Order-parameter symmetries in high-temperature superconductors

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We present two representations of the order-parameter (OP) eigenfunctions for tetragonal cuprate superconductors, and for orthorhombic YBa₂Cu₃O_{7- δ} and Bi₂Sr₂CaCu₂O_{8+ δ} (BSCCO). Recent BSCCO *c*-axis twist Josephson junction experiments of Li *et al.* demonstrate that the tunneling is strongly incoherent, and that the OP contains an isotropic *s*-wave component, but not any $d_{x^2-y^2}$ -wave component. We propose a *c*-axis twist tetracrystal ring experiment.

Several developments have reopened the question of the symmetry of the order parameter (OP) in the high transition temperature T_c superconductors.^{1–5} Although phasesensitive tricrystal experiments were interpreted in terms of an OP in $YBa_2Cu_3O_{7-\delta}$ (YBCO) dominated at low temperature *T* by a $d_{x^2-y^2}$ -wave component,⁶ other YBCO experiments indicated a substantial *s*-wave component,⁷ or even a nodeless OP.^{4,5} *c*-axis Pb/Bi₂Sr₂CaCu₂O_{8+ δ} (BSCCO) Josephson demonstrated the existence of an *s*-wave OP component in BSCCO at low *T*. 3

In BSCCO *c*-axis twist experiments, the critical current densities J_c^J and J_c^S of each twist junction and of the constituent single crystals were *the same*, regardless of twist angle ϕ_0 ¹ As shown in the following, this implies that BSCCO has strongly incoherent *c*-axis tunneling, and its OP contains an isotropic *s*-wave component for $T \le T_c$, but no $d_{x^2-y^2}$ -wave component.

The CuO₂ plane bands cross the Fermi energy E_F .^{8,9} The crystal point group of a tetragonal $CuO₂$ plane is C_{4v} .¹⁰ The group operations are the identity *E*, mirror reflections σ_x , σ_y in the *ac*,*bc* planes, respectively, mirror reflections σ_{d1} , σ_{d2} in the d_1c, d_2c planes containing the diagonals d_1, d_2 , respectively, and rotations $C_4^{\pm 1}$, and $C_2 = C_4^2$ by $\pm 90^\circ$ and 180° about the *c* axis, respectively.¹⁰ The bases of spin singlet functions belonging to the one-dimensional irreducible representations (IR's) Γ of C_{4v} are denoted $|s\rangle$, $|d_{x^2-y^2}\rangle$, $|d_{xy}\rangle$, and $|g_{xy(x^2-y^2)}\rangle$. Under a given symmetry operation \mathcal{O}_s , each function $|f\rangle$ within a given basis satisfies $\mathcal{O}_s|f\rangle = \lambda_{sf}|f\rangle$, where λ_{sf} is the corresponding group character or eigenvalue. Functions within the same basis are *compatible*. Elements of different bases are *incompatible*.

We use two representations of the OP basis sets. For Fermi-surface (FS) restricted pairing (BCS) on a cylindrical FS, the intralayer-paired quasiparticle momenta are described by $\phi_{\mathbf{k}}$, where $\mathbf{k}_F = k_F(\cos \phi_{\mathbf{k}}, \sin \phi_{\mathbf{k}})$. The OP basis is then described using angular momentum quantum numbers *l*. The respective four eigenfunction bases are represented as a constant, $\cos(2\phi_k)$, $\sin(2\phi_k)$, and $\sin(4\phi_k)$, each multiplied by some function $\sum_{l=0}^{\infty} A_l \cos(4l\phi_k)$, behaving as a constant under C_{4v} .

For a tight-binding band structure, we use the real-space representation, in which the paired quasiparticles are located on distinct lattice sites, their separations along the Cu-O bond directions counted by the quantum numbers (*n*,*m*). The wave vectors $\mathbf{k}=(k_x,k_y)$ are within the first Brillouin zone (BZ) given by $|k_x|, |k_y| \le \pi/a$. The respective four eigenfunction bases are represented as a constant, $cos(k_xa)$ $-\cos(k_y a)$, $\sin(k_x a) \sin(k_y a)$, and $\sin(k_x a) \sin(k_y a) [\cos(k_x a) - \cos(k_y a)]$, each multiplied by some function each multiplied by some function $\sum_{n,m=0}^{\infty} A_{nm} \cos(n k_x a) \cos(m k_y a)$ with $A_{nm} = A_{mn}$, behaving as a constant under C_{4v} .

