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Broken time-reversal symmetry and superconducting states in the cuprates

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Recently, Kaminski *et al.* have reported that time-reversal symmetry is broken in the pseudogap phase in the high-temperature superconducting material $Bi_2Sr_2CaCu_2O_{2+\delta}$ (Bi-2212). Here we examine the role of translationally invariant broken time-reversal states on $d_{x^2-y^2}$ superconductors. In particular, we determine the change in the superconducting order parameter structure. We find that the broken time-reversal pseudogap state that is consistent with the experiment of Kaminski *et al.* gives rise to a mixed singlet-triplet pairing $d+ip$ phase. This $d+ip$ state is shown to give rise to a helical superconducting phase. Consequences of this $d+ip$ state on Josephson experiments are discussed.

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The origin of the pseudogap regime in the cuprates has been the subject of controversy. A variety of probes reveal a suppression of the single-particle density of states in this regime.[1,2] A natural explanation is that the pseudogap phase is a precursor superconducting state; a phase in which there are Cooper pairs but no superconducting phase coherence.[3] The recent experimental results of Kaminski *et al.*[4] and of Alff *et al.*[5] provide evidence for a very different explanation of the pseudogap phase: it marks a new phase in which a symmetry is broken. In particular, the results of Ref. 4 have reported that left-circularly polarized photons give a different photocurrent from right-circularly polarized photons in the pseudogap phase. This, combined with a mirror plane symmetry, implies the breaking of time-reversal symmetry in the pseudogap phase. Varma[6] has proposed translationally invariant orbital current states that may account for the observed results. If it is indeed the case that the pseudogap phase does not break translational symmetry and breaks time-reversal symmetry, then the classification of the superconducting pairing symmetry will differ from previous classifications. Figure 1 shows the resulting phase diagram. In this paper, we determine the structure of the superconducting gap function in the pseudogap phase when time-reversal symmetry is broken and translational symmetry is preserved. Note that this implies that we do not consider the *d*-density wave state of Chakravarty *et al.*[7] The reason for this restriction is that a pseudogap phase which breaks translational symmetry will not alter the superconducting state as strongly as the case considered here. The layout of the paper is as follows: first we find the possible superconducting states in the pseudogap phase by using both corepresentation theory and phenomenological Ginzburg Landau arguments. This is done for all pseudogap symmetries that retain translational symmetry. Then we focus on the pseudogap phase that is consistent with the experimental results of Kaminski *et al.* This phase is determined by the requirement that the pseudogap phase breaks time-reversal symmetry, breaks the fourfold rotation symmetry of the CuO_2 plane, and also breaks the mirror plane symmetry with normal along the Cu-O diagonal. This leads to the two possible pseudogap order parameters that have been examined by Varma and Simon.[6,8] A detailed symmetry analysis of the photoemission matrix elements rules out one of these two order parameters.[8] We develop a Ginzburg-Landau theory of the remaining pseudogap order parameter and the interplay of this order parameter with superconductivity. We discuss some observable consequences of the superconducting gap components induced by the pseudogap order parameter. In particular, we show that a helical superconducting phase is a consequence of broken time-reversal symmetry in this case. Finally, we discuss the *c*-axis Josephson current that has been observed in strongly underdoped Bi-2212-Pb junctions.[9] We show how this can possibly be explained by the coupling of the superconducting order parameter to domain walls in the pseudogap order.

Here we describe the possible magnetic point groups of the pseudogap phase and their respective free energies. The symmetry group of the pseudogap phase can be written as $\mathcal{G}=G_M\times U(1)$, where G_M is the magnetic space group and U(1) is the gauge group (which is not broken in the pseudogap phase). The group G_M is the group that leaves both the charge density and the magnetization density **M** invariant. We will be interested in the possible magnetic point groups that arise when time-reversal symmetry is broken in Bi-2212. We will focus only on transitions that do not break translational invariance and thus focus on the 4/*mmm*

