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PHYSICAL REVIEW B

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## Theory of Electron Spin Resonance in Type-I Superconductors\*

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In order to determine whether electron spin resonance in type-I superconductors is experimentally possible, we have extended Dyson's theory to the case of a superconductor, taking into account the penetration depth of electromagnetic fields, the change in phase of the surface impedance, and the change in group velocity and relaxation time of the quasiparticles compared to the normal case. We have calculated line shapes and magnitudes of power absorbed and rf field transmitted for various frequencies and temperatures for thin-film superconductors. From our calculation, we determine the optimum temperatures and frequencies, and conclude that even for these conditions, the experiment would be difficult.

#### I. INTRODUCTION

Three years ago Schultz and co-workers<sup>1</sup> observed conduction electron spin resonance (CESR) in normal aluminum, the first such observation in a metal which undergoes a superconducting transition. Hence, the possibility of observing CESR in the superconducting state was raised. If CESR is observed in a superconductor, we might be able to obtain a key to the spin relaxation mechanism in metals. Below 25  $^{\circ}$ K, the spin relaxation time U in normal Al was found to be temperature independent and values for U were essentially the same for different samples with different resistivity ratios. This leaves the possibility of other mechanisms besides impurity scattering and spin-lattice interactions,<sup>1</sup> Kaplan<sup>2</sup> calculated the shape of CESR absorption curves for superconducting films of thickness less than or the order of the penetration depth  $\lambda$  by using a phenomenological modified Bloch equation. He concluded that CESR should be observable in the superconducting state. Here we extend Dyson's theory<sup>3,4</sup> to the case of a thin-film BardeenCooper-Schrieffer (BCS) superconductor, taking into account changes in the diffusion and spin relaxation of the quasiparticles in the superconducting state. We have calculated not only the line shapes, but also the magnitudes of the power absorbed and rf fields transmitted. We give the temperature and frequency regions for which the resonance will be the largest, and we find that even in the optimum region the resonance will be difficut to observe experimentally.

We consider a film of thickness L parallel to the y-z plane and with surfaces at  $x = -\frac{1}{2}L$  and  $+\frac{1}{2}L$ . A static magnetic field  $H_0$  is applied in the z direction. The linearly polarized rf with its magnetic field  $H_1$  in the y direction will be considered for two situations. (a) The rf field impinges on the film from the negative x region. In this case we shall be interested in the power transmitted through the film into the positive x region. (b) The rf field is set up on both sides of the film with  $H_1(\frac{1}{2}L)$  $=H_1(-\frac{1}{2}L)$ . In this case we are interested in the power absorbed in the film. We shall refer to these cases as (a) and (b), respectively. Both cases are used experimentally.<sup>1,5</sup>

The rf fields then depend only on x and t with

$$\vec{\mathbf{H}}_{1}(x,t) = \hat{y} (H_{1}(x)e^{-i\omega t} + c. c.) ,$$

$$\vec{\mathbf{E}}_{1}(x,t) = -\hat{z} (E_{1}(x)e^{-i\omega t} + c. c.) .$$
(1)

The average power P absorbed by the film per unit area is

$$P = (c^2/8\pi^2) \left| H_1(-\frac{1}{2}L) \right|^2 \operatorname{Re} Z , \qquad (2)$$

where Z is the surface impedance,

$$Z = (4\pi/c) E_1(-\frac{1}{2}L) H_1^*(-\frac{1}{2}L) / \left| H_1(-\frac{1}{2}L) \right|^2.$$
(3)

### **II. SPIN POLARIZATION IN A SUPERCONDUCTOR**

The main differences in properties between the normal and superconducting states are that in a superconductor (i) the static magnetic field  $H_0$  penetrates only a penetration depth  $\lambda$ ; (ii) for the rf field,  $\lambda$  takes the place of the skin depth  $\delta$  of the normal metal and there is a resulting change in phase of the surface impedance; and (iii) the velocity  $v_{\vec{k}}$  and the spin-relaxation time  $U_{\vec{k}}$  of the quasiparticle with wave vector  $\vec{k}$  are  $(|\epsilon_{\vec{k}}|/E_{\vec{k}})v_F$  and  $(|\epsilon_{\vec{k}}|/E_{\vec{k}})U$ , respectively, where  $v_{\vec{k}}$  and U are the Fermi velocity and spin-relaxation time for elastic scatterings in the normal state. Here  $\epsilon_{\vec{k}}$  and  $E_{\vec{k}}$ are energies of the normal and the superconducting quasiparticles, respectively.  $E_{\vec{k}} = (\epsilon_{\vec{k}}^2 + \Delta^2)^{1/2}$ , where  $\Delta$  is the superconducting gap.