In orthorhombic YBCO, the b (or y) axis is longer than the *a* (or *x*) axis. The C_{2v}^1 crystal point group of the CuO₂ planes only allows the group operations $E, \sigma_x, \sigma_y,$ and C_2 . The two allowable OP eigenfunctions denoted |s $+d_{x^2-y^2}$ and $d_{xy}+g_{xy(x^2-y^2)}$ are listed for both representations in Table I. In YBCO, *s*- and $d_{x^2-y^2}$ -wave functions are compatible.

In BSCCO, the orthorhombic axes are the *diagonals* d_1 (*a*) and d_2 (*b*). The *bc* plane containing the periodic lattice distortion **Q** forms a crystallographic σ_{d2} mirror plane.^{2,11} The C_{2v}^{13} point group of the CuO₂ planes allows the

TABLE I. Spin singlet OP eigenfunctions in the FS restricted (l) and real space (*n*,*m*) representations, their IR notations, and character table for the orthorhombic point group C_{2v}^1 , appropriate for YBCO.

| IR | OP Eigenfunction | σ_x, σ_y (σ_a, σ_b) |
|---------|--|--|
| A_1 | $ s+d_{x^2-y^2}\rangle$ | $+1$ |
| | $\Rightarrow a_0 + \sqrt{2} \sum_{l=1}^{\infty} a_l \cos(2l\phi_k)$ $\Rightarrow \sum_{n,m=0}^{\infty} a_{nm} \cos(n k_x a) \cos(m k_y b)$ n.m | |
| A_{2} | $ d_{xy} + g_{xy(x^2-y^2)} $ $\Rightarrow \sqrt{2} \sum_{l=1}^{\infty} b_l \sin(2l\phi_k)$ $\Rightarrow \sum_{n=m=1}^{\infty} b_{nm} \sin(nk_x a) \sin(mk_y b)$ n,m | -1 |

TABLE II. Spin singlet OP eigenfunctions in the FS restricted (l) and real space (n,m) representations, their IR notations, and character table for the orthorhombic point group C_{2v}^{13} , appropriate for BSCCO, where $k_{d1}a = \tilde{k}_x + \tilde{k}_y$ and $k_{d2}b = \tilde{k}_x - \tilde{k}_y \cdot \tilde{k}_x$ and \tilde{k}_y are dimensionless coefficients of the appropriate basis vectors of the reciprocal lattice, which point along the Cu-O bond directions.

symmetry operations E , σ_{d2} , and C_2 . The OP eigenfunctions $|s+d_{xy}\rangle$ and $|d_{x^2-y^2}+g_{xy(x^2-y^2)}\rangle$ are represented both ways in Table II. In BSCCO, *s*- and $d_{x^2-y^2}$ -wave OP functions are *incompatible*.

The orthorhombic crystal symmetry requires the interaction for spin singlet pairing to have the form

$$
\lambda(\mathbf{k}, \mathbf{k}') = \sum_{ij} \sum_{\Gamma = A_1}^{A_2} \varphi_{i\Gamma}(\mathbf{k}) \lambda_{ij, \Gamma} \varphi_{j\Gamma}(\mathbf{k}'), \tag{1}
$$

where each φ_i ^G is a basis element from either Table I or II. An OP $\Delta(\mathbf{k}, T) = \sum_{i=1}^{2} \Delta_i(T) \varphi_{iA_i}(\mathbf{k})$ with two incompatible components arises when one element φ_{iA_i} appears from each IR basis A_i , or $\lambda_{ij,\Gamma} = \lambda_{ii,A_i} \delta_{ij}$. The Ginzburg-Landau (GL) free energy \mathcal{F}_0 , from which the *C* numbers $\Delta_i(T)$ can be determined, is

$$
\mathcal{F}_0 = \sum_{i=1}^2 \mathcal{F}_{0i} + \epsilon |\Delta_1 \Delta_2|^2 + \delta \operatorname{Re}(\Delta_1^2 \Delta_2^{*2}),\tag{2}
$$

