# Oxide Buffer Layer With Perovskite Structure for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> Coated Conductors Prepared by Metal-Organic Deposition Method

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Abstract—For fabricating YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> (YBCO) coated conductors on metal substrates with high critical current density  $(J_c)$ , deposition of oxide buffer layers is very important. BaZrO<sub>3</sub> (BZO) and BaSnO<sub>3</sub> (BSO) with a perovskite structure have excellent chemical compatibility with YBCO and good lattice matching with oxide substrates, including NiO buffer by surface oxidation epitaxy process. We have fabricated BZO or BSO buffer layers on SrTiO<sub>3</sub> single-crystal substrates by metal-organic deposition (MOD) method, in order to investigate the effects of buffer layers on superconducting properties of YBCO films.  $J_c$  of YBCO films deposited by pulsed laser deposition on BZO reached 0.29 MA/cm<sup>2</sup> (77 K, 0 T).

*Index Terms*—BaSnO<sub>3</sub>, BaZrO<sub>3</sub>, MOD, perovskite buffer layer, YBCO.

#### I. INTRODUCTION

ARIOUS substrates have been investigated for a coated conductor application of  $YBa_2Cu_3O_{7-x}$  (YBCO) superconductor. Rolling-assisted bi-axially textured substrates (RABiTS) [1], ion-beam-assisted deposition (IBAD) [2] and surface-oxidation epitaxy (SOE) [3] have been developed for high  $J_c$  conductor applications. These methods require oxide buffer layers on metal substrates to prevent interdiffusion between YBCO and underlying metals. YSZ and CeO<sub>2</sub> buffers deposited by physical vapor deposition have been investigated in many studies. However, it has been reported that BaZrO<sub>3</sub> and BaCeO<sub>3</sub> compounds are likely to be formed at the interface between YBCO and the buffers at high temperature [4] and thus to reduce  $J_c$  values of the YBCO films.

BaZrO<sub>3</sub> (BZO: a = 0.4193 nm) [5] and BaSnO<sub>3</sub> (BSO: a = 0.4116 nm) with perovskite structure are candidates of buffer layers due to their excellent chemical compatibility with YBCO and also to their good lattice matching with SOE-NiO (a = 0.4177 nm) substrate. Both BZO and BSO include a BaO layer in them, so these buffer layers have possibility to suppress the formation of Ba compounds resulting from the reaction between the YBCO film and the buffer.

Another interesting route to prepare buffer layer is metal-organic deposition (MOD) method, which has high potentiality for many applications. This process does not require a high vacuum

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Fig. 1. Annealing profile of MOD process for crystallization of calcined films.

system and a complex apparatus, so that we can fabricate buffer layers with low-cost.

In this work we have prepared BZO and BSO buffer layers with perovskite structure by MOD method on  $SrTiO_3$  (STO) single-crystal substrate and investigated the effects of these buffer layers on structural and superconducting properties of YBCO films deposited by pulsed laser deposition (PLD).

## II. EXPERIMENTAL

We prepared two homogeneous solutions having molar ratios of Ba : Zr = 1 : 1 and Ba : Sn = 1 : 1 by dissolving metal naphthenates in toluene. Subsequently the solutions were diluted to the 0.15 mol/l. Using these solutions precursor films were spin-coated onto STO (001) single-crystal substrates for 30 seconds and at 3500 rpm. After drying, the films were calcined at 450 °C for 20 minutes in air. The calcined films were then crystallized in air with annealing procedure as shown in Fig. 1. Crystallization temperatures were set at 700 °C, 800 °C and 900 °C. Film thickness of these buffer layers were about 100 nm.

YBCO films were deposited on these perovskite buffer layers using PLD with a KrF excimer laser ( $\lambda = 248$  nm). Deposition conditions were as follows: laser energy of 340 mJ, laser pulse repetition rate of 10 Hz, laser pulses of 10 000 shots and an oxygen pressure of 200 mTorr. Substrate temperatures during deposition were varied from 690 °C to 750 °C. After laser ablation the samples were cooled to room temperature, keeping in O<sub>2</sub> pressure of 500 Torr in the chamber. The thickness of YBCO films was about 500 nm.

Crystal orientation of buffer layers and YBCO films were determined from  $\theta$ -2 $\theta$  scans and rocking-curves using X-ray



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Fig. 2. XRD profiles of BaZrO\_3 films on STO(001) substrate annealed at 700  $^\circ\text{C}\text{-}900\ ^\circ\text{C}\text{.}$ 



Fig. 3.  $\phi$ -scan result of BaZrO<sub>3</sub> (111) plane annealed at 700 °C.

diffraction (XRD), and in-plane orientation was confirmed by  $\phi$ -scans using four-circle XRD. Surface morphology of the films was observed by atomic force microscopy (AFM) and scanning electron microscopy (SEM).  $J_c$  values of YBCO films were measured by dc four-probe method.

