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## LETTER TO THE EDITOR

## Pair breaking and bound states in disordered superconductors

A K Chattopadhyay, R A Klemm and D Sa

Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Straße 38, D-01187 Dresden, Germany

E-mail: akc@mpipks-dresden.mpg.de, rklemm@mpipks-dresden.mpg.de and deba@mpipks-dresden.mpg.de

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### Abstract

We study the effects of inhomogeneous pairing interactions and impurities in short-coherence-length superconductors. Within the Born approximation, the effects of pairing disorder and magnetic impurities are identical. The  $T$ -matrices for pairing disorder sites with and without an impurity give rise to bound states within the BCS (Bardeen–Cooper–Schrieffer) gap, consistent with scanning tunnelling microscopy results on  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  with Zn or Ni impurities.

The effect of disorder in superconductors has long been a subject of considerable interest. A generally accepted physical picture is that magnetic impurities destroy superconductivity by locally breaking the pairs [1–4], whereas non-magnetic impurities are not pair breaking, according to Anderson's theorem [5]. This is true for an isotropic s-wave BCS superconductor, in which the order parameter is uniform and momentum independent. Since in most high-transition-temperature ( $T_c$ ) cuprates, the suppressions of  $T_c$  with Zn and Ni doping are comparable [6], there were proposals to explain this in terms of d-wave superconductivity [7]. However, nuclear magnetic resonance (NMR) experiments indicated that the nominally non-magnetic  $\text{Zn}^{2+}$  ions polarize the spin background in the  $\text{CuO}_2$  planes upon substitution of the  $S = 1/2$   $\text{Cu}^{2+}$  sites [6, 8]. Moreover, recent scanning tunnelling microscopy (STM) measurements directly above the Zn or Ni impurity sites observed strong resonance peaks [9, 10]. Very recently, several groups noticed from STM measurements that the non-stoichiometric underdoped  $\text{Bi}_2\text{Sr}_2\text{Cu}_2\text{O}_{8+\delta}$  (BSCCO) and  $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Cu}_2\text{O}_{8+\delta}$  are extremely disordered on a scale of a few nanometres [11–14]. This disorder is characterized by two gaps: one corresponding to the superconducting gap, with characteristic superconducting peaks, and a non-superconducting gap.

There is now a large body of evidence that the pseudogap observed in cuprates above  $T_c$  is not superconducting [12, 15–19]. In particular, the pseudogap regime is field independent

until one reaches the Zeeman field for breaking up chargeless spin-zero pairs [15]. This is precisely consistent with the pseudogap being particle–hole pairs, such as in a charge-density wave (CDW). Thus, if indeed the disorder involves a problem of percolation between superconducting and density-wave regions on the scale of the superconducting coherence length, then the phase coherence can only arise from Josephson coupling of the superconducting grains, which could exist without trapped flux on a nanoscale for an *s*-wave superconductor. In addition, one would expect the *c*-axis tunnelling to be very incoherent. Both of these properties were inferred in BSCCO from *c*-axis twist Josephson junction experiments [20]. In addition, features in the density of states (DOS) expected for a  $d_{x^2-y^2}$ -wave superconductor were not observed [21].

In this letter, we assume that the superconductor is electronically disordered on the scale of the coherence length. We further assume that the essential ingredient in the disorder is not one of impurities, but rather disorder in the pairing interaction itself. Thus, we expect  $T_c$  to vary from site to site, as does the resulting order parameter amplitude [22]. We treat this type of disorder using a Bogoliubov–de Gennes procedure, assuming that the order parameter amplitude varies locally [23–27].

Here we show that in the Born approximation, this problem maps exactly onto that of pair breaking in a superconductor, with all of the features of that model, including gapless superconductivity [1–4]. At a particular defect site, the  $T$ -matrix gives rise to bound states within the gap, even without magnetic impurities.

