Systematic study of anisotropic Josephson coupling between $YBa_2Cu_3O_{7-x}$ **and PbIn using in-plane aligned** *a***-axis films**

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(Received 30 March 1998)

We have fabricated YBa₂Cu₃O_{7-*x*} (YBCO)/Au/Ag/PbIn edge junctions using in-plane aligned *a*-axis oriented films in order to study the anisotropic coupling between YBCO and PbIn. Seven junctions were made in different directions spanning the *b*-*c* plane of YBCO on each chip. Current-voltage characteristics show a systematic change as a function of the angle of the junction direction relative to the YBCO crystal axes. We analyze the results using a superconductor–insulator–normal-metal interface model with anisotropic YBCO/Au interface resistance and discuss implications for measuring the out-of-CuO₂ plane order parameter of YBCO. [S0163-1829(99)01410-1]

Previously, researchers have studied the anisotropic coupling between $YBa_2Cu_3O_{7-x}$ (YBCO) and Nb or Pb in vertical superconductor-normal-metal-superconductor (SNS') geometries using YBCO thin films of various orientations including (001) , (100) , (103) , and (110) with noble metals as the N layer.¹ In these early studies the consensus was that there was no coupling in the *c* direction of YBCO. Even when critical currents were observed in junctions with *c*-axis oriented films, $²$ the common interpretation was that the cou-</sup> pling is in the lateral $(CuO₂)$ plane) direction through lattice steps resulting from screw dislocations or other defects.³

This interpretation was consistent with overwhelming evidence from experiments sensitive to the phase of the order parameter, which show the existence of $d_{x^2-y^2}$ pairing symmetry in YBCO.⁴ Conventional metallic superconductors such as Nb and Pb have isotropic *s*-wave symmetry. When a pure *s*-wave superconductor is brought to contact with a pure *d*-wave superconductor, the symmetry dictates that Josephson coupling should disappear only in specific directions; along the k_z direction, one does not expect to observe any Josephson coupling.

In direct contrast to these results are some recent tunneling studies into the c direction of YBCO.⁵ In single crystals and films, using Pb as the counterelectrode, clear Fraunhofer diffraction patterns have been observed indicating that the current flow is uniform across the junction in the *c* direction. This has been attributed to existence of mixed *d*- and *s*-wave pairing in YBCO arising from its orthorhombicity.⁶ But it remains to be explained why such results are observed in heavily twinned samples where contribution from the *s*-wave order parameter should cancel (for order parameter components $\Delta_d > \Delta_s$).⁷

While many experiments have probed the in-plane anisotropic order parameter symmetry of YBCO, 4.8 it is also of great interest to investigate the order parameter in the out-of- $CuO₂$ -plane direction. In this paper, we report on the experimental attempt to systematically probe the in- $CuO₂$ -plane versus out-of- $CuO₂$ -plane coupling of YBCO with a conventional superconductor PbIn $(5 \text{ wt. } %1)$ In.

The simplest three-dimensional form of a $d_{x^2-y^2}$ state contains a $\cos^2 \theta$ dependence of the order parameter in the k_z direction⁹ (where θ is measured from the plane). This is shown in the upper inset of Fig. 1 (solid line). But the highly two-dimensional nature of the cuprates might lead to a distortion of the order parameter in the k_z direction resulting in a faster suppression of the order parameter as θ increases. The dashed line in the upper inset of Fig. 1 shows, for example, the order parameter in the k_z direction with compression by a factor of 3. Some cuprates such as the Bi-Sr-Ca-Cu-O compounds are more two dimensional than others, and it is expected that this is reflected in the order parameter in the out-of-CuO₂ plane (k_z) direction. Also, orthorhombicity in YBCO might provide an additional θ dependence depending on the amplitude ratio of d and s components.⁷

FIG. 1. Current-voltage characteristics of seven *a*-axis $YBa_2Cu_3O_{7-x}/Au/Ag/PbIn$ edge junctions facing different directions on a single chip at 4.2 K. Note the systematic decrease in the critical current and increase in the junction resistance as a function of the junction direction. The upper inset is the *d*-wave superconducting order parameter in momentum space in the (k_z, k_y) plane. The solid line is the isotropic *d*-wave, and the dashed line is the *d*-wave compressed in the k_z direction by a factor of 3. The lower inset is a schematic (top view) of the chip. Junction directions span the *b*-*c* plane, 15° away from each other. The top surface of $YBa₂Cu₃O_{7-x}$ is covered with SiO₂ and SrTiO₃.

