Progress in Superconductivity Vol.2 No.2 pp65-70 30 June 2001

Tunneling Characteristics in Pb/$Bi_2Sr_2CaCu_2O_{8+\delta}$ Junctions as an Evidence for a *d*-wave Order Parameter Symmetry in $Bi_2Sr_2CaCu_2O_{8+\delta}$ Superconductors

Hyun-Sik Chang and Hu-Jong Lee*
Pohang University of Science and Technology, Pohang 790-784, Korea
Received 31 May 2001

$Bi_2Sr_2CaCu_2O_{8+\delta}$ 고온초전도체의 *d*-파 대칭성 증거로서의 Pb/$Bi_2Sr_2CaCu_2O_{8+\delta}$ 접합 투과전도특성

장현식, 이후종*

Abstract

Pb/$Bi_2Sr_2CaCu_2O_{8+\delta}$-single-crystal junctions with the tunneling direction along the *c* axis of the crystal were fabricated to obtain an *s*-wave-superconductor/*d*-wave-superconductor Josephson junctions. The tunneling $R(T)$ curves and current-voltage characteristics show distinct features which can be explained only under the assumption that the order parameter of high-T_c $Bi_2Sr_2CaCu_2O_{8+\delta}$ superconductors has a pure *d*-wave symmetry, which is in contrast to the case of $YBa_2Cu_3O_{7+\delta}$ superconductors where a minor *s*-wave component is also present.

Keywords : *d*-wave symmetry, $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal, tunnel junction

I. Introduction

Until recently many experimental and theoretical works on high-T_c superconductors (HTSC's) have been focused on the symmetry of the order parameter (OP) which is the superconducting condensate wavefunction of the Cooper pairs. Group symmetry arguments [1] enforce the OP symmetry to match the crystallographic structure of the HTSC's which is two-dimensional (2D) in nature since the superconductivity is confined in the layered Cu-O planes. Among numerous experiments, phase sensitive measurements using π-junctions, especially in $YBa_2Cu_3O_{7+\delta}$ (Y123) single crystals [2,3], have revealed direct evidences that the predominant OP symmetry in the Y123 crystals is *d*-wave (to be more precisely $d_{x^2-y^2}$-wave) with a minor *s*-wave component [3-6]. At present, there is no doubt that the OP of the high-T_c superconducting Y123, whose Cu-O planes are orthorhombic in structure, has

*Corresponding author. Fax: +82 54 279 3099
e-mail: hjlee@postech.ac.kr

d+*s*-wave-type symmetry with a predominant *d*-wave component, consistent with the theoretical prediction that the crystallographic structure should match the OP symmetry [1].

In the case of other HTSC's such as $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) similar phase-sensitive measurements to determine the OP symmetry as in Y123 are more difficult to perform, mainly due to the lack of twin boundaries and/or clean lateral crystal surfaces. However, no OP symmetry other than the *d*-wave can explain the results of *ab*-plane quasiparticle tunneling spectrum of Bi2212 single crystals [8-10]. In the case of *c*-axis tunneling measurements, either by STM (scanning tunneling microscope) or by PCT (point contact tunneling) [11], the cusp-like quasiparticle density of states (DOS) with finite states inside the gap edge are characteristic to *d*-wave OP symmetry. The shape of those quasiparticle DOS is also consistent with the computed results assuming a *d*-wave OP [13]. However, since STM measurements probe only the surface characteristics of the crystal and PCT picks up unwanted contributions from *ab*-plane tunneling components, a sandwich-type planar junction free of other side effects is advantageous in analyzing the measurement results.