The proof concerning  $U_{\vec{k}}$  is the following: The interaction responsible for spin-flip scattering is

$$\mathcal{H}_{sf} = \sum_{\vec{k},\vec{k}^{*}} v(\vec{k} - \vec{k}')(c_{\vec{k}^{*}}^{*}, c_{\vec{k}^{*}} + c.c.) , \qquad (4)$$

where  $c_{k}^{\dagger}$ , is the creation operator for the electronic Bloch state  $\bar{k}$ , spin up, and with wave function  $\psi_{\vec{r}_1}(x)$ . This state refers to a normal quasiparticle. One can show that

$$\sum_{\vec{\mathfrak{g}},\vec{\mathfrak{k}}'} \langle 0 \mid \gamma_{\vec{\mathfrak{q}}' 1} c_{\vec{\mathfrak{k}}' 1}^* c_{\vec{\mathfrak{g}}}, \gamma_{\vec{\mathfrak{q}}0}^* \mid 0 \rangle = 1, \qquad (5)$$

where  $|0\rangle$  is the BCS<sup>6</sup> superconducting ground state, and  $\gamma_{q0}$  and  $\gamma_{q1}$  are Bogoliubov operators<sup>7</sup> defined by

$$\gamma_{\vec{q}\,0} = u_{\vec{q}} c_{\vec{q},} - v_{\vec{q}} c_{\vec{q},}^{+} ,$$

$$\gamma_{\vec{q}\,1} = u_{\vec{q}} c_{-\vec{q},} + v_{\vec{q}} c_{\vec{q},}^{+} ,$$
(6)

with  $v_{\mathbf{d}}^2 = \frac{1}{2} (1 - \epsilon_{\mathbf{d}} / E_{\mathbf{d}})$ , and  $u_{\mathbf{d}}^2 = 1 - v_{\mathbf{d}}^2$ . Then the time dependence of the superconducting quasiparticle distribution function  $f_1(\vec{k})$  for state  $(\vec{k}, 1)$  is

$$\frac{\partial f_1(\vec{\mathbf{k}})}{\partial t} = -\frac{2\pi}{\hbar} \sum_{\mathbf{k}'} |v(\vec{\mathbf{k}} - \vec{\mathbf{k}}')|^2 \delta (E_{\vec{\mathbf{k}}'} - E_{\vec{\mathbf{k}}}) \\ \times \{f_1(\vec{\mathbf{k}})[1 - f_0(\vec{\mathbf{k}}')] - f_0(\vec{\mathbf{k}}')[1 - f_1(\vec{\mathbf{k}})]\} \\ = -\frac{\pi}{\hbar} N(0) \int |v(\vec{\mathbf{k}} - \vec{\mathbf{k}}')|^2 [f_1(\vec{\mathbf{k}}) - f_0(\vec{\mathbf{k}}')] \\ \times \delta(\epsilon_{\vec{\mathbf{k}}'} - \epsilon_{\vec{\mathbf{k}}}) \left| \frac{\partial \epsilon_{\vec{\mathbf{k}}'}}{\partial E_{\vec{\mathbf{k}}'}} \right| d\Omega' d\epsilon' .$$

Noting the factor  $\partial \epsilon_{\vec{k}'} / \partial E_{\vec{k}'} = E_{\vec{k}'} / \epsilon_{\vec{k}'}$  within the integral, it is straightforward to show

$$\frac{\partial f_1(\vec{k})}{\partial t} = -\frac{f_1(\vec{k}) - f_0(\vec{k})}{(|\epsilon_{\vec{k}}|/E_{\vec{k}})U} \quad . \tag{7}$$

Thus, we have shown that  $U_{\vec{k}} = (|\epsilon_{\vec{k}}|/E_{\vec{k}})U$ .

