Angular dependence of the Josephson critical current across c-axis twist junctions of layered superconductors

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ABSTRACT

We studied theoretically the c-axis Josephson critical current for bicrystals of high temperature superconductors twisted an angle ϕ_0 about the c-axis with respect to each other. We used the effective Lawrence-Doniach models appropriate for the cases of pure s-wave or $d_{x^2-y^2}$ -wave order parameters, and of a dominant $d_{x^2-y^2}$ -wave order parameter combined with a subdominant one of either s-wave or d_{xy} -wave form, as a function of the temperature and ϕ_0 . Our results demonstrate that this new phase-sensitive experiment can serve as a very useful test of order parameter symmetry. In particular, the recent zero-field experiments in the vicinity of T_c by Li *et al.* on c-axis twist junctions of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ are very difficult to explain with a dominant $d_{x^2-y^2}$ -wave order parameter.

Keywords: superconductivity, Josephson junctions, order parameter symmetry, critical currents

1. INTRODUCTION

Recently, there has been a raging controversy regarding the orbital symmetry of the superconducting order parameter (OP) in the high temperature superconductors (HTS). Many experiments¹⁻³ were interpreted in terms of a $d_{x^2-y^2}$ -wave OP, but many others⁴⁻⁷ were interpreted in terms of a more conventional s-wave OP. In the last year or so, an increasing number of these experiments appear to have been easiest to explain from a predominant $d_{x^2-y^2}$ -wave OP, most likely accompanied by a substantial subdominant s-wave OP.⁸ However, nearly all of the important experiments purporting to provide evidence regarding the orbital symmetry of the OP were performed on the single material $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO). Unfortunately, YBCO is always distinctly orthorhombic, due to the inescapable presence of the conducting CuO chains. Hence, both the s-wave and the $d_{x^2-y^2}$ -wave OPs belong to the same representation of the relevant crystal group C_{2v} , and can mix freely at all temperatures T . Thus, before one becomes too prejudiced by the apparent results on a single material, one ought to examine the available experimental evidence which might be relevant to this question in a different material.

To date, the only other materials for which Josephson junction experiments, which are the most sensitive experiments to determine the phase of the OP, have been performed are the single experiment on $\text{Tl}_2\text{Ba}_2\text{CuO}_{4+\delta}$ (Tl2201)⁹ and a few on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212).¹⁰⁻¹⁴ Until recently, Tl2201 was thought to have a crystal structure that was tetragonal, but very recent neutron diffraction results have determined that most samples of Tl2201 are rather orthorhombic, as is YBCO.¹⁵ Although there has never been any consensus on the actual crystal structure of Bi2212,¹⁶ most samples are thought to be orthorhombic. However, in this case the orthorhombicity is different, with a distortion along the diagonal between the crystal *a*- and *b*-axes.¹⁷ Such a distortion would not lead to the coexistence of a $d_{x^2-y^2}$ -wave OP with either an s-wave or a d_{xy} -wave OP, except below a second phase transition, since these OPs are manifestations of different representations of the crystal group.

Until recently, the only published experiments in Bi2212 relevant to the OP symmetry of which we were aware were made by Josephson tunneling into the *ab*-plane and into a mixed c-axis, *ab*-plane configuration, and by scanning tunneling microscopy (STM) onto the top (c-axis) surface.^{13,18-20} These experiments led to inconsistent conclusions, with STM measurements appearing to give "d-wave-like" results when the tip was above a (nominally semiconducting) BiO layer, but "s-wave-like" results when it was above a (superconducting) CuO_2 layer. While apparent Josephson tunneling into the c-axis did not produce any measureable values of the product $I_c R_n$ of the critical current and

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Figure 1. Left: Sketch of the twisted bicrystal. A single crystal is cleaved in a plane normal to the c -axis, the two cleaved sections are rotated by an angle ϕ_0 with respect to each other, and then fused together as pictured. Right: Cross-section, with the darkly shaded indicating the twist junction between layers 1 and -1 with pair tunneling strength η' , and the other junctions with pair tunneling strength η .

the quasiparticle resistance, Josephson tunneling into the ab -plane gave a very large $I_c R_n$ value, which has recently also been seen consistently in c -axis point contact measurements,²¹ although the direction of the Josephson currents in that latter experiment was unknown. In addition, c -axis tunneling between Bi2212 and doped-Bi2212 sandwich thin films was measured, giving very large $I_c R_n$ values for underdoped samples.²² Such experiments are easiest to explain with an s -wave OP. However, angle-resolved photoemission spectroscopy (ARPES) experiments on Bi2212 have been interpreted as being consistent with an OP of the $d_{x^2-y^2}$ form.²³ We remark that ARPES experiments are insensitive to electronic properties of the sample arising from states physically deeper than about 10 Å from the surface. Although one cannot determine the phase of the OP through ARPES experiments, one would have to conclude from the data that the OP was locked onto the crystal lattice on the top atomic layer normal to the c -axis, provided that the ARPES experiments are indeed measuring the superconducting and not some other OP such as that pertaining to a charge-density wave (CDW) or spin-density wave (SDW).²⁴

Very recently, some preliminary results relevant to the symmetry of the OP in Bi2212 have become available.^{10,11,14} First, c -axis Josephson tunneling between Pb and Bi2212 was finally observed, with a beautiful Fraunhofer diffraction pattern (in the most recent, as yet unpublished, results), and Shapiro steps indicating that the tunneling was of first order only. This demonstrates conclusively that there is indeed an s -wave component to the OP in Bi2212. However, since $I_c R_n$ was 1-2 μ V, the magnitude of the measured s -wave component at 2 K was very small.¹⁴

In addition, a high-quality single crystal of Bi2212 was cleaved mechanically between neighboring BiO layers, the two cleaved surfaces were then rotated by an angle ϕ_0 with respect to each other, and heat treated to fuse them back together, as pictured in Fig. 1.¹⁰ Miraculously, transmission electron microscope (TEM) studies showed that the fused boundary appeared in many samples to be essentially perfect.¹¹ To measure the I_c across the twist boundary, the authors attached two current leads far from the twist, and four voltage leads near to it. Due to the large values of I_c at low T , it was necessary to apply a substantial magnetic field along the c -axis to reduce I_c . The $I_c(\phi_0)$ they obtained were identical to those obtained for single crystal Bi2212, without any twist boundaries, and were essentially independent of ϕ_0 .¹⁰ For T close to the transition temperature T_c it is possible to measure I_c without applying a magnetic field. First results also show no significant variation of I_c with the twist angle ϕ_0 .¹² Very recent zero field measurements with the current leads on the top and bottom of the twisted bicrystal appear to be much more conclusive, as discussed in the following.¹² These observations appear to be incompatible with a dominant $d_{x^2-y^2}$ -wave OP, and are difficult to explain without a *substantial* s -wave OP component in Bi2212.