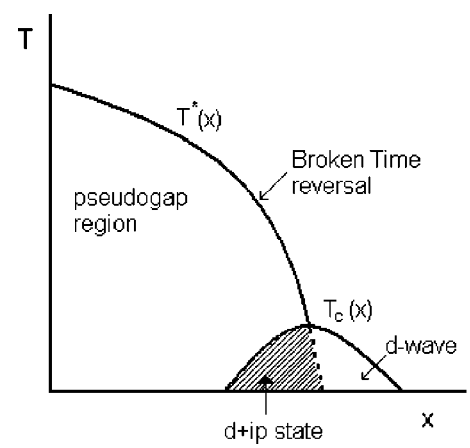

FIG. 1. Possible phase diagram, temperature T as a function of hole doping, of the cuprate Bi-2212. Here T^* is the critical temperature where time-reversal symmetry is broken and T_c is the transition temperature of superconducting phase.

TABLE I. Superconducting COREP basis functions for different magnetic point-group symmetries of the pseudogap phase. Induced REP refers to the superconducting component induced by the pseudogap order. The magnetic point groups for two-dimensional pseudogap REPS depend upon the form of η_1, η_2, here we take $\eta_1 = \pm\eta_2$.

Pseudogap Phase (REP)	Induced (REP)	Basis functions (COREP)
$A_{1g}(4/mmm)$	B_{1g}	$(k_x^2-k_y^2)[1+ia(k_x^2+k_y^2)]$
$A_{2g}(4/m\underline{mm})$	B_{2g}	$(k_x^2-k_y^2)+iak_xk_y$
$B_{1g}(\underline{4}/mm\underline{m})$	A_{1g}	$(k_x^2-k_y^2)+ia(k_x^2+k_y^2)$
$B_{2g}(\underline{4}/mm\underline{m})$	A_{2g}	$(k_x^2-k_y^2)+iak_xk_y(k_x^2-k_y^2)$
$E_g(m\underline{mm})$	E_g	$(k_x^2-k_y^2)+ia(\eta_1k_y-\eta_2k_x)k_z$
$A_{1u}(4/\underline{mmm})$	B_{1u}	$(k_x^2-k_y^2)+ia(\hat{\sigma}_1k_x-\hat{\sigma}_2k_y)$
$A_{2u}(4/\underline{m}mm)$	B_{2u}	$(k_x^2-k_y^2)+ia(\hat{\sigma}_1k_y+\hat{\sigma}_2k_x)$
$B_{1u}(\underline{4}/\underline{m}m\underline{m})$	A_{1u}	$(k_x^2-k_y^2)+ia(\hat{\sigma}_1k_x+\hat{\sigma}_2k_y)$
$B_{2u}(\underline{4}/\underline{m}m\underline{m})$	A_{2u}	$(k_x^2-k_y^2)+ia(\hat{\sigma}_1k_y-\hat{\sigma}_2k_x)$
$E_u(\underline{m}mm)$	E_u	$(k_x^2-k_y^2)+ia(\eta_1k_y-\eta_2k_x)\hat{\sigma}_3$

(D_{4h}) point group which is defined through the elements $\{E,C_{2x},C_{2y},C_{2z},i,\sigma_x,\sigma_y,\sigma_z,C_{da},C_{db},\sigma_{da},\sigma_{db},\pm C_{4z},\pm iC_{4z}\}$. Magnetic point groups are defined as $S_M = H + \theta(G-H)$, where H is a halving subgroup of the ordinary point group G. In magnetic symmetry groups, the crystal point-group operation R, which belongs to $G-H$ group, transforms the magnetization density $\mathbf{M}$ to $-\mathbf{M}$, but the product of the time-reversal operation θ and the operation R leaves $\mathbf{M}$ invariant. In Table I, a list of magnetic point groups corresponding to pseudogap phase has been given. According to the experimental observations of Kaminski *et al.*,[4] the fourfold rotation about z axis (C_{4z}) and the diagonal mirror planes (σ_{da},σ_{db}) are no longer symmetry operations in the pseudogap state. The requirement that these two symmetries are no longer present reduces the number of possible magnetic point groups of the pseudogap phase to two: $\underline{4}/mm\underline{m}$ $\{E,C_{2x},C_{2y},C_{2z},i,\sigma_x,\sigma_y,\sigma_z,\theta C_{da},\theta C_{db},i\theta C_{da},\theta\sigma_{db},\pm\theta C_{4z},\pm i\theta C_{4z}\}$ or $\underline{m}mm$ $\{E,C_{da},\sigma_z,\sigma_{db},i\theta,\theta\sigma_{da},\theta C_{2z},\theta C_{db}\}$. The symmetries $\underline{4}/mm\underline{m}$ and $\underline{m}mm$ agree with the proposed orbital current patterns of Ref. 6.[10] Consequently, we use the same notation: we label the group $\underline{4}/mm\underline{m}$ as type I and the group $\underline{m}mm$ as type II. A detailed symmetry analysis of the photoemission matrix elements rules out the type I state. So we will consider the type II phase in more detail (note that Stanescu and Phillips have also examined the type II phase microscopically[11]).