We now consider the field equations. We shall consider a supercurrent as a real current, and hence the only magnetization  $\overline{\mathbf{M}}$  in the superconducting material is from the paramagnetism of the spins. For case (a) we must find the transmitted fields. For that we assume a uniform rf magnetization  $\vec{\mathbf{M}}$  within the film such that  $M \ll |H_1|$ , where  $M = \vec{\mathbf{M}} \cdot \hat{\mathbf{y}}$ . As in Ref. 4, we split the rf fields into  $E^{(1)}$  and  $H^{(1)}$ , that directly associated with the magnetization, and the remainder which is due to the external fields and the electron currents in the sample. It can be shown<sup>4</sup> that  $H^{(1)} = -4\pi M$  and  $E^{(1)} = 0$ . Thus we can set

$$\begin{array}{l} H_1(x) = H_3(e^{ikx} - R'e^{-ikx}) \\ E_1(x) = H_3(e^{ikx} + R'e^{-ikx}) \end{array} \right\} \quad x \le -\frac{1}{2}L \ , \tag{8}$$

$$\begin{array}{c} H_{1}(x) = H_{4}(e^{-qx} - Re^{qx}) - 4\pi M \\ E_{1}(x) = -(i\omega/cq)H_{4}(e^{-qx} + Re^{qx}) \end{array} \right\} \quad -\frac{1}{2}L \leq x \leq \frac{1}{2}L,$$

$$(9)$$

$$\begin{array}{c} H_{1}(x) = H_{5} e^{ikx} \\ E_{1}(x) = H_{5} e^{ikx} \end{array} \right) \quad \frac{1}{2} L \leq x, \tag{10}$$

where

$$q = \left[ \frac{1}{\lambda^2 - 2i} \left( \frac{T}{T_c} \right)^4 \left( \frac{1}{\delta^2} \right) \right]^{\frac{1}{2}}$$
(11)

according to the London equations and the two-fluid model, and the temperature dependence of the penetration depth is assumed to be

$$\lambda = \frac{\lambda_0}{\left[1 - (T/T_c)^4\right]^{1/2}} \quad . \tag{12}$$

Here  $\delta$  is the skin depth in the normal state, while  $H_3$  is the amplitude of the incoming rf field, and  $R', R, H_4$ , and  $H_5$  are determined by matching the boundary conditions at  $x = \pm \frac{1}{2}L$ .

From these equations, we find the transmitted field,  $H_1(\frac{1}{2}L)$ , is, to second order in M and  $(i\omega/cq)$ , for  $L \approx \lambda$ ,

$$H_{1}(\frac{1}{2}L) \approx \frac{i\omega}{cq} \left[ -\frac{H_{1}(-\frac{1}{2}L)}{\sinh qL} + \left(1 - \frac{\exp(-\frac{1}{2}qL)}{\cosh(\frac{1}{2}qL)}\right) 4\pi M \right] .$$
(13)

Defining

$$H_b = -\frac{(i\omega/cq)H_1(-\frac{1}{2}L)}{\sinh(qL)}$$

and

$$H_s = \frac{i\omega}{cq} \left(1 - \frac{\exp(-\frac{1}{2}Lq)}{\cosh(\frac{1}{2}qL)}\right) 4\pi M,$$

the signal-to-background ratio is  $H_s/H_b$ .

We now set  $Z = Z_0 + Z_M$ , where  $Z_0$  is the surface impedance for spinless electrons and  $Z_M$  is the magnetization-dependent part. From Eq. (9) we get for case (a)

$$Z_{0}^{(a)} = -\frac{4\pi}{c^{2}} \left(\frac{\omega}{q}\right) i \left[1 + \frac{i\omega}{cq} \left(\frac{1}{\cosh qL} + \frac{1}{\sinh qL}\right)\right]$$
  
×  $\operatorname{coth} qL$  (14)

and

$$Z_{M}^{(a)} = \frac{4\pi M H_{1}^{*}(-\frac{1}{2}L)}{|H_{1}(-\frac{1}{2}L)|^{2}} \left[2 \sinh(\frac{1}{2}qL) - \tanh qL\right] \\ \times \exp(-\frac{1}{2}qL)Z_{0}^{(a)} .$$
(15)