where $\mathcal{F}_{0i} = \alpha_i |\Delta_i|^2 + \beta_i |\Delta_i|^4$ and $\alpha_i(T) = \alpha_{i0}(T - T_{ci})$. In weak coupling (BCS) theory, the bare transition temperatures T_{ci} are obtained from the λ_{ii,A_i} , and the constants satisfy β_i >0, ϵ > δ >0. Assuming T_{c1} > T_{c2} , the S₁ regime, $T_{c2}^{\leq} \leq T < T_c = T_{c1}$, has $|\Delta_1(T)| \neq |\Delta_2(T)| = 0$. In regime S_2 , $T < T_{c2}^<$, both $|\Delta_1| \neq 0$ and $|\Delta_2| \neq 0$, and the OP is the nodeless $\Delta_1 + i\Delta_2$ state. This *only* occurs below the second phase transition at T_{c2}^{\leq} .

An OP having two compatible components arises when both basis elements $i, j = 1,2$ belong to the *same* IR, A_{α} , where α is either 1 or 2. Then, $\lambda(k, k')$ $=\sum_{i,j=1}^{2} \varphi_{iA_{\alpha}}(\mathbf{k}) \lambda_{ij,A_{\alpha}} \varphi_{jA_{\alpha}}(\mathbf{k}'), \text{ where } \lambda_{ij,A_{\alpha}} = \lambda_{ji,A_{\alpha}}. \text{ Di-}$ agonalizing $\lambda(\mathbf{k}, \mathbf{k}')$, we obtain $\lambda^2(\mathbf{k}, \mathbf{k}')$ $\tilde{\lambda}$ **(k,k')** $= \sum_{i=1}^{2} \tilde{\varphi}_{iA_{\alpha}}(\mathbf{k}) \tilde{\lambda}_{i\dot{i},A_{\alpha}} \tilde{\varphi}_{iA_{\alpha}}(\mathbf{k}')$ and the OP eigenfunction, $\Delta(\mathbf{k}, T) = \sum_{i=1}^{a} \Delta_i(T) \tilde{\varphi}_{iA_\alpha}^a(\mathbf{k})$. The GL free energy \mathcal{F}_1 then acquires a term additional to \mathcal{F}_0 , Eq. (2),

FIG. 1. Plot of $I_c(\phi_0)/I_c(0)$ for the case considered in Ref. 14 of a dominant $d_{x^2-y^2}$ -wave and subdominant d_{xy} -wave OP, the relative amounts varying with layer index away from the twist junction. The parameters for these curves are $T_{c2}/T_c = 0.2$, T_{c2}/T_c $= 0.1304, \epsilon/6\beta = 0.5, \delta/6\beta = 0.1,$ and the curves for $\eta_d = \eta'_d = 0.1$ and $\eta_d = \eta'_d = 0.001$ are indicated. Curves for $t = T/T_c$ $= 0.99, 0.9, 0.5$ are presented.

$$
\mathcal{F}_1 = \mathcal{F}_0 + \text{Re}(\Delta_1 \Delta_2^*) \sum_{i=1}^2 \mu_i |\Delta_i|^2.
$$
 (3)

The compatible OP components mix in *two* ways. First, the original basis functions $\varphi_{iA_{\alpha}}(\mathbf{k})$ mix via the linear transformation which diagonalizes $\lambda(k, k')$. Second, the subdominant OP Δ_2 is coupled to Δ_1 just below $T_c = T_{c1}$ via the μ_1 term. Depending upon the sign of μ_1 , the phases of Δ_2 and Δ_1 differ by 0 or π , and $|\Delta_2(T)| \propto |\Delta_1(T)|^3 \propto (T_c)$ $(T-T)^{3/2}$ just below T_c . This modifies the relative *T* dependences of Δ_1 , Δ_2 in the " $\Delta_1 + \Delta_2$ " state. Both mixings occur *without* a second phase transition. Thus, the **k** dependence of Δ (**k**,*T*) changes smoothly with *T*.¹³