For the one-dimensional pseudogap order parameters [A or B irreducible representations (REPS) of the D_{4h} point group in Table I], the Ginzburg Landau free energy is simply given as

$$F_{pg,I}[\eta] = \alpha_1\eta^2 + \frac{\beta_1}{2}\eta^4. \tag{1}$$

Two degenerate states $\eta = \pm\sqrt{-\alpha_1/\beta_1}$ minimize this free energy.

For the two-dimensional pseudogap order parameters (E_u or E_g REPS of D_{4h} point group in Table I), the corresponding Ginzburg Landau free energy is

$$F_{pg,II}[\eta_x,\eta_y] = \alpha(\eta_x^2+\eta_y^2) + \frac{\beta}{2}(\eta_x^4+\eta_y^4) + \gamma\eta_x^2\eta_y^2. \tag{2}$$

Minimization of the free energy with respect to η_x and η_y gives the following sets of degenerate states:

$$(\eta_x,\eta_y) = (0,0); \qquad \sqrt{\frac{-\alpha}{\beta}}\{(0,\pm1),(\pm1,0)\};$$

$$\sqrt{\frac{-\alpha}{\beta+\gamma}}\{(\pm1,\pm1),(\pm1,\mp1)\}. \tag{3}$$

Of these states, $\eta_0(\pm1,\pm1)$ and $\eta_0(\pm1,\mp1)$, where $\eta_0 = \sqrt{-\alpha/(\beta+\gamma)}$, minimize the free energy if $\gamma<\beta$. The other four degenerate states $\acute{\eta}_0(0,\pm1), \acute{\eta}_0(\pm1,0)$, where $\acute{\eta}_0 = \sqrt{-\alpha/\beta}$, minimize the free energy when $\gamma>\beta$. Simon and Varma have depicted the (1,1) state to represent the type II current pattern,[6] but there are three more states which are degenerate with this state. In these states, the tetragonal symmetry is lost as $C_{4z}(1,1) = (1,-1)$. Each domain has symmetry $\underline{m}mm$ with the $\underline{m}$ oriented along the different diagonals. The $\sigma_{da(db)}$ symmetry will be lost if there are multiple domains. Note that the observed incommensurate modulation in Bi-2212 implies the existence of an ϵ_{xy} strain.[9] This gives rise to an additional $\epsilon_{xy}\eta_x\eta_y$ invariant in the pseudogap free energy. This prefers the $(\pm1,\pm1)$ and $(\pm1,\mp1)$ states and also breaks the degeneracy of these two states so that they are now twofold degenerate.

To classify the possible superconducting states in the presence of broken time-reversal symmetry we require the use of corepresentation theory. The usual representation theory must be extended because the time-reversal operator is antilinear and antiunitary. The magnetic point group can be written as $S_M = H + A\times H$, where A is antiunitary operator such that all elements of the coset AH are antiunitary. The corepresentations (COREPS) $D\Gamma$ of S_M can be found from REPS Γ of the corresponding normal group H in one of three ways [labeled (a), (b), or (c)]. This approach is explained in Ref. 12 and we use their notation. Recently, similar considerations have appeared in the classification of superconducting states in ferromagnets.[13–15] The superconducting gap function is defined as[16,17] $\hat{\Delta}(\mathbf{k}) = i[\psi(\mathbf{k}) + i\mathbf{d}(\mathbf{k})\cdot\hat{\sigma}]\hat{\boldsymbol{\sigma}}_2$. In Table I we give representative gap functions $\psi(\mathbf{k})$ and $\mathbf{d}(\mathbf{k})$ for various pseudogap symmetries.

