

## Theory of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ cross-whisker Josephson junctions

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Takano *et al.* [Phys. Rev. B **65**, 140513 (2002); J. Low Temp. Phys. **131**, 533 (2003)] made Josephson junctions from single crystal whiskers of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  crossed an angle  $\phi_0$  about the  $c$  axis. From the mesa structures that formed at the cross-whisker interface, they inferred a critical current density  $J_c(\phi_0)$ . As with the single crystal results of Li *et al.* [Phys. Rev. Lett. **83**, 4160 (1999)], we show that the whisker data are unlikely to result from a predominantly  $d$ -wave order parameter. However, unlike the single crystals, these results, if correct, require the whisker  $c$ -axis transport to be coherent.

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Recently, there have been a number of phase-sensitive experiments relevant to the orbital symmetry of the superconducting order parameter (OP) in the high transition temperature  $T_c$  superconductor  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212).<sup>1–10</sup> It was claimed that the tricrystal experiment demonstrated a dominant  $d_{x^2-y^2}$ -wave OP component in Bi2212 at low temperature  $T$  for both underdoped and overdoped samples.<sup>1</sup> However, the Pb/Bi2212  $c$ -axis Josephson junction experiments demonstrated in many samples that at least a small  $s$ -wave component was present for  $T$  below the  $T_c$  of Pb (or Nb).<sup>2,3</sup> In the bicrystal  $c$ -axis twist experiment,<sup>4</sup> a dominant  $s$ -wave OP for  $T \leq T_c$  was claimed.<sup>4,11</sup> The superb quality of the junctions was supported by extensive experimental and simulation analyses including high resolution transmission electron microscopy (HRTEM) studies,<sup>5</sup> demonstrating that the junctions were atomically clean over tens of  $\mu\text{m}$  along the junction direction. The twist angle  $\phi_0$  independence of the  $c$ -axis Josephson critical current density  $J_c$  across the twist junction for  $T$  just below  $T_c$  was interpreted in terms of a dominant  $s$ -wave OP for all  $T \leq T_c$ ,<sup>4,11</sup> in apparent contradiction to the results of the tricrystal experiment.<sup>1</sup> However, these experiments would be compatible if Bi2212 were mostly  $s$  wave in the bulk and  $d$  wave on the surface.<sup>12</sup>

Single crystal Bi2212 consists of a stack of intrinsic Josephson junctions, and low- $T$  measurements of the critical current  $I_c$  and the normal resistance  $R_n$  across a single  $c$ -axis junction led to  $I_c R_n$  values  $\approx 1/3$  the Ambegaokar-Baratoff (AB) result.<sup>10,13,14</sup> This is consistent with nonmetallic and incoherent  $c$ -axis transport in Bi2212,<sup>15–17</sup> and an  $s$ -wave OP.<sup>14</sup> Upon intercalation with  $\text{HgBr}_2$ , mesa studies revealed that  $I_c$  and  $R_n$ , respectively, decreased and increased by two orders of magnitude, but their product  $I_c R_n$  remained about  $1/3$  of the AB value.<sup>10</sup> For incoherent transport,  $I_c$  for an  $s$ -wave or  $d$ -wave OP is, respectively, proportional to the  $s$ -wave ( $1/\tau_s$ ) or  $d$ -wave ( $1/\tau_d$ ) interlayer scattering rate, but  $R_n \propto \tau_s$  for both OP's. Hence, the invariance of  $I_c R_n$  upon intercalation is strong evidence for an  $s$ -wave OP.<sup>18</sup>

Recently, Takano *et al.* crossed two single crystal Bi2212 whiskers an angle  $\phi_0$  about the  $c$  axis and fused them together,<sup>7,8</sup> using a technique similar to that of Li *et al.*<sup>4</sup> In addition to the nominal composition of overdoped Bi2212, with  $T_c \approx 80$  K, such whiskers usually have a second transition at  $T_{c2} \approx 105$  K, due to  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$  (Bi2223)

contamination.<sup>6,9</sup> For a  $90^\circ$  cross-whisker junction, a HRTEM picture revealed that this junction was uniform over 100 nm, but the expected periodic lattice distortion was difficult to discern.<sup>9</sup> No HRTEM pictures of other cross-whisker junctions were made.<sup>19</sup>

