

The c -axis transport in naturally-grown $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ cross-whisker junctions

Yu. I. Latyshev,^{1,*} A. P. Orlov,¹ A. M. Nikitina,¹ P. Monceau,² and R. A. Klemm^{3,†}

¹*Institute of Radio-Engineering and Electronics RAS, Mokhavaya 11-7, 101999, Moscow, Russia*

²*Centre des Recherches sur les Très Basses Températures CNRS, Grenoble 38042, BP 166 Cedex 9, France*

³*Department of Physics, University of North Dakota, Grand Forks, ND 58202-7129 USA*

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We studied the c -axis transport of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) cross-whisker junctions formed by annealing “naturally” formed whisker crosses. These frequently appear during growth when the ab -faces of neighboring whiskers come in contact. We obtained Fraunhofer patterns of the cross-junction critical currents in a parallel magnetic field, and found a sharp increase in the quasiparticle tunneling conductance at $eV = 50-60$ mV, indicating high junction quality. For our weak junctions, the interface critical current density is about 3% of the critical current density across the stack of bulk intrinsic junctions, as is the room temperature conductivity, and both are independent of the twist angle, in contrast to most of the data reported on “artificial” cross-whisker junctions [Y. Takano *et al.*, Phys. Rev. B **65**, 140513(B) (2002)]. As a minimum, our results provide strong evidence for incoherent tunneling at least at the interface, and for at least a small s -wave order parameter component in the bulk of Bi2212 for $T \leq T_c$. They are also consistent with the bicrystal twist experiments of Li *et al.* [Phys. Rev. Lett. **83**, 4160 (1999)].

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I. INTRODUCTION

Since the discovery of the first high temperature superconducting compounds (HTSC),¹ there has been a huge amount of activity to understand why this occurs. Although there is nearly universal agreement that the superconductivity arises from the spin-singlet pairing of holes (or possibly electrons, in some cases), there is no agreement as to the mechanism for this pairing. It is also agreed by nearly all workers that the most likely place in the HTSC for this pairing to take place is in the ubiquitous CuO_2 layers. Although many proposed pairing mechanisms varying widely in exoticy (and correspondingly inversely in likelihood) have been suggested, it has so far been exceedingly difficult to eliminate many of them, and it has been especially difficult to obtain incontrovertible evidence in support of only a single possible mechanism. In essence, the wide variety of proposed mechanisms falls into two classes: those in which the pairing interaction is attractive or repulsive in sign. Although there is still no definite method to distinguish these pairing interaction signs, at least it has generally been agreed that important information in this regard can be obtained if there were to be a strong consensus as to the orbital symmetry of the superconducting order parameter (OP). Since the superconducting coherence length ξ is comparable to a few lattice constants at low temperatures T , one would further expect the orbital symmetry of the OP to reflect the underlying point group symmetry of the CuO_2 planes.²

For the tetragonal point group C_{4v} appropriate for some HTSC containing a single CuO_2 layer, the relevant group operations for a spin singlet superconductor are: (a) reflections about the planes normal to the layers containing the directions along the Cu-O bond directions, (b) reflections about the planes normal to the layers con-

taining the diagonals bisecting neighboring Cu-O bond directions, and (c) rotations by 90° about the c -axis. Based upon oddness or evenness about these group operations, there are four OP irreducible representations of C_{4v} , which are denoted s , $d_{x^2-y^2}$, d_{xy} and $g_{xy(x^2-y^2)}$, respectively.² For example, if the pairing interaction were attractive, as in the Bardeen-Cooper-Schrieffer (BCS) model, one would expect the OP to most likely have an orbital symmetry invariant under all of the crystal point group operations, the s -wave OP. Although this OP could be highly anisotropic, and could even change sign at certain points on the Fermi surface, it necessarily has a non-vanishing Fermi surface average. On the other hand, pairing mechanisms based upon a repulsive interaction necessarily lead to a vanishing Fermi surface average, and generally lead to an OP that is consistent with $d_{x^2-y^2}$ -wave orbital symmetry, which changes sign on opposite sides of the diagonals between the Cu-O bond directions and under 90° rotations about the c -axis. For a tetragonal crystal, the OP must have only one of the four symmetries, except below a second phase transition at $T_{c2} < T_c$, for which a mixed OP form such as $d_{x^2-y^2} + is$ can occur.

In orthorhombic $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the b axis parallel to the CuO chain direction is longer than the a axis normal to it, and hence group operations (b) and (c) no longer apply. In this case, the point group is the orthorhombic C_{2v}^1 , for which the only effective group operation is (a), reflections in the mirror planes normal to the layers and containing either of the Cu-O bond directions.² In this case, s - and $d_{x^2-y^2}$ -wave OP's can mix without a second phase transition, as can the d_{xy} - and $g_{xy(x^2-y^2)}$ -wave OP's, although the relative weight of each component might depend upon T . Although, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) is also orthorhombic, the b axis containing the orthorhombic distortion and the periodic lattice distor-

tion is along a diagonal bisecting neighboring Cu-O bond directions, leading to an effective point group C_{2v}^{13} , with only the group operation (b) remaining.² In this case, the relative mixed OP forms in the absence of a second phase transition are either mixtures of s - and d_{xy} -wave OP's, or mixtures of $d_{x^2-y^2}$ - and $g_{xy(x^2-y^2)}$ -wave OP's. Hence, to the extent that the crystal is perfect, a mixture of s - and $d_{x^2-y^2}$ -wave OP's could only occur as a $d_{x^2-y^2} + is$ OP below a second phase transition at $T_{c2} < T_c$.