Similarly,  $Z_0$  and  $Z_M$  for case (b) are

$$Z_0^{(b)} = - \left(4\pi/c^2\right) \left(i\omega/q\right) \tanh(\frac{1}{2}qL)$$
(16)

and

$$Z_{M}^{(b)} = \left[ 4\pi M H_{1}^{*} \left( -\frac{1}{2}L \right) / \left| H_{1} \left( -\frac{1}{2}L \right) \right|^{2} \right] Z_{0}^{(b)} \quad .$$
 (17)

Note that Eqs. (14)-(17) are the same as for the normal case except for the fact that q is different for the superconducting case as given by Eq. (11).

Our plan is first to find the behavior of a single quasiparticle, and then to add up the effects from all the quasiparticles. Suppose that at time t = t' the system is in the state  $\gamma_{\mathbf{k}0}^* | 0 \rangle$ , which is the state with one quasiparticle in  $(\mathbf{k}, 0)$ . We wish to calculate the transition probabilities to other states  $\gamma_{\mathbf{k}'0}^* | 0 \rangle$  and  $\gamma_{\mathbf{k}'1}^* | 0 \rangle$  due to the rf field.

The perturbing Hamiltonian  $\hat{\mathcal{K}}_1$  is

$$\widehat{\mathscr{H}}_1 = \sum_i \left[ \mu \vec{\sigma}_i \cdot \vec{\mathbf{H}}_1(x_i) e^{-i \,\omega t} + \mathrm{c. c.} \right] ,$$

where  $\vec{H}_1(x) = H_1(x)\hat{y}$ , or in the second quantization notation

$$\mathcal{W}_{1} = \sum_{k\sigma} \sum_{k'\sigma'} (\psi_{\vec{k}'\sigma'}, \hat{\mathcal{W}}_{1}\psi_{\vec{k}\sigma}) c^{\dagger}_{\vec{k}'\sigma'} c_{\vec{k}\sigma} ,$$

where  $\sigma$  refers to spin  $\dagger$  or  $\dagger$  with respect to the z axis. We get for our configurations

$$\mathcal{H}_{1} = \mu \sum_{\vec{k}} (e^{-i\omega t} \vec{\mathbf{H}}_{1} \cdot \vec{\mathbf{s}} * \vec{c}_{\vec{k}} \cdot \vec{c}_{\vec{k}}, + e^{-i\omega t} \vec{\mathbf{H}}_{1} \cdot \vec{\mathbf{s}} \cdot \vec{c}_{\vec{k}} \cdot \vec{c}_{\vec{k}}) + \mathbf{c. c.}$$

where  $\vec{s} = (\psi_{\vec{k}}, \vec{\sigma} \psi_{\vec{k}})$ , independent of k.

By using the Bogoliubov transformation, the matrix elements are found to be

$$\langle \mathbf{0} | \gamma_{-\vec{\mathbf{k}}'} \mathcal{H}_{1} \mathcal{H}_{1} \gamma_{\vec{\mathbf{k}}0}^{\dagger} | \mathbf{0} \rangle = \mu (e^{-i\omega t} \mathbf{\vec{s}}^{*} \cdot \mathbf{\vec{H}}_{1} + e^{i\omega t} \mathbf{\vec{s}}^{*} \cdot \mathbf{\vec{H}}_{1}^{*}) \delta_{\vec{\mathbf{k}}',\vec{\mathbf{k}}}$$

$$= \langle 0 | \gamma_{\vec{k}0} \mathcal{H}_1 \gamma_{-\vec{k}'1}^* | 0 \rangle^*,$$

while

$$\langle 0 | \gamma_{\vec{k}'0} \mathcal{H}_1 \gamma_{\vec{k}0}^* | 0 \rangle = \langle 0 | \gamma_{\vec{k}'1} \mathcal{H}_1 \gamma_{\vec{k}1}^* | 0 \rangle = 0$$

for  $\mathbf{k}' \neq \mathbf{k}$ . Thus, we see that  $\mathcal{R}_1$  induces transitions only between  $(-\mathbf{k}, 1)$  and  $(\mathbf{k}, 0)$ .

