Intrinsic BSCCO Josephson Junctions on off-axis Substrates

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Abstract **– Anisotropic transport and microwave properties of intrinsic $Bi_2Sr_2CaCu_2O_x$(2212) Josephson junctions fabricated on off-axis substrates were studied. The twin-free thin film 2212 Josephson junctions have been prepared by molecular beam epitaxy (MBE) on $LaAlO_3$ (001) substrates tilted 6° toward the [110] direction, and were patterned into a line shape along [110] $LaAlO_3$ with 20 μm in width. We found that the anisotropy parameter γ was increased from 76 to 120 by the annealing at temperature of 320 – 350 °C in the MBE chamber. The coherent Josephson radiation from stacked 2212 intrinsic junctions and their modulation under applied magnetic field were observed at frequency f_{REC}=1.7 GHz by using a non-resonant detection method. These results confirmed the occurrence of the mutual phase locking of the stacked series array of intrinsic junctions along the c-axis.**

I. INTRODUCTION

Recently, the intrinsic Josephson junction effect in a high-T_c superconductor has attracted considerable interest of many groups. It has been found that the intrinsic Josephson effect was observable in highly anisotropic materials such as Bi- or Tl- based oxide superconductors [1]-[4]. Kleiner et al. observed Josephson coupling between CuO_2 double layers in 2212 single crystal using the direct measurements of ac and dc Josephson effects for the first time [1],[2]. Most researches on 2212 materials have been focused on 2212 single crystals.

Despite a number of studies on the atomically controlled growth technique of 2212 thin films [5]-[9], no clear evidence of the intrinsic Josephson effect has been observed in those thin films, except for those fabricated on granular crystals. For practical applications, the intrinsic junction should be fabricated with epitaxial thin films.

In this paper, we report on the anisotropic transport and microwave properties of the twin-free thin film 2212 Josephson junctions. We have performed the Hall measurement by way of the control of carrier density in films using the 2212 thin films grown on MgO (001) substrates. The anisotropic transport properties were investigated for twin-free 2212 thin films grown on tilted $LaAlO_3$ (001) substrates, and the microwave Josephson radiation was observed by using a non-resonant detection method and their modulation under applied magnetic field.

II. EXPERIMENTAL

The off-axis 2212 thin films have been grown on tilted $LaAlO_3$ (001) and MgO (001) substrates by molecular beam epitaxy (MBE) with a sequential deposition technique. The optimum deposition interruption sequence, i.e. the deposition period of 10s and the interruption period of 60s, was adapted. The growth temperature was 740°C, and the oxygen/ozone mixture with the ozone content of ~10% was provided to the substrate at the background pressure of 2.5×10^{-5} Pa. For epitaxial growth of the 2212 thin films, the $Bi_2Sr_2CuO_x$ (2201) buffer layer of 3nm thick was predeposited. The thickness of 2212 films was 120nm, and the growth rate was 29nm/h. The details of the growth techniques and epitaxial structures were described elsewhere [10],[11].

The sample was mounted in a microwave cavity equipped with a microstrip connecting to a coaxial cable. The radiation power from the junction was measured by a superheterodyne detection technique with a non-resonant broad-band matching system at receiving frequency f_{REC}=1.7 GHz with a bandwidth ΔB=800 MHz. The sensitivity of the receiver was $\delta S \cong 3\times10^{-24}$ W/Hz at an integrating time τ=1s. The absolute values of the self-radiation power emitted from the junctions were exactly calibrated by a standard noise source installed inside the microwave receiver system. All electrical connections were carefully filtered by a low pass

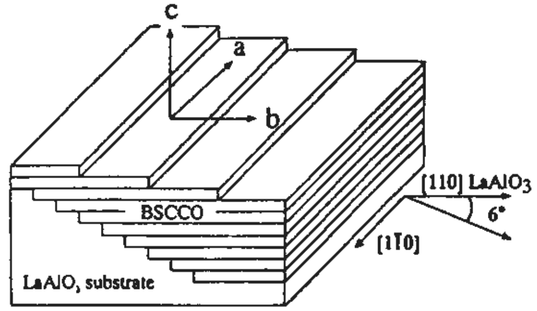

Fig.1 Schematic picture of intrinsic 2212 junction structure on a tilted $LaAlO_3$ substrate.

