Magnetic pair breaking in disordered superconducting films

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I. INTRODUCTION

The physics of the observed $T_c$ suppression in superconductors that contain nonmagnetic disorder has been the subject of much debate in recent years. Let us focus on homogeneously disordered thin superconducting films, with the disorder parametrized by the normal-state sheet resistance $R_\Omega$, and on the BCS-like quasi transition that is well pronounced in these films although the true superconducting transition is of Kosterlitz-Thouless nature. In these systems, the BCS transition temperature $T_c$, defined as the mid-point of the resistive transition, is observed to decrease monotonically with increasing disorder. A complete quantitative understanding of this effect within a microscopic strong-coupling theory has proven difficult, although the first perturbative calculations within a phenomenological BCS model were rather promising. Qualitatively, disorder-induced changes in the electron-phonon coupling in the Coulomb repulsion between the constituents of the Cooper pairs, and in the normal-state density of states, have all been identified to be important. The difficulty lies in the fact that some of these effects tend to suppress $T_c$ while others tend to enhance it, and $T_c$ depends exponentially on all of them so that subtle balancing effects result. Also, the number of parameters that acquire a disorder dependence is quite large, and fits of theoretical results to experimental data are therefore not necessarily very conclusive. Indeed, theories that are structurally quite different, and mutually inconsistent, have been shown to fit the same $T_c$ data equally well.

In this situation it is obvious that one should study the disorder dependence of quantities other than the transition temperature in an attempt to discriminate between various theories, and to obtain independent information about the disorder dependence of the various parameters that determine $T_c$. One possibility is to measure the inelastic lifetime of quasiparticles. The experiment by Pyun and Lemberger on amorphous InO has been analyzed by the present authors in the framework of a strong-coupling theory for disordered superconductors, and quantitative agreement between theory and experiment has been achieved. Another possibility is to study the influence of nonmagnetic disorder on the pair-breaking rate. An attempt to understand this behavior by a phenomenological modification of the Abrikosov-Gorkov result for $\alpha$, using the renormalization of the density of states inherent in Refs. 6,12 and 8, failed. This poses the important question whether the success of these theories in describing the $T_c$ suppression and the disorder...
and temperature dependence of the inelastic lifetime was fortuitous, and whether they are lacking some important physical ingredient that manifests itself in the pairbreaking rate.

It is the purpose of the present paper to analyze these questions. What we will find is that one needs to take the strong-coupling corrections to the Abrikosov-Gorkov expression into account before one generalizes to the disordered case in order to obtain the correct structure of the theory. Once this is done, we find that our previous theory accounts very well for the observed effect.

II. FORMALISM

Our starting point is our theory for the suppression of the superconducting $T_c$ and the enhancement of the inelastic scattering rate by nonmagnetic disorder. In this section we recall the most salient features of this theory. First one uses an exact eigenstate formalism to derive generalized Eliashberg equations for the normal Green function $G(\epsilon, \omega)$, and the anomalous Green function $F(\epsilon, \omega)$. Since the wavenumber is not a good quantum number in the presence of static impurities, $G$ and $F$ are functions of energy and frequency, rather than wavevector and frequency as in Eliashberg theory. The Green functions are expressed, as usual, in terms of an anomalous self-energy, $W(\epsilon, i\omega)$, and a normal one. The latter is split into a piece $i\omega Z(\epsilon, i\omega)$ that is an odd function of frequency, and a piece $Y(\epsilon, i\omega)$ that is even in $\omega$. In terms of these quantities, $G$ and $F$ read,

$$G(\epsilon, i\omega) = \frac{i\omega Z(\epsilon, i\omega) + \epsilon + Y(\epsilon, i\omega)}{|i\omega Z(\epsilon, i\omega)|^2 - |\epsilon + Y(\epsilon, i\omega)|^2},$$

and

$$F(\epsilon, i\omega) = \frac{-W(\epsilon, i\omega)}{|i\omega Z(\epsilon, i\omega)|^2 - |\epsilon + Y(\epsilon, i\omega)|^2}. \tag{1b}$$