Let  $|\vec{k}, 0; t, t'\rangle$  be the time-dependent state vector of the system at time t, knowing that at time  $t' \leq t$  there was only one quasiparticle in the system and that it was in state  $(\vec{k}, 0)$ . Assuming that the quasiparticle is localized in a wave packet so that  $H_0(x)$  and  $H_1(x)$  are essentially constant over the wave packet, the time-dependent state vector can be written

$$\begin{aligned} |\vec{\mathbf{k}},0;t,t'\rangle &= \exp\left(-i\frac{\mu}{\hbar}\int^{t}\vec{\sigma}\cdot\vec{\mathbf{H}}_{0}(\vec{x}) dt''\right)\gamma_{\mathbf{k}0}^{*}|0\rangle \\ &+a_{1}(t)\exp\left(-i\frac{\mu}{\hbar}\int^{t}\vec{\sigma}\cdot\vec{\mathbf{H}}_{0}(\vec{x}) dt''\right)\gamma_{\mathbf{k}1}^{*}|0\rangle , \end{aligned}$$
(18)

where  $\overline{x}$  is the x coordinate of the center of the quasiparticle wave packet.  $\overline{x}$  depends on time and traces out the motion of the quasiparticle. The coefficient  $a_1(t)$  is determined by time-dependent perturbation theory.

In order to obtain the spin expectation value, we follow the same development as in Ref. 3. The Fermi function for state  $(\vec{k}, i)$  in the presence of a magnetic field  $H_0$  is (as can be shown by minimizing the free energy of the BCS superconductor in the presence of  $H_0$ )

$$f_{\vec{k}i} = \left\{ \exp[(E_{\vec{k}} \pm \mu H_0)/k_B T] + 1 \right\}^{-1}, \qquad (19)$$

where the sign in the exponent is plus for i = 0 and is minus for i = 1. After a spin-dependent collision, it is assumed that the quasiparticle will be in equilibrium in terms of its spin. That is, it will be in state *i* with probability  $P_{ki}$  with

$$P_{\mathbf{k}0}(x) - P_{-\mathbf{k}1}(x) = \frac{f_{\mathbf{k}0} - f_{-\mathbf{k}1}}{f_{\mathbf{k}0} + f_{-\mathbf{k}1}} \approx \frac{\mu H_0(x)}{f_{\mathbf{k}}} \frac{\partial f_{\mathbf{k}}}{\partial E_{\mathbf{k}}}, \quad (20)$$

where  $f_{\vec{k}}$  is the Fermi function for state  $(\vec{k}, i)$  in the absence of a magnetic field.

Using the arguments above, we obtain for the rf part cf the complex spin expectation value

$$\overline{\sigma}\left(\overline{\mathbf{k}},x\right) = \frac{i\mu^{2}\overline{\mathbf{s}}^{*}}{\hbar U_{\mathbf{k}}f_{\mathbf{k}}} \frac{\partial f_{\mathbf{k}}}{\partial E_{\mathbf{k}}} \int_{-\infty}^{t} du \int_{L/2}^{L/2} dx'$$

$$\times \overline{\mathbf{s}} \cdot \overline{\mathbf{H}}_{1}(x')G(x',u,x,t) A(x',u,t)$$

$$\times \langle \exp\left\{i \int_{u}^{t} dt' \alpha[\overline{x}(t')]\right\} \rangle_{\mathbf{av}}, \qquad (21)$$

where

$$A(x', u, t) = \int_{-L/2}^{L/2} dx'' \int_{-\infty}^{u} du' G(x'', u', x', u)$$
$$\times H_0(x'') \exp[-(t - u')/U_{\vec{k}}] ,$$

 $\alpha(x) = \omega - 2\mu H_{a}(x)/\hbar$ 

(23)