Unfortunately, the dependence of I_c upon the junction area A and on the current distribution, which could be very inhomogeneous, is presently unknown. However, we expect that these and other potential ambiguities inherent in the present experiments will be removed in the future.¹² We have therefore studied the problem of the critical supercurrent along the c -axis in a highly anisotropic layered superconductor (i.e. Bi2212) with a c -axis twist junction angle ϕ_0 . We restrict our considerations to the temperature regime near T_c , where the experiments can be performed in the absence of a complicating magnetic field, which we neglect. It is our aim to establish whether d -wave pairing can be ruled out conclusively by measuring the supercurrent through a c -axis twist junction. To

Figure 2. Configuration expected across a 45° twist junction for a (a) pure $d_{x^2-y^2}$ -wave OP, (b) a real, mixed dominant $d_{x^2-y^2}$ - and subdominant d_{xy} -wave OP combination, and (c) a real, mixed dominant $d_{x^2-y^2}$ - and subdominant s -wave OP combination. Dark (light) figures represent the spatial form of the model OPs on the lower (upper) half-crystals, respectively. The in-plane axes in the two crystal halves are indicated.

achieve this aim it is necessary to take into consideration admixtures of OPs with different symmetries in such a way that the interpretation of other experiments is not affected. If the pair state, locked onto the crystal lattice, had $d_{x^2-y^2}$ -symmetry, as pictured in Fig. 2a, it is obvious that the supercurrent would vanish for $\phi_0 = 45^\circ$. The presence of a subdominant OP with d_{xy} -symmetry would allow the overall d -wave OP to rotate and thus compensate for the twist, as pictured in Fig. 2b. In addition, a subdominant s -wave OP could dominate the supercurrent because of differences in the tunneling matrix elements. Such a configuration is pictured in Fig. 2c.

2. THE MODEL

The details of this calculation have been presented elsewhere.²⁵ We assume that the system consists of $2N \gg 1$ layers, which are labelled with integers n such that $-N \leq n \leq -1$, $1 \leq n \leq +N$. The two sections $n \geq 1$ and $n \leq -1$ of the crystal are assumed to be rotated relative to one another by an angle ϕ_0 , creating a [001] twist grain boundary between layers $n = 1$ and $n = -1$, as in Fig. 1. For simplicity, we treat each microscopic CuO_2 double layer of Bi2212 as a single conducting (or superconducting) layer. We assume that the charge carriers on these planes are fermions described by the standard Hamiltonian $\mathcal{H}$ of layered superconductors. We assume both coherent and incoherent interlayer tunneling with matrix element $t_{n,n\pm 1}(\mathbf{k} - \mathbf{k}')$, with its incoherent part allowing for a small but finite amount of interlayer tunneling of $d_{x^2-y^2}$ -wave pairs.^{25,26} Assuming the interlayer tunneling process is 'weak', we expand the anomalous Green's function $\mathcal{F}_{n,n}(\mathbf{k}, \omega)$ in powers of t . For coherent tunneling in the bulk of a layered superconductor, it is elementary to include the tunneling to all orders in t ,^{27,28} but when a surface (such as a twist boundary) is introduced, the OPs on each layer are inequivalent, and the Fourier transform technique does not work along the c -axis direction.^{29,30} We thus rely on this expansion technique, although it is only valid for $t \ll T_c$ in the absence of intralayer scattering. We calculate $\mathcal{F}_{n,n}(\mathbf{k}, \omega)$ to order t^2 , which leads to the linear coupling of the OPs on adjacent layers, the Josephson tunneling term in the Lawrence-Doniach model.²⁵

The gap equation is obtained by inserting the anomalous Green's function evaluated to second order in t into the mean field equation for $\Delta_n(\mathbf{k})$. Depending on the range over which the twist grain boundary affects the intralayer OPs, we need to solve a rather large number of coupled two-dimensional nonlinear integral equations. (The total number of layers considered in most of our numerical calculations was 160.) This problem is so formidable that one needs to make some simplifying assumptions to make it amenable to numerical solution.

We first make the standard approximation of expanding about the Fermi Energy E_F . Second, we approximate the pairing interaction λ_n by a sum of separable terms: $\lambda_n(\phi_{\mathbf{k}}, \phi_{\mathbf{k}'}) = \sum_i \lambda_i \varphi_n^{(i)}(\phi_{\mathbf{k}}) \varphi_n^{(i)}(\phi_{\mathbf{k}'})$, where the basis functions $\{\varphi_n^{(i)}(\phi_{\mathbf{k}})\}$ are chosen in accordance with the symmetry of the system. The functions we shall consider are $\varphi_n^{(d1)}(\phi_{\mathbf{k}}) = \sqrt{2} \cos 2\phi_{\mathbf{k}}$, $\varphi_n^{(d2)}(\phi_{\mathbf{k}}) = \sqrt{2} \sin 2\phi_{\mathbf{k}}$, and $\varphi_n^{(s)}(\phi_{\mathbf{k}}) = 1$, corresponding to the $d_{x^2-y^2}$ -wave state, the d_{xy} -wave state, and the s -wave state. Since the OPs are expected to lock onto the lattice, the arguments of the basis functions have to be shifted by $+\phi_0/2$ ($-\phi_0/2$) for positive (negative) values of n on different sides of the junction twisted by the angle ϕ_0 . Otherwise, the basis functions are independent of the layer index. For this type of

pairing interaction, the momentum dependence of the OP is of the form $\Delta_n(\phi_{\mathbf{k}}) = \sum_i \Delta_n^{(i)} \varphi_n^{(i)}(\phi_{\mathbf{k}})$, where the $\Delta_n^{(i)}$ are C -numbers, which we calculate.