Recently Möβle and Kleiner [14] showed that a component of *s*-wave OP, which is about three orders of magnitude smaller than the dominant *d*-wave OP, exists in the tetragonal Bi2212 single crystal. They observed a Fraunhofer-diffraction-pattern-like magnetic field dependence of the critical current below the transition temperature of Pb in a Pb/Bi2212 Josephson junction (JJ) with the tunneling current parallel to the *c* axis of Bi2212. Such a field dependence is not possible if the OP of Bi2212 were pure *d*-wave, since the positive and negative contributions to the critical current of the *s*-wave-SC/*d*-wave-SC JJ would completely cancel each other, resulting in a zero critical current for any values of magnetic field. Later, Rae [15] argued that the observed critical current could have resulted from the anisotropy of the OP in Pb rather than from a tiny *s*-wave OP component in the tetragonal Bi2212 which should have been of pure *d*-wave to satisfy the matching condition of the crystallographic structure with the OP symmetry [1].

In this work tunneling characteristics of Pb/Bi2212 single crystal sandwich-type planar JJ with the tunneling current along the *c* axis of the Bi2212 crystal will be presented. Depending on the particular temperature regime, the $R(T)$ curve and current-voltage characteristics (IVC) are analyzed in terms of NId, dId', and d'Is junctions, where N, I, d, d', and s stand for normal metal, insulator, *d*-wave HTSC, *d*-wave HTSC with suppressed superconductivity [16], and conventional *s*-wave SC, respectively. The results are consistent with a pure *d*-wave OP in Bi2212 single crystals.

II. Sample Preparation

Bi2212 single crystals were grown by solid-state-reaction method, where powders of Bi_2O_3, $SrCO_3$, $CaCO_3$, and CuO were mixed, thoroughly ground, and heat treated in an alumina crucible. The mixing molar ratio of Bi:Sr:Ca:Cu was 2.3:2:1:2. During the crystal growth process, a constant oxygen gas was provided and a temperature gradient of about 5 °C/cm was set up across the diameter of the crucible to enhance the growth rate. As-grown single crystals with proper size were glued on MgO substrates using negative photoresist (OMR-83) and cleaved with 3M (Scotch Magic) tape until optically smooth surfaces were obtained. Upon cleaving, 200 Å Ag (99.99%) and 200 Å Au (99.9%) were thermally deposited in succession on the top of the crystal to protect the surface from contamination during further fabrication process as well as to obtain a clean interface between the normal electrodes and the Bi2212 single crystal. Three mesas (mesa A, B, C of Fig. 1) of size ~200×200 μm^2 were then patterned using the conventional photolithography with positive photoresist (+PR, Microposit 1400-23) and the Ar-ion-etching technique. After covering the crystal with +PR to avoid sharp edges and removing the +PR at the mesa positions (except mesa B), about 1-μm-thick Ag was evaporated and chemically etched with KI/I_2 solution (56%) to form extension pads. Then covering the whole sample with +PR again and patterning three windows (B, D, E of Fig. 1), one on the middle mesa (B) and the other two (D, E) on the extension pads. The whole Au film and about 50-100Å-thick Ag film deposited on the top of mesa B were removed by Ar-ion-etching. Finally the three windows on B, D, and E were linked to form a Pb/Ag/Bi2212 junction by evaporating a strip of ~5000- Å-thick Pb film using a metal mask as illustrated in Fig 1(b). Since Pb is known to diffuse easily into Au, Ag was preferred as the normal-metal electrode between Bi2212 and Pb. An optical micrograph of a completed junction is shown in Fig. 1(c). Immediately after completing the sample fabrication, it was sealed in a copper can and immersed in a μ-metal shielded LN/LHe (liquid nitrogen/liquid helium) dual dewar to prevent further thermal diffusion of Pb into the Bi2212 crystal. All measurements were performed using conventional four-probe method as shown in Fig. 1(a).