As an example, consider the $\underline{4}/mm\underline{m}$ magnetic point group. The pseudogap order parameter in this case corresponds to a one-dimensional REP of D_{4h}. For this magnetic point group, $H = D_{2h}$ and $A = \theta C_{4z}$. For a $d_{x^2-y^2}$ pairing symmetry when time-reversal symmetry is not broken, we are interested in the A_{1g} REP of D_{2h}. The resulting pairing state is a real linear combination of $\psi(\mathbf{k}) = k_x^2 + k_y^2$ and $\psi(\mathbf{k}) = i(k_x^2 - k_y^2)$ which can be denoted as a $d+is$ pairing state.

TABLE II. The COREPS of $\underline{m}mm$ group with $H=C_{2v}$. a and b are arbitrary constants, $k_{da}=k_x+k_y$, $k_{db}=k_x-k_y$, and σ_1,σ_2 and σ_3 are Pauli matrices. The COREPS are all type (a).

	E $i\theta$	C_{da} $\theta\sigma_{da}$	σ_z θC_{2z}	σ_{db} θC_{db}	$\psi(\mathbf{k})+\mathbf{d}(\mathbf{k}).\hat{\sigma}$ Basis (COREP)
A_1	1	1	1	1	$(k_x^2+k_y^2)+ak_xk_y+ib\sigma_3k_{da}$
A_2	1	1	-1	-1	$k_{db}k_z+ia(\sigma_1k_x+\sigma_2k_y)$ $+ib(\sigma_1k_y+\sigma_2k_x)$
B_1	1	-1	1	-1	$(k_y^2-k_x^2)+ia\sigma_3k_{db}$
B_2	1	-1	-1	1	$k_{da}k_z+ia(\sigma_1k_y-\sigma_2k_x)$ $+ib(\sigma_1k_x-\sigma_2k_y)$

The magnetic point group $\underline{m}mm$ deserves further consideration since it is consistent with the experimental observations of Kaminski *et al.*[8,4] For this group, $H=C_{2v}$ and $A=i\theta$. Due to the broken parity symmetry in the pseudogap phase, the pairing gap function is a mixture of spin-singlet $[\psi(\mathbf{k})]$ and spin-triplet components $[\mathbf{d}(\mathbf{k})]$. For a $d_{x^2-y^2}$ pairing symmetry when time-reversal symmetry is not broken, we are interested in the B_1 REP of C_{2v} (given in Table II). The corresponding COREP is a real linear combination of the spin-singlet $\psi(\mathbf{k})=(k_y^2-k_x^2)$ and the spin-triplet $\mathbf{d}(\mathbf{k})=i(k_x-k_y)\hat{z}$ gap functions. We label this state the $d+ip$ phase. In Table II, we have also provided representative basis functions for the other COREPS corresponding to the different REPS of C_{2v}. From the form of the gap functions found here we can deduce whether there is possibility of nodes in the gap. We find that the superconducting gap will vanish along the $k_x=k_y$ line in this case (the gap does not vanish along the $k_x=-k_y$ line). Note that similar considerations appear in a recent symmetry analysis of a d-wave superconductor in a uniform current.[18]

