$$

where the arguments of the  $\psi$  functions are  $1/2 + \rho/2$  and

$$f_1 = \sum_{n=0}^{\infty} \frac{n + 1/2}{(n + 1/2 + \rho/2)^4} = -\frac{\psi_3}{2} - \frac{\rho}{2} \frac{\psi_4}{6},$$

as introduced by Maki.<sup>[2]</sup>

Thus the value of  $H_C S$  is predicted to be just one-half of the value obtained in the dirty limit.<sup>[1]</sup> (The factor of  $1/2$  arises because  $\rho$  is quadratic in  $H$  in the dirty limit and linear in  $H$  here.) The dependence of  $D(V)$  on  $V$  should be the same as in the dirty limit.

For cleaner films ( $l \sim \xi_0$ ,  $eH_c d^2 \sim 1$ ) the critical field is determined from Shapoval's equation (3.6)<sup>[5]</sup> and has a smaller value than given by (24). To determine  $\Delta$  we again make use of the fact that the same logarithmic term occurs in the equation for  $H_C$ :

$$\frac{f^3 \Delta^2}{2\pi^2 T^2} \frac{f_1}{8} = (j - f_c) \left\{ \frac{f}{2\tau\pi T} \frac{\psi_2}{4} + \frac{1}{2f} \ln(1.22eHd\xi_0) \right\}, \quad (27)$$

$$S = 2 \frac{f'}{f^2} \frac{\psi_3}{2f_1} \left\{ \frac{f^2}{4\tau\pi T} \psi_2 + \ln(1.22eHd\xi_0) \right\}, \quad (28)$$

where the arguments of the  $\psi$  functions are  $1/2 + (1-f)/4\tau\pi T$ . (28) reduces to (15) for absolutely clean samples and to (25) for the dirtier ones. ( $\xi_0 \gg l \gg \sqrt{\xi_0 d}$ ). For temperatures low enough so that  $(1-f)/4\tau\pi T \gg 1$  we may insert the asymptotic forms of the  $\psi$  functions:

$$S \rightarrow 3 \frac{f'}{f^2} \left\{ \frac{f^2}{1-f} + \ln(1.22eHd\xi_0) \right\}. \quad (29)$$

For small fields  $eH_c d^2 \ll 1$   $S \rightarrow 3/H_C$ , which decreases as the film becomes cleaner and  $H_C$  increases. However, when  $eH_c d^2$  becomes much larger than one  $S \rightarrow 3ed^2 \ln eH_c d\xi_0$  and increases logarithmically.

### 3. EXPERIMENTAL DISCUSSION

From an experimental point of view confirmation of the theory is difficult. This is because of the difficulty of producing thin films without defects in their interior. For example the bulk mean free path for the pure film No. 46 of our earlier work<sup>[1]</sup> as well as the width may be inferred from the values of the parallel and perpendicular critical fields measured near  $T_C$ :  $d \approx 300 \text{ \AA}$ ,  $l \approx 700 \text{ \AA}$ . Since  $\xi_0 \approx 2000 \text{ \AA}$ ,  $l \sim \sqrt{\xi_0 d} \approx 800 \text{ \AA}$ , and this film is in between the range of applicability of the theory of the dirty limit and the present theory. Thus the dependence of  $H_C$  on  $[\tau_u(t_r)]^{-1}$  is intermediate between a square root and a linear character (Figure 5 of<sup>[1]</sup>). Similarly the slope of the differential conductivity has a character intermediate between the value applicable to the dirty limit and its half (26), although it is seen that  $S$  approaches the new limit faster than  $H_C$  does (Figure 6 of<sup>[1]</sup>).

If we consider that  $l$  is always limited to approximately  $2.5d$ , the most favorable experimental situation should be  $d \approx 800 \text{ \AA}$  so that  $l \approx \xi_0 \approx 1.6\sqrt{\xi_0 d}$ . Then using Shapoval's Eq. (3.6)<sup>5</sup>  $eH_c d^2$  would be 0.64, and (28) must be used to find  $S$ . If a thicker film is chosen, say  $d \approx 1600 \text{ \AA}$ ,  $l \sim 2\xi_0$ , a higher value of  $eH_c d^2$  is predicted ( $\approx 1.07$ ), but the condition  $d \ll \xi_0$  is not well satisfied.

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