Remarkably, Takano *et al.* observed branch structures in the current-voltage ( $I$ - $V$ ) characteristics of their cross-whisker junctions at 5 K, indicating that the insulating edge regions of the two whiskers had somehow fused into a mesa structure,<sup>7</sup> consisting of a stack of Josephson junctions.<sup>10,13</sup> By assuming  $I_c$  of the central  $V=0$  branch corresponded to that of the twist junction and that the junction area was equal to the entire whisker overlap, Takano *et al.* inferred a value for the junction  $J_c$  for each of their 10–16 samples.<sup>7,8</sup> In sharp contrast to the single crystal twist experiments,<sup>4</sup>  $J_c(\phi_0)$  obtained in this way for the cross-whisker junctions varied substantially with  $\phi_0$ .<sup>7,8</sup>

Nevertheless, for the  $45^\circ$  cross-whisker junction in a parallel magnetic field, a Fraunhofer-like diffraction pattern consistent with the long-junction limit was observed for their junction with width  $36.7 \mu\text{m}$  and Josephson length  $\lambda_J \approx 3 - 4 \mu\text{m}$ .<sup>8,13</sup> In addition, Shapiro step analysis of the  $45^\circ$  cross-whisker junction indicated that only first-order tunneling is present.<sup>20</sup> If we assume those results are correct, then the data for the crucially important region  $\phi_0 \approx 45^\circ$  arises from weak first-order tunneling only. Since at 5 K,  $I_c(45^\circ) = 0.227$  mA is orders of magnitude larger than the minimum measurable  $I_c$ ,<sup>8</sup> we use a logarithmic scale to fit the  $J_c$  data. Since the overdoped whiskers had  $T_c \approx 80$  K,<sup>8</sup> we take the maximum gap value in our fits to be  $\Delta_0 = 22$  meV, consistent with single crystal point contact tunneling values.<sup>21</sup>

Although  $J_c(45^\circ) \neq 0$ , Takano *et al.* nevertheless claimed that the strong, fourfold periodic  $\phi_0$  dependence of  $J_c(\phi_0)$  was evidence for a “ $d$ -like” OP.<sup>7,8</sup> Here we show that such  $J_c(\phi_0)$  behavior, if correct, is merely a signal of coherent tunneling in a nearly tetragonal crystal with a noncircular Fermi surface. Then, only the behavior of  $J_c(\phi_0)$  for  $\phi_0 \approx 45^\circ$ , where the Josephson tunneling can safely be taken to be weak and first order, is sensitive to the OP symmetry. Quantitative fits to the data of Takano *et al.* are obtained with either a very anisotropic  $s$ -wave gap function on the tight-binding Fermi surface, or a constant gap  $\Delta_0$  limited to the extended Van Hove saddle bands,<sup>22</sup> but the only possibilities of a predominantly  $d$ -wave OP to fit the data are very unlikely.

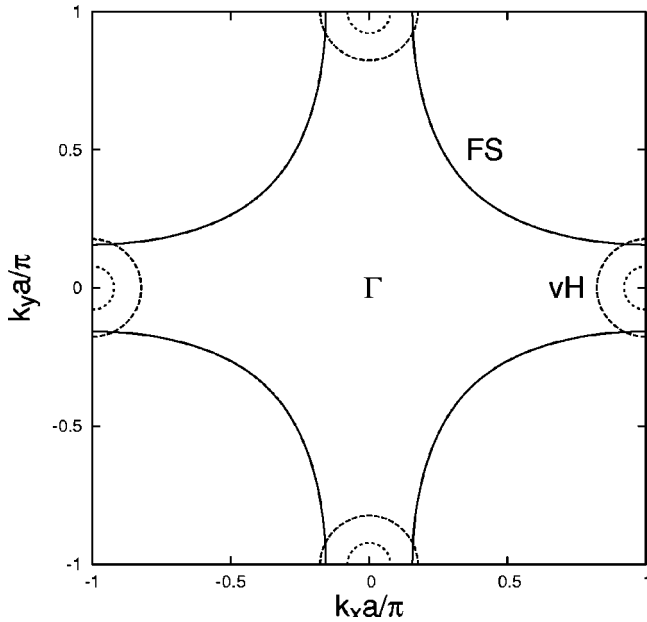


FIG. 1. Quasiparticle excitation features used in the fits. Solid: tight-binding Fermi surface. Dotted and dashed: Van Hove lines of constant energy -25 and -90 meV, respectively, relative to  $E_F$ .