For the pure $d_{x^2-y^2}$ -wave state, we then have

$$\Delta_n(\phi_{\mathbf{k}}) = A_n \sqrt{2} \cos(2\phi_{\mathbf{k}} \pm \phi_0), \quad (d) \quad (1)$$

where the $\pm$ refers to opposite sides of the twist. This is pictured for $\phi_0 = 45^\circ$ in Fig. 2a, at which the critical current vanishes.

Next, we consider a combination of states with $d_{x^2-y^2}$ - and d_{xy} -symmetry,

$$\Delta_n(\phi_{\mathbf{k}}) = A_n \sqrt{2} \cos(2\phi_{\mathbf{k}} \pm \phi_0) + B_n \sqrt{2} \sin(2\phi_{\mathbf{k}} \pm \phi_0), \quad (dd) \quad (2)$$

which can also be written as $C_n \sqrt{2} \cos(2\phi_{\mathbf{k}} \pm [\phi_0 - \delta_n])$, provided A_n and B_n are real and B_n changes sign across the twist grain boundary. This situation is pictured for $\phi_0 = 45^\circ$ in Fig. 2b. We see that such a superposition of the two d -wave states will reduce the mismatch between the OPs across the twist grain boundary and thus will lead to an increase in the coupling between layers and hence also in $I^J C$. A complicating feature is that in the presence of two nearly degenerate pairing channels, weak coupling theory predicts a time reversal symmetry breaking state like $A_n \sqrt{2} \cos(2\phi_{\mathbf{k}} \pm \phi_0) + i B_n \sqrt{2} \sin(2\phi_{\mathbf{k}} \pm \phi_0)$ far from the twist. Near the twist and below the transition temperature of the subdominant pair state we would, therefore, expect A_n and B_n to be both complex.

Finally, we consider a combination of states with $d_{x^2-y^2}$ - and s -wave symmetry,

$$\Delta_n(\phi_{\mathbf{k}}) = A_n \sqrt{2} \cos(2\phi_{\mathbf{k}} \pm \phi_0) + B_n, \quad (ds) \quad (3)$$

For real OPs, this case is pictured in Fig. 2c. We only consider two OP components, so we can use A_n to designate the average amplitude of the dominant OP component (taken to be $d_{x^2-y^2}$ -wave) and B_n to designate that of the subdominant OP component, which may be either d_{xy} - or s -wave. Since there appears to be no thermodynamic evidence for a second zero-field phase transition, and hence the presence of two or more OP components in Bi2212, we shall study the variation of the OP components and the Josephson critical current with layer index for several values of the temperature at which the transition to a two-component state takes place, including very low ones.

There is no need to consider the case of a pure s -wave OP. Although very important, this situation does not require any calculations, since the twist junction does not necessarily affect the OP, which is generally independent of n ,²⁹ and the critical current is independent of ϕ_0 . It obviously fits the existing c -axis twist data very well.

Third, we only consider the Ginzburg-Landau regime.

3. LAWRENCE-DONIACH PHENOMENOLOGICAL MODEL

The Ginzburg-Landau theory of superconductivity has been generalized to layered materials by Lawrence and Doniach.³¹ The corresponding equations for multicomponent unconventional pair states in the presence of a twist grain boundary are obtained by expanding the first term in the gap equation for Δ_n to third order in Δ_n while keeping only first order contributions in the terms coupling near-neighbor layers. Linearizing the resulting equations and ignoring contributions from tunneling, one finds the bare transition temperatures T_{ci}^0 . The d -wave transition temperatures are suppressed from their bare values by scattering, leading to modified T_{ci} values. We shall thus treat T_{cA} and T_{cB} as independent parameters. For the Josephson tunneling across neighboring layers other than across the twist junction, we write the relevant coupling strengths as η_d and η_s , which contain identical contributions from coherent tunneling, but different contributions from incoherent tunneling. Across the twist junction, we write the analogous quantities as η'_d and η'_s , allowing for the possibility that tunneling matrix elements representing this process might be different. This possibility arises from the fact that the atomic orbitals entering in a microscopic calculation of the tunneling matrix elements would be rotated with the lattice. Depending on the anisotropy of the orbitals involved, their overlap will thus depend on the twist angle ϕ_0 . In addition to this intrinsic effect the quality of the interface could be different, especially if the samples were treated chemically or physically after cleavage and before fusing together to form the twist junctions. This possibility could be of great experimental importance, and will be discussed further in the conclusions.

In order to introduce dimensionless quantities and to reduce the number of parameters, we normalize all OP components with respect to $|A|$, the amplitude of the dominant OP in the bulk far from the twist junction in the

absence of additional pairing channels, given by $|A|^2 = -2t_A/[3b_0(T)] \approx 2/[3b_0(T_{cA})][1 - (T/T_{cA})]$, where $\frac{3}{2}b_0(T)$ is the strength of the leading non-linear (cubic) term in each of the gap equations. We write $X_n = A_n/|A|$ and $Y_n = B_n/|A|$ for the normalized OP components, and introduce the phenomenological parameters β , δ , ϵ which, in the weak coupling theory developed above, have the values $\beta_d = \frac{3}{2}\beta_s = 1$, $\epsilon_d = \frac{1}{2}\epsilon_s = \frac{2}{3}$, and $\delta_{d,s} = \frac{1}{4}\epsilon_{d,s}$. Different values can result from strong-coupling effects. We shall consider the regime $T_{cA} \geq T \geq 0.5T_{cA}$.