We now consider the case of a high-temperature superconducting (HTSC) Josephson junction formed by twisting bicrystal halves an angle ϕ_0 about the *c* axis, as pictured in Fig. 1 of Ref. 14. For weak tunneling between adjacent layers *n* and *n'*, $J_c^i = |4eT\Sigma_{\omega}\langle f^i(\mathbf{k}-\mathbf{k}')F_n(\mathbf{k}_i)F_{n'}(\mathbf{k}'_i)\rangle_{n\cap n'}|$ for $i = J, S$, where $\mathbf{k}_{S} = \mathbf{k}$, $\mathbf{k}'_{S} = \mathbf{k}'$, but $\mathbf{k}_{J} = \mathbf{k}_{+}$ and $\mathbf{k}'_{J} = \mathbf{k}'_{-}$ are rotated by $\pm \phi_0/2$ about the *c*-axis, respectively. $\langle \cdot \cdot \cdot \rangle$ \cdot $\rangle_{n\cap n'}$ is an integral over the overlapping first BZs, and $f^i(q)$ is the spatial average of the quasiparticle tunneling matrix element squared.¹⁴ For J_c^i to $\mathcal{O}(f^i)$, $F_n = \Delta_n / [\omega^2]$ $+\xi_n^2 + |\Delta_n|^2$, etc., where ω is a Matsubara frequency, and the quasiparticle dispersions ξ_n and OPs Δ_n are *independent* of f^i . In this limit, Bloch's theorem and group theory require each component of the $\Delta_n(\mathbf{k}_i, T)$ and the $\Delta_{n'}(\mathbf{k}'_i, T)$ to lock onto the local Cu-O bond orientation. Thus, $\Delta_n(\mathbf{k},T)$ $=\sum_{j=1}^{2} \Delta_{nj}(T) \varphi_{jA_j}(\mathbf{k})$ for two incompatible OP components on the *n*th layer. Although $f^{i}(\mathbf{q})$ can contain both coherent $(q=0)$ and incoherent (q arbitrary) parts, for purely incoherent tunneling (AB), $f^{i}(\mathbf{q}) = f_0^{i}$, $\langle F_n \rangle = \langle F_{n'} \rangle$, and each J_c^{i} 50, *except* for an *s*-wave OP, projecting out its FS average near T_c .¹²

If the FS were cylindrical and $f^J(q)$ were coherent, then the twist experiments would infer an isotropic *s*-wave OP. However, this scenario is unlikely, since the FS is tight binding.^{8,9,13} The dominant coherent tunneling processes involve quasiparticle states near the FSs on each side of the junction. For both the BCS and most strong coupling models, $\Delta_n=0$, except for a narrow region of the BZ near the FS. Since the intersection of tight-binding FSs twisted $\pm \phi_0/2$ about the *c* axis is vanishingly small, *c*-axis twisting reduces such coherent processes, causing $J_c^J(\phi_0)/J_c^J(0)$ to be largest for $\phi_0 = 0.90^\circ$, even for an isotropic *s*-wave OP. The twist experiments thus imply strongly incoherent tunneling.¹

Previously, we investigated whether in the GL regime tunneling to higher order in the $fⁱ$ might allow a dominant $d_{x^2-y^2}$ -wave OP to twist, by locally mixing in a subdominant *s*- and/or d_{xy} -wave OP component by the proximity effect, thereby compensating for the junction twist. 14 We found that such OP proximity twisting is possible, allowing $J_c^J(\phi_0)$ $\neq 0$ for all ϕ_0 . However, the amplitude of the d_{xy} OP component is very small for $T \geq T_{c2}^<$, and $T_{c2}^<$ must be so low as to be unobservable. Also, the proximity effect locally suppresses the dominant OP component, reducing $J_c^J(\phi_0)$ for $\phi_0 \neq 0$.