We use the Nambu representation:

$$\Psi^\dagger(\mathbf{r}) = (c_\uparrow^\dagger(\mathbf{r}), c_\downarrow^\dagger(\mathbf{r}), c_\uparrow(\mathbf{r}), c_\downarrow(\mathbf{r})), \quad (1)$$

where  $c_\downarrow(\mathbf{r})$  ( $c_\uparrow^\dagger(\mathbf{r})$ ) annihilates (creates) a quasiparticle with spin eigenstate  $\downarrow$  ( $\uparrow$ ) at the position  $\mathbf{r}$ . We set  $\hbar = c = k_B = 1$ . The Hamiltonian under study is  $H = H_0 + H_1 + H_2 + H_3$ , where

$$H_0 = \int d^3\mathbf{r} \Psi^\dagger(\mathbf{r}) [\hat{\xi}(\mathbf{r})\rho_3\sigma_0 + \Delta_0\rho_2\sigma_2] \Psi(\mathbf{r}), \quad (2)$$

$$H_i = \frac{1}{2} \int d^3\mathbf{r} \Psi^\dagger(\mathbf{r}) \hat{V}_i(\mathbf{r}) \Psi(\mathbf{r}), \quad (3)$$

where  $\hat{V}_1(\mathbf{r}) = U_1(\mathbf{r})\rho_3\sigma_0$ ,  $\hat{V}_2(\mathbf{r}) = U_2(\mathbf{r})\mathbf{S} \cdot \vec{\alpha}/[S(S+1)]^{1/2}$ ,  $\vec{\alpha} = \hat{x}\rho_3\sigma_1 + \hat{y}\rho_0\sigma_2 + \hat{z}\rho_3\sigma_3$ ,  $\hat{V}_3(\mathbf{r}) = U_3(\mathbf{r})\rho_2\sigma_2$ ,  $\rho_j\sigma_{j'} \equiv \rho_j \otimes \sigma_{j'}$  is a rank-4 tensor composed of two Pauli matrices for  $j, j' = 1, 2, 3$  and  $\rho_0, \sigma_0$  are rank-2 identity matrices, respectively.  $H_0$  is the Bogoliubov–de Gennes version of the BCS Hamiltonian, with momentum space quasiparticle energy dispersion  $\xi_k$  relative to the Fermi energy  $\mu$ ,  $\Delta_0(T)$  is the real bare uniform BCS order parameter, and  $H_1$  and  $H_2$  are the interactions due to scattering off random non-magnetic and magnetic impurities with effective potentials  $U_1(\mathbf{r})$  and  $U_2(\mathbf{r})/[S(S+1)]^{1/2}$ , respectively.  $H_3$  with effective potential  $U_3(\mathbf{r})$  is the usually neglected interaction arising from random variations in the pairing interaction [22, 24–27]. In  $H_2$ ,  $\mathbf{S}$  and  $S$  are the spin vector and quantum number of the magnetic impurities, respectively, and  $\vec{\alpha}$  represents the quasiparticle spin eigenvector. We assume the spatial average of each random potential satisfies  $\langle U_i(\mathbf{r}) \rangle = n_i U_i(0)$  for  $i = 1, 2, 3$ , where  $n_i$  is the density of defects of type  $i$ . In the absence of all defects, the order parameter  $\Delta_0(T) = V \langle c_\uparrow(\mathbf{r})c_\downarrow(\mathbf{r}) \rangle$  satisfies the standard BCS gap equation:

$$\Delta_0 = -VT \sum_{|\omega_n| \leq \omega_0} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \text{Tr}[\rho_2\sigma_2 \hat{G}_0(\mathbf{k}, \omega_n)], \quad (4)$$

$$\hat{G}_0^{-1}(\mathbf{k}, \omega_n) = i\omega_n\rho_0\sigma_0 - \xi_k\rho_3\sigma_0 - \Delta_0\rho_2\sigma_2,$$

where  $\hat{G}_0$  is the bare Green function matrix,  $V < 0$  is the uniform (BCS) part of the pairing interaction,  $N(0)$  is the single-spin quasiparticle DOS,  $\omega_0$  is a BCS-like cut-off, and the  $\omega_n$  are the Matsubara frequencies.

