Spontaneous Time Reversal and Parity Breaking in a $d_{x^2-y^2}$-Wave Superconductor with Magnetic Impurities

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It is shown that patches of *complex* d_{xy} components are generated around a magnetic impurity in the presence of coupling between the orbital moment of the condensate and the impurity spin S_z. The locally induced d_{xy} gap leads to the fully gapped quasiparticle spectrum near impurity. It is suggested that at low temperatures the well defined patches of d_{xy} are formed, possibly leading to a phase locked state due to Josephson tunneling. Violation of time-reversal symmetry and parity occurs spontaneously at this point via a second order transition. In the ordered phase both the impurity magnetization and the d_{xy} component of the order parameter develop and are proportional to each other. [S0031-9007(98)05427-1]

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It is well known that magnetic impurities destroy the singlet superconducting state due to spin scattering which breaks pair singlets [1]. In the case of the gapless (with the nodes of the gap) d-wave superconductor both magnetic and nonmagnetic impurities produce a finite density of states at zero energy. These are well known "incoherent" effects of impurities in unconventional superconductors. After recent experiments by Movshovich *et al.* [2] we are led to believe that another phenomenon is possible, namely, the transition to the second superconducting phase as a result of condensate interactions with magnetic impurities. The time-reversal violating state is formed at low energy, and the order parameter of the new phase is $d_{x^2-y^2} + id_{xy}(d + id)$. In this phase the impurity spins acquire nonzero spin density along the z axis, i.e., out of plane.

The physical origin of the instability comes from the fact that the $d + id$ state has an orbital moment which couples to the magnetic impurity spins. The relevant interaction is the $\hat{L}_z S_z$ coupling between impurity spin S_z and the conduction electron orbital moment L_z,

$$H_{\text{int}} = g \sum_i \int d^2r \, \frac{S_z(\mathbf{r}_i)}{|\mathbf{r} - \mathbf{r}_i|^3} \, \psi^\dagger_{\mathbf{r}\sigma} [\mathbf{r} \times i\partial_{\mathbf{r}}]_z \psi_{\mathbf{r}\sigma} \,, \quad (1)$$

where g is the coupling constant, $\psi_{\mathbf{r}\sigma}$ is the electron annihilation operator, and the summation is over impurity sites i. In the pure phase one can think of d-wave state as an equal admixture of the orbital moment $L_z = \pm 2$ pairs,

$$\Delta_0(\Theta) = \Delta_0 \cos 2\Theta = \frac{\Delta_0}{2} [\exp(2i\Theta) + \exp(-2i\Theta)] \,. \quad (2)$$

Here Θ is the 2D planar angle of the momentum on the Fermi surface, and Δ_0 is the magnitude of the $d_{x^2-y^2}$ component. We consider the 2D $d_{x^2-y^2}$ superconductor, motivated by the layered structure of the cuprates. In the presence of the (ferromagnetically) ordered impurity spins S_z the coefficients of the $L_z = \pm 2$ components will shift *linearly* in S_z with *opposite* signs,

$$\begin{aligned} \Delta_0(\Theta) \to \frac{\Delta_0}{2} [&(1 + gS_z)\exp(2i\Theta) \\ &+ (1 - gS_z)\exp(-2i\Theta)] \\ = \Delta_0(\Theta) &+ iS_z \Delta_1(\Theta) \,, \end{aligned} \quad (3)$$

where $\Delta_1(\Theta) \propto g/2 \sin 2\Theta$ is the d_{xy} component. The relative phase $\pi/2$ of these two order parameters comes out naturally because the $d + id$ state has a noncompensated orbital moment $L_z = +2$.