In clean systems, the normal self-energy piece $Y$ is a constant that just shifts the chemical potential and can be omitted. In the presence of disorder, however, $Y$ has been found to be of crucial importance, and to reflect the physics of the Coulomb anomaly in the density of states in the context of superconductivity. The generalized Eliashberg equations then take the form of integral equations in both energy and frequency for the three functions $Z$, $W$, and $Y$. A solution of these equations has been obtained by means of various approximations. In particular, the frequency dependence of $Y$ was found to be weak and could be omitted, and its energy dependence was approximated by the first term in a Taylor expansion about a characteristic energy $\bar{\omega}$,

$$Y(\epsilon, i\omega) \simeq (\epsilon - \bar{\omega})Y', \tag{2}$$

with $Y' = \partial Y/\partial \epsilon|_{\epsilon = \bar{\omega}}$, and $\bar{\omega}$ an average phonon frequency. With some further approximations, the energy integrations could then be performed, and the theory be cast in the same form as Eliashberg theory. A two-square well approximation then leads to a $T_c$ formula that has the structure of a generalized McMillan or Allen-Dynes formula, viz.

$$T_c = \frac{\omega_{\text{log}}}{1.2} \exp \left[ -1.04(1 + \bar{\lambda} + Y') \frac{\ln(1 + 0.62\bar{\lambda}/(1 + Y'))}{\lambda - \bar{\mu}^*} \right]. \tag{3}$$

Here $Y'$ is the normal self-energy piece mentioned above, and $\bar{\lambda}$ and $\bar{\mu}^*$ are disorder dependent generalizations of the electron-phonon coupling constant $\lambda$ and the Coulomb pseudopotential $\mu^*$, respectively, in Eliashberg theory. Explicit expressions for all three of these quantities have been given in Ref. and will be evaluated for the case of thin films below.

In the presence of magnetic impurities, $T_c$ is reduced by pair breaking. Abrikosov-Gorkov theory has been modified to allow for strong-coupling effects with the only resulting change being a factor of $1/Z$ in the pair breaking parameter. The result is

$$- \ln(T_c/T_{c0}) = \psi(\alpha/2\pi T_c + 1/2) - \psi(1/2), \tag{4}$$

with $T_{c0}$ the value of $T_c$ in the absence of the magnetic impurities, $\psi$ the di-gamma function, and $\alpha$ the pair breaking parameter $\alpha = (1/Z)(1/\tau_s)$, with $1/\tau_s$ the spin-flip scattering rate. In the case of clean superconductors, $Z = 1 + \lambda$. In the presence of nonmagnetic disorder, and with the same approximations that lead to the $T_c$ formula given by Eq. (3), it is straightforward to repeat the calculation of Allen and Mitrovic within the framework of the theory of Ref. The result is again Eq. (4), but with $\alpha$ replaced by a disorder dependent $\tilde{\alpha}$ which in turn is related to disorder dependent parameters,

$$\tilde{\alpha} = \frac{1/\tilde{\tau}_s}{1 + \lambda}. \tag{5}$$

Here $\tilde{\lambda}$ is the same quantity as in Eq. (3), and $1/\tilde{\tau}_s$ is the disorder dependent spin flip-scattering rate. Throughout this paper, we choose units such that $\hbar = k_B = 1$. In the next section, we derive an explicit form for $\tilde{\alpha}$ in a disordered thin superconducting film.