Here we determine the superconducting gap structures in the pseudogap phase using Ginzburg Landau theory. This approach provides the same results as those found above using the less familiar corepresentation theory. Using corepresentation theory, it has been shown that the existence of d-wave order parameter ψ_d and pseudogap order parameter η ensures the existence of an induced superconducing order parameter ψ. In Table I, the different possible combinations of order parameters of the pseudogap order η and of induced superconducting order ψ have been listed. These same states can be found by examining Ginzburg Landau theory. In particular, the invariance of the free energy with respect to time-reversal symmetry requires any free energy invariant corresponding to any one-dimensional pseudogap REP to have the form $i\epsilon\eta(\psi_d^*\psi-\psi^*\psi_d)$, where ϵ is real coupling coefficient. Therefore, the superconducting Ginzburg Landau free energy corresponding to the one-dimensional pseudogap REPS of Table I (up to second order in ψ) is

$$F_I(\eta,\psi_d,\psi)=F_{pg,I}+\alpha_d|\psi_d|^2+\beta_d|\psi_d|^4+\tilde{\alpha}|\psi|^2+i\epsilon\eta(\psi_d^*\psi-\psi^*\psi_d). \tag{4}$$

The relation between the order parameters can be obtained by minimizing $F(\eta,\psi_d,\psi)$ with respect to ψ^*. The relation is $\psi=\pm i\epsilon\eta\psi_d/\tilde{\alpha}$, where $\pm$ sign is due to the degeneracy of ψ and $-\psi$ states. From the relation it is clear that for the nonzero η and ψ_d order parameters, ψ must be nonzero, which ensures that the symmetry of superconducting state in pseudogap phase is $d+i\psi$.

For a type II pseudogap phase, the order parameter belongs to the two-dimensional E_u REP of the D_{4h} point group. Since the product of the d-wave order parameter ψ_d and pseudogap order parameter (η_x,η_y) transforms as a E_u REP, the induced superconducting order parameter also belongs to the E_u REP. The induced superconducting order parameter can be written as $\psi=(p_x,p_y)$. To construct a nontrivial invariant, we decompose the product of representations $E_u\otimes E_u\otimes B_{1g}$. The relevant invariant is $i\tilde{\epsilon}[\psi_d^*(\eta_xp_x-\eta_yp_y)-\psi_d(\eta_xp_x^*-\eta_yp_y^*)]$, where $\tilde{\epsilon}$ is a real positive coefficient. The Ginzburg Landau free energy [up to second order in (p_x,p_y)] is

$$F_{II}(\psi_d,\eta,p)=F_{pg,II}+\alpha_d|\psi_d|^2+\beta_d|\psi_d|^4+\alpha_p(|p_x|^2+|p_y|^2)+i\tilde{\epsilon}[\psi_d^*(\eta_xp_x-\eta_yp_y)-\psi_d(\eta_xp_x^*-\eta_yp_y^*)]. \tag{5}$$

Minimizing the free energy with respect to p_x^* and p_y^* gives $p_x=i\tilde{\epsilon}\psi_d\eta_x/\alpha_p$ and $p_y=-i\tilde{\epsilon}\psi_d\eta_y/\alpha_p$. Thus we have $(p_x,p_y)=i\tilde{\epsilon}(\eta_x,-\eta_y)\psi_d/\alpha_p=i\tilde{\epsilon}\eta_0(1,-1)\psi_d/\alpha_p$, where $(\eta_x,\eta_y)=\eta_0(1,1)$ has been used.

The appearance of the induced superconducting gap functions in the pseudogap phase leads to a variety of observable consequences. In the pseudogap phase having $d+ip$ state, the breaking of parity symmetry gives rise to Lifshits invariants in the free energy which give rise to a spatially varying (helical) superconducting phase.[19] This behavior can be readily explained in terms of the Ginzburg Landau free energy. In particular, the relevant Lifshitz invariant is

$$F_L=F_{II}+i\acute{\epsilon}[\psi_d^*(D_xp_x-D_yp_y)-\psi_d(D_x^*p_x^*-D_y^*p_y^*)], \tag{6}$$