For the last decade, there has therefore been a raging debate with regards to this s -wave/ d -wave controversy.^{3,4} However, as the HTSC exhibit a non-superconducting pseudogap in addition to this OP,⁵ many experiments cannot distinguish them very well, complicating the analysis. In particular, angle-resolved photoemission spectroscopy (ARPES) and point contact tunneling experiments primarily measure the quasiparticle density of states, and can infer an overall gap in its spectrum, but cannot infer any information about the phase of the OP. Although such experiments can infer that both the pseudogap and the superconducting gap arising from the non-vanishing OP below the superconducting transition temperature T_c can be highly anisotropic, they cannot distinguish if the combined superconducting gap and pseudogap actually vanishes at some positions in the first Brillouin zone, or is just less than the experimental resolution there, and they certainly cannot provide any information as to whether it might change sign there. However, phase-sensitive experiments based upon Josephson junctions are not affected by the pseudogap, and can distinguish a $d_{x^2-y^2}$ -wave OP from a highly anisotropic s -wave OP form, such as an “extended- s ”-wave OP. Recently, scanning tunneling microscopy (STM) with atomic resolution have examined surfaces of Bi2212 cleaved at low T , and determined that a disordered array of pseudogap and superconducting regions of characteristic size $2\xi \approx 3$ nm is stable for long times.⁶ If true, this would suggest that there might not be any preferred underlying symmetry relevant for the OP, so that a mixture of all four OP forms would be possible below T_c . Furthermore, this observation would lend strong support to the notion that the c -axis tunneling across the intrinsic layers in Bi2212 ought to be strongly incoherent.

c -Axis bicrystal twist junctions⁷ and more recently artificial cross-whisker junctions (CWJ's)⁸ have attracted considerable attention because of the possibility of providing phase-sensitive tests of the orbital symmetry of the order parameter (OP) in (Bi2212).^{9,10,11} With incoherent c -axis quasiparticle tunneling, the c -axis critical current density J_c across a junction twisted an angle φ is a constant for s -wave or $\propto |\cos(2\varphi)|$ for d -wave OP's, respectively.^{7,12} For coherent tunneling, an anisotropic Fermi surface causes both OP forms to exhibit a strong, four-fold dependence of $J_c(\varphi)$, but a d -wave OP leads for weak, first-order tunneling to $J_c(45^\circ) = 0$, whereas $J_c(45^\circ) \neq 0$ for an s -wave OP. The vanishing of J_c for a predominantly d -wave OP with weak, first-order coherent tunneling is a consequence of

the fact that J_c must change sign at about 45° , even in the presence of weak orthorhombicity effects. The experimental $J_c(\varphi)$ results are still controversial: in the bicrystal experiments of Li *et al.*,⁷ a constant $J_c(\varphi)$ was found, but in the artificial CWJ experiments of Takano *et al.*,⁸ a strong, non-vanishing four-fold $J_c(\varphi)$ was observed. The quality of the Josephson effects on 45° CWJ's subsequently studied by Takano *et al.* was imperfect.¹³

II. BI2212 NATURALLY-GROWN CROSS-WHISKER JUNCTIONS

Here we report on experiments on a new type of twist junction, naturally grown CWJ's. We found Fraunhofer-like oscillations of the critical currents I_c of our CWJ's in parallel magnetic fields \mathbf{H} that clearly indicate dc Josephson behavior across the interface thickness $\ell \approx 4\text{\AA}$. This suggests that the naturally grown CWJ interface represents a single tunnel junction with a small thickness ℓ . We also found an increase in the quasiparticle tunneling conductivity near $eV = 2\Delta \approx 50 - 60$ mV that is much sharper than for intrinsic stacks of Bi2212 junctions. We also found J_c to be reduced from the bulk value, but independent of φ . These results provide strong evidence for the existence of at least a small s -wave OP component in the bulk of Bi2212.

Bi2212 single crystal whiskers¹⁴ are known to possess a high degree of crystalline order.¹⁵ They grow along the a -axis direction, independent of the crucible or substrate. The thin whiskers (with thicknesses $d < 0.3\mu\text{m}$ and b -axis widths $w \leq 10 - 20\mu\text{m}$) are often free of growth steps, macroscopic defects, and dislocations.¹⁵ That motivated us to use whiskers to fabricate junctions with small twist junction areas. Takano *et al.* prepared their CWJ's by placing one whisker upon a MgO substrate, a second atop its ab face, and annealing them together.^{8,13} They reported $J_c(90^\circ)$ values of their CWJ's comparable to the intrinsic J_c of a Bi2212 junction stack, with a rapid decrease in $J_c(\varphi)$ with decreasing φ , followed by a plateau in $J_c(\varphi)$ for $30^\circ < \varphi < 60^\circ$.^{8,13} However, the $I-V$ characteristics of their junctions revealed multi-branched structures, suggesting that the interfaces themselves consisted of rather ill-defined stacks of about 10 intrinsic junctions.