For weak tunneling across a cross-whisker Josephson junction with cross angle  $\phi_0$ ,  $J_c$  is given by<sup>11</sup>

$$J_c(\phi_0) = \left| 4eT \sum_{\omega} \langle f^J(\mathbf{k}, \mathbf{k}') F_{\omega}(\mathbf{k}) F_{\omega}^*(\tilde{\mathbf{k}}') \rangle \right|, \quad (1)$$

where we have set  $\hbar = c = k_B = 1$ ,  $\langle \dots \rangle$  represents the integrations over the first Brillouin zones (BZ's) on each side of the junction,  $F_{\omega}(\mathbf{k}) = \Delta(\mathbf{k}, T) / [\omega^2 + \xi^2(\mathbf{k}) + |\Delta(\mathbf{k}, T)|^2]$  is the anomalous Green function,  $\Delta(\mathbf{k}, T)$  is the OP with wave vector  $\mathbf{k}$ ,  $\xi(\mathbf{k})$  is the quasiparticle dispersion, the  $\omega$  are the Matsubara frequencies, and  $\tilde{\mathbf{k}}$  represents the wavevectors  $\mathbf{k} = (k_x, k_y)$  rotated by  $\phi_0$  about the  $c$  axis.<sup>11</sup> Here we set  $T_c = 80$  and  $T = 9$  K, sufficiently close to 5 K, allowing us to include only 500 Matsubara frequencies in the sum in Eq. (1). For overdoped cross whiskers, the normal state is rather metallic, and the BCS-like model, employed above, with general orbital OP symmetry, is a good approximation.

For strongly incoherent tunneling,  $J_c(\phi_0)/J_c(0) = 1$  for all  $s$ -wave OP forms, and although for all  $d$ -wave forms,  $J_c(0)$  is vanishingly small,  $J_c(\phi_0)/J_c(0) \approx |\cos(2\phi_0)|$ , regardless of the details of the quasiparticle states.<sup>11</sup> Neither form fits the cross-whisker data.<sup>7,8</sup> However, for the quasiparticle states pictured in Fig. 1, a substantial fraction of coherent tunneling necessarily leads to a strong, fourfold  $\phi_0$  dependence to  $J_c(\phi_0)$  for  $\phi_0 \approx 0^\circ, 90^\circ$ ,<sup>11,23</sup> even for an isotropic  $s$ -wave OP, and  $J_c(\phi_0)$  is mainly sensitive to the OP symmetry for  $\phi_0 \approx 45^\circ$ .<sup>11,23</sup> Regardless of the tunneling coherence, for weak tunneling between single component, nearly tetragonal  $d$ -wave superconductors, at some particular  $\phi_0$  value  $\phi_0^* \approx 45^\circ$ , the quantity inside the absolute value signs in Eq. (1) changes sign, and hence  $J_c(\phi_0^*) = 0$ .<sup>11,24</sup>

In order to fit the whisker data, we therefore assume coherent tunneling  $f^J(\mathbf{k}, \mathbf{k}') = f_0 \delta^{(2)}(\mathbf{k} - \mathbf{k}')$ , and take  $\xi(\mathbf{k})$  to

have either the tightbinding form  $\xi(\mathbf{k}) = -t[\cos(k_x a) + \cos(k_y a)] + t' \cos(k_x a) \cos(k_y a) - \mu$ , where  $t = 306$  meV,  $t'/t = 0.90$ , and  $\mu/t = -0.675$ ,<sup>11</sup> or to represent the extended “van Hove” saddle bands,  $\xi(\mathbf{k}) = -t[|\cos(k_x a) - \cos(k_y a)| - \nu]$ , as shown in Fig. 1. Here we take  $t = 500$  meV and  $\nu = 2.02$ , so that the extended van Hove states never cross the Fermi energy  $E_F$ , but lie just below it in the vicinity of the  $\bar{M}$  points at  $(0, \pi/a)$ , etc. This form is a rather crude approximation to the actual van Hove states, but serves to illustrate the effects of a cross-whisker junction rather well. Since both of these quasiparticle state forms are periodic, umklapp processes are automatically included in our calculations.<sup>23</sup> For simplicity of notation, we set  $\phi(\mathbf{k}) \equiv [\cos(k_x a) - \cos(k_y a)]N$ , so the ordinary  $d$ -wave OP is  $\Delta_0 \phi(\mathbf{k})$ , where  $N = 0.5$  for the van Hove case, and  $N = 0.5315$  for the tight-binding case, which lead to the maximum value of the gap  $\Delta_0 = 22$  meV at the respective closest approaches to the  $\bar{M}$  points in the BZ.