From these equations one can construct a dimensionless Lawrence-Doniach free energy F_{LD} such that variations with respect to each of the OPs and their complex conjugates reproduce the Lawrence-Doniach (gap) equations as well as the equations for all other layer indices. Also, combinations of two d -wave states, Eq. (2), can easily be treated simultaneously with the combination of d - and s -wave states, Eq. (3). We find

$$F_{LD} = \sum_{n=1}^N (F_n + F_{-n}) + \sum_{n=1}^{N-1} (F_{n,n+1} + F_{-n,-n-1}) + F_{-1,+1}, \quad (4)$$

where

$$F_n = -|X_n|^2 - \frac{t_A}{i_A}|Y_n|^2 + \frac{1}{2}|X_n|^4 + \beta_{d,s}\frac{1}{2}|Y_n|^4 + \epsilon_{d,s}|X_n|^2|Y_n|^2 + \delta_{d,s}(X_n^2 Y_n^{*2} + X_n^{*2} Y_n^2), \quad (5)$$

$$-t_A F_{n,n+1} = \eta_d |X_n - X_{n+1}|^2 + \eta_{d,s} |Y_n - Y_{n+1}|^2 \quad (6)$$

$$-t_A F_{-1,+1} = \begin{cases} \eta'_d \left(\left[|X_1 + X_{-1}| \sin \phi_0 - [Y_1 - Y_{-1}] \cos \phi_0 \right]^2 + \right. \\ \left. \left[|X_1 - X_{-1}| \cos \phi_0 + [Y_1 + Y_{-1}] \sin \phi_0 \right]^2 \right) & \text{for } dd \\ \eta'_d \left(|X_1 - X_{-1}|^2 \cos^2 \phi_0 + |X_1 + X_{-1}|^2 \sin^2 \phi_0 \right) + \eta'_s |Y_1 - Y_{-1}|^2 & \text{for } ds, \end{cases} \quad (7)$$

are the terms arising from intralayer pairing, interlayer coupling, and twist grain boundary coupling, respectively.

F_{LD} remains the same if we change ϕ_0 from zero to $\pi/2$, provided the signs of X_n and Y_n for all $n \leq -1$ are changed. The same is true if we replace an arbitrary ϕ_0 by $\pi/2 - \phi_0$. To render $F_{-1,+1}^{(dd)}$ invariant, we also have to change the sign of ϕ_0 taking into account the fact that one is now rotating the sample sections in opposite directions. The effect of a twist grain boundary for tetragonal systems is thus seen to be symmetric with respect to the twist angle $\phi_0^{\max} = \pi/4$. Henceforth, we shall only consider the range $0 \leq \phi_0 \leq \pi/4$.

The free energy describing the twist grain boundary coupling is not symmetric with respect to the interchange of layer indices in the dd -case, unless $X_1 = X_{-1}$ and $Y_1 = Y_{-1}$. This choice, however, does not minimize $F_{-1,+1}$ for $\phi_0 \neq 0$. By analyzing the relevant symmetries, we conclude that the solutions of the Lawrence-Doniach equations for the dd -case have the symmetry $X_{-n} = X_n^*$ and $Y_{-n} = -Y_n^*$ for all $1 \leq n \leq N$. This conclusion can also be reached by studying the Lawrence-Doniach equations for the real and imaginary parts of the OPs, for which the computer code was written, and it is supported by the results of our numerical calculations. For $\pi/2 \geq \phi_0 \geq \pi/4$, the minus sign would appear in the relation for the dominant OP. Using this symmetry, $F_{-1,+1}$ reduces to

$$F_{-1,+1}^{(dd)} = -\frac{4\eta'_d}{t_A} \{ |X'_1 \sin \phi_0 - Y'_1 \cos \phi_0|^2 + |X'_1 \cos \phi_0 + Y'_1 \sin \phi_0|^2 \} \quad (8)$$

where a prime (double prime) denotes the real (imaginary) part. We see that, with $X'_1 = -Y'_1 \tan \phi_0$ and $Y'_1 = -X'_1 \tan \phi_0$, $F_{-1,+1}^{(dd)}$ would vanish for any ϕ_0 . No such cancellation can occur for $F_{-1,+1}^{(ds)}$ so that the only way to reduce this (positive) contribution to the free energy is to suppress the d -wave OP near the twist, while the (isotropic) s -wave OP, unaffected by the presence of the twist, should show the bulk behavior $Y_{-n} = Y_n$.

A complication can be expected in the dd -case at temperatures $T < T_{cB}$ and for $\delta_d < 0$. Then the subdominant OP is real and finite for all twist angles. To minimize $F_{-1,+1}^{(dd)}$ one would need to have the symmetry $Y_1 = Y_{-1}$ for $\phi_0 = 0$ and $Y_1 = -Y_{-1}$ for $\phi_0 = \pi/2$.

4. CRITICAL CURRENT

In the Ginzburg-Landau regime we find for the combination of two d -wave states given in Eq. (2) that the total Josephson current across the twist grain boundary

$$I_0' = 4eN_{2D}(0)\eta'_d \text{Im} \{ (A_1 A_{-1}^* + B_1 B_{-1}^*) \cos 2\phi_0 - (A_1 B_{-1}^* - B_1 A_{-1}^*) \sin 2\phi_0 \} \quad (9)$$

The corresponding result for the combination of a $d_{x^2-y^2}$ -state and an s -wave state given in Eq. (3) reads

$$I_0^J = 4eN_{2D}(0)\text{Im}\{\eta'_d A_1 A_{-1}^* \cos 2\phi_0 + \eta'_s B_1 B_{-1}^*\} \quad (10)$$

The current between any other pair of neighboring layers is formally obtained from these equations by setting $\eta'_d = \eta_d$, $\eta'_s = \eta_s$, $\phi_0 = 0$, and by replacing $(1, -1)$ by any pair $(n, n \pm 1)$ of positive (upper) or negative (lower) integers.