We assumed a real bare uniform order parameter  $\Delta_0$ , and restricted our consideration in  $H_3$  to spatial fluctuations of the amplitude of  $\Delta_0$ . The model can also treat spatial fluctuations of the phase of  $\Delta_0$  by letting  $\Delta_0\rho_1\sigma_2$  and  $U_3(\mathbf{r})\rho_2\sigma_2$  be generalized to  $\Delta_{01}\rho_1\sigma_2 + \Delta_{02}\rho_2\sigma_2$  and  $U_{31}(\mathbf{r})\rho_1\sigma_2 + U_{32}(\mathbf{r})\rho_2\sigma_2$ , respectively.

Our main interest lies in studying  $H_3$ . Using quantum Monte Carlo techniques to study a two-dimensional square lattice with an on-site attractive Hubbard pairing interaction in  $H_0$  [24], Ghosal *et al* [12] obtained interesting results in excellent qualitative agreement with those obtained from STM measurements. We also consider  $H_1$  and  $H_2$  for comparison, because the combination of one or both of them with  $H_3$  can lead to novel behaviour. In the Born approximation, these interactions add or subtract in a simple fashion. However, in the  $T$ -matrix approximation for a single defect site, they combine in a highly non-trivial manner.

In the self-consistent Born approximation, the quasiparticle self-energy  $\hat{\Sigma} = \hat{\Sigma}_1 + \hat{\Sigma}_2 + \hat{\Sigma}_3$ , where

$$\hat{\Sigma}_i(\mathbf{k}, \omega_n) = n_i \sum_{\mathbf{k}'} \hat{V}_i(\mathbf{k} - \mathbf{k}') \hat{G}(\mathbf{k}', \omega_n) \hat{V}_i(\mathbf{k}' - \mathbf{k}), \quad (5)$$

$$\hat{G}^{-1}(\mathbf{k}, \omega_n) = \hat{G}_0^{-1}(\mathbf{k}, \omega_n) - \hat{\Sigma}(\mathbf{k}, \omega_n), \quad (6)$$

$\hat{V}_i(\mathbf{k})$ ,  $U_i(\mathbf{k})$  are the spatial Fourier transforms of  $\hat{V}_i(\mathbf{r})$ ,  $U_i(\mathbf{r})$ , respectively. Neglecting any possible anisotropy arising from Fermi surface integrations, the effective rates of the three processes are  $1/\tau_i = 2\pi n_i N(0) |U_i(\mathbf{k}_F)|^2$ .

As in the usual pair-breaking theory [1–4],  $\hat{G}$  has the same form as  $\hat{G}_0$ , except that  $\omega_n$  and  $\Delta_0$  are replaced by their renormalized equivalents  $\tilde{\omega}_n$  and  $\tilde{\Delta}$ , respectively. We then obtain the *standard* equations for the renormalized gap and Matsubara frequency:

$$\tilde{\omega}_n = \omega_n + (1/\tau_1 + 1/\tau_{\text{pb}}) \frac{\tilde{\omega}_n}{2[\tilde{\omega}_n^2 + \tilde{\Delta}^2]^{1/2}}, \quad (7)$$

$$\tilde{\Delta} = \Delta_0 + (1/\tau_1 - 1/\tau_{\text{pb}}) \frac{\tilde{\Delta}}{2[\tilde{\omega}_n^2 + \tilde{\Delta}^2]^{1/2}}. \quad (8)$$

$$1/\tau_{\text{pb}} = 1/\tau_2 + 1/\tau_3 \quad (9)$$

is the total pair-breaking rate. The new physics arises from  $H_3$ . Evidently, within the self-consistent Born approximation, the effects of the random interactions are *exactly equivalent* to those of magnetic impurities.