Here I argue that time reversal (T) and parity (P) symmetries can be broken *spontaneously* in the bulk of the d-wave state due to coupling to the impurity spins. Original $d_{x^2-y^2}$ is unstable towards the formation of the bulk $d_{x^2-y^2} + id_{xy}$ phase. To show how complex the d_{xy} component appears, I first consider the single magnetic impurity and find that the spin-orbit interaction generates a finite *complex* d_{xy} anomalous amplitude near impurity in the $d_{x^2-y^2}$ state. This patch of d_{xy} state is formed near the impurity site, as long as the $d_{x^2-y^2}$ amplitude is finite, and has a spatial extent of coherence length $\xi_0 = 20$ Å. It is therefore possible for these patches to form a long range phase coherent state at some lower temperature as a result of Josephson tunneling between different patches. Then I present a macroscopic Ginzburg-Landau (GL) functional and find that there is a *linear* coupling between the original $d_{x^2-y^2}$ order parameter $\Delta_0(\Theta) = \Delta_0 \cos 2\Theta$ and the spontaneously induced d_{xy} component $\Delta_1(\Theta) = \Delta_1 \sin 2\Theta$,

$$F_{\text{int}} = -\frac{b}{2i} (\Delta_0^* \Delta_1 - \text{H.c.}) S_z \,, \quad (4)$$

where $b \propto n_{\text{imp}} g$ is the macroscopic coupling constant, n_{imp} is the impurity concentration per unit cell of linear size a, $\Delta_{0,1}, g/2$ have dimension of energy. The time-reversal violation is natural in this case as it allows the order parameter $\Delta_0 + i\Delta_1$ to couple directly to the impurity spin. This coupling is possible only for $d + id$ and not for $d + is$ symmetry of the order parameter. From the GL description it follows that instability develops as a *second order* phase transition where both the out-of-plane

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magnetization S_z and d_{xy} component developed together and are proportional to each other [3].

Recent experimental observation of the surface-induced time-reversal violating state in YBCO suggests that the secondary component of the order parameter $(d + is)$ can be induced [4]. Theoretical explanation, based on surface-induced Andreev states, has been suggested by Fogelstrom and co-workers [5]. The source of the secondary component is the bending of the original $d_{x^2-y^2}$ order parameter at the surface.

In a different approach Laughlin [6] argued that the $d_{x^2-y^2}$ state is unstable toward the $d + id$ state in the bulk in the perpendicular magnetic field at low enough temperatures. The time reversal and parity are broken by the external field in this case. This transition was suggested to be responsible for the kinklike feature in the thermal conductivity in experiments by Krishana *et al.* [7].

Recent experiments reported the anomaly in the thermal conductivity in Bi2212 at low temperatures: The thermal conductivity of the Bi2212 with Ni impurities was observed to have a sharp reduction at $T_c^* = 200$ mK [2]. These data indicate the possible superconducting phase transition in the Bi2212 in the presence of the magnetic impurities. So far the transition has been seen only in the samples with magnetic impurities, e.g., Ni as opposed to the nonmagnetic impurities such as Zn [2]. It was reported that the feature in the thermal conductivity is completely suppressed by applying the field of $H \sim 200$ G. The low field and the fact that the feature disappears is consistent with the superconducting transition into second phase. Results presented here might be relevant for the experimentally observed transition at T_c^* in Bi2212 with Ni.

I begin by considering one impurity at site $\mathbf{r}_i = 0$ and interacting with conduction electrons via H_{int} in Eq. (1). Similar to the approach of [8], one can find the anomalous propagator in the presence of the single impurity scattering potential: $F_{\omega_n}(\mathbf{k}, \mathbf{k}') = F^0_{\omega_n}(\mathbf{k})\delta(\mathbf{k} - \mathbf{k}') + F^1_{\omega_n}(\mathbf{k}, \mathbf{k}')$, where $F^0 = \frac{\Delta_0 \cos 2\Theta}{\omega_n^2 + \xi_\mathbf{k} + \Delta_0^2 \cos^2 2\Theta}, G^0 = -\frac{i\omega_n + \xi_\mathbf{k}}{\omega_n^2 + \xi_\mathbf{k} + \Delta_0^2 \cos^2 2\Theta}$ are the pure system propagators, $F^1_{\omega_n}(\mathbf{k}, \mathbf{k}')$ is the correction due to impurity scattering, $\mathbf{k} = (k, \Theta)$ are the magnitude and angle of the momentum $\mathbf{k}$ on the cylindrical Fermi surface, ω_n is the Matsubara frequency, and $\xi_\mathbf{k} = \epsilon_\mathbf{k} - \mu$ is the quasiparticle energy, counted from the Fermi surface. We take S to be a classical variable and ignore spin flip scattering. To linear order in small g one finds