In order to obtain CWJ's with more definite interface properties, we studied naturally-grown whisker crosses. Many of these form when the ab faces of two whiskers come in contact during their growths,¹⁵ as pictured in Figs. 1a,b. The results of an analysis of 267 natural crosses shown in Fig. 1c reveal a greater abundance of crosses with $\varphi > 20^\circ$, with abundance maxima at 30° , 60° , and 90° . However, these as-grown crosses have quite high interface resistances R of several tens of $\text{k}\Omega$ at 300 K, with semiconducting $R(T)$ behavior, and without any sign of a superconducting transition temperature

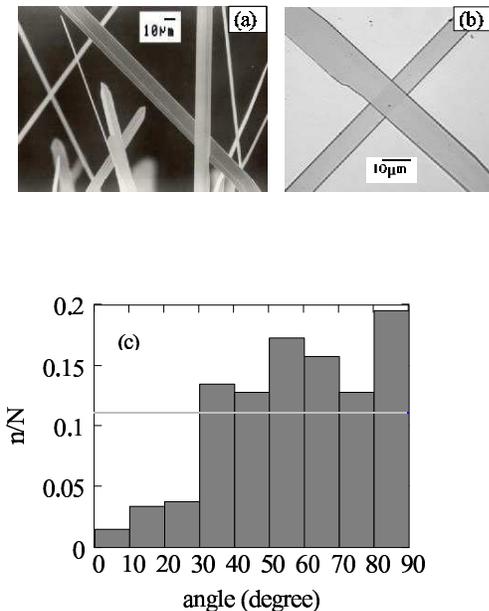


FIG. 1: SEM pictures of (a) a batch of Bi2212 whiskers containing natural crosses, (b) of an individual cross-whisker junction, (c) a histogram of the cross-angle distribution of $N = 267$ natural whisker crosses, where n/N is the relative fraction of the crosses found within different 10° intervals, and the straight line is the weighted average value.

| # | φ (Deg) | S μm^2 | T_{ann} $^\circ\text{C}$ | R Ohm | I_c μA |
|---|--------------------|------------------------|-------------------------------|------------|------------------------|
| 1 | 56 | 309 | 845 | 33.0 | 180.0 |
| 2 | 80 | 138 | 845 | 76.0 | 62.0 |
| 3 | 50 | 183 | 847 | 50.0 | 200.0 |
| 4 | 30 | 201 | 845 | | 80.0 |
| 5 | 70 | 341 | 840 | 25.0 | |
| 6 | 89 | 119 | 840 | 65.0 | 172.0 |
| 7 | 38 | 1696 | 843 | 7.5 | 450.0 |

TABLE I: Natural cross-whisker junction data. φ is the twist angle, S is the junction area, T_{ann} is the annealing temperature, R is the room temperature resistance of the cross-whisker junction, and I_c is the critical current at 4.2 K of the the cross-junction.

T_c . After annealing in flowing oxygen at $\approx 845^\circ\text{C}$ for 20 minutes, $R(300\text{K})$ decreased by 2-3 orders of magnitude and the barrier became transparent to a supercurrent below T_c . Some parameters of seven “natural” CWJ’s selected for study are listed in Table 1. These cross whiskers were grown in the slightly overdoped oxygen stoichiometry regime.

To demonstrate the electrical uniformity of our fabricated CWJ’s, a log-log plot of the four-probe interface

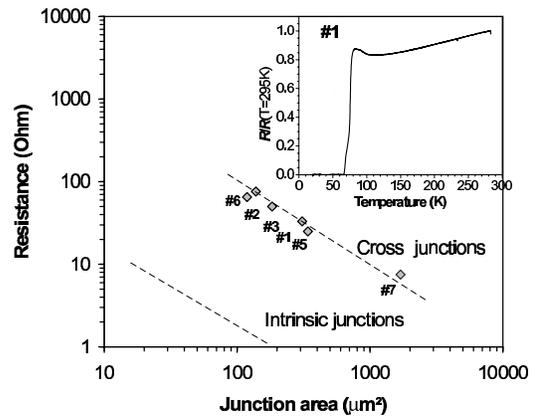


FIG. 2: The dependence of the cross-junction resistance on its area for the junctions listed in Table 1. The inset shows $R(T)$ for a natural cross-whisker junction.