The amplitude of the d_{xy} OP component is determined somewhat by the ratio T/T_{c2}^{\lt} , but mostly by the *d*-wave Josephson coupling strengths η_d' and η_d obtained from the $\langle f^i(\mathbf{k} - \mathbf{k}') \varphi_{jA_j}(\mathbf{k}_i) \varphi_{j'A_{j'}}(\mathbf{k}'_i) \rangle_{n \cap n'}$ with $j, j' = 1, 2$ at E_F for $i=J,S$, respectively. The weaker the η_d^j , the lower the *T* at which OP twisting can partially compensate for the junction twist. For $T_{c2}/T_c = 0.2$ and strong ($\eta_d = \eta'_d = 1$) coupling, we found a small but finite $J_c^J(45^\circ)$ at $t = T/T_c = 0.7$. However, reducing η_d' to 0.01 with $\eta_d=1$ dramatically reduced $J_c^J(45^\circ)$ at $t=0.7$.¹⁴

Moreover, since BSCCO is highly anisotropic, $2,3$ we have recalculated $J_c^J(\phi_0)/J_c^J(0)$ for $\eta_d = \eta'_d = 0.1,0.001$, and plotted the results for $t=0.5,0.9,0.99$ in Fig. 1. For $\eta_d=0.1$, these effects are small but clearly visible. For η_d =0.001, the suppression of the dominant $d_{x^2-y^2}$ -wave OP component would be noticeable only very close to T_c . For *T* below the fluctuation regime, $J_c^J(\phi_0)/J_c^J(0) \approx |\cos(2\phi_0)|$. Clearly, *d*-wave OP twisting *cannot* explain the twist experiments.¹

We therefore consider only one OP eigenfunction constructed from *compatible* functions. A full calculation in the real space representation will be presented elsewhere.¹⁶ In the FS restricted representation, $f^J(q)$ contains coherent $|J|^2 \delta^{(2)}(\mathbf{k}-\mathbf{k}')$ and incoherent $\sum_{l=0}^{\infty} f_{l0} \cos[l(\phi_{\mathbf{k}}-\phi_{\mathbf{k}'})]$ parts.^{14,15} For either YBCO or BSCCO, when the OP eigenfunctions belong to the same IR, A_1 or A_2 in Tables I and II, $J_c^J(\phi_0)/J_c^J(0)$ is the same. Just below T_c , the integrals can be performed exactly. Since the single crystal and twist junctions are chemically identical, with identical *c*-axis spacings, we take $|J|^2$ and the f_{l0} to be independent of ϕ_0 .^{[2,11} We define $\eta_0^{\text{inc}} = e m^2 f_{00} / (4 T_c)$ and $\eta_l = f_{l0} / f_{00}$ $1 + C|J|^2/(mf_{00}T_c)$ for $l \ge 0$, where $C = 7\zeta(3)/(2\pi^5)$ and *m* is the planar effective mass. We note that $\eta_2 \propto \eta_d'$ in Fig. 1. We find

$$
J_c^J(\phi_0)/\eta_0^{\text{inc}} = \left| \alpha_0 + \sum_{l=1}^{\infty} \alpha_l \cos(2l\phi_0) \right|, \tag{4}
$$

where for $l \ge 1$, $\alpha_l = \eta_{2l} a_l^2$, $\eta_{2l} b_l^2$ for the A_1 , A_2 states, respectively. The a_l and b_l are the OP eigenfunction expansion

FIG. 2. Plots of $|A + \cos(2\phi_0) + B\cos(4\phi_0)|/|1+A+B|$, along with the *A*,*B* values, of the lowest *l* contributions to $J_c^J(\phi_0)/J_c^J(0)$ in Eq. (4). Upper two curves: A_1 states. Lower three curves: A_2 states.

coefficients in the FS restricted representation in Tables I and II. Most important, $\alpha_0 = a_0^2(1 + \eta_0)$ can be finite for the *A*₁ states, but for the A_2 states, $\alpha_0=0!$

For pure *l*-wave states, $J_c^J(\phi_0)/J_c^J(0) = |\cos(l\phi_0)|$, as shown for $l=0,1,2$ in Fig. 4 of Ref. 1. For an isotropic (*l* =0) *s*-wave state, $J_c^J(\phi_0)/J_c^J(0) = 1$, consistent with the data.¹ The other pure *l*-wave states have some ϕ_0^* at which $J_c^J(\phi_0^*)=0$. For $l=2$, $\phi_0^* = 45^\circ$, and for $l=4$, ϕ_0^* $=22.5^{\circ},67.5^{\circ}$. Including higher *l* compatible components will not change these qualitative results. As shown in Fig. 2, an A_2 state always has some ϕ_0^* at which $J_c^J(\phi_0^*)=0$. If the isotropic component of an A_1 state were nearly negligible, then $J_c^J(\phi_0)/J_c^J(0)$ could also be highly anisotropic. Otherwise, an A_1 state gives the least anisotropic $J_c^J(\phi_0)$ behavior. Thus, the observation $J_c^J(\phi_0)/J_c^S = 1$ at and below T_c in BSCCO is *prima facie* evidence that the dominant OP is $|s|$ $+d_{xy}$, with an isotropic component.¹