2558 and

$$G(x', t', x, t) = \frac{1}{L} \sum_{n=-\infty}^{+\infty} \cos[\mu_n(x + \frac{1}{2}L)] \cos[\mu_n(x' + \frac{1}{2}L)]$$

$$\times \exp\left[-\frac{1}{3}v_{\vec{k}}\,\Lambda\mu_n(t-t')\right]\,,\tag{22}$$

with  $\mu_n = n\pi/L$  and  $\Lambda$  the mean free path for non-spinflip scattering. The factor  $\int_{u}^{t} dt' \alpha[\overline{x}(t')]$  in the exponent of Eq. (21) is the spin phase of the quasiparticle. This phase depends on the path of the individual quasiparticle due to the fact that  $H_0$  is a function of x, unlike the normal conducting case.  $\langle \; \rangle_{av}$  in Eq. (21) means an average over all possible paths from (x', u) to (x, t). Different quasiparticles with different paths will have different phases, and this will tend to make the magnitude of  $\overline{\sigma}(k, x)$  smaller. In fact, if there is much variation of  $H_0$  over the sample the phases would be random and  $\overline{\sigma}$  would vanish. To see this, note that  $\overline{x}(u) = x'$  and  $\overline{x}(t) = x$ , and define  $\alpha_{av}(x')$  to be the average  $\alpha$  of the quasiparticles which move from x' at time u to x at time t. Then

$$\langle \exp \{ i \int_{u}^{t} dt' \alpha[\overline{x}(t')] \} \rangle_{av} = D \exp[i\alpha_{av}(x')(t-u)],$$

where

$$D = \langle \exp\left\{i \int_{u}^{t} dt' [\alpha(\bar{x}(t')) - \alpha_{av}(x')]\right\} \rangle_{av} \quad (24)$$

Since

$$\left| \alpha[\overline{x}(t')] - \alpha_{av}(x') \right| \leq 2(\mu/\hbar) \Delta H_0,$$

where  $\Delta H_0$  is the maximum variation of  $H_0$  in the sample, we see that  $D \ge 1 - \frac{1}{2} (\mu \Delta H_0 U_k^*/\hbar)^2$  if  $(\mu \Delta H_0 U_k^*/\hbar) \ll 1$ , and  $D \approx 0$  if  $(\mu \Delta H_0 U_k^*/\hbar) \ge 1$ . Hence, we must restrict ourselves to a thin film such that  $L \le \lambda$ . For such a film it is enough to take only the n = 0 term in G(x', t', x, t) defined by Eq. (22). Also

$$\alpha_{av}(x) = \alpha_{av} = \omega - (2\mu/\hbar)H_{0av} , \qquad (25)$$

where  $H_{0av}$  is the spatial average of  $H_0(x)$  and is equal to

$$H_{0 \text{ av}} = (2\lambda/L)H_0 \tanh(L/2\lambda) . \qquad (26)$$

Substituting Eq. (23) into (21) we have

$$\overline{\sigma}(k) = \frac{i\mu^2}{\hbar} \, \overline{s} * \frac{1}{f} \, \frac{\partial f}{\partial E} \, \overline{s} \cdot \overline{H}_{1 \text{ av}} \, H_{0 \text{ av}} \, \frac{DU_{\vec{k}}}{1 - i\alpha_{\text{ av}} \, U_{\vec{k}}} \, , \quad (27)$$

where  $H_{1 \text{ av}}$  is the spatial average of  $H_1(x)$ .

Now the magnetization is given by

$$\vec{\mathbf{M}} = -\mu N(0) \int_{-\infty}^{\infty} f_{\vec{\mathbf{k}}} \, \overline{\sigma}(\vec{\mathbf{k}}) \, d\epsilon \,, \tag{28}$$

where N(0) is the normal density of states at the Fermi surface for both spins. Notice that since the right-hand side of Eq. (27) is independent of x, the spin magnetization is independent of x, even for

case (a) where the rf field has a considerable change through the film. The absorption  $P_M$  due to the magnetization for case (b) (rf field equal on both sides of the film) is obtained from Eqs. (2), (16), (17), and (28):

$$P_{M} = \frac{8\omega}{L} \left\| \vec{\mathbf{s}} \cdot \vec{\mathbf{H}}_{1}(-\frac{1}{2}L) \right\|^{2} \operatorname{Re}\left(\frac{\tanh^{2}(\frac{1}{2}qL)}{q^{2}} F(T)\right),$$
(29a)