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(a) $LaAlO_3$ substrate

(b) buffer layer (2201 / $LaAlO_3$)

(c) 2212 / 2201/ $LaAlO_3$

Fig.2 RHEED patterns observed during the growth of 2212 thin films with electron beam in the [1$\bar{1}$0] azimuth of the $LaAlO_3$ substrate:(a) $LaAlO_3$ substrate (b) the 2201 buffer layer (c) the 2212 film.

filter and the sample holder was magnetically shield by a μ-metal. The detailed microwave experimental setup has been described in [12].

III. RESULTS AND DISCUSSION

Figure 2 shows reflection high energy electron diffraction (RHEED) patterns of $LaAlO_3$ substrate, 2201 buffer layer and 2212 thin film, which was observed with electron beam incident [1$\bar{1}$0] $LaAlO_3$ azimuth. The inclination of substrate surface with 6° toward [110] formed periodic surface step structure with 2 nm interval, as observed in (a). (b) was observed during the growth of buffer 2201 phase with thickness of about 3 nm. Clear streak patterns without additional spots indicate that the films have very smooth surface and contain no precipitates. (c) shows the RHEED pattern for 2212 phase, which indicates a superstructure pattern caused by modulating b-axis, and epitaxy was restricted so as to tilt the c-axis by 6° from the substrate normal.

A. Electric Properties

The c-axis-oriented 2212 thin films grown on MgO (001) substrates were used in the Hall measurement although they had a twin structure. The estimation of the carrier density in 2212 on the tilted substrate is thought to be complex because the Hall coefficients show opposite signs for H⊥c and H//c in

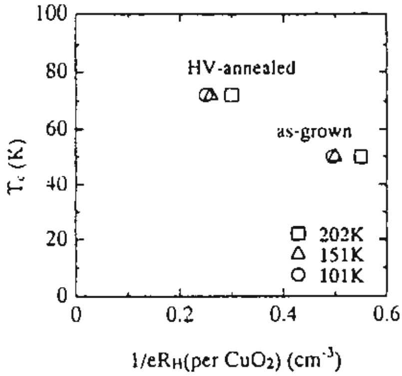


Fig.3 Critical temperature depending on $1/eR_H$ for an as-grown sample and HV-annealed sample prepared on MgO (001) substrates.

single crystals [13]. The Hall coefficients were measured with the magnetic field of 0-7 T parallel to the c-axis of 2212 film, and at temperatures between 101 and 202K. To change the carrier density, postannealing was done in a high vacuum (HV) of 10^{-6} Pa at temperature of 350°C. In Fig.3, the critical temperature is plotted against the carrier density per CuO_2 plane. In contrast to the bulk 2212 sample which shows a maximum $1/eR_H$ at 0.1-0.2 [14]-[16], our as-grown film (T_c=50K) can be assumed to be in an overdoped region. After HV-annealing at 350°C, T_c was increased to 71K and $1/eR_H$ was reduced to 0.3 at 202K. It is clear that the carrier density was reduced by HV-annealing and was optimized.

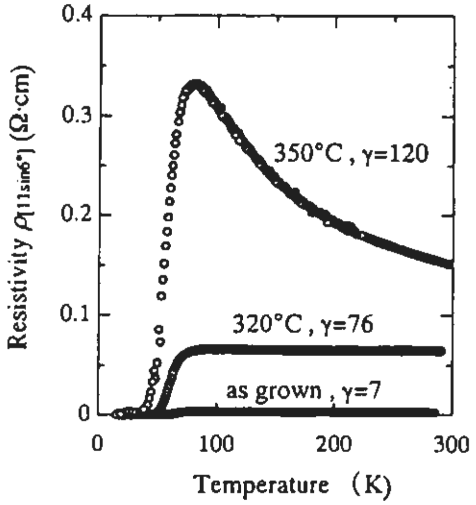
Note that, for 2212 films on tilted $LaAlO_3$ substrate in order to change the carrier density, postannealing was done in a high vacuum of 10^{-6} Pa at temperature of 320-350 °C for 1 hour in a MBE chamber. The thin films were patterned using standard photolithography and wet-etching process into a line shape along [110] $LaAlO_3$ with 20 micron in width. A 150 nm thick Au layer was evaporated on the surface of 2212 thin film for electrical contacts, and the transport properties were measured by four probe method.

