Superconducting Pairing Symmetry in $YBa_2Cu_3O_{7-x}$ from a Study of Josephson Tunnel Junctions

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The biepitaxial $YBa_2Cu_3O_{7-x}$(YBCO)/CeO_2/MgO Josephson junctions and c-axis YBCO/Au/Pb junctions have been fabricated to investigate the pairing state in YBCO. As a result, for the in-plane biepitaxial YBCO grain boundary junctions, both the integral and half-integral Shapiro steps are observed under zero applied magnetic field, suggesting the possibility of d-wave pairing in YBCO. On the other hand, Josephson tunneling currents have been observed on the YBCO/Au/Pb junctions with tunneling along the c axis of the YBCO films. The dependence of the Josephson critical current on the applied field for the YBCO/Au/Pb junctions exhibits an unconventional diffraction pattern which is close to the characteristic of a predominantly d-wave superconductor. The results are discussed in terms of the existing s-wave and d-wave symmetry theories.

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I. Introduction

Since the discovery of high-temperature superconductors (HTSC), the issue of whether the pairing symmetry is conventional or unconventional has been hotly debated. Primarily, the most discussed candidate for unconventional pairing is the $d_{x^2-y^2}$ state which was demonstrated by some experiments [1-6]. However, c-axis tunneling experiments between YBCO and Pb showed that there must be a non-vanishing s component of the superconducting order parameter in YBCO [7, 8]. Recently, several groups show strong evidences of mixed d- and s-wave in YBCO [9] and BSCCO [10]. The mixing of s- and d-wave due to the structural distortion has been confirmed by the results from c-axis tunneling experiments with detwinned or twinned crystals of YBCO or BSCCO [9-11]. However, evidences for d-wave component from heavily twinned samples remain rare.

For distinguishing the pairing states, a more clear approach is to probe the phase of order parameter. It is known that the Josephson effect is a direction-sensitive phenomena connected with the orientation of the junction and with the crystal axis of the superconductor on each side. To measure the phase of the order parameter directly from a Josephson junction consisting of a HTSC and an ordinary superconductor could be based on the idea that the tunneling Cooper pairs will pick up the local phase of the order parameter as they move across the junctions, so that the phase difference in the different faces can be checked by the nodal points of the maximum supercurrent as a function of applied flux. In the c-axis tunneling between YBCO and Pb, for instance, if YBCO were a conventional s-wave superconductor, the critical current, $I_c(B)$, would exhibit a Fraunhofer-like dependence on magnetic field. If, on the other hand, YBCO were predominately d wave, a local minimum in I_c at a $B = 0$ should be observed, as in the d-wave corner junction experiment [1-3]. Moreover, a d-wave order parameter could be characterized by a nonzero Josephson current which results from the second-order tunneling processes. Such processes can be identified by measuring microwave-induced steps in the V-I characteristics. In this paper, we report the V-I characteristics within microwave irradiation for the biepitaxial YBCO/CeO_2/MgO Josephson junctions and the measurements on pair tunneling from the c axis of YBCO films to Pb to clarify the pairing symmetry in the YBCO superconductor.

II. Experiment

Epitaxial c-axis YBCO and CeO_2 films were grown onto rotated MgO substrates by a magnetron sputtering system. For the fabrication of biepitaxial YBCO Josephson Junctions, CeO_2 was chosen to be a seed layer to create the biepitaxial grain boundary. Fig. 1 shows the schematic diagram of a YBCO Josephson junction with a seed CeO_2 layer which covers half of the MgO substrate. The in-plane Josephson junction between YBCO grown onto MgO and CeO_2 with their axes rotated to $45^{\pm}$ to each other is shown in Fig. 1 also. The biepitaxial YBCO/CeO_2/MgO films were first characterized by the x-ray diffractionmeter and the atomic force microscope. The fabrication and characteristics of biepitaxial YBCO/CeO_2/MgO Josephson junctions have been described in detail elsewhere [12].