$$F^1_{\omega_n}(\mathbf{k}, \mathbf{k}') = -i2\pi g S_z G^0_{\omega_n}(\mathbf{k}) F^0_{\omega_n}(\mathbf{k}') \frac{[\mathbf{k} \times \mathbf{k}']_z}{|\mathbf{k} - \mathbf{k}'|}, \tag{5}$$

where $F^1_{\omega_n}(\mathbf{k}, \mathbf{k}')$ is the function of incoming and outgoing momenta because of broken translation symmetry. Upon integrating F^1 over $\mathbf{k}'$ and going to integrated over $\xi_\mathbf{k}$ propagator one finds

$$\begin{aligned} F^1_{\omega_n}(\Theta) &= \int N_0 \, d\xi_\mathbf{k} \, F^1_{\omega_n}(\mathbf{k}) \\ &= i\Lambda_{\omega_n}(N_0 g S_z)(N_0\Delta_0) \sin 2\Theta \,. \end{aligned} \tag{6}$$

Here $\Lambda_{\omega_n} \simeq k_F(\pi^2/2\sqrt{2}) \ln(W/\sqrt{\omega_n^2 + \Delta_0^2}) \langle 1/\sqrt{\omega_n^2 + \Delta_0^2 \cos^2 2\Theta} \rangle_\Theta$ is the model dependent coupling, W is the energy cutoff, N_0 is the density of states at the Fermi surface, and $\langle\,\rangle_\Theta$ stands for Fermi surface averaging.

The angle dependence of $F^1_{\omega_n}(\Theta) \sim igS_z \sin 2\Theta \propto k_x k_y$ is the one of d_{xy}. Together with the fact that this amplitude is complex, it indicates the existence of the id_{xy} component in the vicinity of magnetic impurity. This result also shows that incoming $d_{x^2-y^2}$-wave state electrons have a finite amplitude, linear in gS_z, to be scattered into the d_{xy} outgoing state via the $\hat{L}_z S_z$ coupling. From the solution Eq. (5) it is easy to see that the typical size of the patch, ignoring nodal directions, is given by superconducting coherence length $\xi_0 = 20$ Å. For the relevant concentration of Ni $n_{\text{imp}} \sim 1\%$ the Ni-Ni distance is about 35 Å. The patches are thus well overlapping in this limit, making phase ordering due to tunneling from patch to patch possible. These patches work as a microscopic seed of the d_{xy} component which grows into a true long range state at low temperatures $T \leq T_c^*$. A similar result for the d_{xy} patches in the mixed state of pure Bi2212 was shown in [9], where the role of impurities is assumed by vortices.

An important observable consequence of the *local* d_{xy} component near impurity is that the gap, as seen in STM tunneling near Ni impurity, will increase and the low energy part of density of states will be suppressed because of finite gap everywhere on the Fermi surface, as opposed to nodes for the pure $d_{x^2-y^2}$ state. The increase of the gap in the Ni-doped Bi2212, compared to the pure case, was observed in STM tunneling [10].