III. DISORDER DEPENDENCE OF $\alpha$

A. Electron-phonon coupling $\tilde{\lambda}$

The electron-phonon coupling strength $\tilde{\lambda}$ is defined as an integral over the Eliashberg function $\alpha^2 F$,

$$\tilde{\lambda} = 2 \int \frac{d\nu}{\nu} \alpha^2 F(\nu). \tag{6}$$

$\alpha^2 F$, and hence $\tilde{\lambda}$, are disorder dependent due to effects first discussed by Pippard in the context of ultrasonic
attenuation, and by Schmidt for the electron-phonon inelastic lifetime. The main physical point is that disorder decreases the coupling of the electrons to longitudinal phonons due to collision drag, but increases the coupling to transverse phonons due to the breakdown of momentum conservation. For realistic parameter values the latter effect is stronger than the former, leading to an overall increase of $\lambda$ with disorder. For Debye phonons in $3-d$ systems, $\lambda$ has been calculated in Ref. 12. Repeating that calculation in $d=2$ is straightforward, and very similar to the corresponding calculation of the electron-phonon inelastic lifetime. The result is

$$\lambda = 2 \int_0^{\omega_D} \frac{d\nu}{\nu} \sum_{\nu m} \frac{d_{\nu}}{c_b} f_b(\nu/c_b) ,$$

(7)

for a system with mean free path $l$. Here $c_{\nu L,T}$ are the longitudinal and transverse speeds of sound, respectively, $\omega_D$ is the Debye frequency, the dimensionless constant $d_\nu = k_F^2/16\pi\rho_ic_b$ with $\rho_i$ the ion density and $k_F$ the Fermi wave number, and the functions $f_{T,L}$ are given by

$$f_T(x) = \frac{8}{x^4} \left( 1 + \frac{x^2}{2} - \sqrt{1 + x^2} \right) ,$$

(8a)

$$f_L(x) = 2 \left( \frac{1}{\sqrt{1 + x^2}} - \frac{2}{x^2} \right) .$$

(8b)

Substituting Eqs. (8) into Eq. (7) we obtain the disorder dependence of $\lambda$,

$$\lambda = \frac{\lambda R_D E_F c_L}{\pi \omega_D v_F} \left[ F_L \left( \frac{\pi \omega_D v_F}{R_D E_F c_L} \right) + 2 \frac{c_T^2}{c_L^2} F_T \left( \frac{\pi \omega_D v_F}{R_D E_F c_T} \right) \right] ,$$

(9a)

where we have defined two functions,

$$F_L(x) = \sqrt{1 + x^2} - 1 - \ln(\sqrt{1 + x^2} + 1)/2 ,$$

$$F_T(x) = \frac{1 - \sqrt{1 + x^2}}{2(1 + \sqrt{1 + x^2})} + \ln((\sqrt{1 + x^2} + 1)/2) ,$$

(9b)

with $\lambda = 4\omega_D d_L/\pi mc_L^2$ the electron-phonon coupling in a clean $2-d$ system. The dimensionless resistance $R_D$ is $R_D c^2/h \approx R_D/4.1$ kΩ is a measure of the disorder in the material.

As in three dimensions, the size of the disorder renormalization of $\lambda$ depends on the ratio of the longitudinal to the transverse speed of sound. This is a result of the abovementioned competition between an increase in the coupling between electrons and transverse phonons and a decrease of the coupling to longitudinal phonons. Since the transverse speed of sound is invariably smaller than the longitudinal one, $\lambda$ increases with increasing disorder. This effect tends to reduce the pair breaking rate, Eq. (5). However, we also have to calculate the disorder dependence of the spin-flip scattering rate in order to obtain the disorder dependence of $\bar{\alpha}$.

**B. Spin-flip scattering rate $1/\tau_s$**

The interaction between the electron spin and an impurity spin $\vec{S}(\vec{r})$ at site $\vec{r}$ is described by a Hamiltonian,

$$H_S = \sum_{\vec{k},\vec{k}',\mu,\nu} J_{\vec{k},\vec{k}'} \vec{S}(\vec{k} - \vec{k}') \cdot (c_{\nu}^\dagger c_{\mu}^\dagger \vec{\sigma}) c_{\mu} c_{\nu} .$$

(10)