where $\mathbf{D}=(D_x,D_y)$, $D_j=-i\boldsymbol{\nabla}_j-2eA_j/\hbar c$ and $\mathbf{A}$ is the vector potential. The helical superconducting phase can be found by setting $\mathbf{A}=0$ and considering the spatial variation of order parameters as $\psi_d=\psi_{d0}e^{i\mathbf{q}\cdot\mathbf{r}}$ and $(p_x,p_y)=(p_{x0},p_{y0})e^{i\mathbf{q}\cdot\mathbf{r}}$. Minimizing with respect to q_x and q_y gives $q_x=q_y=-\acute{\epsilon}\epsilon\eta_0/\alpha_p\tilde{\kappa}$ (where $\tilde{\kappa}$ is the defined through the gradient term $\tilde{\kappa}|\mathbf{D}\psi_d|^2$ in the free energy). Note that gauge invariance and minimization of the free energy with respect to $\mathbf{q}$ implies that the current in helical phase is zero. The helical structure of the order parameter can be verified by Josephson junction experiments. We refer to Ref. 20 for details. The existence of gap nodes found in the last section assumed a uniform (non-helical) order parameter. If $1/q_x\gg\xi_0$, where ξ_0 is the coherence length, then the nodes will presumably still provide a reasonable description of the low-energy excitations of the superconductor.

It is interesting to note that a Josephson current has been observed through c-axis Josephson junctions between *underdoped* Bi-2212 and Pb. Mößle and Kleinger *et al.* have demonstrated this through a series of very careful experiments.[9] They have shown that the junctions are homogeneous and the coupling is a conventional lowest-order Josephson coupling. This experiment is difficult to explain with a pure *d*-wave order parameter in Bi-2212. Rae has pointed out that this current may exist if the Pb superconductor contains a *d*-wave contribution.[21] This is possible if the junction is made from a low-symmetry orientation of lead (a [110] face, for example). However, as pointed out by Rae, this explanation requires that there is a systematic bias towards [110] Pb faces in the junctions which remains to be verified. Given the possibility of a $d+ip$ phase, it is natural to ask if this can be related to the observed Josephson current. Here we show that while a $d+ip$ state as described above does not have a Josephson current, a domain wall in the pseudogap order parameter will. This can be understood by considering surface energy at a *c*-axis junction between a $d+ip$ superconductor and a conventional *s*-wave superconductor;

$$F_{sur}=\int d^2S[\psi_s^*(\eta_x p_x+\eta_y p_y)+\text{c.c.}]. \tag{7}$$

If we consider $(\eta_x,\eta_y)=\eta_0(1,1)$ then, as shown above, $p_x=-p_y$ and F_{sur} will be zero. But consider a domain wall of the type $(\eta_x,\eta_y)=\eta_0[1,\tanh(x/\xi)]$ (here η_x remains constant but η_y varies from $-\eta_0$ to η_0 across the domain wall), the current density in this case is given as

$$j=j_0[1-\tanh^2(x/\xi)]\sin(\theta_d-\theta_s), \tag{8}$$

where $j_0=4e\eta_0^2\tilde{\epsilon}|\psi_d||\psi_s|$. A Fraunhofer pattern can be obtained by applying the field along the normal to the domain walls. However, as the angle between the field and domain walls decreases, the current pattern will deviate from the usual Fraunhofer pattern. In particular, for the field along the domain wall, only the central peak of the original Fraunhofer pattern remains. Note that these considerations require the junction size to be much smaller than the period of the helical order, so that the *p*-wave order parameter is approximately uniform. Furthermore, in Bi-2212, the incommensurate modulation may alter this analysis. If there is a strong interaction between this modulation and the pseudogap order parameter, then any domain walls in the pseudogap order parameter will be tied to domain boundaries of incommensurate modulation.

In conclusion, we have determined the superconducting gap structure in translationally invariant pseudogap phases that break time-reversal symmetry. It has been shown that a $d+ip$ state is the superconducting ordered state for the pseudogap state that agrees with the experimental results in Bi-2212. The induced ip component removes two of the four nodes associated with a $d_{x^2-y^2}$ order parameter. The consequences of the induced ip order parameters on Josephson experiments have been explored. The induced ip phase can explain the observed Josephson current through a *c*-axis junction between underdoped Bi-2212 and Pb only if there are the domain walls in the pseudogap order parameter. It has also been shown that the $d+ip$ state will give rise to a helical superconducting phase.

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