A mixed  $d+is$  OP could occur below a second phase transition, provided that the  $bc$  plane containing the periodic lattice distortion is indeed a good mirror plane.<sup>24</sup> However, if the disorder suggested by the STM measurements on cleaved single crystal Bi2212 were present in the Bi2212 whiskers,<sup>17</sup> then there might not be any relevant mirror plane, and a mixed  $d \pm s$  type of OP could occur without a second phase transition. Hence, we considered both OP forms. A  $d_{x^2-y^2} + id_{xy}$  OP is inconsistent with both the single crystal twist experiment and with the Pb(Nb)/Bi2212 Josephson junction experiments,<sup>2-4</sup> with consequences very similar to those of the  $d_{x^2-y^2} + is$  state.

The  $d+is$  scenario might at first sight appear to be consistent with the observation of an effect claimed to be due to spontaneous time-reversal symmetry breaking below the pseudogap onset in underdoped Bi2212.<sup>25</sup> However, in the overdoped regime of the cross-junction experiments, the angle-resolved photoemission spectroscopy (ARPES) measurements were only made above  $T_c$ , and the effect was not observed.<sup>25</sup> There is very strong evidence that the pseudogap exists above  $T_c$  for all Bi2212 dopings, and that it is distinct from the superconducting gap.<sup>17,26-28</sup> Whatever the source of the effect, if it were simply a property of the pseudogap, it should have been seen for all Bi2212 dopings. Since the effect was only seen in the underdoped regime, where Bi2212 is known to be very strongly disordered,<sup>17</sup> the effect itself might be a combined property of the pseudogap and the disorder. In any event, it is unlikely to be relevant to the superconducting OP in the overdoped regime.

In Fig. 2, we show our results for the best  $d$ -wave fits to the data, assuming the tightbinding  $\xi(\mathbf{k})$  form. In this and subsequent figures, we assumed coherent  $c$ -axis tunneling  $T = 9$  K and took the value  $\langle J_c(90^\circ) \rangle$  to be the average of the three data points in that vicinity. We then fit the data to the ordinary  $d_{x^2-y^2}$ -wave form  $\Delta_0 \phi(\mathbf{k})$ , a cubed  $d$ -wave OP  $\Delta_0 \phi^3(\mathbf{k})$ , as suggested by recent ARPES measurements on underdoped  $(\text{Pb}, \text{Bi})_2 \text{Sr}_2 \text{CaCu}_2 \text{O}_{8+\delta}$ ,<sup>29</sup> a single-domain time-reversal symmetry broken  $d+is$  state  $\Delta_0[\phi(\mathbf{k}) + i\epsilon]/(1 + \epsilon^2)^{1/2}$  and a single-domain  $d \pm s$  state  $\Delta_0[\phi(\mathbf{k}) \pm \epsilon]/(1 + |\epsilon|)$ . For the  $d \pm s$  state, the best fit was for  $\epsilon$

