Sputter Synthesis of c-axis YBCO Films with Excellent Surface Smoothness and Fabrication of Sandwich type Junctions with Interface Engineered Barrier

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Abstract. Flat surface is essential to base electrodes of sandwich type Josephson junctions. In the present study, c-axis YBa2Cu3O7 (c-YBCO) films with excellent surface smoothness were fabricated by off-axis sputtering. For the flat surfaces, key parameters in the sputtering process were growth temperature, atmosphere conditions and surface nature of substrates. The correlation between the sputtering conditions and the characteristic of the c-YBCO films reveal the following phenomena; i) The films deposited at low temperature of 660 °C showed a low zero-resistance temperature because of a poor connection between the c-axis crystallites. Surfaces of these low temperature grown films involve large and rectangular shaped a-axis grains, thereby the peak-to-valley (PV) amplitude of the surface was beyond 80 nm. The experiments indicate that the growth of the grains should be caused by low surface diffusion of adatoms at the low growth temperature. ii) Contrarily, at high $T_s$ above 770 °C, a serious deviation of film composition from the stoichiometry took places, which also promoted an outgrowth of a-axis grains. Consequently, the high temperature grown films had seriously rough surfaces (PV amplitude > 25 nm). iii) The growth at the optimised $T_s$ of 765 °C led a remarkable reduction of surface roughness (root-mean-square (RMS) of the roughness < 1.8 nm) without any degradation of superconducting properties. The achieved surface morphology is classified as one of the smoothest surfaces of the YBCO film grow by sputtering. iv) A density of the a-axis outgrowth was related to the surface defects of the substrate. The usage of SrTiO3 (100) plane consists of atomically flat and wide terraces and unit-cell high steps resulted in almost outgrowth-face surface with an excellent smoothness (PV amplitude < 10 nm, RMS of the roughness < 1.0 nm).

1. Introduction

High $T_c$ Superconductor Josephson junction device is expected as next-generation device, since Josephson junction has high potentials such as a high-speed switching and low-power consumption. In particular, sandwich type junctions are attractive for applying to integrated circuits [1-3]. Sputtering is a suitable technique for the HTS electrodes, because of its simplicity and potentials for wide area and uniform films. It is, however, repeatedly reported that surface of the sputtered c-YBCO films were
rougher than other methods such as co-evaporation. This rough surfaces mainly originated in both a mixed growth of a-axis crystallites in the c-axis matrix and compositional deviations due to re-evaporations of adatoms. These reports indicate that there may be narrow window of sputtering conditions for the flat surfaces. In the present study, for the excellent smooth surface suitable to engineered interface barriers of the sandwich type junctions, growth of the flat c-YBCO films have been attempted by detailed controls of sputtering conditions around the critical regions where surface diffusion and re-evaporation of the adatoms apparently change.

In order to make smooth surfaces, we have clarified correlations of the characteristics of the YBCO films and the growth conditions, such as growth temperature \( T_g \), growth atmosphere and substrates. The present study reveals that a sputter growth of c-YBCO films at the temperature just below the re-evaporation region and utilization of so-called “step-and-terrace” substrate are useful for realize excellent surface smoothness and superconducting properties. Continuous and non-superconducting engineered barriers are successfully obtained by utilizing the obtained flat surfaces.

2. Experiment

c-YBCO films were grown on (100) SrTiO\(_3\) by an off-axis rf sputtering. For obtaining smooth surface, surface diffusion of adatoms on the growing surface was controlled by growth atmosphere and temperature. Sputtering atmosphere and growth temperature were varied in the range from 20 to 40 Pa and from 760 to 770 °C, respectively. A gas mixture ratio of O\(_2\) : Ar was fixed 30 : 25. In the present study, sputtering plasma was turned on prior to a rising the substrate temperature, in order to maintain original surface features of the substrate. After the deposition, the films were annealed at 400 °C for 1 hour in oxygen atmosphere. Surface morphology of the films was characterized by an atomic force microscope (AFM). Peak-to-valley (PV) amplitude and root-mean-square (RMS) evaluate the region of width of 2 μm in the scan area of 4 μm x 4 μm. Temperature dependent of resistivity was measured by standard four probe measured. Some of the film surfaces were subsequently engineered by irradiation of Ar ion beam; kinetic energy of 500 eV, flux density 400 μA/cm\(^2\), irradiation time of 180 sec.

3. Result and Discussion

In the present study, sputtering pressure and growth temperature were firstly optimized. Then, the effect of the surface morphology of SrTiO\(_3\) with step-and-terrace on roughness was examined.

In the first step of the experiment, films were deposited on as-polished (as-received, commercial) SrTiO\(_3\) substrates at a temperature of 765 °C and pressure of 20 - 40 Pa. The films deposited at 20 Pa were non-superconducting, though surfaces of the films were rather smooth (PV amplitude < 20 nm). The film deposited at 40 Pa exhibited a superconducting transition temperature above 80 K. Their surfaces were, however, rough (PV amplitude > 90 nm). These results indicated that the low-pressure deposition should result in a degradation of superconducting property whereas the high-pressure growth was harmful to the surface smoothness. For realizing both smooth surface and high \( T_c \), films were deposited at an intermediate pressure of 25 – 30 Pa. These films showed \( T_c > 70 \) K and relatively smooth surface (PV amplitude < 40 nm). In the next-step experiment, films were, therefore, deposited at the pressure of 25 Pa.

