Josephson Properties of $Bi_2Sr_2CaCu_2O_y$ Bicrystal Junctions Grown by a Sequential Deposition Technique Using Molecular Beam Epitaxy

Kiejin Lee and Ienari Iguchi

Department of Physics, Tokyo Institute of Technology, Oh-Okayama, Meguro-ku, Tokyo 152, Japan

Takayuki Ishibashi and Katsuaki Sato

Faculty of Technology, Tokyo University of Agriculture and Technology, Koganei, Tokyo 184, Japan

Hiroaki Soutome and Mitsuo Kawabe

Institute of Materials Science, University of Tsukuba, Tsukuba, Ibaraki 305, Japan

Abstract-We report Josephson and crystallographic properties of the $Bi_2Sr_2CaCu_2O_y$ junctions fabricated on MgO(100) bicrystal substrates by a molecular-beam-epitaxy method incorporating co-evaporation and sequential deposition techniques. With the sequential deposition technique which has the advantage of promoting the surface diffusion of adatoms, we obtained the highly growth-controlled films without precipitation of any second impurity phases. During the film growth, the sharp reflection high-energy electron diffraction (RHEED) patterns were also observed. However, the RHEED patterns showed the *a-b* twin structures due to the lattice mismatch, which influenced the Josephson transport properties at the junction boundary. The normal resistance of the bicrystal junction was 1.5 Ω and the I_cR_n product was 0.75 mV at 4.2 K. The Shapiro steps under millimeter-wave irradiation were clearly observed up to 65 K. We also observed the Josephson microwave self-radiation spectra at receiving frequency $f_{REC}=22$ GHz. The observed Josephson transport properties are discussed in relation to the microscopic crystallograpic properties.

I. INTRODUCTION

A detailed study of microstructures and device characteristics of high-T_c superconducting(HTSC) bicrystal Josephson junctions is very important for understanding the transport mechanism of high T_c oxide superconductor and the application to electronic devices. In contrast to YBCO bicrystal Josephson junctions, there are only a few reports on the Josephson microwave properties of the Bi-Sr-Ca-Cu-O (BSCCO) junctions [1], [2] because the epitaxial growth of the single phase thin film is more difficult than that of YBCO films and three different superconducting phases of $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ (n=1-3) are easy to be produced during the thin-film processing. Most measurements on the Josephson effect in BSCCO junctions were performed on the magnetic-field dependence of critical current. Thus, it is necessary to demonstrate whether the BSCCO Josephson junction are useful devices for microwave applications. As

comparing to $SrTiO_3$ substrates, in reality, it is more difficult to control the microstructures of the junction boundary on a MgO bicrystal substrate.

Among various techniques to grow a Bi-Sr-Ca-Cu-O thin film, molecular beam epitaxy (MBE) is the most promising method to control the microstructure in an atomic scale including epitaxial orientation, and it allows *in-situ* observation of the crystal structure by reflection high-energy electron diffraction (RHEED) during the film growth. There are two methods for preparing BSCCO thin film by MBE: coevaporation [4] and sequential evaporation [5]. In the previous report [2], we have investigated the Josephson transport and crystallographic properties of $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ (2212) bicrystal junctions using a coevaporation technique by MBE. However, it yielded the strong *a-b* twin structure and the bumpy-line boundaries including impurity phases. In this work, we report the crystallographic properties, the Josephson transport, and the microwave properties of the 2212 bicrystal junctions fabricated on MgO substrates by a sequential deposition method. We also discuss the intrinsic problems of the micro-crystal structures of 2212 film at the junction boundary. We used (100) MgO bicrystal substrate, instead of $SrTiO_3$ [3],[6], for the purpose of the high frequency microwave applications.

II. EXPERIMENTAL

A sequential deposition technique is an effective method to suppress the precipitation of impurity phases and enhance the two-dimensional growth because the surface migration of adatoms can be enhanced during the interruption period [7]. The layered 2212 films can be controlled sequentially by controlling the shutters of four Knudsen cells. The 2212 film was grown on (100) MgO bicrystal substrate with the misorientation angle 24° (5 x 5 mm^2 in size, Dowa Mining Co., Ltd.) by introducing the oxygen/ozone mixture gas at a background pressure of 2.5×10^{-3} Pa with ozone content of 10 % generated by an ozonizer. The growth temperature was 740 °C and the growth rate was kept at about 0.075 nm/s for 2212 phase. The growth interruption of 60 s after 10 s and 60 s deposition was adopted. During the film growth, the crystal structure was monitored by *in-situ* RHEED measurement. The c-axis oriented films of 30-300 nm thick
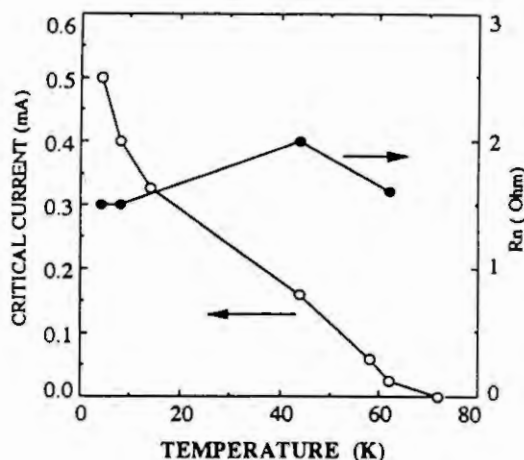
Fig. 3. Temperature dependence of R_n and I_c of a 2212 bicrystal junction prepared by the sequential deposition method on a (100) MgO bicrystal substrate.