Next, I consider a simple example of magnetically ordered state: the ferromagnetically ordered impurity spins. Calculation, similar to the one above, yields

$$F^1_{\omega_n}(\mathbf{k}) = -in_{\text{imp}} \frac{2\pi}{a} g S_z G^0_{\omega_n}(\mathbf{k}) [\partial_\mathbf{k} \times \mathbf{k}]_z F^0_{\omega_n}(\mathbf{k}) \,. \tag{7}$$

The existence of the homogeneous d_{xy} component $F^1_{\omega_n}(\mathbf{k}) \sim -in_{\text{imp}} \frac{2\pi}{a} g S_z \Delta_0 \sin 2\Theta$ is evident from this equation. The relative phase of the d_{xy} component with respect to $d_{x^2-y^2}$ is determined by the sign of S_z. The gap Δ_1 has to be determined self-consistently provided there is interaction in the xy channel: $\Delta_1(\mathbf{k}) = T \sum_{\omega_n, \mathbf{k}'} V^1(\mathbf{k}, \mathbf{k}') F^1_{\omega_n}(\mathbf{k}')$. $V^1(\mathbf{k}, \mathbf{k}')$ does not have to be attractive, since $d_{x^2-y^2}$ plays the role of the source and Δ_1 will be generated for any sign and for any strength of interaction. I will assume there is such interaction, and results below will be expressed in terms of the induced gap Δ_1. With the help of this equation I find for the energy change due to Δ_1 : $\delta F = 1/2T \sum_{\omega_n, \mathbf{k}} F^1_{\omega_n}(\mathbf{k}) \Delta_1^*(\mathbf{k}) + \text{H.c.}$

$$\delta F = -i/2(N_0 \Delta_1^*)(N_0 \Delta_0) \frac{2\pi}{a} (gS_z) n_{\text{imp}} + \text{H.c.} \tag{8}$$

Equation (8) and single impurity result Eq. (6) are the main results of this section. From this equation I find the linear term Eq. (4) with

$$b = N_0^2 g \frac{2\pi}{a} n_{\rm imp} . \qquad (9)$$

So far the spin flips were ignored. The relative phase of the second component is determined by the sign of S_z. At high temperatures $T \gg T_c^*$, when spins are strongly fluctuating, the relative phase of the d_{xy} component is fluctuating strongly as well. This phenomenon is an interesting new realization of the superconducting phase (d_{xy}) coupled to the heat bath (fluctuating spins).

If and when the impurity spins are slowing down or even freezing out, then the phase scattering time becomes large and the phase ordering of the patches is possible; see Fig. 1. Measurements indicate that spin flips of Ni spins are slowing down at low temperatures $T \leq 2$ K. Specific heat measurements on Ni-doped Bi2212 indicate additional entropy, compared to undoped Bi2212, of the order of $n_{\rm imp} R \ln 3$, accumulated near 1 K [11]. The general shape of the specific heat, associated with the impurity spins is strikingly similar to the specific heat, observed in spin glasses [12]. A broad peak in the specific heat might indicate the glassy behavior of spins at lower temperatures.

Next I will consider simple mean field theory of the coupled magnetic impurities and superconducting condensate. I ignore the fluctuations in the magnetic subsystem and will assume that Ni impurities would order ferromagnetically at T_m in the absence of the interaction with condensate. This is a drastic oversimplification because of the possible spin-glass ordering discussed above. Nevertheless, the model presented below is useful in understanding the coupling between impurity spins and condensate and the $d + id$ instability of the original state.

I introduce the GL theory of the secondary superconducting transition: $\Delta_0 \to \Delta_0 + \Delta_1$ at T_c^*, where both $\Delta_{0,1}$ are homogeneous variables corresponding to the macroscopic ordering. The relative phase of Δ_1 with respect to the phase of Δ_0 *is not fixed* and will be determined by the free energy minimization. Assume that the second transition, if at all, occurs at $T_c^* \ll T_c$, where $T_c \sim 90$ K is the first transition temperature. Hence the order parameter Δ_0, which can be assumed to be real, is robust, and its free energy $F(\Delta_0)$ cannot be expanded in Δ_0.