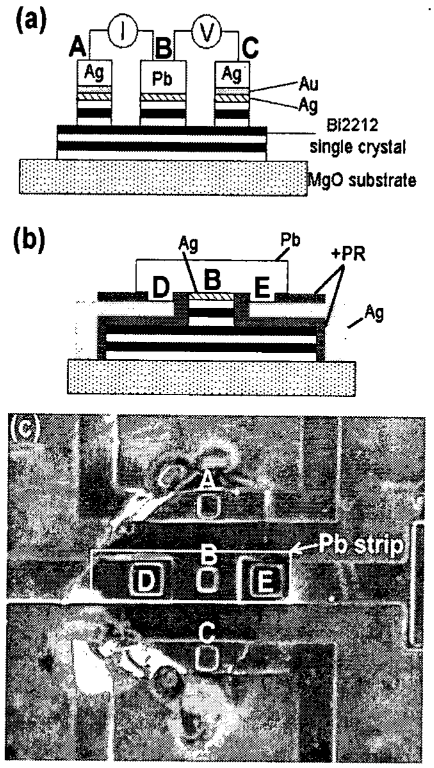

Fig. 1. Schematic illustration of (a) the measurement configuration and (b) the connection of the Pb/Ag/Bi2212 junction to the extension pads; (c) an optical microscope photograph of the measured sample. The mesa corresponding to the junction is connected to the extension pads via a Pb strip (surrounded by a white box for clarity) as shown in the photograph.

III. Results and Discussion

The tunneling current in a junction with tunneling barrier can be simplified as [17]

$$I(V,T) = \frac{1}{eR_N}\int_{-\infty}^{\infty} dE[f(E-eV,T) - f(E,T)]n_A(E)n_B(E-eV) \qquad (1)$$

where $f(E,T)=1/[1+\exp(E/k_BT)]$ is the Fermi distribution function, R_N the normal resistance of the junction, and $n_A(E)$ ($n_B(E)$) is the normalized quasiparticle DOS in A (B) given by [13],

$$n(E) \equiv \frac{N(E)}{N_0} = \frac{1}{2\pi}\int_0^{2\pi} d\phi \frac{E}{\sqrt{E^2-\Delta^2}} \qquad (2)$$

The subscript A (or B) stands for N, d, d', and s which represent, as previously mentioned, normal metal, *d*-wave HTSC, *d*-wave HTSC with suppressed superconductivity, and conventional *s*-wave SC, respectively. If A (or B)=d or d', Δ is chosen as [13],

$$\Delta = \Delta(T)\cos(2\phi) \qquad (3)$$

to account for the *d*-wave OP symmetry. For A (or B)=s, $\Delta=\Delta(T)$ is adopted, and for A (or B)=N, $n_N(E)$ is just set to unity. For the temperature dependence of the energy gap $\Delta(T)$ itself an empirical approximation [18] of the form

$$\frac{\Delta(T)}{\Delta_0} = \tanh\left(1.74\sqrt{\frac{T_c}{T}-1}\right) \qquad (4)$$

is adopted. It agrees very well, except for some minor negligible digressions, with the numerical solution of the BCS gap function. Here Δ_0 denotes the zero temperature gap value and T_c the transition temperature of d, d' and s. If the finite quasiparticle lifetime is taken into account, E on the right hand side of Eq. (2) should be replaced by $(E-i\Gamma)$ where Γ

is the smearing parameter.

Figure 2 shows the measured $R(T)$ curve of the Pb/Bi2212 junction from room temperature down to liquid helium temperature. The $R(T)$ curve itself can be divided into the following four regions;

(i) Region I ($T>T_c$): the temperature is above the bulk transition temperature T_c of Bi2212 and the whole junction is in the normal state;

(ii) Region II ($T'_c<T<T_c$): Bi2212 is in the superconducting state except for the surface Cu-O bilayer the superconductivity of which is suppressed [16] and will be called the surface Bi2212' with transition temperature T'_c ($<T_c$). Pb is still in the normal state;

(iii) Region III ($T_c^{Pb}<T<T'_c$): the whole Bi2212 crystal is superconducting except for the Pb electrode whose transition temperature is $T_c^{Pb}<T'_c$);

(iv) Region IV ($T< T_c^{Pb}$) : the whole junction is in the superconducting state.