The OP amplitudes A_n and B_n are calculated by minimizing Eq. (4), which describes a large but finite stack of Josephson coupled superconducting layers. Without attaching current leads, no net current can flow perpendicular to the layers so that I_n^J must vanish for all n . This means that the expression inside the braces must be real, even though A_n and B_n may be complex. We can thus interpret the expressions on the right hand sides of Eq. (9) and Eq. (10) without the imaginary part being taken as the *critical Josephson current* I_n^{JC} .

5. BOUNDARY CONDITIONS

The influence of the twist grain boundary should be restricted to some limited number of adjoining layers so that on the extremal layers $n = \pm N$ of a large ($N \gg 1$) but finite stack considered in the numerical calculations, the OPs should attain their bulk values X_∞ and Y_∞ , which are characterized by their independence of the layer index n . If $T_{cA}^0 > T > T_{cB}^0$, only the dominant OP is present. Because of our normalization we have the simple boundary conditions $X_N = X_{-N} = 1$ and $Y_N = Y_{-N} = 0$ for twist angles $\phi_0 \leq \pi/4$.

At sufficiently low temperatures the subdominant OP becomes nonvanishing. From Eq. (5) it is obvious that for $\delta_{d,s} > 0$ the free energy is minimized if the phase difference between X_∞ and Y_∞ is $\pm\pi/2$, corresponding to a pair state with an anisotropic but nodeless energy gap. For $\delta_{d,s} < 0$ the phase difference is zero. Since the overall phase factor is arbitrary we can choose X_∞ real and Y_∞ real or imaginary, depending on the sign of $\delta_{d,s}$. For all the cases under discussion, X_∞ and Y_∞ can be determined by minimizing F_n (F_{LD} with all interlayer coupling terms removed), resulting in $0 = 1 - X_\infty^2 - \gamma|Y_\infty|^2$, $0 = \frac{t_B}{t_A} - \beta|Y_\infty|^2 - \gamma X_\infty^2$, where $\gamma = \epsilon_{d,s} - 2|\delta_{d,s}| = \frac{1}{2}\epsilon_{d,s}$ for weak coupling. The weak coupling expressions for the parameters are given in the paragraph preceding Eq. (4). The equations for X_∞^2 and $|Y_\infty|^2$ are easily solved with the results, $|Y_\infty|^2 = (t_B - \gamma t_A)/[-t_A(\gamma^2 - \beta)]$ and $X_\infty^2 = 1 - \gamma|Y_\infty|^2$.

Y_∞ becomes finite below the transition temperature $T_{cB}^<$, obtained from the condition $t_B - \gamma t_A = 0$, which gives

$$\frac{T_{cB}^<}{T_{cA}} = \frac{T_{cB}}{T_{cA}} \frac{1 - \gamma}{1 - \gamma \frac{T_{cB}}{T_{cA}}} \quad (11)$$

As compared with the bulk transition temperature, $T_{cB}^<$ is reduced by the presence of the dominant OP.

For $\delta_{d,s} > 0$ it would seem natural to use the boundary condition $X_{\pm N} = X_\infty$ and $Y_{\pm N} = i|Y_\infty|$, which is symmetric about the twist. When the Lawrence-Doniach equations for the dd -case are solved with these boundary conditions, one finds a finite Josephson current across the sample, which is unphysical. The source of this problem is that, with a complex d -wave OP locking onto the lattice, there exists a finite average phase difference across the twist grain boundary. The unphysical Josephson current can be made to vanish by compensating for this average phase difference. The correct boundary conditions are, therefore, $X_{\pm N} = X_\infty e^{\pm i\theta_\infty}$ and $Y_{\pm N} = i|Y_\infty| e^{\pm i\theta_\infty}$, where

$$\theta_\infty = -\frac{1}{2} \tan^{-1} \left(\frac{2 \tan 2\phi_0 X_\infty |Y_\infty|}{X_\infty^2 + |Y_\infty|^2} \right). \quad (12)$$

For $\phi_0 = \pi/4$ one finds $\theta_\infty = -\pi/4$. The sign of the phase angle is such that $X_{+N}'' < 0$, in agreement with the discussion below Eq. (8). It is clear from Eq. (10) that no such problems arise for the ds -case Eq. (3), so that we can take the phase $\theta_\infty = 0$ for all twist angles ϕ_0 .

The situation is even more complex in the dd -case with $\delta_d > 0$ for temperatures in the range $T_{cB}^< \leq T \leq T_{cB}$, where $Y_\infty = 0$. It would appear that one possible solution would be to have both X_n and Y_n real for all n . However, since the dominant OP is suppressed near the twist grain boundary, it is less effective in suppressing the subdominant OP. We can thus expect to find a complex subdominant OP for small layer indices. A complex Y_n varying from layer to layer results in a finite Josephson current. This is compensated by a finite imaginary part X_n'' . While $\lim_{n \rightarrow \infty} Y_n = 0$, there is no reason for X_∞'' to vanish, because the phase of the dominant OP on either side of the twist grain boundary is arbitrary. Imposing $X_{\pm N}'' = 0$ has the consequence that X_n'' varies linearly with n when Y_n is already negligible. This, again, results in an unphysical Josephson current, which has to be removed by a judicious choice of the phase for the dominant OP. For $\phi_0 = \pi/4$ one can see from Eq. (9) that $I_1^J = 0$ if we put $\theta_\infty = -\pi/4$. For arbitrary twist angles ϕ_0 there seems to be no analytic result for θ_∞ .

Figure 3. The Josephson critical current [Eq. (9)] with $\eta_d = \eta'_d$ between neighboring pairs of layers with $\phi_0 = 45^\circ$, normalized by its value in the bulk. The layer index # 0 designates the junction formed by the twist grain boundary between layers # - 1 and # + 1, and the layer index # 1 designates the junction between layers # 1 and # 2, etc. Because I_n^{JC} is symmetric, we only show results for junctions on one side of the twist grain boundary. The other parameters are $T_{cB} = 0.2T_{cA}$, $T_{cB}^< = 0.1304T_{cA}$, $\epsilon_d = 0.5$, and $\delta_d = 0.05$.