where F(T) is defined as

$$F(T) = \frac{\mu^{3} N(0) H_{0 \text{ av}}}{\hbar} \frac{U}{k_{B} T} \int_{0}^{\infty} \frac{d\epsilon \, \epsilon \exp(E/k_{B} T)}{E[\exp(E/k_{B} T) + 1]^{2}} \\ \times \frac{[1 + i\alpha_{\text{av}} U_{\vec{k}}]}{[1 + (\alpha_{\text{av}} U_{\vec{k}})^{2}]}, \qquad (29b)$$

and where D has been set equal to one. The signalto-background ratio of the transmitted field for case (a) (rf field on one side) is obtained from Eqs. (13) and (28):

$$H_{s}/H_{b} = -16\pi(i/q)\tanh(\frac{1}{2}qL) \sinh^{2}(\frac{1}{2}qL) F(T) .$$
(30)

It should be noted that in the limit  $T - T_c$ , Eqs. (29) and (30) give correct expressions for  $P_M$  and  $H_s/H_b$  in the normal state.

#### III. NUMERICAL RESULTS AND DISCUSSION

Equations (29) and (30) have been evaluated numerically. The absorption curve for case (b) as plotted in Fig. 1 is essentially Lorentzian with asymmetry due to the factor  $H_{0 \text{ av}}$  in Eq. (29b). In the normal-state CESR, this factor does not affect the shape very much since the variation of  $H_0$  over the linewidth is small compared to the magnitude of  $H_0$  at resonance. However, in the superconducting state, the maximum  $H_0$  is restricted by the superconducting critical field  $H_c$  and the peak spreads over the whole region of  $H_0$ . Figure 2 shows peak



FIG. 1. Shape of an absorption curve for case (b), the case for which the rf field is equal on both sides of the film. It is essentially Lorentzian except for the extra factor of  $H_{0 \text{ av}}$  which causes asymmetry.  $H_c$  for Al with these parameters is 247 G.



FIG. 2. Maximum absorption for case (b) divided by  $|H_1(-\frac{1}{2}L)|^2$  as a function of temperature for various frequencies. Note that  $H_1(-\frac{1}{2}L)$  is given by Eq. (8) and is not  $H_3$ , the amplitude of rf field in the absence of the sample. If the quantity on the ordinate is divided by  $c/2\pi$ , it gives a rough estimate of absorptivity. For Al only the part below the dashed line is experimentally accessible, since the maximum static field is restricted below  $H_c$ .  $H_c(T=0)=99$  G for bulk material. The dashed curve takes into account the fact that  $H_c$  is a function of T and also the fact that  $H_c$  is higher for a thin film. It is assumed  $N(0) = 10^{34} (\text{cm}^3 \text{ erg})^{-1}$ .

heights of absorption curves as a function of temperature for various frequencies for the convenience of determining the most favorable experimental conditions. The dashed curve represents conditions under which the peak appears around  $\frac{1}{2}H_c$  for aluminum, taking into account the fact that  $H_c$  depends on both the temperature and the thickness of the sample. We are not taking into account the fact that the density of states in superconductors depends on the static magnetic field. The dependence is rather small in clean superconductors<sup>8</sup> unless  $H \approx H_c$ , where our calculation probably fails to give correct results. Thus, the dashed curve in Fig. 2 gives the highest temperature to which one can go for a given frequency.

The temperature dependence is caused mainly by the change of the number of quasiparticles with temperature. Very close to  $T_c$ , the normal current contribution becomes important and changes q as is seen from Eqs. (11) and (12), and hence gives an additional temperature dependence. However, for  $T/T_c < 0.96$ , the normal current contribution to the determination of  $H_1(-\frac{1}{2}L)$  is negligible for the frequencies and normal-state skin depths we are considering. In fact, there is only little change in power absorbed when we change the normal skin depth from 1000 to 5000 Å for  $T/T_c = 0.9$  and  $\omega = 10^9 \text{ sec}^{-1}$ . Thus, there is no point in considering a more sophisticated approach than the two-fluid model for the penetration of the rf fields, Eqs. (8)-(11).