Fig. 4. Typical I-V characteristics of a 2212 junction prepared by the sequential deposition method on a (100) MgO under microwave irradiation of the frequency f=36 GHz at 52 K. The numbers denote the attenuation level in dB.

Fig. 2 shows the RHEED intensity variation during the growth of the junction B, where the repeated deposition and interruption times were 10 and 60 s, respectively. In order to compare the RHEED intensity between the two methods, the RHEED of the junction A is shown in the inset of Fig.2. At the beginning, both of the RHEED intensities decreased because the crystallization is disturbed by the large lattice mismatch between 2212 phase and MgO substrate and then the RHEED intensity recovered for a while. As the deposition thickness was increased, the RHEED intensity of the junction B showed periodic oscillations and became fully recovered to the level before the deposition. At the end of the deposition, the films grew with smooth surface on an atomic scale. However, the RHEED pattern of the junction A did not recover the original level. These facts clearly showed the difference in the film quality between two methods. Thus, we may conclude that the sequential evaporation method is quite suitable for the growth of the high quality epitaxial 2212 thin films without impurity phases, and it is expected that the grain boundary is also aligned epitaxially according to the MgO bicrystal misorientation angle without secondary phases.

The improved microstructure of the 2212 films led to about four times higher critical current density and the transition temperature up to 72-75 K as shown in Table I. Fig. 3 shows the temperature dependence of the critical current and the normal resistance(R_n) of the junction B with dimensions of 20 μm wide and 150 nm thick. R_n was determined by suppressing the critical current by microwave radiation. The R_n value of the junction B was about three times larger as compared with that of the junction A in the whole temperature region. Comparing to the YBCO bicrystal junction [9], the junction B showed higher R_n values. These facts provide the good impedance matching for microwave circuits.
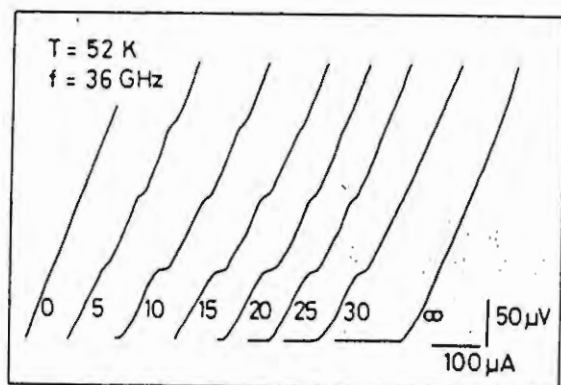
The typical I-V characteristics under microwave irradiation with the frequency of f=36 GHz at 52 K are shown in Fig. 4. The R_n of the junction B was about 1.5 Ω. The I_cR_n product and the characteristic frequency were 0.75 mV and 376 GHz at 4.2 K, respectively, which were also higher than those of the junction A. The Shapiro steps also showed rather sharp behavior. The Shapiro steps were observed for the wide range of temperature from 4.2 K up to 68 K clearly. For the Shapiro-step height dependence on the microwave irradiation, a good agreement have been achieved for small microwave power. However, for higher microwave power, significant supression of the step amplitude and critical current, as compared to the simulation based on RSJ model, has been observed. For the YBCO bicrystal junctions [9], when the higher microwave field was irradiated, the oscillation of Shapiro steps was clearly observed for step numbers much higher than those expected for the RSJ model. These facts indicate the additional mechanism of the suppression of Shapiro-steps in BSCCO junctions.