In studying in-plane versus out-of-plane properties of YBCO, it is necessary to compare results from samples of different orientations. Although single crystals provide good surfaces for fabricating junctions, it is difficult to create multiple angle junctions on a single sample. Since junctions made with YBCO in general suffer from a certain degree of irreproducibility and device parameter spread, it is desirable to directly compare different junctions made in different directions on a single chip. To do this we used in-plane aligned *a*-axis oriented YBCO films to fabricate edge junctions in different directions relative to the crystallographic axes of YBCO. This configuration allows single-chip measurements of the Josephson coupling in different directions.

The *a*-axis films used here are fabricated by pulsed laser deposition on (100) LaSrGaO₄ substrates using a modified template method.¹⁰ These films provide a unique configuration in which in-CuO₂-plane versus out-of-CuO₂-plane properties of YBCO can be studied in the substrate plane.^{10,11} θ -2 θ x-ray diffraction scans of the films show better than 99% *a*-axis orientation. ϕ scans of the (102) planes of YBCO display a complete twofold symmetry indicating total inplane separation of the *b* and *c* axes of YBCO. Planar transmission electron microscopy indicates that less than 2% of the grains are in-plane misaligned.¹⁰

The lower inset of Fig. 1 shows a schematic of our edge junctions. Seven junctions were fabricated spanning the substrate plane from the *b* and *c* directions of YBCO with 15° between each junction. In fabricating the devices, we deposit 500 Å of SrTiO₃ on top of 2300 Å YBCO above a 400 Å $PrBa_2Cu_3O_{7-x}$ template layer. The transition temperature of the *a*-axis YBCO is typically 88 K before patterning.¹¹ 3500 Å $SiO₂$ is deposited and patterned by lift-off in order to provide a sharp-edged ion milling mask for the multilayer. A 500 eV Ar beam is directed normal to the substrate for ion milling. The resulting edge profile is measured by atomic force microscopy, and the edge angle was typically 60° to 70° from the substrate plane. No difference in the edge profile for edges facing different directions was observed, and we concluded that the edge milling was uniform for different angles. It is desirable to have the edge as sharp as possible here, and our angles compare favorably with the commonly observed angles of 30° or less for standard edge junctions milled with a photoresist mask. After milling, 1000 Å of Au is evaporated, followed by annealing at 350 °C for an hour in flowing oxygen to form good electrical contacts at the edge junction faces. Ag/PbIn $(500 \text{ Å}/2000 \text{ Å})$ layers are then deposited and patterned to complete the devices. Evaporation of Au and Ag/PbIn are performed with the chip at an angle and rotating in order to provide equal deposition to the edge surfaces for all seven junctions. Finally PbIn/Ag/Au layers are patterned by ion-milling, leaving a junction width of 100 μ m.

Figure 1 shows current-voltage (*I*-*V*) characteristics of the seven junctions at 4.2 K. The characteristics show a striking systematic change with angle. The critical current decreases, and the normal resistance increases as the junction angle goes from 0° to 90° (where the angle is measured from the direction parallel to the b direction. We also note that the junctions displayed good microwave response, with a clear ac Josephson effect being observed.

FIG. 2. Junction resistance versus angle. Data points are from two chips. The lines are their fit to Eq. (1) . Inset is a magnetic diffraction data from a b -direction junction $(4.2 K)$.

The critical currents of the junctions were completely suppressible by magnetic field. The inset to Fig. 2 shows the magnetic field dependence of a *b*-direction junction with the field applied parallel to the substrate and from the side of the junction. The period of the critical current modulation calculated using $\lambda_{ab} \approx 2000 \text{ Å}$ (at 4.2 K) is in good agreement with the observed magnetic field dependence.