Using standard pair-breaking theory [1–4], one finds

$$\frac{\omega_n}{\Delta_0} = u \left( 1 - \frac{\zeta}{\sqrt{1+u^2}} \right), \quad (10)$$

where  $u = \tilde{\omega}_n/\tilde{\Delta}$  and  $\zeta = 1/(\tau_{\text{pb}}\Delta_0)$ . The spatial average gap  $\Delta(T)$  is then

$$\Delta = \pi |V| N(0) T \sum_{|\omega_n| \leq \omega_0} \frac{1}{\sqrt{1+u^2}}, \quad (11)$$

leading to the standard equation for  $T_c/T_{c0} = t$ :

$$0 = \ln(t) + \psi \left( \frac{1}{2} + \frac{\alpha_{\text{pb}}}{2\pi t} \right) - \psi \left( \frac{1}{2} \right), \quad (12)$$

where  $\alpha_{\text{pb}} = 1/(\tau_{\text{pb}}T_{c0})$  and  $\psi(x)$  is the digamma function. For small  $\alpha_{\text{pb}}$ , as assumed in conventional superconductors [25, 26],  $T_c \approx T_{c0} - \pi/4\tau_{\text{pb}}$ . However, in extremely

disordered superconductors such as BSCCO,  $T_c/T_{c0}$  can be suppressed to zero even *without any magnetic impurities*, for  $1/\tau_3 \geq 1/\tau_{3c} = \pi T_{c0}/2\gamma$ , where  $\gamma = 1.781$  is the exponential of Euler's constant. In addition, the superconductivity becomes gapless for  $1 > \tau_{3c}/\tau_3 \geq 12 \exp(-\pi/4) \approx 0.912$ . Thus, even an isotropic, *s*-wave superconductor can become gapless, as observed in the cuprates with STM [9, 10, 13].

This can only occur in short-coherence-length superconductors with strong local inhomogeneities in the pairing interaction, as is a likely explanation for the vanishing of the  $T_c$  in the highly underdoped region of the cuprate phase diagram, although that region is also complicated by the simultaneous appearance of local CDW order at the non-superconducting regions not included in this calculation [12].

In order to make direct comparison with STM experiments, we solve the  $T$ -matrix for a single defect site. We assume that the site has all three types of defect associated with it. We approximate the effects of the magnetic impurity by assuming that its spin behaves classically [28]. Then, the  $T$ -matrix equation can be solved exactly:

$$\hat{T}(\omega_n) = \frac{\hat{V}(0)}{\hat{1} - \hat{g}_0(\omega_n)\hat{V}(0)}, \quad (13)$$

where  $\hat{V}(0) = \sum_{i=1}^3 \hat{V}_i(0)$  and  $\hat{g}_0(\omega_n) = -\pi N(0)[i\omega_n\rho_0\sigma_0 + \Delta_0\rho_2\sigma_2]/[\omega_n^2 + \Delta_0^2]^{1/2}$  is the bare Green function at the origin,  $\hat{1} = \rho_0\sigma_0$  is the rank-4 identity matrix, and the effects of a finite quasiparticle energy bandwidth have been neglected. Bound states within the gap at  $T = 0$  at the frequency  $\omega$  are obtained from

$$\det[\hat{1} - \hat{g}_0(i\omega)\hat{V}(0)] = 0. \quad (14)$$

Solving equation (14) exactly, we generally obtain four bound states within the gap at  $\omega = \bar{\omega}_b\Delta_0$ , where

$$\bar{\omega}_b = \pm[A + sR]^{1/2}/B, \quad (15)$$

$A = 16v_2^2v_3^2 + a_+^2(a_-^2 + 4v_1^2)$ ,  $B = a_+^2 + 4v_2^2$ ,  $C = a_+^2 - 4v_3^2$ ,  $R = [A^2 - B^2C^2]^{1/2}$ ,  $a_{\pm} = v_1^2 + v_3^2 - v_2^2 \pm 1$ ,  $v_i = \pi N(0)U_i(0)$ , and  $s = \pm 1$ .