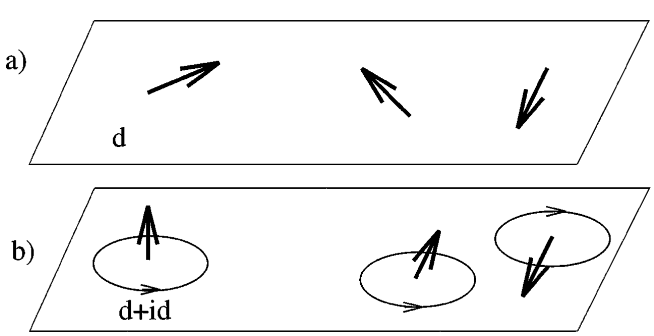

FIG. 1. (a) Impurity sites with random spins at high temperatures are shown. Spin flips lead to the d_{xy} component averaged to zero. (b) Upon slowing down and freezing of impurity spins the patches of $d \pm id$ with $L_z = \pm 2$ state near each Ni site are formed, shown with right (left) circulating current near each impurity site. At low temperatures Josephson tunneling locks the phase between patches, leading to the global $d + id$ state.

Assuming expansion in powers of small S_z, Δ_1 near second transition, the GL functional $F = F_{sc} + F_{\rm magn} + F_{\rm int}$ is

$$\begin{aligned} F_{sc} &= F(\Delta_0) + \tfrac{\alpha_1}{2} |\Delta_1|^2 + \tfrac{\alpha_2}{4} |\Delta_1|^4, \qquad \alpha_{1,2} \geq 0 , \\ F_{\rm magn} &= \tfrac{\alpha_1(T)}{2} |S_z|^2 + \tfrac{\alpha_2}{4} |S_z|^4, \qquad (10) \\ F_{\rm int} &= -\tfrac{b}{2i} (\Delta_0^* \Delta_1 - {\rm H.c.}) S_z . \end{aligned}$$

Δ_0 should enter in $F_{\rm int}$ for it to be invariant under the global $U(1)$ symmetry $\Delta_{0,1} \to \Delta_{0,1} \exp(i\theta)$; see Eq. (8) [13]. The homogeneous solution will have the lowest energy, and gradient terms are taken to be zero.

All but $F_{\rm int}$ terms in the free energy Eq. (10) are positive and cannot produce the instability of the original $d_{x^2-y^2}$ state. $F_{\rm int}$ can be negative since it is linear in Δ_1 and S_z, and this term is crucial in producing second transition.

Magnetic energy $F_{\rm magn}$ has a temperature dependent coefficient

$$a_1(T) = a_1 n_{\rm imp}(T - T_m) \qquad (11)$$

and vanishes at T_m; a_1 is dimensionless.

Consider F_{sc}. The second and third terms in F_{sc} describe the energy cost of opening the fully gapped state with Δ_1 when the interaction prefers to keep the node, i.e., pure $d_{x^2-y^2}$, state. The change in free energy due to the secondary order parameter is given by the difference in the energy of quasiparticles before and after the Δ_1 component is generated. One can calculate the change in the energy of the superconductor subjected to the *homogeneous* external d_{xy} source field and find an increase of energy at $\Delta_1 \ll T \ll \Delta_0 : \delta F_{sc} = \frac{\alpha_1}{2} |\Delta_1|^2 + \frac{\alpha_2}{4} |\Delta_1|^4$ with $\alpha_1 \simeq N_0$. Amplitude Δ_1 is taken to be constant on the Fermi surface [3].

Fix Δ_0 to be real positive and the relative phase of $\Delta_1 = |\Delta_1| \exp(i\nu)$ without loss of generality. Minimizing the functional Eq. (10) I find that the $\pi/2$ relative phase of Δ_1 comes out naturally,

$$\nu = \pi/2 \, {\rm sgn}(b S_z) . \qquad (12)$$

This choice takes the maximum advantage of the $L_z S_z$ coupling, and minimization *requires* a complex order parameter $\Delta_0 + i\Delta_1$ in the low temperature phase. T and P are violated *spontaneously* even at $T_m = 0$. The minimization yields $S_z = \frac{b}{a_1 n_{\rm imp}(T - T_m)} \sin \nu \Delta_0 |\Delta_1|$ with