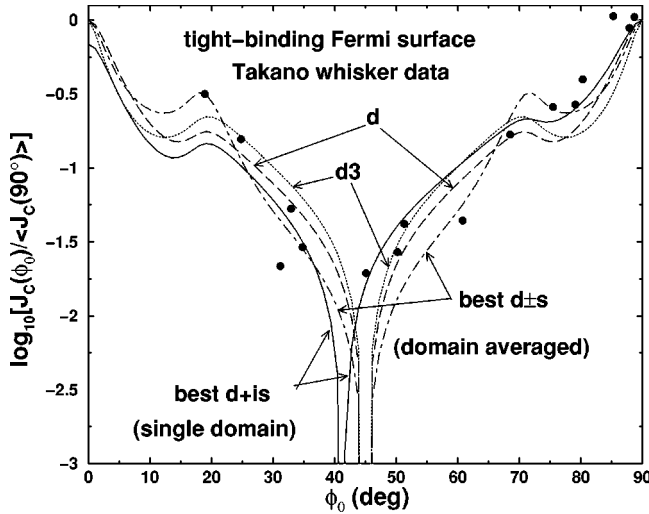


FIG. 2. Plot of  $\log_{10}[J_c(\phi_0)/\langle J_c(90^\circ) \rangle]$  versus  $\phi_0$  obtained from Ref. 8 (solid circles). Also shown are the fits at 9 K assuming coherent  $c$ -axis tunneling, the tight-binding  $\xi(\mathbf{k})$ , and OP's of the ordinary  $d$ -wave ( $d$ , long-dashed) and  $d$ -wave cubed ( $d3$ , dotted) forms, along with the best  $d+is$  single domain fit  $\epsilon=0.15$  (solid) and the domain-averaged result of the best single domain  $d\pm s$  fit (dot-dashed). See the text.

$=0.25$ . Over the region of available data, this curve was nearly indistinguishable from the  $d+is$  curve with  $\epsilon=0.15$  shown in Fig. 2. In Fig. 2, we also show the result of a multidomain average of the  $d+s$ ,  $d-s$  domains with  $\epsilon=0.25$ , which includes the identical contributions of  $d\pm s|d\pm s$  domains across the cross-whisker junction, and the inequivalent  $d\pm s|d\mp s$  domain contributions.

The best single domain  $d+is$  and  $d\pm s$  fits straddled the dataless region near to  $40^\circ$ , and hence could be consistent with the data for  $0.1 \leq \epsilon \leq 0.25$ . However, this time-reversal symmetry breaking  $d+is$  state has a vanishing  $J_c(\phi_0)$  in the dataless regime in the vicinity of  $\phi_0=40^\circ$ , but not also at the crystallographically identical cross-whisker angle  $50^\circ$ , for which nonvanishing data were available. Hence, this state would require a single  $d+is$  domain, in apparent contradiction with STM studies.<sup>17</sup> For a mixed  $d+is$  or  $d\pm s$  state with multiple domains, averaging over the domains always leads to  $J_c(45^\circ)=0$  for  $\epsilon < 1$ , as pictured in Fig. 2. We note that averaging over multiple  $d+is$  domains with the optimal  $\epsilon=0.15$  results in a curve that is nearly indistinguishable over the region of available data from the  $d$ -cubed state plotted in Fig. 2.

In Fig. 3, we show the best  $s$ -wave fits using the same tight-binding  $\xi(\mathbf{k})$ . Here we show fits to the isotropic  $s$ -wave OP  $\Delta_0$ , the extended- $s$ -wave OP  $\Delta_0|\phi(\mathbf{k})|$ , and several values of the highly anisotropic- $s$ -wave OP  $(\Delta_0 - \Delta_c)\phi^8(\mathbf{k}) + \Delta_c$  (indicated in Fig. 3 by “ext8+c-s”) for  $\Delta_c=0, 1, 1.2, 1.4$ , and  $2$  meV, as indicated. The best fits are for  $\Delta_c \approx 1.2 - 1.4$  meV, but  $1 \leq \Delta_c \leq 2$  meV is acceptable. These values are compatible with the resolution of recent ARPES experiments.<sup>29</sup> The flat  $\mathbf{k}$  dispersion away from the minimum gap position on the tight-binding Fermi surface is also compatible with the ARPES data. We note that ARPES data are complicated by the nonsuperconducting pseudogap, which