Twin-free 2212 thin film on tilted substrate should exhibit an anisotropic transport behavior, because the a-b plane of these 2212 thin films are parallel to the (001) plane of the substrate and are inclined to the substrate surface. Therefore, the resistivity along the c-axis could be estimated from the resistivity along the [11sin6°] direction which has both b- and c-axis components. Although $LaAlO_3$ has a twinned structure, the orientation relationship between 2212 film and a tilted $LaAlO_3$ substrate is the same as in the case of $SrTiO_3$ reported in [6],[10],[11]. The a-axis of 2212 thin films is parallel to the step edges ([1$\bar{1}$0] $LaAlO_3$), and the b-axis accompanying incommensurate modulation is

Table I

Anisotropic transport properties of 2212 thin films on tilted $LaAlO_3$ substrates.

Sample	Annealing Temperature (°C)	$\rho_{[11\sin 6°]}$ (mΩ-cm)	γ
A	as grown	7.2	7
B	320	64	76
C	350	157	120



Fi The temperature dependence of $\rho_{[11\sin\theta]}$ for the as-grown and HV-annealed samples.

perpendicular to the step edge ([110] $LaAlO_3$).

In Table I, We present the data of anisotropic transport properties of as-grown and HV-annealed 2212 thin films prepared on the tilted $LaAlO_3$ (001) substrate. The anisotropy parameter $\gamma=(\rho_c/\rho_a)^{1/2}$ is given by [17]

$$\rho_{[11\sin\theta]}=\rho_{[110]}\cos^2\theta+\rho_c\sin^2\theta$$
$$=\rho_{[110]}(\cos^2\theta+\gamma^2\sin^2\theta), \qquad (1)$$

where θ is the tilt angle, $[11\sin\theta]$ and [110] are directions of the substrate, and $\rho_{[11\sin\theta]}$ is the resistivity obtained experimentally. Equally, it is assumed that $\rho_{[110]}=1$ mΩ-cm. A large anisotropy in resistivity was observed for HV-annealed samples, as shown in Fig.4, and the anisotropic parameters were calculated to be 76 and 120 using (1). These values are larger than that of as-grown sample, indicating that the oxygen in crystal can be removed and the anisotropic parameter can be changed. They were comparable to those of single crystals (γ=50-280) [1],[2].

B. Microwave Properties

Figure 5 (a) shows the typical current-voltage (*I-V*) characteristic and the voltage-dependent Josephson radiation spectral power at receiving frequency f_{REC}=1.7 GHz for the junction annealed at 320°C for 1 hour. The *I-V* characteristic was flux-flow type without hysteretic behavior. The reproducible Josephson emission power *P(V)* appeared at the voltage range *V*=0.22 - 0.37 mV. Note that, for single junction at receiving frequency f_{REC}=1.7 GHz, the Josephson radiation peak should appear at *V*=3.5μV, corresponding to the Josephson fundamental voltage-frequency relation. This fact reflects that the Josephson emission power may originate from the mutual phase locking of the series-connected Josephson junctions formed along the tilted substrate. As regards to our sample in the resistive state, the net voltage dropping across the junction is the total sum of the voltage drops in individual stacked intrinsic junctions along the current path. Thus, the Josephson radiation power spectrum of the junction should be delivered from the total voltage across the series array junctions according to the Johnson-Nyquest formula. From the peak voltage, we deduce that the coherence state may come from the phase-locking of N=63-106 junctions. It is estimated that 7% of total junctions are in phase.

The observed intensity of spectral power was $P(V)=7\times10^{-11}$ W. Note that, for a single 2212 bicrystal junction, the observed maximum output level for this frequency was recorded about $P(V)=1.6\times10^{-13}$ W [18]. The Josephson radiation power of 2212 intrinsic Josephson junction has about two orders of magnitude higher than that of the single bicrystal junction.