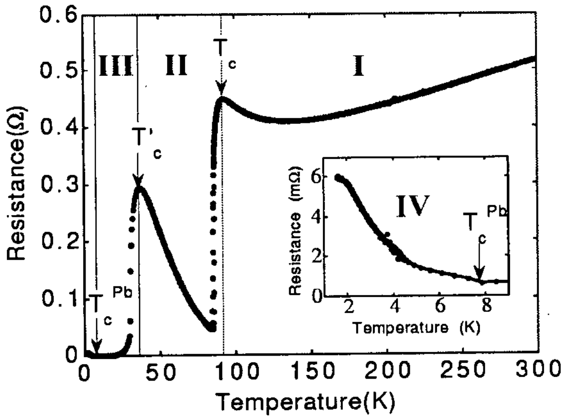

Fig. 2. $R(T)$ curve of the Pb/Bi2212 junction from room temperature down to liquid helium temperature. The curve can be divided into four regions separated by T_c, T'_c, and T_c^{Pb}, which are the transition temperatures of bulk Bi2212, surface Bi2212', and Pb, respectively. Inset shows the $R(T)$ curve in a narrower temperature range below T_c^{Pb}.

Region I is the typical normal region above T_c of c-axis tunneling in Bi2212. Region II is quite surprising and it appears as if another phase other than the Bi2212 were present in the crystal. However, the only other possible phase is the 2201 phase which has a maximum transition temperature of about 20 K [19]. In our previous work Kim *et al.* [16] interpreted the state in Region II in terms of suppressed superconductivity of the surface Cu-O bilayer. Since the Cu-O bilayer at the surface of the Bi2212 single crystal is in a proximity contact with the normal metal (Ag in this case), its boundary condition is quite different from the Cu-O bilayers beneath it. As a result the OP of the surface bilayer gets suppressed without any change in the symmetry, and results in a lower transition temperature $T'_c(<T_c)$. Also the critical current of the surface junction I'_c between the surface Cu-O bilayer and the Cu-O bilayer next to it is much suppressed compared to the critical current $I_c(\approx 10I'_c)$ of the inner intrinsic Josephson junctions. In Region III the whole Bi2212 single crystal is superconducting and the resistance is nearly constant except near T'_c as should be for a pure tunnel junction. Below T_c^{Pb} which corresponds to the temperature Region IV, $R(T)$ shows a reentrant behavior as shown in the inset of Fig. 2, consistent with the fact that the OP in Bi2212 is of pure d-wave (or at least no s-wave component of the OP is present). If an s-wave component of the OP were present, due to a finite critical current between the surface Cu-O bilayer and Pb, $R(T)$ would fall to zero in that temperature region. In the case of pure d-wave OP and pure c-axis tunneling the positive [$\sin(\theta)$] and negative [$\sin(\theta+\pi)=-\sin(\theta)$] contributions to the critical current of the s-wave-SC (Pb)/I/d-wave-SC (Bi2212) JJ should be completely cancelled, resulting in a zero critical current state. Here θ is the gauge-invariant phase difference between Pb and Bi2212 and the factor π accounts for the phase difference of the d-wave OP with relative orthogonal momenta [according to Eq. (3) $\cos 2(\phi+\frac{\pi}{2}) = -\cos(2\phi)$].

To analyze the two anomalous Regions II and IV, Eq. (1) is adopted to describe the tunneling resistance of a tunneling junction as