Here $c^\dagger$ and $c$ are fermion operators, $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ denotes the Pauli matrices, and $J_{\vec{k},\vec{k}'}$ denotes the electromagnetic impurity exchange integral. We now calculate the electron self energy contribution, $\Sigma$, due to this interaction in Born approximation. It is most convenient to do this in an exact eigenstate representation, in analogy to the calculation of the Coulomb self energy in Ref. 26. The calculation is straightforward, and we obtain

$$\Sigma(\epsilon, i\omega) = \int \frac{d\epsilon'}{N_F} G(\epsilon', i\omega) \sum_q V_S(q) F_s(q, \epsilon - \epsilon') .$$

(11)

Here, $N_F$ is the free electron density of states per spin at the Fermi level. We only retain the $s-$wave component of the interaction so that $V_S(q) = n_p S(S+1)J^2$, where $n_p$ is the concentration of paramagnetic impurities and $J$ is a measure of the exchange interaction strength.

In a clean system, the spin-density correlation function $F_s(q, \omega)$ is given by

$$\frac{1}{\tau_s} = \frac{1}{2} \text{Im} \Sigma(\omega, \omega + i0) .$$

(12)

In a disordered system, $F_s$ is diffusive and in the Green function $G$ we must keep the self energy piece $\Sigma'$ as discussed above. We thus obtain

$$\frac{1}{\tau_s} = \frac{2n_p S(S+1)J^2}{N_F [1 + Y']} \sum_q F_s \left( \frac{q \omega}{1 + Y'} \right) ,$$

(14a)

where
\[ F_s(\mathbf{q}, \omega) = g(q) \frac{Dq^2}{\omega^2 + (Dq^2)^2} \]  

(14b)

with \( g(q) \) the Lindhard function, which for simplicity we replace by \( N_F \Theta(2k_F - q) \). Here \( D \) denotes the normal phase spin density diffusion coefficient, which in the noninteracting electron approximation coincides with the mass or charge diffusion coefficient, so \( D = \pi/mR_\square \). Performing the wavenumber integral in Eq. (14a) we finally obtain

\[
\frac{1}{\tau_s} = \frac{1}{\tau_s} \left\{ 1 + \frac{1}{1 + Y} \frac{\tilde{R}_\square}{8\pi} \ln \left[ 1 + \left( \frac{8\pi \epsilon_F}{\tilde{R}_\square \omega^*} \right)^2 \right] \right\},
\]

(15a)

with

\[
\omega^* = \frac{\lambda}{1 + Y},
\]

(15b)

and \( 1/\tau_s \) given by Eq. (13). \( 1/\tilde{\tau}_s \) depends on disorder both explicitly, and implicitly through \( Y' \). Our final task is therefore to calculate the dependence of \( Y' \) on \( \tilde{R}_\square \).

C. Normal Self Energy Piece \( Y' \)

In order to calculate \( Y' \) we again have to repeat the calculations of Ref. 12 in \( d = 2 \). Both the electron-electron and the electron-phonon contributions to the self energy contribute to the self energy piece \( Y \). Performing a Taylor series expansion in energy around \( \epsilon = \omega \) of Eq. (2.12) of Ref. 12 we obtain

\[
Y'(\omega) = \delta U'_C(\omega) + 4 \int \frac{d\nu}{\nu} \delta \alpha^2 F^H(\omega, \nu).
\]

(16)

\( \delta U'_C(\omega) \), which describes the Coulomb contribution to \( Y' \), is taken from Ref. 12.

\[
\delta U'_C(\omega) = \frac{1}{\pi N_F} \sum_q g(q) \frac{Dq^2}{(Dq^2)^2 + \omega^2} \times
\]

\[
\left\{ V_C(\mathbf{q}) - \frac{2}{g(q)^2} \sum_{k,p} \sum_{k',p'} g_{k,k'}(\mathbf{q}) g_{p,p'}(\mathbf{q}) V_C(\mathbf{k} - \mathbf{p}) \right\},
\]

(17)

with the statically screened Coulomb potential,

\[
V_C(\mathbf{q}) = \frac{1}{2N_F} \frac{\kappa}{\kappa + q}, \quad \kappa = 4\pi e^2 N_F.
\]