resistance R at 300 K of each sample versus the junction area S determined using a high resolution optical microscope is presented in Fig. 2. Although the R , S , and φ values of our samples varied widely, the junctions we tested were all consistent with the simple formula $R = R_{\square}/S$ expected for an electrically uniform set of junctions with a best fit to the constant interface resistance per unit square $R_{\square} = 10^{-4}\Omega\text{cm}^2$. The consistency of this R_{\square} value suggests that it is independent of φ and that the electrical contact area is consistent with the optically determined S . For comparison, we estimate the resistance per square R_{\square}^{intr} at 300 K for intrinsic c -axis junctions of single crystal Bi2212 by $\rho_c s$, where ρ_c is the c -axis resistivity and s is the spacing between conducting layers. Using the typical values $\rho_c = 10\Omega\text{cm}$ and $s = 1.5$ nm, we estimate $R_{\square}^{intr} = 1.5 \times 10^{-6}\Omega\text{cm}^2 \approx R_{\square}/60$.

III. SUPERCONDUCTING RESULTS

A typical $R(T)$ for a CWJ measured with an ac current $\sim 1\mu\text{A}$ is shown in the inset of Fig. 2. This T dependence is typical of that for $\rho_c(T)$ for slightly overdoped single crystals.¹⁶ Below T_c , the low- T $I - V$ characteristics of CWJ’s pictured in Fig. 3a show a long, linear $I(V)$ quasiparticle branch region of the tunneling type at low bias voltages V , followed by a sharp rise in I at $V_g = 50 - 60$ mV, accompanied by a switch to the normal, resistive state at some current I_{sw} . The current density corresponding to this switch $J_{sw} = I_{sw}/S$ was found to be $\sim 2\text{kA}/\text{cm}^2$ for three CWJ’s studied at high currents. This J_{sw} value corresponds to J_c for the intrinsic junctions in slightly overdoped Bi2212 stacks,^{17,18} suggesting that the switching may be associated with the spreading of the resistive state inside the bulk of the whiskers in contact. In the subgap bias region $V < V_g$, the quasiparticle branch exhibits fine structure

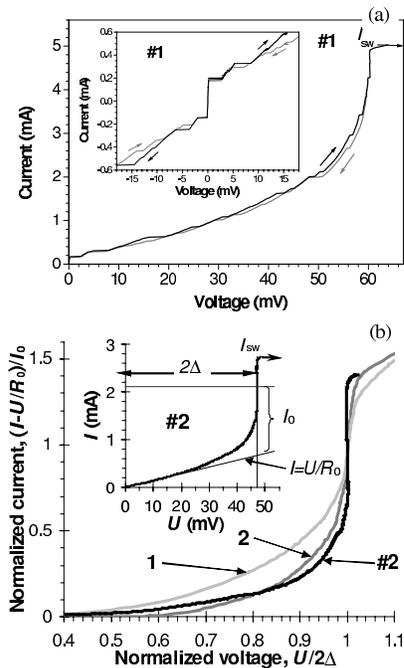


FIG. 3: The I - V characteristics of Bi2212 cross-whisker junctions: (a) for junction #1 at large and small voltage scales; (b) a comparison of the normalized I - V characteristics with the initial linear parts subtracted of Bi2212 junctions of different types: an intrinsic junction within a Bi2212 stack (curve 1),¹⁵ a Bi2212/Dy2278/Bi2212 junction (curve 2),²² and our cross-whisker junction #2, where I_c and the subgap structure are suppressed by a 4T parallel magnetic field. The inset shows the original, unsubtracted I - V curve of sample #2 in the same field. See text.

characterized by 10-20 jumps in V with increasing I which are 1-2 mV in magnitude. Application of an H of several T parallel to the layers removes these V jumps, as shown in Fig. 3b. More details of this fine structure will be published elsewhere.

One of the most remarkable features of natural CWJ's is the very sharp increase in $I(V)$ at $eV \approx 50 - 60$ mV, pictured in Figs. 3a,b, accompanied by a vanishing of the dynamic resistance. This behavior is expected for superconducting-insulating-superconducting (SIS) junctions at $eV = 2\Delta$. Values of $2\Delta = 50 - 60$ mV were obtained both from intrinsic tunneling experiments on slightly overdoped Bi2212 mesas using a pulsed voltage technique to avoid self-heating effects,¹⁹ and from scanning tunneling microscopy (STM) measurements.²⁰ Hence, our CWJ interfaces are also likely to be elementary single junctions with highly suppressed self-heating effects. Experiments on mesas of $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ containing a single intrinsic junction led to the same conclusion.²¹

In Fig. 3b, we compare our CWJ interface I/V char-

acteristics of sample #2 with those of two other sample types, plotting the curves with the linear part of the quasiparticle branch subtracted. The subtracted quasiparticle branch of the intrinsic stacked junction I/V data, curve 1, differs considerably from our subtracted CWJ interface data, and shows a much more smooth increase near $eV = 2\Delta$. In curve 2 we show the earlier data obtained from artificial Bi2212 structures containing a single insulating $\text{Bi}_2\text{Sr}_2\text{Dy}_x\text{Ca}_{7-x}\text{Cu}_8\text{O}_{20+y}$ (Dy2278) layer.²² In that experiment the I - V characteristics also have a subtracted, long linear initial part and a very sharp quasiparticle current increase at $V_g = 50$ mV, corresponding to $eV = 2\Delta$.