In addition, if the tunneling across the twist and intrinsic junctions were equal and purely incoherent, $(f_{l0}\rightarrow 0$ for *l* \neq 0), then the $|d_{x^2-y^2}+g_{xy(x^2-y^2)}\rangle$ state would have $J_c^J(\phi_0) = J_c^S = 0 \forall \phi_0$, and the $|s + d_{xy}\rangle$ state would have $J_c^J(\phi_0)/J_c^S$ = 1, as observed, provided that it had an isotropic *s*-wave component.¹ An example of an OP consistent with many experiments is the ''extended *s*-wave'' OP, $\{[\cos(k_x a) - \cos(k_y a)]^2 + \epsilon^2\}^{1/2}$, which is expandable as a tetragonal *s*-wave function with near nodes for $\epsilon \ll 1$. Although the *c*-axis tunneling in BSCCO must be strongly incoherent, it need not be purely incoherent. If it is not purely incoherent, however, then the OP must be isotropic.

Since the *c*-axis twist and tricrystal experiments are incompatible, $1,6$ we propose an experiment to settle the issue. The new geometry is the tetracrystal ring shown in Fig. 3. A single crystal of BSCCO is cleaved twice into three pieces, shown as dark, medium, and light. The medium piece is cut into two equally thick pieces, which are placed across the dark crystal, forming angles ϕ_{12} and ϕ_{23} . The light crystal is then placed atop the medium ones, forming a straight $(0^{\circ}$ or 180°) angle ϕ_{34} with one of them, and $\phi_{41} = \pm \phi_{31}$ with the other, the sign depending upon ϕ_{34} . Then, the entire triangular ring is fused together.

FIG. 3. Proposed configuration of a *c*-axis version of the tricrystal ring experiment. Dark crystal: bottom. Light crystal: top. Medium shading: equal thickness crystals. Arrows indicate the direction of a given single crystal axis.

One calculates the $I_c(\phi_{ij})$ for each OP eigenfunction. Whenever $I_c(\phi_0^*)=0$, I_c changes sign at ϕ_0^* , and $I_c<0$ implies a π junction. The phase of a single junction can be chosen to minimize the free energy, thereby changing I_c to $|I_c|$ when $I_c < 0$. But, for the tetracrystal, the relative junction phases are fixed by the loop.

The equations for a ring of *ab*-planar junctions also apply

to a ring of c -axis junctions.⁶ The self-inductance L should satisfy the requirement $\beta = LI_c^J/\Phi_0 \ge 1$ with large single crystals, where I_c^J is the minimum junction critical current value.¹⁷ However, for high flux detector sensitivity, the area inside the ring should be sufficiently small. If the *s*-wave OP component dominates, there would be no π junctions in the ring, and integral multiples of the flux quantum $\Phi_0 = hc/2e$ would be trapped inside. However, if the $d_{x^2-y^2}$ OP component dominates, then if all three ϕ_{ij} satisfy $\cos(2\phi_{ij})$ < 0 (as in an equilateral triangle), the ring would contain an odd number of π junctions, trapping half-integral multiples of Φ_0 . Other OP scenarios can be studied by varying the ϕ_{ij} .

Thus, the *c*-axis twist experiments provide strong evidence that the OP is $|s+d_{xy}\rangle$, with a nonvanishing isotropic component, and that the *c*-axis tunneling is strongly incoherent. In addition, either the OP is isotropic or the *c*-axis tunneling is purely incoherent, as in AB.12 We propose a *c*-axis tetracrystal experiment to settle the conflict with the tricrystal experiment.

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