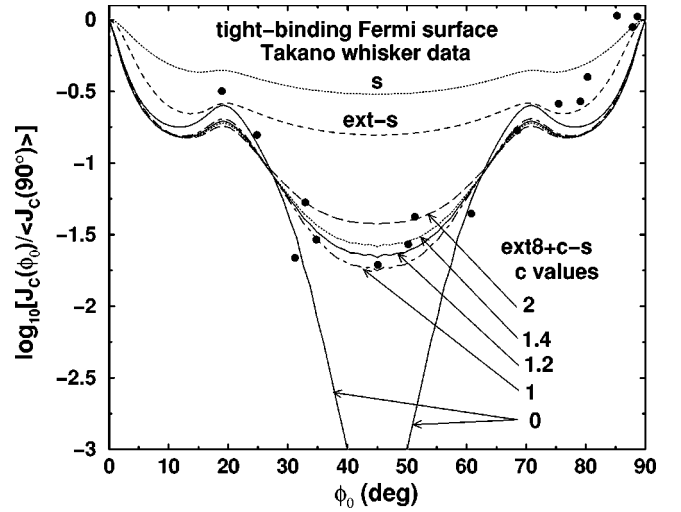


FIG. 3. Plot of  $\log_{10}[J_c(\phi_0)/\langle J_c(90^\circ) \rangle]$  versus  $\phi_0$  obtained from Ref. 8 (solid circles). Also shown are the fits obtained at 9 K with the tight-binding  $\xi(\mathbf{k})$ , and OP's of the isotropic  $s$ -wave (“ $s$ ,” dotted) and extended- $s$ -wave (“ext- $s$ ,” dashed) forms, and of the anisotropic “ext8+c- $s$ ,”  $(\Delta_0 - \Delta_c)\phi^8(\mathbf{k}) + \Delta_c$ , forms with the  $\Delta_c$  values in meV of 2 (long-dashed), 1.4 (dotted), 1.2 (solid), 1 (dot-dashed), and 0 (solid), as indicated.

appears at  $T^* > T_c$  for all Bi2212 dopings,<sup>26,27</sup> so that the superconducting gap at each  $\mathbf{k}$  of observation is merely constrained to be less than or equal to the gap observed by ARPES. We note that the curves in Figs. 2 and 3 for the ordinary  $d$ , ordinary  $s$ , and extended- $s$ -wave OP's differ slightly from those presented previously,<sup>11</sup> since those curves were calculated just below  $T_c$ , and these results are for  $9 \text{ K} \ll T_c$ .

Finally, we present our fits relevant to the van Hove scenario. Here we adjusted the bandwidth  $t$  and the maximum of the saddle bands  $-t\nu$  relative to  $E_F$  to obtain the best fit for the ordinary  $s$ - and  $d$ -wave OP's. The values shown here,  $t=500$  meV and  $\nu=2.02$ , are intermediate to both optimal values. In addition, we showed the calculations for the single domain  $d+is$  (or  $s+id$ ) state  $\Delta_0[\phi(\mathbf{k}) + i\epsilon]/(1 + \epsilon^2)^{1/2}$  for  $\epsilon=0.75, 1$ , and  $2$ , respectively. For  $\epsilon=1$ , we also showed the results for the single domain  $s\pm d$  state  $\Delta_0[\phi(\mathbf{k}) \pm \epsilon]/(1 + |\epsilon|)$ . We note that the ordinary  $d$ -wave curve and the three curves with  $\epsilon=0.75, 1$  are inconsistent with the data, but the curve with  $\epsilon=2$  is consistent with the data. However, this (predominantly  $s$  wave)  $d+is$  state exhibits a strong amount of time-reversal symmetry breaking, and is hence unlikely to be compatible with a variety of other experiments, as noted above.

We remark that the highly anisotropic OP “ext8+c- $s$ ” used phenomenologically in Fig. 3 to fit the data could arise from a van Hove scenario, in which the dominant pairing occurs over the van Hove bands pictured in Fig. 1, and appears on the tight-binding Fermi surface by weak coupling of the electronic states, as discussed elsewhere.<sup>22,30</sup> We note that a good fit could be obtained within the van Hove scenario using an isotropic  $s$ -wave OP, as shown in Fig. 4, so the physics of the generalized  $s$ -wave OP's in Figs. 3 and 4 need not be substantially different or exotic.