Studies of I_c in a parallel magnetic field \mathbf{H} show Fraunhofer-like oscillations,

$$I_c = I_{c0} \left| \frac{\sin x}{x} \right| + I_{c1}, \quad (1)$$

where $x = \pi H w \ell / \Phi_0$, Φ_0 is the flux quantum, and w and ℓ are the in-plane junction width $\perp \mathbf{H}$ and effective junction thickness, respectively. The non-oscillation background part of $I_c(H)$, I_{c1} , was only 10% of $I_c(0)$ for the best junction, #2, but can be as large as 50% of $I_c(0)$ for a lower quality junction such as #6, as shown in Fig. 4. In the intrinsic stacked junction $\ell = c/2 = 15.6\text{\AA}$, one-half the c -axis lattice parameter.¹⁷ Remarkably, for natural CWJ's we reproducibly found $\ell \approx 4\text{\AA}$, about 4 times smaller than $c/2$. That indicates that the CWJ interfaces contain only one insulating layer. It is well known that a regular Bi2212 crystalline structure contains two insulating distances between elementary conducting layers. One is a short distance of 3\AA formed by the Ca layer between single CuO_2 layers. The larger distance of 15.6\AA is associated with the coupling of the cuprate bilayers.¹⁷ That contains two BiO and two SrO layers. If the only parameter involved in the tunneling matrix elements were the junction thickness, then one might infer that the CWJ interface were related to the shorter elementary CuO_2 interlayer distance, and that the interfaces would be the terminating layer of each contacting whisker. However, since the interfaces lead to a factor 60 weaker transparency in the normal state than do the intrinsic junctions, a more likely scenario is that the interfaces uniformly contain some more strongly insulating oxide barrier.

To test the $J_c(\varphi)$ dependence, we measured J_c for 6 natural CWJ's with various twist angles φ (see Table 1). Our data differ significantly from those found for "artificial" crosses studied by Takano *et al.*,^{8,13} as shown in Fig. 5. We defined J_c in three different ways: (1) from $J_c = I_c/S$ (stars), (2) from $J_c = I_{c0}/S$, where I_{c0} is the amplitude of the oscillating $I_c(H)$ defined in Eq. (1) (filled circles), and (3) from the switching current I_{sw} into the resistive state (filled squares). If I_{sw} corresponds to the bulk intrinsic $J_{cb} \approx 2 \times 10^3 \text{ A/cm}^2$,^{17,18} then $J_c = (I_c/I_{sw})J_{cb}$. As seen from Fig. 5, the values of J_c defined in these three different ways are roughly

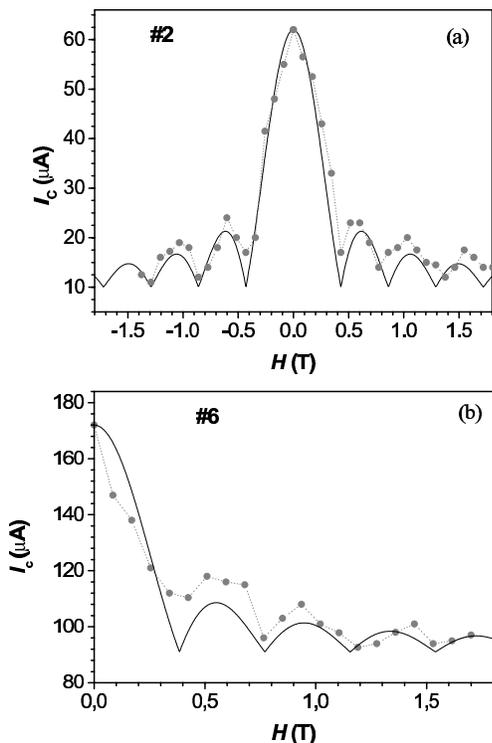


FIG. 4: Dependence of the critical current I_c of cross-junctions #2 (a) and #6 (b) on the parallel magnetic field H at $T = 4.2$ K. The solid lines correspond to the function $I_c = I_{c0}|\sin x/x| + I_{c1}$ with $x = \pi H w \ell / \Phi_0$, where w is the width of the junction and ℓ is the thickness of the junction. For #2 with parameters $\Delta H = 0.39$ T, $w = 13.6 \mu\text{m}$, $\ell = 3.9 \text{ \AA}$, $I_{c0}/I_c = 0.47$.

consistent with each other for each sample. The most reliable data obtained by the second method were available only for samples #2 and #6. However, averaging all of the data for 6 samples shows a φ -independent J_c (dashed line) with the averaged $J_c \approx 50 \text{ A/cm}^2$, a factor 20-40 smaller than J_c for bulk intrinsic junctions.