Additionally, for the fabrication of c-axis YBCO-Pb Josephson Junctions, the YBCO films were first patterned into 1.0-mm-wide stripes by a wet etching. After etching, an insulating 1 50-nm-thick $SrTiO_3$(STO) film was deposited onto the strip to prevent the leakage-current tunneling outside the junction area. To fabricate a junction, one must be able to connect the YBCO and Pb films through a "window" in the STO insulator. We fabricated the window and the electrodes on the YBCO film simultaneously using an ion mill. For milling the STO film, the mill rate of about 25 nm/min was calibrated with a 498 V, 20 mA argon ion beam. After milling, the sample was apace located into the vacuum chamber to evaporate an Au/Pb film. It is known that the junction resistance could be reduced by evaporating a few nm of diffusion barrier before the Pb evaporating [13]. The thin barrier is used to prevent either the depletion of oxygen and the formation of lead oxide or the interdiffusion of Pb and YBCO with subsequent oxidation of the Pb. Further, after

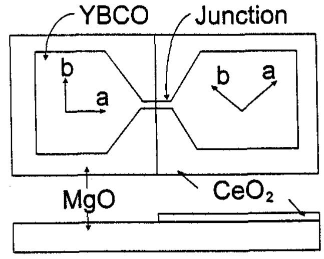

FIG. 1. Configuration of a biepitaxial YBCO/CeO_2/MgO Josephson Junctions. The CeO_2 layer which covers half of the MgO substrate is used as a seed layer. The width of the junction is 5 μm.

the deposition of Au layer the sample was annealed at temperature of 400 ±C in one atmospheric oxygen gas for 1 hour before the Pb evaporation. Thickness of Au barrier was controlled to be about 2 nm by a calibrated mill rate. Fig. 2 shows a scheme of the patterning process.

In Fig. 3, we show the temperature dependence of the resistance for a YBCO/Au/Pb tunnel junction. Tow superconducting transitions occurring around 7.2 and 90 K are corresponding to the critical temperature of Pb and YBC0 respectively. The V-I curve shows a resistively-shuntedjunction-like characteristic as shown in the inset of Fig. 3. The values of the critical current, I_c, are in the range of 50 $\sim$ 330 μA at temperatures between 2 $\sim$ 7 K.

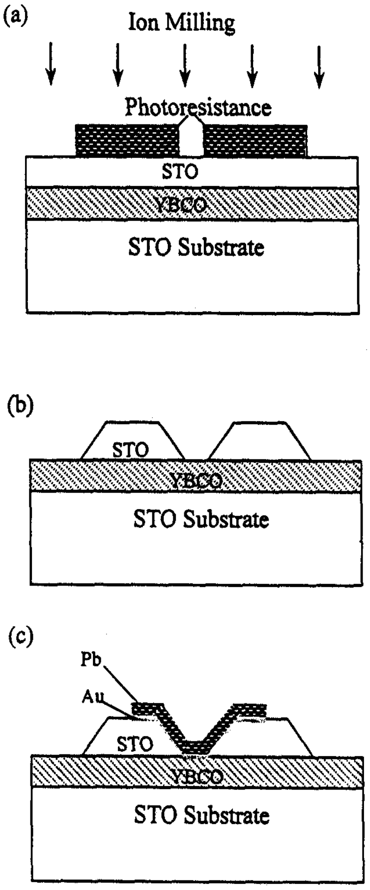

FIG. 2. Fabrication of the YBCO/Au/Pb junction: (a) a mask of photoresistance is photolithographically patterned, (b) $SrTiO_3$ is etched by an ion mill to make the window and the electrodes on YBCO, (c) Au and Pb layers are evaporated on the top of STO layer with an annealing process described in the text.

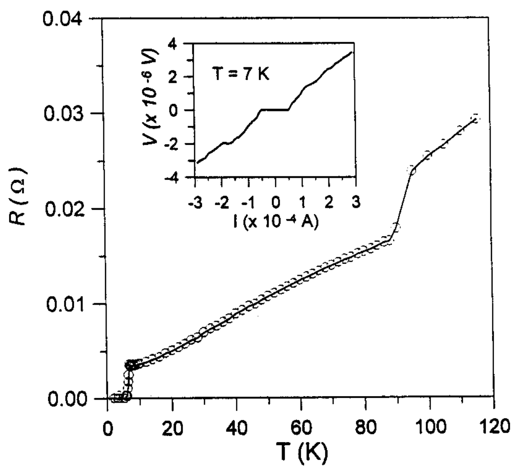

FIG. 3. The temperature dependence of resistance for the YBCO/Au/Pb junction. The inset shows the V-I curve at 7 K.