We rewrite equation (15) for the special cases of two defects only. For non-magnetic defects,  $v_2 = 0$ , there are only two bound states symmetric about zero bias:

$$\bar{\omega}_b \rightarrow \pm \frac{[(1+v_3)^2 + v_1^2][(1-v_3)^2 + v_1^2]^{1/2}}{(1+v_1^2+v_3^2)}. \quad (16)$$

For the trivial case of a site with just a non-magnetic impurity, equation (16) shows that there are no poles inside the gap. However, for a pairing interaction defect alone, there are poles at  $\bar{\omega}_b = \pm|1 - v_3^2|/(1 + v_3^2)$ .

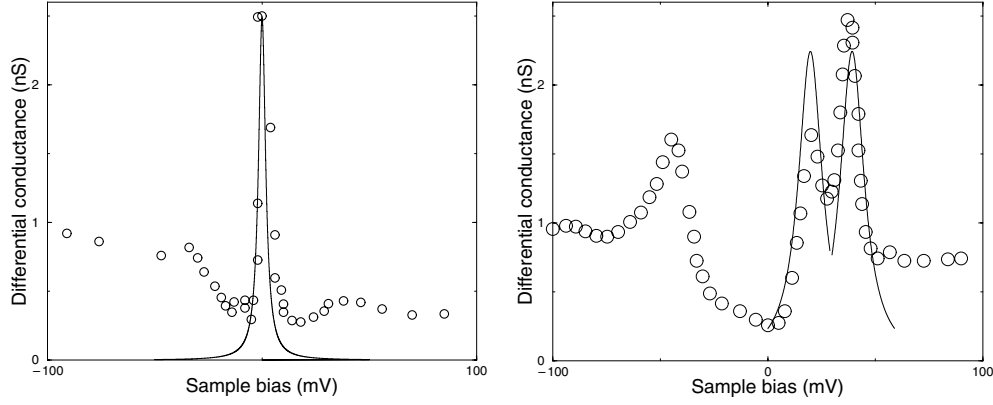
For  $v_3 = 0$ , there are also two bound states symmetric about zero bias, at

$$\bar{\omega}_b \rightarrow \pm \frac{|1 + u_+u_-|}{[(1 + u_+^2)(1 + u_-^2)]^{1/2}}, \quad (17)$$

where  $u_{\pm} = v_1 \pm v_2$ . For  $v_1 = 0$ , this reduces to the result of Shiba for a classical magnetic impurity,  $\bar{\omega}_b = \pm|1 - v_2^2|/(1 + v_2^2)$ , even though we did not average over the classical spin direction before summing the  $T$ -matrix [28].

The most interesting cases arise when  $0 \neq v_2 \neq v_3 \neq 0$ . When  $v_1 = 0$ , there are four bound states symmetric about zero bias at

$$\bar{\omega}_b \rightarrow \pm \frac{(v_+^2 - v_-^2 + s|1 - v_+^2v_-^2|)}{(1 + v_+^2)(1 + v_-^2)}, \quad (18)$$



**Figure 1.** Left: sketch of a bound state with  $v = (0, 0, \pm 1)$ , along with the DOS data from the STM differential conductance above a Zn site in BSCCO [9]. Right: sketch of the two positive-bias bound states obtained with  $v = (0, \pm 0.51, \pm 1.01)$ , along with the data obtained above the Ni site in BSCCO [10].

where  $v_{\pm} = v_2 \pm v_3$ . If either  $v_2$  or  $v_3 = 0$ , or if  $v_2 = v_3$ , there are only two bound states symmetric about zero bias. Equation (18) for either  $v_2 = 0$  or  $v_3 = 0$  reduces to equation (16) or (17) with  $v_1 = 0$ , respectively. For  $v_2 = v_3$ , it also reduces to equation (17) with  $v_1 = 0$  and  $v_2 \rightarrow 2v_2$ , etc. Moreover, setting  $v_2 = v_3$  in equation (15) with  $v_1$  arbitrary leads to only two bound states symmetric about zero bias, at  $\bar{\omega}_b = \pm[|b_-|/b_+]^{1/2}$ , where  $b_{\pm} = (1 + v_1^2)^2 \pm 4v_2^2$ . Thus, we conclude that in order to obtain four bound states symmetric about zero bias, one requires  $0 \neq v_2 \neq v_3 \neq 0$ .