An angularly independent $J_c(\varphi)$ data set was reported for bicrystal twist junctions.⁷ Those authors measured the same J_c as the interface as in the bulk, and attributed this result to s -wave symmetry of the OP in the crystal bulk.⁷ The experiment was done, however, on large crystals with an in-plane cross section $100\text{-}300 \mu\text{m}$ and with a somewhat reduced critical current density $J_c \sim 200 \text{ A/cm}^2$ at low T .⁷ They did not present convincing evidence for Josephson behavior of the interface. However, as they observed the same J_c values for their twist junctions as for their single crystal junctions at $0.9T_c$, their low- T twist junction J_c values were much larger than our CWJ interface J_c values.

The experiments of Takano *et al.*^{8,13} on artificial CWJ's showed high J_c values of $1.5 \times 10^3 \text{ A/cm}^2$ at $\varphi = 90^\circ$ that decreased with decreasing φ , exhibiting

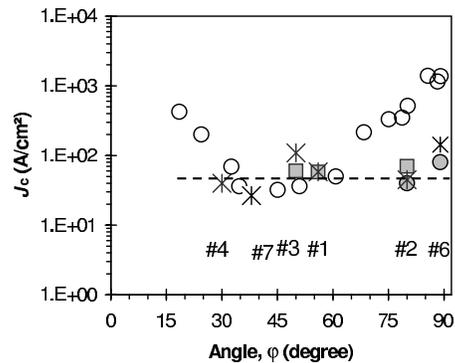


FIG. 5: Dependencies of the critical current density J_c of Bi2212 natural cross-whisker junctions on the twist angle φ . The full symbols and stars correspond to naturally-grown cross-whisker junctions, the different symbols relating to the three different definitions of J_c given in the text. The open circles correspond to the data of Takano *et al.* for “artificial” cross-whisker junctions.¹³

an extended, flat minimum for $30^\circ < \varphi < 60^\circ$. They initially considered that strong φ dependence of $J_c(\varphi)$ to be evidence for a predominant d -wave OP symmetry. However, they subsequently found reproducible (non-vanishing) $J_c(45^\circ)$ values in many artificial CWJ's, and for these $\varphi \approx 45^\circ$ junctions they found Fraunhofer-like $I_c(H)$ patterns with a high background value of $\approx 50\%$ of $I_c(0)$.¹³ For other angles the backgrounds of the Josephson J_c values were not analyzed in this way. Very recently, they also presented Shapiro step data on a 45° artificial CWJ.²³ Combined with the Fraunhofer data, the Shapiro step analysis provided strong evidence that their artificial CWJ's contained only weak, first-order quasiparticle CWJ tunneling. They also showed that the strong $J_c(\varphi)$ they obtained for their artificial CWJ's was independent of T for $5\text{K} \leq T \leq 60\text{K}$. Thus, they concluded that the superconducting gap did not vanish in the bulk, even along the “nodal direction”, for temperatures up to at least 60K .²³

For our natural CWJ's, we found an angularly-independent J_c of about 50 A/cm^2 , 30 times smaller than the Takano *et al.* data for $\varphi = 90^\circ$, but very close to their data for $30^\circ < \varphi < 60^\circ$, as shown in Fig. 5. Our data near $\varphi = 90^\circ$, however, were confirmed by Fraunhofer patterns. Anyway, the J_c values of our junctions are much lower than the intrinsic J_{cb} values measured on mesas fabricated from the same Bi2212 whiskers.¹⁷ The c -axis transport and magneto-transport on those mesas at low temperatures were well described by a d -wave Fermi-liquid model with a significant amount of coherent interlayer tunneling.^{24,25} In that model one would expect to observe a strong four-fold $J_c(\varphi)$ dependence at the interface, which *vanished* at 45° .^{9,26} As one possible qualitative explanation of the reduced and angularly-independent J_c through the interface of

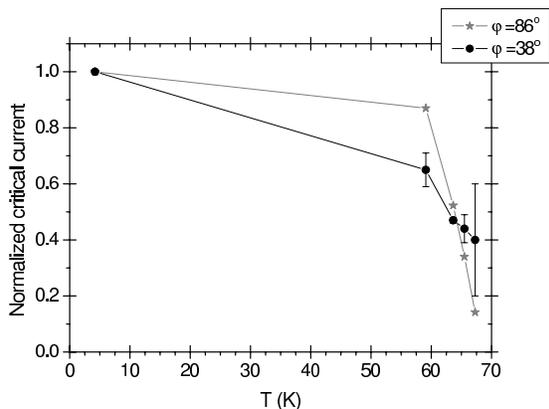


FIG. 6: $J_c(T)/J_c(4.2\text{K})$ for two natural CWJ's with $\varphi = 38^\circ, 86^\circ$.