Fig. 5 shows the typical I-V characteristic and the current-dependent Josephson microwave self-radiation power $P(I)$ at 4.2 K. The distinct peak of the radiation power at receiving frequency f_{REC}= 22 GHz is found at V=45 μV, satisfying the basic Josephson-frequency relation. The calibrated observed radiation power was $P(I)$=6×10^{-13} W. The measured value is one order of magnitude smaller than that of the YBCO bicrystal junction, however, it is about four times higher than that of the junction A. Note, the Josephson microwave self-radiation strongly depends on the junction parameters, such as I_c and R_n. Considering the Josephson parameter set of the junction A, the self-radiation properties should have been much better than those of YBCO junctions. However, we could not observed the characteristics expected for the junction parameters of junction A.

In general, the characteristic frequency of the 2212 bicrystal junctions, which gives the high-frequency limit to the Josephson junction, is smaller than that of the YBCO bicrystal junctions, in spite of the same Josephson junction parameters of R_n and I_c as the YBCO bicrystal junctions as

shown in [9]. In practice, the YBCO bicrystal junctions have much better characteristics than those of BSCCO. The *I-V* characteristics of the YBCO junctions were conventional RSJ-like and the Shapiro steps could be observed up to 8 mV. On the other hand, the high-frequency properties of BSCCO bicrystal junctions appeared to be rather poor. These facts suggest that the microscopic nature of BSCCO Josephson junctions are different from that of the YBCO junctions. The poor nature of high frequency characteristics could be caused by the influence of the BSCCO crystal structure at the grain boundary containing the *a-b* twin structures and the small grain boundary facets. Thus, a special bicrystal substrate may be required in order to overcome these intrinsic limitations.
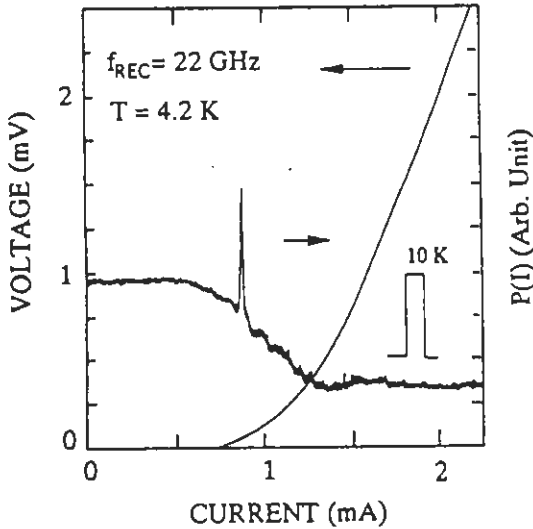

Fig. 5. Typical I-V characteristic and the Josephson microwave self-radiation signal at receiving frequency f_{REC}= 22 GHz for a 2212 bicrystal junction prepared by the sequential deposition method on a (100) MgO crystal substrate at 4.2 K. The inset bar indicates the calibration temperature of the signal of 10 K.

Note that, the *I-V* characteristics close to that predicted by the resistively shunted junction model were obtained above 30-35 K and, as temperature decreased, the *I-V* characteristics gradually changed from RSJ-type to the flux-flow type accompanying the rounding effect. These results reflect that the *I-V* characteristics of the flux-flow type with rounding at low temperature might arise from the anisotropy of the transport characteristics and the microscopic wavy structure at the bicrystal junction boundary. The imperfect crystallites such as *a-b* axes twin structure and small grain boundary facets at the bicrystal junction interface may affect the magnitude of Josephson current and the spatial distribution of Josephson transport current. In particular, the presence of strong in-plane twin structures at the junction boundary, 2212[100]‖MgO [100] and 2212[110]‖MgO [110], should affect the Josephson properties largely since each microscopic interface has two different crystal orientations with respect to the MgO substrate. For the bicrystal junctions, the

magnitude of Josephson current strongly depended on the bicrystal angle. The [100] in-plane direction should align with [100] with the angle of 12 ° and 102 ° with respect to the junction boundary. Thus, it is possible to consider two different current-phase relations and the mixture effect would be expected for the Josephson transport and microwave properties of the 2212 bicrystal junctions, because the Josephson current across the grain boundary strongly depends on the right- and left- crystal angles relatively to the interface boundary.

IV. CONCLUSIONS

We report the Josephson and the crystallographic properties of the 2212 junctions fabricated on MgO (100) bicrystal substrates by a MBE method incorporating sequential evaporation technique. With the sequential deposition technique, we obtained the highly growth-controlled films without precipitation of any second impurity phases. During the film growth, the sharp RHEED patterns were also observed. The Shapiro steps under the millimeter-wave irradiation and the Josephson self-radiation spectra at receiving frequency f_{REC}=22 GHz were clearly observed up to 68 K. In order to improve Josephson transport properties, it is necessary to use a novel substrate which can control the growth of twin structure.

ACKNOWLEDGEMENTS

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