In junctions that contain YBCO–noble-metal interfaces, the normal resistance of the junctions is often dominated by the contact resistance of the interface.^{1,11} Reasonable electrical contacts to YBCO can be obtained by *in situ* deposition of a noble metal following the deposition of YBCO or annealing the contact in oxygen, as done here.¹² The lowest reported experimental values obtained are typically $\rho_{i,ab}$ $\sim 10^{-8}$ to $10^{-9} \Omega \text{ cm}^2$ and $\rho_{i,c} \sim 10^{-7}$ to $10^{-8} \Omega \text{ cm}^2$, for contacts into the $CuO₂$ plane direction and the c direction, respectively.¹³ The theoretical lower limits to the interface resistivities taking into account the mismatch in the Fermi velocities of the materials are $\rho_{i,ab} \sim 10^{-12} \Omega \text{ cm}^2$ and $\rho_{i,c}$ \sim 10⁻¹¹ Ω cm².¹⁴ The discrepancy between theory and experiment comes from residual interface reactivity and oxygen deficiency at the interface.¹² These factors probably also give rise to the anisotropic interface resistivity with a typical ratio $\rho_{i,c}/\rho_{i,ab}$ of $\approx 10^{13}$

In our junctions, the contact resistivity increases as the angle goes from 0° to 90°. The junction resistances correspond to contact resistivities of $\rho_{i,b}$ < 10⁻⁸ Ω cm² and $\rho_{i,c}$ $\langle 10^{-7} \Omega \text{ cm}^2 \text{ for junctions in the } b \text{ and the } c \text{ directions,}$ respectively, and they are among the lowest values reported.

The YBCO–noble-metal contacts used here can be modeled as superconductor-insulator-normal-metal (SIN) interfaces, and the transparency of the insulating barrier strongly affects the Josephson coupling of the device. A recent study of such interfaces has shown evidence of tunneling transport through a barrier even in contacts with interface resistivities as low as $\sim 10^{-9} \Omega \text{ cm}^2$.¹⁵ The barrier causes a discontinuous depression of the order parameter of YBCO at the SIN interface. Kupriyanov and Likharev have calculated how this affects the $I_c R_n$ products of junctions containing such interfaces.¹⁶ They showed that $I_c R_n$ should scale as ρ_i^{-1} to $\rho_i^{-3/2}$ depending on the value of ρ_i and the details of the geometry of the device. R_n is effectively ρ_i times the area of

FIG. 3. Critical current versus angle of the junction direction $(4.2 K)$. Data points are normalized critical currents from two chips. Curve A is $I_c \propto \cos \theta$. Curve B is $I_c \propto R_n^{-2}(\theta)$. Curve C is I_c $\propto R_n^{-2}(\theta)$ combined with the isotropic *d*-wave model. Curve D is $I_c \propto R_n^{-2}(\theta)$ combined with a "compressed" (by a factor of 3) *d*-wave model. The inset shows the $I_c R_n$ products from the two chips.

the junction interface, and this leads to $I_c R_n$ scaling as R_n^{-1} to $R_n^{-3/2}$. Reduction factors in I_cR_n obtained from their calculation are typically on the order of 10^{-2} to 10^{-3} . This is consistent with the experimental $I_c R_n \approx 40 \mu V$ observed here in the *b* direction at 4.2 K in comparison with the expected maximum $I_c R_n \approx 5$ mV for a YBCO–normal-PbIn junction system.¹⁴

To analyze the angular dependence of the *I*-*V* characteristics, we start by considering the junction resistance. For a junction with angle θ from the *b* direction, we consider a simple model in which the interface resistivity consists of two parallel contact resistivity components of $\rho_{i,b}$ and $\rho_{i,c}$ scaled by appropriate areas determined by the angle θ . The resistance is then given by

$$
R_n(\theta) = \frac{\rho_{i,b}\rho_{i,c}}{A(\rho_{i,b}\sin\theta + \rho_{i,c}\cos\theta)}.
$$
 (1)

Here, *A* is the area of the junction interface, which is the same for all seven junctions. Figure 2 shows data from two chips and their fit to $R_n(\theta)$ (solid and broken lines). Despite scatter in the data, Eq. (1) qualitatively describes the angular trend observed here. A particularly good fit was obtained for one of the chips as seen in the lower curve in the figure.

Next we consider the critical current. Figure 3 shows critical currents of junctions from the two chips, normalized by the critical currents of the junctions at $\theta=0^{\circ}$. There is scatter in the data, but both chips clearly show that the critical current decreases as a function of the angle.