$$R(T) = \left(\left[\frac{dI(V,T)}{dV} \right]_{V=0} \right)^{-1} . \quad (5)$$

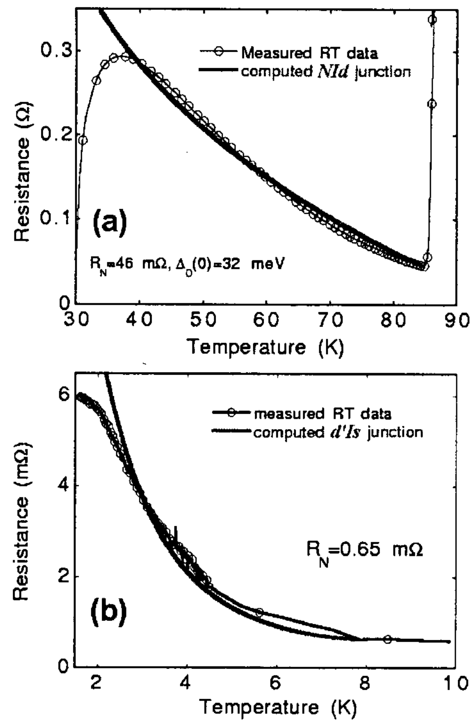

Fig. 3. (a) Computed curve (-) of an NId [Bi2212' ($T>T'_c$)/I/Bi2212] junction fitted to the measured $R(T)$ curve (o) in Region II. (b) Computed curve (-) of a dIs [Bi2212/I/Pb ($T<T_c^{Pb}$)] junction fitted to the measured $R(T)$ curve (o) in Region IV. The fitting parameters are as shown in the figures.

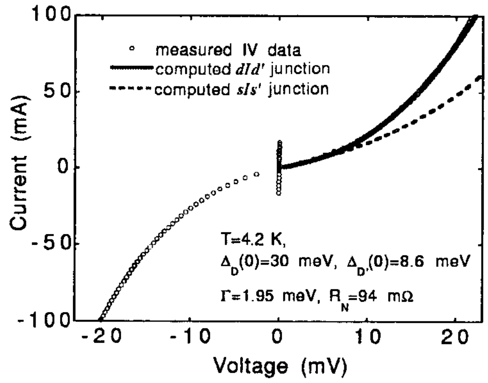

Fig. 4. Experimental IVC (o) at T=4.2 K together with computed curves of dId' (-) and sIs' (- -) junctions. The dId' junction fits far better than the sIs' junction. The fitting parameters are shown in the figure.

The state of the junction in Region II can be represented in terms of a NId (N/I/Bi2212) junction, where N represents the surface Cu-O bilayer with suppressed superconductivity including the Ag/Pb normal metal film in the temperature range $T'_c<T<T_c$ and 'I' represents the layer in between the surface Cu-O bilayer and the next Cu-O bilayer below it.

By assuming the OP of Bi2212 to be of pure *d*-wave, a computed curve together with the measured $R(T)$ data is shown in Fig. 3(a). The normal resistance R_N and $\Delta(0)$=32 meV are used as fitting parameters, which agree well with the measured values of others [20]. A similar computation was also made for Region IV in terms of a d'Is (Bi2212'/I/Pb) junction as shown in Fig. 3(b), where this time 'I' is the nonsuperconducting layer in between Pb and the surface Cu-O bilayer. A rather good fit of the two computed curves to the measured $R(T)$ curve serves as a direct evidence that the OP symmetry of the Bi2212 single crystal (including Bi2212') is indeed the *d*-wave. Especially the absence of a finite critical current and the tunneling behavior in Region VI excludes the possibility of an extended *s*-wave OP symmetry in Bi2212.

Another evidence for the OP symmetry in Bi2212 single crystal can be obtained by fitting the measured IVC in Region III or IV in terms of a dId' junction [21] using Eq. (1). Figure 4 displays the measured IVC at T=4.2 K together with the computed curves by assuming the junction to be of either dId' or sIs' type. The fitting parameters R_N and $\Delta(0)$ are close to the values of Figure 3. To enhance the fitting a finite quasiparticle lifetime Γ has been introduced. As shown in Fig. 4, the dId' junction picture agrees better with the measured IVC than the sIs' junction picture.

V. Summary

In summary, Pb/Bi2212 junction with the tunneling current parallel to the *c* axis of the Bi2212 single crystal were fabricated and used to convince that the OP symmetry of Bi2212 is pure *d*-wave. Dividing the tunneling $R(T)$ curve into four temperature regions and analyzing them in terms of dId' and d'Is tunneling junctions showed agreements with the *d*-wave OP picture. Especially, the quasiparticle tunneling feature instead of a Josephson tunneling behavior in the d'Is junction region below the transition temperature of Pb confirmed that the OP of Bi2212 is of pure *d*-wave without any *s*-wave components. Similar analysis done on the measured IVC also agreed with the picture of pure *d*-wave OP in Bi2212 single crystals.