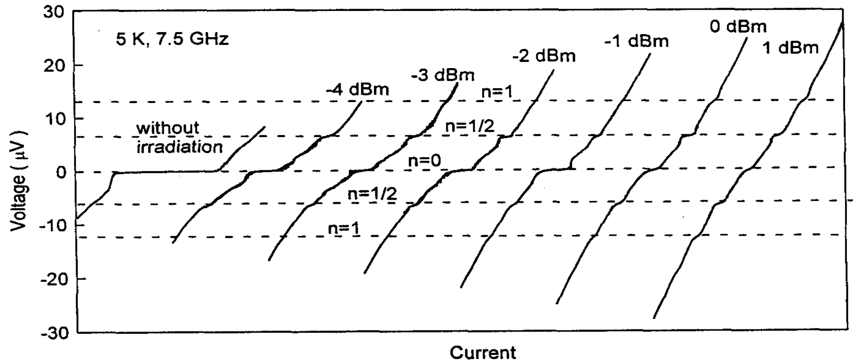

FIG. 4. The microwave responses of V-I curves for the biepitaxial YBCO/CeO_2/MgO junction under zero field at 5 K.

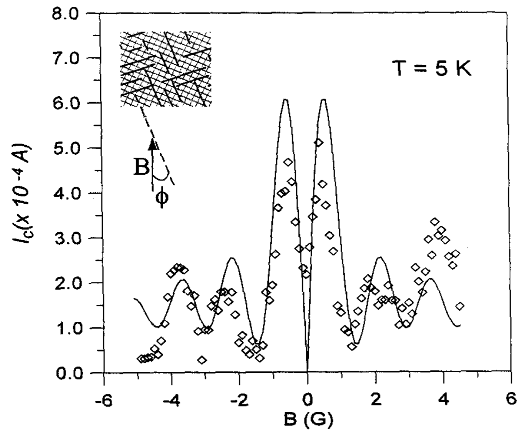

FIG. 5. Magnetic field dependence of Josephson critical current I_c at 5 K for a YBCO/Au/Pb tunnel junction. The solid line is the calculated curve based on Eq. (3). The inset shows the schematic diagram for the junction with an applied field at an angle ϕ relative to the twin boundary.

III. Result and Discussion

Fig. 4 shows the microwave responses of the V-I curves for a biepitaxial YBCO/CeO_2/MgO Josephson junction under zero applied magnetic field at 5 K. The frequency of the incident microwave is 7.5 GHz corresponding to the constant voltage of 15.5 μV for the first order step. However, the V-I curves exhibit both the integral and half-integral Shapiro steps, as shown in Fig. 4. Recently, Zhang [14] predicted that applied microwaves of frequency, f, will induce constant voltage steps at voltages $(n/2)f\Phi_0$ for in-plane Josephson junctions between $d_{x^2 i\, y^2}$ superconductors with their axes rotated to 45$^\pm$ to each other. Thus, according to Zhang's prediction, the possibility of d-wave pairing in YBCO should not be ruled out. Nevertheless, it is worthily notable that the microwave frequency coinciding with a self-resonant frequency of the junction may introduce half-integer steps [15]. If juncction parameters are within certain regimes [16], even first-order tunnelling can produce half-integer steps. However, half-integer steps also can be observed in other samples with different junction parameters. Furthermore, it is necessary to confirm the pairing symmetry for YBCO with other measurements. A more clear approach for distinguishing the pairing state is to measure the Josephson critical current in applied fields. Fig. 5 shows a typical result for the critical current of c-axis YBCO-Pb Josephson junction as a function of magnetic field parallel to the a-b plane at 5 K. Contrary to the conventional Fraunhofer diffraction pattern, the Josephson critical current was suppressed and took on minimum behavior around zero field. The anomalous result can be realized by studying the twin boundaries existing in the junction area. Recently, Konznetsov *et al.* [9] reported a new class of c-axis Josephson tunneling experiments in which a Pb film was deposited across a single twin boundary of a YBCO crystal. Basing on the assumption that YBCO were predominantly d-wave, any s-wave component to the order parameter would change sign across the twin boundary, for fields applied parallel to the boundary, they wrote the critical current of a small, uniform junctions as