6. NUMERICAL RESULTS AND DISCUSSION

Probably the most interesting aspect of the present theory is the creation of a subdominant d -wave OP by the strain associated with the twist grain boundary, which would render the Josephson critical current finite even for a 45° -twist. Since the assumed dominant $d_{x^2-y^2}$ -wave OP is forced to be small near the twist, its suppression of the subdominant d_{xy} or s OP component is reduced near the twist from its larger amount in the bulk. Thus, a finite critical current can exist across even a 45° twist for $T > T_{cB}^<$. This process is, however, possible only if the pairing interaction, characterized by a bare transition temperature T_{cB}^0 , is finite. On the other hand, if one assumes that the penetration depth measurements,³² exhibiting linear in- T behaviors at low T , are related to nodes in the superconducting OP, then one would need to have the actual $T_{cB}^<$ very small, indeed.

We shall begin our discussion with choosing $T_{cB}/T_{cA} = 0.2$, which is large enough to make the expected effects clearly visible but small enough to make the observation of a second superconducting transition in the bulk material unlikely. The other parameters are $\eta = \eta' = 1$, $\epsilon_d = 0.5$, and $\delta_d = 0.05$. For these parameters, the actual bulk transition temperature of the subdominant OP $T_{cB}^< = 0.1304T_{cA}$. This value for T_{cB} is well below the temperatures T for which we can trust our Ginzburg-Landau approximation to give reliable results. We shall mainly consider the regime $T > T_{cB}$ for which both OPs are real. In Fig. 3 we show the Josephson critical current Eq. (9) between neighboring pairs of layers. The prefactor $4e N_{2D}(0) \eta_d$ in Eq. (9) has been omitted and the OP amplitudes have been normalized, so that $\lim_{n \rightarrow \infty} I_n^{JC} = 1$ at all temperatures for which the subdominant OP vanishes in the bulk. Since $\phi_0 = 45^\circ$, the finite value of I_0^{JC} is entirely due to the subdominant OP Y_1 . For very low T_{cB} we can estimate Y_1 by setting $t_B = 0$. The normalized I_0^{JC} Eq. (9) reduces to $2X_1Y_1$ and thus depends linearly on the small parameter t_B^{-1} . Even for T_{cB}/T_{cA} as low as 0.001 we find $I_0^{JC} = 0.067$ for $\eta' = 1$ and temperatures low enough (e. g. $0.5 T_{cA}$) such that X_1 is a sizeable fraction ($X_1 = 0.524$ in this case) of its bulk value.

We see from Fig. 3 that, depending on the temperature, the Josephson critical current can be suppressed on a fair number of junctions. Unlike I_0^{JC} , I_n^{JC} for $n \geq 1$ is primarily determined by the dominant OP, which varies substantially with η' . The number of junctions with severely suppressed I^{JC} would thus be smaller for smaller η' . Remember that large values of η and η' are compatible only with coherent tunneling.

Fig. 4 shows the dependence of I_0^{JC} on the twist angle ϕ_0 for several reduced temperatures and $\eta' = 1$ (solid lines). The other parameters are $\eta = 1$, $T_{cB} = 0.2T_{cA}$, $\epsilon_d = 0.5$, and $\delta_d = 0.05$, as in Fig. 3. Also shown is I_0^{JC} as a

Figure 4. The Josephson critical current Eq. (9) as function of the twist angle ϕ_0 , normalized by its value at $\phi_0 = 0^\circ$, for several reduced temperatures and $\eta' = 1$ (solid lines). The other parameters are the same as in Fig. 3. The various dashed lines show the changes in this angular dependence at $t = 0.7$ as η' is lowered.

function of ϕ_0 for several values of η' at $t = 0.7$. The curve for $\eta' = 0.01$ is indistinguishable from $\cos 2\phi_0$, which is the dependence on twist angle expected at any temperature if only the dominant OP, unaffected by the proximity effect, were present.

So far we have not shown how the results depend on the parameters ϵ and δ which describe the strength of coupling between the dominant and the subdominant OPs. The values used actually differ from the weak coupling result, listed above Eq. (4). We did change ϵ to $2/3$ and varied δ between $+\epsilon/4$ and $-\epsilon/4$. Even changing the sign of δ , which would favor a real subdominant OP, has little effect on the results presented so far. The changes are at most a few percent and would be barely visible in these figures.

For the temperatures $T_{cA} > T > T_{cB}^*$ considered so far, the combination Eq. (3) of d - and s -wave OPs only leads to the trivial solution $B_n = 0$ for all n and $I_0^C(\phi_0 = 45^\circ) = 0$. The proximity effect on A_n is the same as in the dd -case discussed above with $T_{cB} = 0$.

The only other really relevant parameter, in addition to η and η' , therefore is the strength of pairing in the subdominant channel, parametrized by T_{cB} . Results for the ds -case with $T_{cB}/T_{cA} = 0.9$ are presented in Fig. 5, which shows the bulk solutions and the solutions on layer #1 together with I_0^C as function of temperature for the maximum twist angle $\phi_0 = 45^\circ$. For this value of T_{cB} we obtain $T_{cB}^* = 0.8571 T_{cA}$. At this temperature the bulk s -wave order parameter Y_∞ sets in with the usual square root behavior while X_∞ drops with a discontinuity in slope.

As in the dd -case with $T > T_{cB}$ it suffices to consider only two OP amplitudes, X_n which is chosen to be real, and Y_n which is real or imaginary depending on the sign of δ_s in Eq. (5). The phase is of no consequence here, since we consider the coupling between layers and the Josephson current only to lowest order in the OP amplitudes. The difference between a time reversal breaking state $d+is$ without nodes in the energy gap and a real combination $d+is$ with nodes can be expected to show up at low temperatures.

For the parameters chosen, X_1 at T_{cB} is reduced through the proximity effect to about half its bulk value. Replacing X_∞ with $X_1 < X_\infty = 1$ in the relations above Eq. (12) shows why the transition temperature for Y_1 is found to be higher than T_{cB}^* . An accurate estimate of this transition temperature is not possible because the coupling to outer layers also serves to suppress Y_1 . It is only in this rather indirect way that the twist grain boundary has an effect on an isotropic subdominant s -wave OP. Note that, unlike X_∞ , X_1 continues to grow with decreasing temperature, even after the onset of Y_1 .