our natural CWJ's, we suggest that the scattering at the interface of the twist junctions might be highly incoherent due to either the breaking of translational symmetry at the interface, or to junction disorder. The former could impose a mixed order parameter of the $d + is$ type, with a subdominant s -component in the layers near to the interfaces, at least at low T .²⁶ However, such behavior is not expected near to T_c .²⁶ We therefore measured the temperature dependence of J_c for two natural CWJ's with cross-whisker angles 38° and 86° , and the results are presented in Fig. 6. We conclude that there is no qualitative difference in the onset of J_c for these two φ values, arguing strongly against that $d + is$ scenario, as any s -wave component would have to be present at 68K. In addition, translational symmetry breaking would cause the quasiparticles to change their momentum locally in tunneling from one atomic site to another one on the opposite side of the junction, which would be displaced parallel to the junction in real space even for a 90° junction. However, the quasiparticles on each side of the junction have a well-defined wave vectors \mathbf{k} and \mathbf{k}' , respectively, and the ones most likely to contribute to the tunneling have dispersions $\xi(\mathbf{k}) = \epsilon(\mathbf{k}) - E_F$ and $\xi(\mathbf{k}')$ that are small on both sides of the junction. As shown for c -axis twist junctions,^{12,27} for bandwidths consistent with ARPES experiments, it is still possible to have quasielastic coherent tunneling that is only weakly suppressed from that for intrinsic, untwisted junctions for twist angles up to $2\text{-}5^\circ$, regardless of the OP symmetry.¹² Hence, interface imperfections pose a more likely origin for any possible incoherent interface tunneling. However, we do not have any specific experimental evidence to demonstrate conclusively that the interfaces are disordered. In the absence of any such evidence, we have to also consider the possibility that the tunneling between the intrinsic layers of Bi2212 might also be incoherent, consistent with the STM observations of Lang *et al.*⁶

The R_{\square} of our twist junctions is a factor of 60 higher than for the individual intrinsic junctions, in spite of the lower effective barrier thicknesses. However, we note that J_c is only a factor 20-40 lower than for the bulk intrinsic junctions, suggesting that if R were to represent the intrinsic, low- T values of $R_n, I_c R_n$ for our CWJ's would be at least as large as those for intrinsic Bi2212 single crystal junctions. Thus, it seems that a likely explanation for our values of J_c being lower than those obtained from intrinsic bulk junctions is simply due to the weaker tunneling matrix elements, as evidenced by the larger R values of our CWJ's. Because of the presumed strongly incoherent scattering at the interface, any d -wave component to the critical current would be completely suppressed, and the observed small critical current might be due to the remaining s -wave bulk component. This qualitative explanation implies a reduced T_c value of the junction T_{cj} relative to the bulk value T_{c0} .²⁶ In our experiments we observed a reduction of T_{cj} by about 8K below the intrinsic T_{c0} of the whiskers ($T_{c0} = 76\text{K}$), which places a lower limit ($T_{cj} = 68\text{K}$) on the s -wave T_c value. However, this T_{cj} reduction could arise from our annealing process, as unannealed samples were not superconducting. In the Li *et al.* data,⁷ the reduction in T_c from the twist junctions was only about 1K from the bulk values. The fact that our data for J_c are close to the Takano *et al.* data¹³ at $30^\circ < \varphi < 60^\circ$ may be an indication of the presence of an s -wave OP component of the same strength in their cross-whisker junctions as well. On the other hand, both sets of low J_c values could just be due to similar R_{\square} values characteristic of similarly weak tunnel barriers, and that the OP was predominantly s -wave. We remark that the presence of at least a small s -wave component of the OP was also reported at the c -axis interface of Bi2212/Pb Josephson junctions.²⁸

We remark that the sharp increase of the quasiparticle conductivity at $eV = 2\Delta$ may also be a signature of the presence of a rather isotropic s -wave component of the OP in our junctions and in Bi2212/Dy2278/Bi2212 junctions.²² This might suggest that the superconductivity could arise primarily on the saddle bands near the \bar{M} points in the first Brillouin zone, as suggested by Tachiki *et al.*,²⁹ and would appear to be rather constant for either s - or d -wave superconductors. For a substantially d -wave OP with a gap on the regular Fermi surface at the interface, this onset would be expected to be very broad.

IV. CONCLUSIONS

Experiments on naturally-grown and annealed Bi2212 cross-whisker junctions show a small effective interface thickness $\approx 4\text{\AA}$ and a very sharp quasiparticle gap edge in their $I - V$ characteristics, in contrast to intrinsic Joseph-

son junctions in bulk single crystals. We also found that the Josephson critical current density at the interface is significantly reduced from the intrinsic bulk value, and is insensitive to the twist angle. However, this reduction in the critical current density may simply be a consequence of more comparably greatly increased normal state resistance at the interface, which is also independent of the twist angle. As a minimum, we infer incoherent quasiparticle and Josephson tunneling at least at the interface, and the presence of at least a small (3% of the total or greater) *s*-wave component of the order parameter in the bulk of the samples for $T \leq T_{cJ} = 68\text{K}$. Our results on natural CWJ's are also consistent with the the Li *et al.* bicrystal twist experiments.⁷