(18)

Using the prescription for performing momentum sums of this type as described in Ref. 12, the integrals can be done and yield

\[
\delta U'_C(\omega) = \frac{\mu \tilde{R}_\square}{8\pi} \left[ G \left( \frac{\tilde{R}_\square \omega}{8\pi \epsilon_F} \frac{4k_F^2}{\kappa^2}, \frac{\tilde{R}_\square \omega}{8\pi \epsilon_F}, \frac{2k_F}{\kappa} \right) - \frac{2}{\pi} H \left( \frac{\tilde{R}_\square \omega}{8\pi \epsilon_F}, \frac{2k_F}{\kappa} \right) \right],
\]

(19)

with the functions

\[
G(x, y, z) = \frac{z}{1 + x^2} \ln \left( \frac{1 + \sqrt{y + y^2}}{1 + \sqrt{y + y^2}} \right)
\]

\[
+ \sqrt{2x(1 + x) \tan^{-1} \left( \frac{\sqrt{2y}}{y - 1} \right)} \tan^{-1}(1/y)
\]


\[
H(y, z) = \frac{z}{1 + y^2} \ln \left( \frac{1 + \sqrt{z^2 - 1}}{\sqrt{z^2 - 1}} \right).
\]

(20)

The Coulomb pseudopotential \( \mu \) in \( d = 2 \) is given by

\[
\mu = \frac{\kappa}{2\pi k_F} \ln \left( 1 + \frac{2k_F}{\kappa} \right).
\]

(21)

As discussed in Ref. 12, the phonon contribution to \( Y' \), which is given by the second term on the right-hand side of Eq. (16), is related to a stress-stress correlation function and can be calculated in a similar manner as \( \delta U'_C(\omega) \). One obtains

\[
4 \int \frac{d\nu}{\nu} \delta \alpha^2 F^H(\omega, \nu) = \frac{c_L E_F}{2\pi^2 v_F \omega_D} \sin^{-1} \left[ \frac{\omega_D v_F}{4E_F c_L} \right] \ln \left[ 1 + \left( \frac{8\pi E_F}{\tilde{R}_\square \omega^*} \right)^2 \right].
\]

(22)

Finally, both contributions can be collected to give

\[
Y'(\omega) = \tilde{R}_\square \left\{ \mu \left[ G - \frac{2}{\pi} H \right] + \frac{c_L E_F}{2\pi^2 v_F \omega_D} \sin^{-1} \left[ \frac{\omega_D v_F}{4E_F c_L} \right] \ln \left[ 1 + \left( \frac{8\pi E_F}{\tilde{R}_\square \omega^*} \right)^2 \right] \right\},
\]

(23)

where \( G \) and \( H \) have the same arguments as in Eq. (19). The enhancement of \( Y' \) with increasing disorder is due to the opening of a correlation gap in the (normal state) density of states, and contributes to the decrease of \( T_c \).

IV. FINAL RESULT AND DISCUSSION

We are now in position to collect our results and thus obtain the disorder dependence of the magnetic pair breaking rate. Substituting Eqs. (1), (15), and (23) into Eq. (8) yields our final result,

\[
\frac{\tilde{\alpha}}{\alpha} = \frac{1 + \lambda}{1 + \lambda} \left\{ 1 + \frac{1}{1 + Y/8\pi} \frac{\tilde{R}_\square}{\ln \left[ 1 + \left( \frac{8\pi E_F}{\tilde{R}_\square \omega^*} \right)^2 \right]} \right\}.
\]

(24)

The disorder renormalizations of the pair breaking rate appear both in the numerator and denominator and
therefore the rate may either increase or decrease with increasing disorder depending upon the material parameters $\lambda, c_L, c_T, v_F, E_F, \omega_D$, and $\mu$.