$$\begin{aligned} |\Delta_1|^2 &= \tfrac{1}{\alpha^2} \left(\tfrac{b^2}{a_1 n_{\rm imp}(T - T_m)} \Delta_0^2 - \alpha_1\right) = \chi(T_c^* - T) , \\ T_c^* &\simeq T_m + 4\pi^2 (N_0 \Delta_0)^2 n_{\rm imp} \tfrac{g^2 N_0}{a_1 a^2} , \end{aligned} \qquad (13)$$

where I used Eq. (9) in the last line. This is the main result of this paper. Solution Eq. (13) indicates that the transition is of the second order $\delta F = -\frac{\alpha_2}{4} |\Delta_1|^4 \sim |T - T_c^*|^2$ with the jump in the specific heat. I assumed that $|\Delta_0(T)|^2/\alpha_1(T - T_m)$ has a linear temperature slope near T_c^*. Higher order terms in F_{sc}, e.g., $\gamma\Delta_0^{*2}\Delta_1^2$ + H.c. do not change this result and only renormalize T_m in Eq. (13) regardless of the sign of γ. From F_{int}, once the spin field is integrated out, the effective quadratic coupling is b^2/a_1 and dominates near T_m. From the solution of Eq. (13) the following results are noted.

(1) Even if $T_m = 0$ the ordering will occur at $T_c^* = 4\pi^2 n_{\text{imp}}(N_0\Delta_0)^2 \frac{g^2 N_0}{a_1 a^2}$. However, the softness of the spin system near T_m enhances the effect and makes $T_c^* \geq T_m$ within this mean field approach. Taking the typical values for Bi2212 of $\Delta_0 = 450$ K, $E_F = 1/N_0 = 3000$ K assuming $a_1 \sim 1$, and taking the characteristic value of spin-orbit coupling of Ni in these compounds $g/a \sim$ 320 K [14], I find $T_c^*|_{T_m=0} \simeq 0.3$ K. In the real system, if the spin-glass freezing occurs, the spins that are freezing out first will be far apart. This will preclude the Josephson tunneling between the patches, as discussed above. Only at lower temperatures, when the majority of spins are frozen, the tunneling would be able to lock in the superconducting phase. Hence the real phase ordering will occur at temperature, substantially lower than the mean field estimated $T_c^* \leq T_m$.

(2) As the function of impurity concentration two effects occur simultaneously. First, condensate density $|\Delta_0|^2$ decreases. Second, the suppression of Δ_1 due to increased impurity scattering will also lower T_c^*. These effects will lead eventually to the disappearance of the transition. Quick suppression of the transition with impurity concentration should be expected.

(3) Strong magnetic field parallel to the layers, $H \gg H_{c1,ab} \sim 1$ G in plane, will suppress the second phase. In the field Ni spins will be aligned in the layers, the linear coupling term on H_{int} will be zero, and the d_{xy} component will vanish. This effect might explain the suppression of the second transition by magnetic field $H_{c1,ab} \ll H \leq H_{c1,c} \sim 300$ G, seen in experiment [2].

In the superconducting state with nonzero orbital current L_z the dominant fraction of the orbital moment is "stored" at the edge of the sample, similar to ^{3}He-A. The edge currents in the $d + id$ state and their topological characteristics were addressed recently in [6,15].

In conclusion, I presented the mechanism for a second order phase transition of the original d-wave state into the $d + id$ state with spontaneously broken T and P. In the ordered phase both impurity spins S_z and the d_{xy} component of the order parameter develop simultaneously. To test the proposed state the following experiments can be done. The driving mechanism for the second phase clearly distinguishes between magnetic and nonmagnetic impurities, hence more experiments on Bi2212 with nonmagnetic impurities will be helpful [2]. Theory predicts the ordering of Ni moments below T_c^*, and one should be able to detect magnetization in muon spin rotation (μSR) experiments or in ac susceptibility. The increased superfluid density due to the second component translates into the change in the penetration depth below T_c^* which can be detected. These and other experiments will help to resolve whether the proposed mechanism is correct.

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