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- * Electronic address: lat@cplire.ru, yurilatyshev@yahoo.com
 † Electronic address: richard.klemm@und.nodak.edu
- ¹ J. G. Bednorz and K. A. Müller, *Z. Phys. B* **64**, 189 (1986).
 - ² R. A. Klemm, C. T. Rieck, and K. Scharnberg, *Phys. Rev. B* **61**, 5913 (2000).
 - ³ J. R. Kirtley and C. C. Tsuei, *Rev. Mod. Phys.* **72**, 969 (2000).
 - ⁴ K. A. Müller, *Phil. Mag. Lett.* **82**, 279 (2002).
 - ⁵ R. A. Klemm, in K. Morawetz, Ed., *Nonequilibrium Physics at Short Time Scales. Formation of Correlations* (Springer, Berlin, 2004) pp. 381-400.
 - ⁶ K. M. Lang, V. Madhavan, J. E. Hoffman, E. W. Hudson, H. Eisaki, S. Uchida, and J. C. Davis, *Nature (London)* **415**, 412 (2002).
 - ⁷ Q. Li, Y. N. Tsay, M. Suenaga, R. A. Klemm, G. D. Gu, and N. Koshizuka, *Phys. Rev. Lett.* **83**, 4160 (1999).
 - ⁸ Y. Takano, T. Hatano, A. Fukuyo, A. Ishii, M. Ohmori, S. Arisawa, K. Togano, and M. Tachiki, *Phys. Rev. B* **65**, 140513 (2002).
 - ⁹ R. A. Klemm, *Phys. Rev. B* **67**, 174509 (2003).
 - ¹⁰ K. Maki and S. Haas, *Phys. Rev. B* **67**, 020510 (2003).
 - ¹¹ For a recent review, see R. A. Klemm, in *Horizons in Superconductivity Research*, (Nova, New York, 2004).
 - ¹² A. Bille, R. A. Klemm, and K. Scharnberg, *Phys. Rev. B* **64**, 174507 (2001).
 - ¹³ Y. Takano, T. Hatano, M. Ohmori, S. Kawakami, A. Ishii, S. Arisawa, S.-J. Kim, T. Yamashita, K. Togano, and M. Tachiki, *J. Low Temp. Phys.* **131**, 533 (2003).
 - ¹⁴ I. Matsubara, H. Kageyama, H. Tanigawa, T. Ogura, H. Yamashita, and T. Kawai, *Jpn. J. Appl. Phys.* **28**, L1121 (1989).
 - ¹⁵ Yu. I. Latyshev, I. G. Gorlova, A. M. Nikitina, V. U. Antokhina, S. G. Zybtev, N. P. Kukhta, and V. N. Timofeev, *Physica C* **216**, 471 (1993).
 - ¹⁶ T. Watanabe, T. Fujii, and A. Matsuda, *Phys. Rev. Lett.* **79**, 2113 (1997).
 - ¹⁷ Yu. I. Latyshev, S.-J. Kim, V. N. Pavlenko, T. Yamashita, and L. N. Bulaevskii, *Physica C* **362**, 156 (2001).
 - ¹⁸ K. Inomata, T. Kawai, K. Nakajima, S.-J. Kim, and T. Yamashita, *Appl. Phys. Lett.* **82**, 769 (2003).
 - ¹⁹ M. Suzuki, T. Watanabe, and A. Matsuda, *Phys. Rev. Lett.* **82**, 5361 (1999).
 - ²⁰ Ch. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and Ø. Fischer, *Phys. Rev. Lett.* **80**, 149 (1998).
 - ²¹ A. Odagawa, M. Sakai, H. Adachi, and K. Setsune, *Jpn. J. Appl. Phys.* **37**, 486 (1998).
 - ²² I. Bozovic, J. N. Eckstein, and G. F. Virshup, *Physica C* **235-240**, Part 1, 178 (1994).
 - ²³ Y. Takano, T. Hatano, S. Kawakami, M. Ohmori, S. Ikeda, M. Nagao, K. Inomata, K. S. Yun, A. Ishii, A. Tanaka, T. Yamashita, and M. Tachiki, *Physica C* **xxx**, xxx (2004) (in press, doi:10.1016/j.physc.2004.02.091).
 - ²⁴ Yu. I. Latyshev, T. Yamashita, L. N. Bulaevskii, M. J. Graf, A. V. Balatsky, and M. P. Maley, *Phys. Rev. Lett.* **82**, 5345 (1999).
 - ²⁵ N. Morozov, L. Krusin-Elbaum, T. Shibauchi, L. N. Bulaevskii, M. P. Maley, Yu. I. Latyshev, and T. Yamashita, *Phys. Rev. Lett.* **84**, 1784 (2000).
 - ²⁶ R. A. Klemm, C. T. Rieck, and K. Scharnberg, *Phys. Rev. B* **58**, 1051 (1998).
 - ²⁷ G. B. Arnold and R. A. Klemm, *Phys. Rev. B* **62**, 661 (2000).
 - ²⁸ M. Mößle and R. Kleiner, *Phys. Rev. B* **59**, 4486 (1999).
 - ²⁹ M. Tachiki, M. Machida, and T. Egami, *Phys. Rev. B* **67**, 174506 (2003).