At $\phi_0 = 45^\circ$ the Josephson current is carried solely by the s -wave component of the condensate so that, unlike the dd -case considered above, I_0^C is finite only below temperatures at which Y_1 is finite. At temperatures not very far

Figure 5. Order parameter components on layer #1 and in the bulk for the sd -combination Eq. (3) as function of temperature in the vicinity of the s -wave transition temperature. Also shown (solid line) is $I_0^{JC}(\phi_0)/I_0^{JC}(0)$, Eq. (10). The parameters are: $T_{cB}/T_{cA} = 0.9$, $\beta = 1$, $\eta_d = \eta'_d = \eta_s = \eta'_s = 0.2$, and $\phi_0 = 45^\circ$.

below $T_{cB}^<$, Y_1 and X_1 are comparable in size. Since from our discussion of the tunneling matrix elements we would expect $\eta_s \gg \eta'_d$, the Josephson current would flow primarily in the s -channel at any twist angle so that I_0^{JC} would show little variation with ϕ_0 except very close to $T_{cB}^<$. This explanation for the absence of a strong variation of I_0^{JC} with twist angle implies, however, a substantial deviation in the nodal structure of the OP in the bulk material from that of the $d_{x^2-y^2}$ -state.

Finally, we consider the case of two coexisting d -wave states, Eq. (2). As before, we choose $T_{cB}/T_{cA} = 0.9$ and consider the temperature $T = 0.8 T_{cB} < T_{cB}^< = 0.8571 T_{cA}$. $T_{cB}^<$ is the same as for the ds -case because we use the same parameter values for ϵ and δ . When δ_d in Eq. (5) is negative, the phase difference between the $d_{x^2-y^2}$ - and the d_{xy} -state vanishes. As discussed above, in order to relieve the strain induced by the twist grain boundary the subdominant pair state should be antisymmetric with respect to the layer index. Starting the iteration with a trial solution that has this symmetry, we obtain a convergent result for any twist angle. In the left panel of Fig. 6 we show the purely real solution $Y'_1 = -Y'_{-1}$ for the subdominant state on layer #1 as a long dashed line and the Josephson critical current across the twist junction as a short dashed line. However, in the absence of the twist we expect that a solution with the symmetry $Y'_1 = Y'_{-1}$ would minimize the free energy. Starting with a trial solution that has this symmetry we find convergence only up to twist angles $\phi_0 \approx 25^\circ$. For $\phi_0 \neq 0$ the solution actually has no symmetry, as the two lines representing Y'_1 (solid) and Y'_{-1} (dot dot dashed) show. For equal coupling constants $\eta = \eta'$, the minimum Josephson critical current, shown as a dot dashed line, does not occur at the twist junction but a few layers away from it, the distance depending on ϕ_0 . In this case we compared the free energies to establish which is the correct solution. The result is that the most stable state is the one that has the largest *minimum* Josephson critical current. Thus, at the twist angle at which the short dashed and the dot dashed lines cross, the system undergoes a phase transition during which the OP changes its symmetry with respect to the layer index.

When $\delta_d > 0$, as expected from weak coupling theory, the phase difference between the two d -wave states is $\pi/2$. In order to avoid spurious Josephson currents we have to multiply the bulk solutions with a phase factor Eq. (12). The phase angle θ in Eq. (12) is not quite the correct choice when the boundary conditions are imposed not at infinity but at some finite layer index N . For $N = 80$, θ has to be modified by less than one degree to reduce the Josephson current to below the numerical error. The numerical solutions have the symmetry expected according to that describe above Eq. (8) at all twist angles. At $\phi_0 = 0$, the amplitude of the dominant pair state is real, that of the subdominant state is purely imaginary. As ϕ_0 is increased, a real part Y' and an imaginary part X'' are created that serve to rotate the clover leaf of a d -wave state relative to the lattice to minimize the interlayer coupling energy.

Figure 6. The left panel shows the amplitude $Y'_{\pm 1}$ of the subdominant d -wave order parameter on layers #1 and # - 1 as function of ϕ_0 for the case of a real dd -superposition. The two solutions shown have different symmetries with respect to the layer index. In the right panel, the two d -wave OPs differ in phase by $\pi/2$. The symmetry of these states is given above Eq. (8). $I^J_C(\phi_0)$ is included in both panels. It is normalized to unity at $\phi_0 = 0$ when only a single OP is present. The parameters are $T_{cB} = 0.9 T_{cA}$, $T = 0.8 T_{cA}$, $\beta = 1$, $\epsilon = 1/3$, $\delta = \pm \epsilon/4$, and $\eta = \eta' = 1$.

In the intermediate temperature regime $T_{cB}^< < T < T_{cB}$ the subdominant OP has a finite complex amplitude near the twist grain boundary. To cancel the Josephson current that results from $Y'' \neq 0$ the dominant state must acquire an imaginary part. While Y' and Y'' must go to zero far away from the twist since $Y_\infty = 0$, there is no reason for X'' to vanish in the bulk. When one imposes $X'' = 0$ as a boundary condition, one finds that X'' vanishes linearly at the outermost layer, whatever the number of layers considered may be. The result is again a spurious Josephson current. The correct solution is obtained by multiplying X_∞ with a suitable phase factor, Eq. (12). In this case we do not have an analytic formula to calculate the phase angle. The Josephson critical current for two such nearly degenerate d -wave states is very nearly independent of the twist angle at all temperatures.