When the defect is a quantum spin with a single component normal to the surface, the spin operator  $S_z$  commutes with the Hamiltonian, and the spin states are easily described by  $|SM\rangle$ , with  $S_z|SM\rangle = M|SM\rangle$ . Then the magnetic impurity in the presence of the non-magnetic potential and the pairing disorder can all be solved exactly. There are bound states for each of the  $2S + 1$  eigenstates.

In figure 1, we illustrated how this solution can aid in understanding the STM results obtained from the Bi sites directly above Zn and Ni impurity sites in the presumed top underlying  $\text{CuO}_2$  plane of BSCCO [9, 10]. In the left panel of figure 1, we fit the Zn STM data with a single pole obtained from equation (15) with  $v \equiv (v_1, v_2, v_3) = (0, 0, \pm 1)$  (or equivalently with  $v = (0, \pm 1, 0)$ ) and a width  $\delta\omega$  chosen to fit the data. The experimental peak centre appears at a slight offset from zero, which can be understood quantitatively by adjusting  $v_1 \ll 1$  and a width broader than the offset.

In the right panel of figure 1, we used equation (18) with  $v = (0, \pm 0.51, \pm 1.01)$  (or  $v = (0, \pm 1.01, \pm 0.51)$ ) to fit the two peak positions, and adjusted the widths to those of the data obtained above a Ni site [10]. We did not show the other two peaks expected from equation (18). That is because the data not pictured here show that the two peaks present in these data appear at equivalent negative biases on adjacent Bi sites [10]. In any event, our theory suggests that the STM data for Zn are consistent with it behaving either as a strong pairing fluctuation defect or as a strong magnetic impurity, and the Ni data suggests it behaves as both a strong magnetic impurity and a strong pairing defect in BSCCO. The similar pair-breaking defect strengths are consistent with the  $T_c$ -suppressions in BSCCO doped with these elements [6].

Thus, we solved the  $T$ -matrix in this modified BCS model of a local, on-site attractive pairing interaction with three types of defect on a site [24]. For a superconductor with

local, near-neighbour pairing of  $d_{x^2-y^2}$ -wave symmetry, the local gap  $\Delta_{ij}$  at the site  $(i, j)$  on a tetragonal lattice is coupled hierarchically to the  $\Delta_{i'j'}$  at every site  $(i', j')$ . Hence, a generalization of our results to d-wave superconductors would not be straightforward. A recent work neglected these problems with local d-wave pairing, treated Ni as an  $S = 1/2$  Ising impurity, and introduced an electronic filter by neglecting the most important Bi orbitals in the top insulating BiO layer, in order to fit the STM data [29]. In the present work, we fit the Ni and Zn data with s-wave pairing, a classical Ni spin, and without any BiO filter.

In summary, we have shown that disordered short-coherence-length superconductors can exhibit pair breaking from spatial fluctuations in the pairing interaction, in a manner very similar to that found with magnetic impurities. We studied the effects of a single site with up to three types of defect using the  $T$ -matrix approximation, and found bound states within the superconducting gap arising from either pairing fluctuations or magnetic impurities. Our best fits to the STM data above the sites of Zn and Ni impurities suggest that Zn behaves either as a strong pairing fluctuation defect or as a strong magnetic impurity, whereas Ni behaves both as a strong magnetic impurity and as a strong pairing defect.

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