If edge profiles in the *b*-*c* plane in our junctions consisted of microscopic steps with interfaces parallel and perpendicular to the $CuO₂$ planes (as seen in high resolution transmission electron microscopy studies of high- T_c edge junctions made with standard c -axis oriented films¹⁷), one would expect to see a $\cos\theta$ dependence of the critical current arising from the net area facing the *b* direction in each junction. Curve A in the figure shows the angular dependence I_c \propto cos θ . This dependence is clearly not observed in our data, suggesting that the junctions here are not made up of microsteps. This is consistent with the fact that usually such microstepped edges result only after a relatively high temperature $(>700 \degree C)$ process causing the edge surfaces to undergo a regrowth, whereas the edge interfaces in our devices were annealed at only 350 °C following ion milling and deposition of Au. This temperature is too low to cause regrowth.

We now consider an alternative model for $I_c(\theta)$ which uses Eq. (1) for $R_n(\theta)$ and then finds I_c by using the Kupriyanov and Likharev approach. Using $I_c \dot{R}_n \propto R_n^{-1}$, ¹⁶ we plot the angular dependence of $I_c \propto R_n^{-2}(\theta)$ as curve B in Fig. 3. Although it produces the overall decrease in the critical current with angle, the fit is quite poor: despite scatter, the data show a faster reduction of I_c with angle.

To understand what might be causing the rapid suppression, we note that the characteristic of each junction should involve the magnitude of the order parameter in the out-of-CuO₂-plane direction "averaged" near θ . In the present junction system, $I_c R_n \propto (\Delta_{\text{PbIn}} \Delta_{\text{YBCO}})^{1/2}$.¹⁸ In principle, the anisotropic interface resistivity can be combined with a functional form of the k_z direction order parameter to fully describe the angular dependence of I_c . With this in mind, Fig. 3 shows the functional form of $I_c(\theta)$ expected from isotropic $d_{x^2-y^2}$ (curve C) and that from $d_{x^2-y^2}$ compressed in the k_z direction by a factor of 3 (curve D) for YBCO, combined with the $R_n^{-2}(\theta)$ factor. More data are needed in order to identify the exact functional form of the out-of-plane order parameter, but the data are qualitatively suggestive that incorporation of such decreasing functions of θ might account for the I_c reduction.

Finally we note that in an ideal tunnel junction $I_c R_n$ is the main parameter of interest in probing the order parameter and the strength of the Josephson coupling. The inset of Fig. 3 plots the $I_c R_n$ from the two chips without normalization. Although we find a systematic angular dependence in the *I*-*V* characteristics, there is a significant scatter in $I_c R_n$. This prevents us from closely studying the angular dependence. A larger number of junctions would be needed to provide data that will allow better analysis of the amplitude of $I_c R_n$.

In conclusion, we have fabricated anisotropic YBCO/Au/ Ag/PbIn edge junctions using in-plane aligned *a*-axis films. By comparing the characteristics of the junctions made along different directions on single chips, we demonstrated the feasibility of measuring the anisotropic angular dependence of the out-of-plane order parameter in YBCO. Although the *I*-*V* characteristics are dominated by interface resistivity, the $I_c(\theta)$ data are indicative that YBCO has an order parameter that gets suppressed as the angle away from the $CuO₂$ plane is increased. In particular, the data do not appear consistent with the isotropic *s* wave, but the *d*-wave models provide suppression of the order parameter that is consistent. Depending on the degree of anisotropy in such a symmetry, the order parameter can also show a significant reduction in the k_z direction. In the future, junctions with even lower contact resistivity or tunnel junctions with a larger number of devices per chip should provide a better probe of the out-of- $CuO₂$ -plane order parameter.

This project was supported by ONR Grant No. N00014- 96-C-2008.