Figure 1 shows an effect of the growth temperature on surface morphology. In the AFM image of the 200 nm thick films deposited at 760 °C, many rectangular shaped a-axis grains with edge length about some hundreds nm existed with a high density. Furthermore, some large grains, of which edge was along [100] axis of SrTiO\(_3\) surface, were grown, thereby the PV amplitude over 4 μm x 4 μm area was higher than 80 nm. Superconducting transition temperature \( T_c (\rho = 0) \) of the film was low about 78 K as shown in Figure 2(a). This poor superconductivity should be due to a poor inner-grain connections between the c-axis crystallites. These results indicate that an insufficient surface diffusion of adatoms in the low growth temperature of 760 °C should caused non-uniform growth of a-axis grains. In order to eliminate this unfavourable growth mode, the growth temperature \( T_g \) was increased to 770 °C. Figure 1(b) shows the AFM image of surface morphology of YBCO deposited at high
temperature of 770 °C. The prominent grains along the substrate’s [110] axis were completely vanished by the rise of growth temperature by 10 °C. Spirals clearly observed in this figure indicated a promotion of lateral growth of c-axis grains. The a-axis crystallites were still observed. The area density of the a-axis component in this film was almost comparable to that of the 760 °C films. \( T_r \) of the 770 °C films was also about 76 K (Figure 2(b)). In the present experiment, an apparent decrease of growth rate and a compositional deviation from the stoichiometry were observed in the films grown at \( T_r \) of the 770 °C. This selective decrease of sticking coefficients of the constituents might promote an outgrowth of a-axis grains. Consequently, the high temperature grown films had seriously rough surfaces (PV amplitude > 25 nm in the scan area of 4 \( \mu \)m x 4 \( \mu \)m). These results doubly showed that the growth of the thin film strongly depends on the growth temperature. These results also indicated the necessity of simultaneous realization of high surface diffusion of adatoms and suppression of the compositional deviation. Figure 1(c) shows the AFM image of the surface morphology of YBCO deposited at optimized temperature of 765 °C. The above-mentioned optimization of growth conditions resulted in an apparent reduction of surface roughness (RMS < 1.8 nm) without any degradation of superconducting properties as shown in (Figure 2(c)). Since this growth temperature was just below the re-evaporation like region of the growth mode, the surface diffusion of the adatoms should be maximized. An origin of the residual a-axis grains on these films should be attributed to other factors.

In order to pursue further suppression of the density of the a-axis grains, in this study, we utilized SrTiO\(_3\) (100) substrates with step-and-terrace structure. The SrTiO\(_3\) substrates with step-and-structure were prepared by annealing at 1000 °C in oxidizing atmosphere for 3 hours. Figure 3 and 4 shows the AFM image of the surface morphology of the SrTiO\(_3\) with step-and-terrace structure and the film deposited on the substrate, respectively. The usage of the SrTiO\(_3\) substrate resulted in almost outgrowth free surfaces with an excellent smoothness (maximum PV amplitude < 10 nm, RMS < 1.0 nm) without any degradation of superconducting properties as shown Figure 2(d). This surface morphology obtained by the growth at the

![Figure 1](image1.png)  
**Fig. 1** Typical AFM images of YBCO films. The scan area is 4 \( \mu \)m x 4 \( \mu \)m. (a) \( T_r \) = 760 °C, RMS = 14.5 nm (b) \( T_r \) = 770 °C, RMS = 1.2 nm, the density of a-axis grain = 4.6 / \( \mu \)m\(^2\) (c) \( T_r \) = 765 °C, RMS = 1.8 nm, the density of a-axis grain = 1 / \( \mu \)m\(^2\).

![Figure 2](image2.png)  
**Fig. 2** Temperature characteristics of resistance of the films deposited at (a) 760 °C, (b) 770 °C, (c) 765 °C. (d) shows the characteristic of the film deposited on SrTiO\(_3\) substrate with step-and-terrace structure.
critical temperature, where the growth mode changes, is classified as one of the smoothest surfaces of the YBCO film grow by sputtering.

Subsequently, some surfaces of these smooth films were engineered by irradiation of Ar ion beam. After the irradiating, the surfaces of the films became smoother. The irradiated surfaces exhibited an insulative feature in photoemission spectra. These results mean that the films deposited in optimised growth condition are useful for the electrode of the sandwich type junctions.

4. Conclusions

In order to simultaneously realize excellent surface smoothness and superconducting properties in c-axis YBa$_2$Cu$_3$O$_y$ thin films by sputtering, the effect of the growth conditions, such as growth temperature $T_g$, growth atmosphere and surface nature of substrate have been examined in detail. The results reveal that an enhancement of surface diffusion of adatoms on the growth surface without decrease of the sticking coefficient of the sputtered atoms is crucial for it. The film deposited at low temperature showed a low $T_g$ and rough surface because of mixed growth of the a-axis grains. A deviation of film composition promoted by a high growth temperature also resulted in co-existence of the a-axis component. By the precise control of growth temperature just below the latter region, smooth surfaces are realized without degradation of superconducting properties. For the films grown under the optimized sputtering conditions, the utilization of the smoothed SrTiO$_3$ substrate with step-and-terrace surface resulted in further remarkable improvements of surface smoothness; PV amplitude < 10 nm, RMS < 1.0 nm, which is one of the smoothest surface among YBCO sputtered films. Present study revealed that the comprehensive optimization of both sputtering parameters and surface nature of substrate should be a key to simultaneously obtain the YBCO film with excellent surface smoothness and high $T_c$. These results and the further improvement of the smoothness by the Ar ion irradiation indicate that the obtained smooth c-YBCO should be one of the most suitable base electrode materials for sandwich type junctions.

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