7. CONCLUSIONS

If the in-plane OP of high temperature superconductors has d -wave symmetry, the Josephson critical current from one conducting layer to the next is exactly zero^{30,33} unless there is some coherent tunneling,^{27,28} or else there is some momentum dependence to the incoherent tunneling amplitude^{25,26,30,33,34} containing a component with d -wave symmetry. If either one or both of these preconditions is fulfilled, the Josephson critical current I_0^{JC} across a c -axis twist junction varies with twist angle ϕ_0 as $\cos 2\phi_0$ if the pair state has $d_{x^2-y^2}$ -symmetry. Thus, $I_0^{JC}(\phi_0 = 45^\circ) = 0$ for $0 < T \leq T_{cA}$, which is clearly inconsistent with the published data.¹⁰

At intermediate twist angles, I_0^{JC} even falls below $\cos 2\phi_0$ because of the proximity effect, which suppresses the OP on layers close to the twist grain boundary. The size of this suppression, and the number of layers affected, depend on the strength of the Josephson coupling between the layers, which cannot be estimated directly from the normal state resistance. The proximity effect depends via the coherence length strongly on temperature so that changes resulting from variations of the twist angle should be more easily detectable close to T_c . This predicted strong dependence of $I_0^{JC}(\phi_0)$ near to T_c for the $d_{x^2-y^2}$ -wave state is inconsistent with the more recent (unpublished) data.¹²

A nonvanishing Josephson critical current at twist angle $\phi_0 = 45^\circ$ can result if a second pairing channel with d_{xy} - or s -symmetry exists. Even if the d_{xy} -pairing is very weak, so that one would not expect to see a second superconducting transition, we find a finite OP with this symmetry on the layers forming the twist junction at all temperatures at which the dominant OP is nonvanishing. For weak d_{xy} -pairing we would still expect to see a substantial variation of I_0^{JC} with the twist angle. This variation is stronger close to T_c because, as the temperature

is raised, $I_0^{JC}(\phi_0 = 45^\circ)$ will drop much more rapidly than the bulk critical current. This is inconsistent with the recent unpublished zero field data taken near to T_c .¹² If by some miracle the two d -wave states are nearly degenerate, however, the combined state is more or less free to rotate relative to the crystal axes and very little variation of $I_0^{JC}(\phi_0)$ would ensue. Such a state, however, would have a finite and rather wavevector-independent energy gap everywhere on the Fermi surface, which would render it inconsistent with all experiments purporting to give evidence for d -wave superconductivity.

The existence of an s -wave pairing channel has no effect on $I_0^{JC}(\phi_0 = 45^\circ)$ at temperatures above the transition temperature T_{cs} at which this s -wave state would form in the absence of any other pairing. In a narrow temperature range $T_{cs}^< < T < T_{cs}^>$, where $T_{cs}^<$ is the temperature below which a two-component pair state is formed in the bulk, the s -wave component can form near the twist junction and thus render $I_0^{JC}(\phi_0 = 45^\circ)$ finite. If the tunneling is predominantly incoherent, the Josephson coupling between s -wave states is expected to be much stronger than that between d -wave states, unless the quasiparticle momentum is very nearly conserved during the incoherent tunneling process. A small s -wave component, possibly too small to be detected by other means, could thus carry most of the Josephson current, in which case no dependence of I_0^{JC} on the twist angle would be expected. Unless the s - and d -components of the pair state coexist at all temperatures, possibly because the crystal symmetry is monoclinic, or otherwise different than purported, one would expect to see some dramatic change with temperature in $I_0^{JC}(\phi_0)$. Such changes are not seen in the recent (unpublished) data.¹²

It is evident from Fig. 4 that if the dominant OP were the presumed $d_{x^2-y^2}$ -wave OP, decreasing η' , causes strong variations in the angular dependence of the normalized critical current, so that twist junctions with $\phi_0 = 45^\circ$ would have a vanishing critical current for T values down to T_{cB} , the bare transition temperature of the subdominant OP. We remark that η' , the Josephson coupling parameter across the twist junction, can be varied experimentally by chemically reacting the cleaved surfaces prior to forming the twist junction. In addition, since the actual critical current is proportional to η' , weakening the twist junction decreases the critical current for all twist angles ϕ_0 . Note that this would also be the case for a pure s -wave OP, except in that case there would be no dependence upon the twist angle ϕ_0 . Decreasing η' substantially should provide evidence that effects of the twist boundary are indeed observed.

We thus encourage further experiments to be carried out near to T_c with zero applied magnetic field. In principle, one can measure the full $I_c(T)$ for different ϕ_0 values. These are predicted to give the maximum information regarding the symmetry of the superconducting OP and the nature of the tunneling process between layers. A preliminary set of such experiments is currently in progress, and the results will be presented at this meeting.¹² Ideally, one should study groups of samples, each group having the twist junctions deliberately weakened with identical procedures. Furthermore, experiments should be carried out to determine whether the current paths are indeed homogeneous. This could be done by decreasing the area of the junction, and seeing if the critical current scales with the junction area. Also, the placement of the current and voltage leads should be varied. Some of these changes have been made, and the others are planned.¹²

We remark that the very recent experiments of Li *et al.* reported at this conference were performed with $\phi_0 = 0^\circ, 45^\circ$, and 90° .¹² The critical current was measured along the c -axis direction, and the junction area was measured. $I_c(T)$ was measured from T_c down to at least $0.5T_c$. In all samples, no variation with twist angle was seen, and $I_c(T)$ fit the Ambegaokar-Baratoff curve quantitatively.³⁵ Thus, the data appear to be difficult to explain with a purported dominant $d_{x^2-y^2}$ -wave OP in Bi2212. To the contrary, the data are easy to explain with a pure s -wave OP. However, because the crystal symmetry allows mixing of an s -wave with a d_{xy} -wave OP, one cannot rule out the simultaneous presence of a d_{xy} -wave OP, even at or near to T_c .

Finally, we remark that one might wonder if a large s -wave OP component inferred in Bi2212 could be reconciled with the small amplitude of $I_c R_n$ measured in c -axis Pb/Bi2212 Josephson junctions.¹⁴ One possibility is that the 10 Å of Ag in the tunnel junction might greatly reduce the amplitude of the s -wave OP component on the surface from its much larger, bulk value.³⁰ In a fused twist junction, an s -wave OP would behave as if it were in the bulk.

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