For case (a) (rf field on one side), the real and imaginary parts of  $H_s/H_b$  (the parts in phase and out of phase, respectively) are plotted in Fig. 3. The temperature dependence of peak heights for this case is about the same as the temperature dependence of power absorbed for case (b). Unfortunately, even under ideal conditions, the peak of  $H_s/H_b$  is of the order 10<sup>-7</sup>, which indicates that it would be very difficult to observe the peak experimentally by the techniques now used. However, it should be noted that the x component of the magnetization M is a background free signal, and therefore is perhaps easier to observe.

We have used the local London equations rather than the more realistic nonlocal expressions for the penetration of the fields into the superconductor in Eqs. (8)-(17). However, by choosing  $\lambda$  properly, we can get a fairly good description of the penetrating fields. Our primary concern in this paper is the spin magnetization, and since this does not depend crucially on the exact details of the position dependence of the fields, this treatment should be good enough for our purposes.

We have also calculated  $\vec{M}$  by using Kaplan's method of modified Bloch equations to compare it with our results. We found the two methods yield



FIG. 3. Real and imaginary parts of  $H_s/H_b$  as a function of  $H_0$  with  $N(0) = 10^{34} \text{ (cm}^3 \text{ erg})^{-1}$  for case (a) (rf field on one side).  $H_c$  for Al with these parameters is 165 G.

similar results for line shape and power absorbed. We also have found that when the rf frequency is swept by keeping  $H_0$  fixed, the shift of the peak position for thicker samples is to lower frequencies, for both calculations. This is contrary to the graphs given in Ref. 2.

In conclusion, CESR in superconductors is at least theoretically possible and would be experi-

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mentally easier the higher the critical field since the power absorbed  $P_M \propto H_0^2$  and also since the signal-to-background ratio  $H_s / H_b \propto H_0$ .

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### PHYSICAL REVIEW B

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# Breakdown of the Mean Field Theory in the Superconducting Transition Region

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We consider a dirty superconducting material above its transition temperature. Using a perturbation expansion, we study the effect of the fluctuations of the order parameter on the electron Green's function G as well as on the fluctuation Green's function  $\mathfrak{D}$ . Our main results are : (a) G and  $\mathfrak{D}$  can be determined separately. (b) The behavior of  $\mathfrak{D}$  can be analyzed using the Ginzburg-Landau functional where the only nonlinear term present is the lowest-order one. (c) As a result of this analysis, the corrections to the mean field theory are generally small as far as  $\mathfrak{D}$  is concerned. They become important only at the onset of the critical region, where our perturbation approach breaks down. We derive a criterion for the onset of this critical region and compare our estimate to previous ones. (d) On the other hand, G is well described by taking only the lowest-order correction to the electron self-energy due to the fluctuations, even inside the critical region. (e) We propose a scheme for the analysis of the critical region. (f) We establish in this way the complete equivalence between the critical behavior of the interacting Bose system and that of a superconductor.

### I. INTRODUCTION

The fluctuations of the order parameter in "dirty" one- or two-dimensional superconductors (thin films or whiskers) have been studied extensively both theoretically and experimentally. <sup>1-15</sup> Many experiments on the electrical conductivity<sup>2, 3</sup> seem to be successfully accounted for in terms of the Aslamazov-Larkin theory, <sup>9</sup> which is a mean field approach, although there exists still a question about the completeness of that derivation. <sup>11,13</sup>

On the other hand, there has been a number of suggestions<sup>16-23</sup> that the mean field theory will be no longer valid in the immediate vicinity of the transition temperature, where the fluctuation spec-

trum of the order parameter may be modified drastically.

In the present work, we use a perturbation expansion technique to study systematically the effect of the fluctuations of the order parameter on the electron Green's function G as well as on the fluctuation Green's function  $\mathfrak{D}$ . As a preparation of a more complete analysis, we start with a study of the first-order correction to G (Sec. II) and to  $\mathfrak{D}$  (Sec. III). We find that the effect of the first-order correction is much more important on  $\mathfrak{D}$  than on G. A detailed analysis of the higher-order corrections are carried out in Sec. IV, which results in the following conclusions.

(a) G and  $\mathfrak{D}$  can be determined separately.

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