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- ¹H. Akoh, S. Kohjiro, C. Camerlingo, R. Yuasa, and S. Takada, Physica C 180, 227 (1991); J. Yoshida, T. Hashimoto, S. Inoue, M. Sagoi, and K. Mizushima, *ibid.* 185-189, 1923 (1991); Mark Lee, D. Lew, C. B. Eom, T. H. Geballe, and M. R. Beasley, Appl. Phys. Lett. 57, 1152 (1990).
- 2 J. Kwo, T. A. Fulton, M. Hong, and P. L. Gammel, Appl. Phys. Lett. 56, 788 (1990).
- ³C. Gerber, D. Anselmetti, J. G. Bednorz, J. Mannhart, and D. G. Schlom, Nature (London) 350, 280 (1991).
- ⁴A. Mathai, Y. Gim, R. C. Black, A. Amar, and F. C. Wellstood, Phys. Rev. Lett. **74**, 4523 (1995); D. A. Wollman, D. J. Van Harlingen, W. C. Lee, D. M. Ginsberg, and A. J. Leggett, *ibid.* **71**, 2134 (1993); C. C. Tsuei, J. R. Kirtley, C. C. Chi, L. S. Yu-Jahnes, A. Gupta, T. Shaw, J. Z. Sun, and M. B. Ketchen, *ibid.* **73**, 593 (1994).
- ⁵ A. G. Sun, D. A. Gajewski, M. B. Maple, and R. C. Dynes, Phys. Rev. Lett. **72**, 2267 (1994); A. S. Katz, A. G. Sun, R. C. Dynes, and K. Char, Appl. Phys. Lett. 66, 105 (1995); R. Kleiner, A. S. Katz, A. G. Sun, R. Summer, D. A. Gajewski, S. H. Han, S. I. Woods, E. Dantsker, B. Chen, K. Char, M. B. Maple, R. C. Dynes, and J. Clarke, Phys. Rev. Lett. **76**, 2161 (1996).
- 6K. A. Kouznetsov, A. G. Sun, B. Chen, A. S. Katz, S. R. Bahcall, John Clarke, R. C. Dynes, D. A. Gajewski, S. H. Han, M. B. Maple, J. Giapintzakis, J.-T. Kim, and D. M. Ginsberg, Phys. Rev. Lett. **79**, 3050 (1997).
- 7 A. G. Sun, A. Truscott, A. S. Katz, R. C. Dynes, B. W. Veal, and C. Gu, Phys. Rev. B 54, 6734 (1996).
- ⁸Y. Gim et al., IEEE Trans. Appl. Supercond. 7, 2331 (1997).
- ⁹ Melvin H. Hanna, *Quantum Mechanics in Chemistry* (Benjamin-Cummings, New York, 1981).
- 10Z. Trajanovic, I. Takeuchi, P. A. Warburton, C. J. Lobb, and T. Venkatesan, IEEE Trans. Appl. Supercond. 5, 1237 (1995); Physica C 265, 79 (1996).
- ¹¹ I. Takeuchi, C. J. Lobb, Z. Trajanovic, P. A. Warburton, and T. Venkatesan, Appl. Phys. Lett. 68, 1564 (1996).
- 12S. E. Russek, S. C. Sanders, A. Roshko, and J. W. Ekin, Appl. Phys. Lett. **64**, 3649 (1994); J. W. Ekin, C. C. Clickner, S. E. Russek, and S. C. Sanders, IEEE Trans. Appl. Supercond. **5**, 2400 (1995); M. A. M. Gijs, D. Scholten, Th. van Rooy, and R. Ijsselsteijn, Solid State Commun. **71**, 575 (1989).
- ¹³ I. Takeuchi, J. S. Tsai, H. Tsuge, N. Matsukura, S. Miura, T. Yoshitake, Y. Kojima, and S. Matsu, IEEE Trans. Magn. **27**, 1626 (1991).
- ¹⁴ Mark Lee and M. R. Beasley, Appl. Phys. Lett. **59**, 591 (1991).
- 15S. C. Sanders, S. E. Russek, C. C. Clickner, and J. W. Ekin, IEEE Trans. Appl. Supercond. **5**, 2404 (1995); Yizi Xu, J. W. Ekin, S. E. Russek, R. Fiske, C. C. Clickner, I. Takeuchi, Z. Trajanovic, and T. Venkatesan, *ibid.* 7, 2836 (1997).
- 16M. Yu. Kupriyanov and K. K. Likharev, Sov. Phys. Usp. **160**, 49 (1990); M. Yu. Kupriyanov and K. K. Likharev, IEEE Trans. Magn. 27, 2460 (1991), and references therein.
- 17S. Rozeveld, K. L. Merkel, and K. Char, Physica C **252**, 348 $(1995).$
- 18 V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963); **104**, 11(E) (1963).