We now address the experiment on Pb$_{0.9}$Bi$_{0.1}$ by Chervenak and Valles [13]. To estimate the parameters entering into Eq. (24), let us first consider the parameter values for bulk PbBi, as far as available, given in Ref. 14. Thereby we have $\omega = 56K$, and $\omega_D = 108K$. The Bohn-Staver relation gives

$$\frac{\omega_D}{E_F c_L} = 2(2/Z)^{1/3}.$$  

(25)

For clean bulk Pb one has $E_F = 1.1 \times 10^5K$, $c_L = 2050$ m/s, $c_T = 710$ m/s, and $Z = 4$. However, we do not expect the actual parameter values to correspond to those for either bulk Pb or bulk Pb$_{0.9}$Bi$_{0.1}$. First of all, the material in question is a thin film, and moreover the substrate is expected to modify its properties, in particular the acoustic ones. Evidence for this is provided by the fact that the parameter values quoted above do not give the correct value of the clean limit $T_c$ as measured in Ref. 13. It is therefore likely that the substrate on which the thin layer of Pb is deposited strongly affects the phonon spectra of the film, altering both $\lambda$ and the ratio $c_L/c_T$ compared to bulk Pb. Accordingly, we choose a value for the bare $\lambda = 1.12$ and $\mu = 0.1$ which (in the absence of disorder) reproduces the highest $T_c$ as measured in Ref. 13. Lastly, since $c_L/c_T$ is not known even for Pb$_{0.9}$Bi$_{0.1}$, we let $c_L/c_T$ be determined by a fit to the data.

These parameters provide the curves shown in Figure 1. The solid curves are the results for the disorder dependence of the normalized pair breaking rate as given by Eq. (5). The points represent the data taken from Ref. 13 for two different runs. The decrease at small $R_D$ is due to the fact that for small disorder, $\lambda$ and $Y'$ grow more rapidly than $1/\tau_s$. The normalized rate goes through a shallow minimum at roughly $R_D \sim 0.3$ at which point the disorder renormalizations of $Y'$, $\lambda$, and $1/\tau_s$ are balanced and offset each other. With further increasing disorder the enhancement of $1/\tau_s$ dominates, and leads to a slowly increasing pair breaking rate.

We remark that the point at which the minimum occurs depends sensitively on the ratio of the longitudinal and transverse speeds of sound. To obtain the solid lines in Fig. 1a, $c_L/c_T = 1.9$ was used while 2.1 was used for Fig. 1b. These values lie between the value for bulk Pb (2.88) and the substrate (similar to pyrex, 1.72) used in Ref. 13 and thus does not seem unreasonable. Larger values of $c_L/c_T$ yield a more drastic reduction of the rate for small disorder and the region of increasing $\alpha$ occurs at larger values of $R_D$. This sensitivity of the overall shape of the curve to the material parameters may be reflected in the slightly different results obtained for the two experimental runs in Ref. 13 as shown in Fig. 1. It would therefore be very interesting to repeat the experiments using substrates with different acoustic properties.

In summary, we have presented a theory for the paramagnetic pair breaking rate in disordered superconducting films and have shown that the disorder dependence of the rate depends delicately on the disorder renormalizations of $Y'$, $\lambda$, and $1/\tau_s$. As a result, the rate can either increase or decrease with disorder, depending upon material parameters, and in general it is not a monotonic function of disorder. Our conclusion is that the disorder dependence of the rate as observed by Chervenak and Valles in Pb$_{0.9}$Bi$_{0.1}$ films can be quantitatively understood via an application of the microscopic theories developed in Refs. 1 and 3.

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**FIG. 1.** Plot of the pair breaking rate as a function of disorder for two different Pb$_{0.9}$Bi$_{0.1}$ films. The symbols are data from Ref. 29, the solid line is obtained from Eq. (5). For Fig. 1a(b) we have chosen $c_L/c_T = 1.9$ (2.1), and $\alpha = 1/\tau_s(1 + \lambda) = 90.5$K (70K). All other parameters are the same for both figures and are given in the text.