Figure 5 (b) shows microwave emission under applied magnetic field of 80G at 4.2K. For the magnetic field perpendicular to c-axis, we found clear modulation of the critical current on applied magnetic field before [19]. Hence, it is possible to optimize the maximum emission power by applying a small magnetic field. We found that the power increased about three orders of magnitude larger than that of the junction without magnetic field. The results are probably due to the tilted junction structure and the pinned vortices.

Figure 6 shows the typical *I-V* characteristics of an intrinsic 2212 junction on a tilted $LaAlO_3$ substrate under

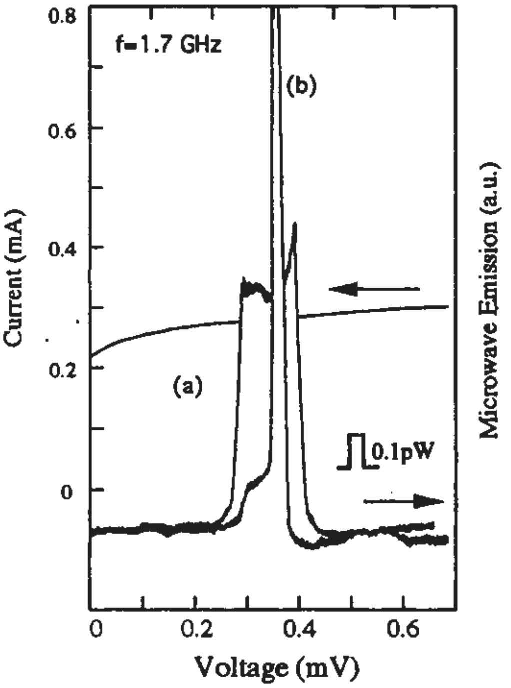


Fig.5 Typical I-V characteristics and Josephson microwave radiation power P(V) of f_{REC}=1.7 GHz for (a) B=0 and (b) B=80 G for an intrinsic 2212 junction on tiled $LaAlO_3$ substrate at 4.2K.

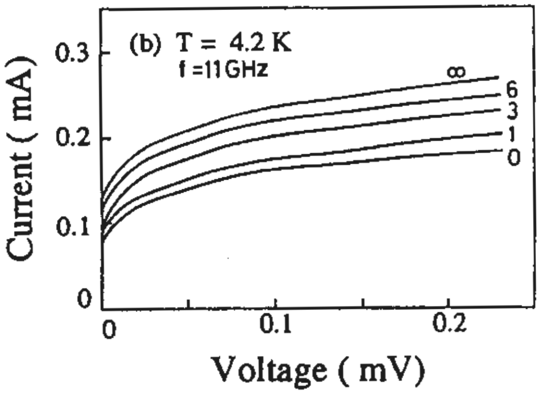


Fig.6 Typical *I-V* characteristics of an intrinsic 2212 junction on tilted $LaAlO_3$ substrate under microwave irradiation of frequency f=11 GHz at temperature T=4.2K. The numbers denote the attenuation level in dB.

microwave irradiation of frequency *f*=11 GHz at T=4.2K. We found that the Shapiro steps were smeared out, while the critical current modulation was observed. This fact indicates that the coupling between the external source and the intrinsic Josephson junction is very poor. Note that, for the series junction array model, the Shapiro step characteristics strongly depend on the junction which represents the critical current. For our intrinsic Josephson junction, we estimate that the I_cR_n product was too small to respond to the external microwave fields, suggesting that the normal resistance was very small. Such behavior was sometimes observed in 2212 bicrystal junctions [20].

IV. CONCLUSION

In summary, we have observed a large anisotropy in resistivity, and have investigated the intrinsic Josephson effect for HV-annealed 2212 thin films on tilted $LaAlO_3$ (001) substrates with 6° toward [110] direction. The observed coherent Josephson microwave radiation at frequency f_{REC}=1.7 GHz showed the occurrence of mutual phase locking of the stacked series array of intrinsic junctions along the c-axis.

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