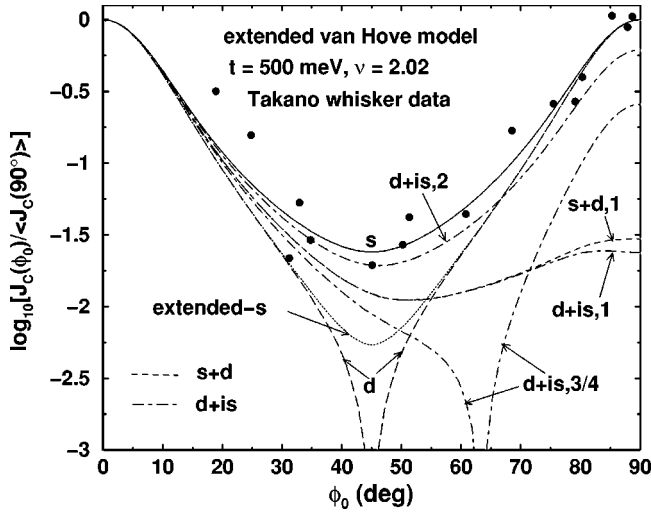


FIG. 4. Plot of  $\log_{10}[J_c(\phi_0)/\langle J_c(90^\circ) \rangle]$  versus  $\phi_0$  obtained from Ref. 8 (solid circles). Also shown are the fits obtained from the extended van Hove  $\xi(\mathbf{k})$ , for OP's of the ordinary  $d$ -wave (long dashed), ordinary  $s$ -wave (solid), extended- $s$ -wave, (dotted), and the  $d+is$  forms (dot dashed) for the  $\epsilon$  values indicated. For  $\epsilon = 1$ , results for the  $s+d$  state (short dashed) are also shown.

Since this work was submitted for publication, it came to our attention that evidence exists that might cause one to suspect that some of the  $J_c(\phi_0)$  values reported by Takano *et al.* might not represent the intrinsic  $J_c(\phi_0)$  of the cross-whisker junctions.<sup>9</sup> For the same  $90^\circ$ ,  $75^\circ$ , and  $60^\circ$  cross-whisker junctions for which the mesa branchings were shown,<sup>7</sup> the measured resistances  $R$  in the  $T$  region  $70$ – $75$  K  $\approx T_c \leq T \leq 105$  K revealed significant complications, suggestive of the presence of more superconducting Bi2223 at the  $90^\circ$  cross-whisker interface than for the  $75^\circ$  cross whisker, and what might be evidence for a nonsuperconducting or possibly even *insulating* barrier at the junction of the  $60^\circ$  cross whisker.<sup>9</sup> Extensive studies of Bi2212 mesas cut from single crystals established that the central  $V=0$  branch, with the smallest  $I_c$ , is frequently associated with the junction

closest to the current lead, and hence furthest from the mesa center.<sup>13</sup> In addition, the branch with the lowest  $I_c$  corresponds to the true critical current of the stack.<sup>13</sup> These properties might lead to an overestimate of  $J_c(90^\circ)$ , and an underestimation of  $J_c(60^\circ)$ , for example. Hence it is possible that the intrinsic  $\phi_0$  dependence of  $J_c$  might be weaker than reported.

In summary, we have shown that the recent cross-whisker Josephson junction results of Takano *et al.*, while different in detail from the single crystal results of Li *et al.*, also render a predominantly  $d$ -wave OP form unlikely. As for a mixed  $s$ - and  $d$ -wave OP, there is a narrow window of 10–25%  $s$  wave that would be allowed in a particular single domain of mixed OP's, but otherwise, it appears that one is forced to accept the result that the dominant OP is indeed  $s$ -wave, although it could be highly anisotropic. Although the results are compatible with an isotropic OP on an extended van Hove saddle band of quasiparticle states, if the pairing were to take place mainly on the tightbinding Fermi surface, the  $s$ -wave gap functions would have to be highly anisotropic, with a minimum value in the range 1–2 meV, consistent with ARPES experiments.<sup>29</sup>

In order to strengthen these conclusions, we urge that additional data points in the range  $30^\circ \leq \phi_0 \leq 60^\circ$  be taken, and that the temperature of the measurement be raised up near to  $T_c$ .<sup>24</sup> A few more junctions with  $\phi_0 \approx 45^\circ$  are currently under study, and the preliminary results appear to be consistent with the above data.<sup>19</sup> We would also like to see other measurements to investigate if the quasiparticle  $c$ -axis transport is indeed coherent, unlike Bi2212 single crystals. We also urge that resistivity measurements between  $T_c$  and 105 K be made on cross-whisker junctions with  $\phi_0 \approx 45^\circ$ , and that alternative fabrication procedures be investigated, in order to guarantee their uniformity.

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