$$I_c^2(\Phi,\gamma) = (I_0\Phi_0/\pi\Phi)^2\{1+\cos^2(\pi\Phi/\Phi_0) - \cos(2\gamma\pi\Phi/\Phi_0) - \cos[2(\gamma-1)\pi\Phi/\Phi_0]\}. \quad (1)$$

Here, I_0 is the maximum critical current of the junction, Φ, is the magnetic flux penetrating the junction, and γ is defined as $\gamma \equiv A_1/A$, where A_1 is the area of the smaller twin domain and A is the total area of the junction. If the fields are applied with an angle ϕ between the twin boundary and B, the flux will be replaced by $\Phi\cos\phi$ in Eq. (1). In our case, for a heavily twinned YBCO film, the geometric asymmetry parameter should be taken by γ = 0.5 and Eq. (1) is reduced as

$$I_c = I_0 \sin^2(\pi\Phi/2\Phi_0)/|\pi\Phi/2\Phi_0|, \quad (2)$$

which is identical to the result for the d-wave corner junction [1-3]. However, as the fields are applied parallel to the ab plane of the heavily twinned YBCO film, there are two orthogonal field directions, i.e., ϕ and ϕ + 90$^{\pm}$, in the tunneling process. Thus, the Josephson critical current is given by the sum of the two contributions:

$$I_c = I_{0,\phi}\sin^2(\pi\Phi\cos\phi/2\Phi_0)/|\pi\Phi\cos\phi/2\Phi_0| + I_{0,\phi+90^{\pm}}\sin^2(\pi\Phi\sin\phi/2\Phi_0)/|\pi\Phi\sin\phi/2\Phi_0| \quad (3)$$

Here, $I_{0,\phi}$, $I_{0,\phi+90^{\pm}}$ and ϕ are the parameters that can be adjusted (In fact, the fields are applied nearly along the a or b axis, i.e., $\phi \approx 0^{\pm}$). The solid line shown in Fig. 5 is the calculated curve for $I_{0,\phi} \approx 5.83 \times 10^{i\ 4}A$, $I_{0,\phi+90^{\pm}} \approx 1.03 \times 10^{i\ 4}A$ and $\phi = 9.2^{\pm}$. The agreement between the experimental data and theoretical calculation looks qualitatively good. The $I_c(B)$ shows regular periodicity of minimum values which occur whenever $\Phi\cos\phi$ = 2Φ_0, i.e.,

$$\Delta B = 2\Phi_0/\cos\phi[w(\lambda_{pb} + \lambda_{YBCO} + d)],$$

where w is the junction width, d is the thickness, λ_{Pb} iS the penetration depth in Pb and λ_{YBCO} is the penetration depth in YBCO. Taking w = 0.25 mm and $\lambda_{Pb} + \lambda_{YBCO} + d \approx$ 100 nm to make an approximatic calculation, we obtain $\Delta B \approx$ 1.6 G which is near the experimental values of $\Delta B \approx$ 1.5 G. This result suggests the presence of mixed d- and s-wave pairing in YBCO.

IV. Conclusion

We have fabricated the biepitaxial YBCO/CeO_2/MgO Josephson junction and the tunnel junction between the conventional superconductor Pb and the high-Tc YBCO film to probe the pairing symmetry in HTSC. In the microwave responses of the V-I curves for the biepitaxial YBCO/CeO_2/MgO Josephson junction, both the integral and half-integral Shapiro steps are observed. The presence of half-integral steps implies the possibility of d-wave pairing in YBCO. The admixture of d- and s-wave pairing in YBCO is suggested by the measurement of critical current in applied fields on the c-axis YBCO-Pb Josephson junction. A qualitatively good agreement between the data and theory for $I_c(B)$ was obtained. The results are consistent with the suggestion that there is a dominant d-wave pairing symmetry existing in YBCO.

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