References

[1] M. B. Walker, Phys. Rev. B 53, 5835 (1996); J. Lesueur, Phys. Rev. B 55, R3398 (1997).

[2] D. A. Wollman *et al.*, Phys. Rev. Lett. 71, 2134 (1993); D. A. Wollman *et al.*, ibid. 74, 797 (1995); D. J. Van Harlingen, Rev. Mod. Phys. 67, 515 (1995).

[3] K. A. Kouznetsov *et al.*, Phys. Rev. Lett. 79, 3050 (1997).

[4] P. Chaudhari and S.-Y. Lin, Phys. Rev. Lett. 72, 1084 (1994).

[5] A. G. Sun *et al.*, Phys. Rev. Lett. 72, 2267 (1994); A. G. Sun *et al.*, Phys. Rev. B 52, R15 731 (1995); A. G. Sun *et al.*, ibid. 54, 6734 (1996).

[6] R. Kleiner *et al.*, Phys. Rev. Lett. 76, 2161 {1996}; R. Kleiner *et al.*, Physica C 282-287, 2435 (1997); K. A. Muller, Nature 377, 133 (1995).

[7] H. Z. Durusoy *et al.*, Physica C 266, 253 (1996).

[8] Y. Tanaka and S. Kashiwaya, Phys. Rev. Lett. 74, 3451 (1995).

[9] S. Sinha and K.-W. Ng, Phys. Rev. Lett. 80, 1296 (1998).

[10] J. Y. T. Wei *et al.*, Phys. Rev. Lett. 81, 2542 (1998).

[11] H. Hancotte *et al.*, Physica C 282-287, 1487 (1997); H. Hancotte *et al.*, Phys. Rev. B 55, R3410 (1997).

[12] N. Miyakawa *et al.*, Physica C 282-287, 1519 (1997); N. Miyakawa *et al.*, Phys. Rev. Lett. 80, 157 (1998); Y. DeWilde *et al.*, *ibid.* 80, 153 (1998); J. Y. T. Wei *et al.*, Phys. Rev. B 57, 3650 (1998); L. Ozyuzer *et al.*, Phys. Rev. B 57, R3245 (1998).

[13] H. Won and K. Maki, Phys. Rev. B 49, 1397 (1994).

[14] M. Möβle and R. Kleiner, Phys. Rev. B 59, 4486 (1999).

[15] A. I. M. Rae, Phys. Rev. Lett. 84, 2235 (2000).

[16] N. Kim *et al.*, Phys. Rev. B 59, 14639 (1999).

[17] E. L. Wolf, *Principles of Electron Tunneling Spectroscopy*, (Oxford Univ. Press, New York, 1985).

[18] J. H. Xu *et al.*, Phys. Rev. Lett. 73, 2492 (1994).

[19] C. C. Torardi *et al.*, Phys. Rev. B 38, 225 (1988); K. Byczuk and J. Spalek, *ibid.* 53, R518 (1996).

[20] Renner *et al.*, Phys. Rev. Lett. 80, 149 (1998); Krasnov *et al.*, Phys. Rev. Lett. 84, 5860 (2000).

[21] Region IV was analyzed in terms of dId' rather than d'Is. The gap structure of Pb gets shadowed by the gap structure of Bi2212', *i.e,* as the bias current is increased the Josephson coupling between the surface Cu-O bilayer and the next bilayer breaks down before the breakdown of the coupling between Pb and the surface Cu-O bilayer. Thus the voltage first jumps to the characteristic voltage of Bi2212' which is much larger than the gap voltage of Pb and the resulting quasiparticle branch in the IVC is that of a dId' junction.