### **III. RESULTS AND DISCUSSION**

## A. BaZrO<sub>3</sub> and BaSnO<sub>3</sub> Buffer Layers

The  $\theta$ -2 $\theta$  profiles of BZO buffer layers annealed at 700 °C, 800 °C and 900 °C and the  $\phi$ -scan profile of BZO buffer layer annealed at 700 °C are shown in Figs. 2 and 3. All the BZO films presented *c*-axis and in-plane orientations. For the BZO film annealed at 700 °C with the smoothest surface, the full width at half maximum (FWHM) of BZO (002) plane rocking-curve was 0.49°, and the FWHM of BZO (111) plane  $\phi$ -scan was 1.22°. Fig. 4 shows the surface morphology of BZO buffer layers observed by SEM. The grain size of the BZO layer annealed at higher temperature was larger than that annealed at lower temperature. The surface of BZO annealed at 700 °C was highly smooth and the surface roughness (Ra) was 1.8 nm.

*c*-axis and in-plane orientations of BSO films are shown in Figs. 5 and 6. The change of surface morphology with temperature is also shown in Fig. 7. The smoothest surface was obtained



Fig. 4. SEM images of the BaZrO\_3 films annealed at (a) 700  $^\circ C$ , (b) 800  $^\circ C$ , and (c) 900  $^\circ C$ .



Fig. 5. XRD profiles of  $BaSnO_3$  films on STO(001) substrate annealed at 700  $^\circ\text{C}\text{-}900\ ^\circ\text{C}\text{.}$ 

at 900 °C. The FWHM of BSO (002) plane rocking curve was  $1.68^{\circ}$  and the FWHM of BSO (112) plane  $\phi$ -scan was  $2.71^{\circ}$  for the BSO buffer annealed at 900 °C. The crystalline quality of BSO buffer layer was worse than that of BZO buffer layer, although their misfit values with STO substrate and thermal expansion coefficients were not so different. The grain size of BSO annealed at higher temperature (900 °C) was larger than that annealed at lower temperature, also observed in the case of BZO. Ra of BSO annealed at 900 °C was 3.4 nm.



Fig. 6.  $\phi$ -scan result of BaSnO<sub>3</sub> (112) plane annealed at 900 °C.



Fig. 7. SEM images of BaSnO\_3 films annealed at (a) 700  $^\circ C$  (b) 800  $^\circ C$  , and (c) 900  $^\circ C.$ 

#### B. YBCO on BZO/STO and BSO/STO

YBCO films were deposited by PLD at substrate temperatures of 690-750 °C on both BZO and BSO buffers, in order to investigate the effects of the buffers on superconducting properties of YBCO. Here, BZO buffers annealed at 700 °C and BSO buffers annealed at 900 °C were used. The YBCO film deposited at 730 °C on BZO showed the highest  $J_c$  value of 0.29 MA/cm<sup>2</sup> (77 K, 0 T). On the other hand,  $J_c$  of YBCO deposited at 730 °C on BSO was only 0.03 MA/cm<sup>2</sup>. FWHM



Fig. 8. XRD profiles of YBCO deposited at  $730\,^{\circ}\text{C}$  (a) on BZO and (b) on BSO.



Fig. 9. SEM images of YBCO films deposited at 730  $^\circ C$  (a) on BaZrO\_3 and (b) on BaSnO\_3.

values of YBCO (005) plane rocking curves for YBCO/BZO and YBCO/BSO were  $0.93^{\circ}$  and  $1.12^{\circ}$ , respectively. The difference of the FWHM values between them was smaller than that between BZO and BSO buffers. XRD profiles of YBCO/BZO and YBCO/BSO deposited at 730 °C are shown in Fig. 8, indicating the existence of *a*-axis oriented grains in the YBCO layer on the BSO buffer. Fig. 9 shows SEM images of YBCO surfaces deposited at 730 °C on BZO and BSO buffer layers. The surface of YBCO deposited on BSO was rougher than the surface of YBCO deposited on BZO. The rectangular grains and disk-shaped grains were also observed in YBCO deposited on BSO.

YBCO films deposited on the buffer layers with a perovskite structure are easily bi-axially oriented due to the excellent compatibility between YBCO and the buffers, provided that the surface roughness of the buffer layers is low. The surface of the YBCO film on the BZO buffer was relatively smooth, but the surface of YBCO on BSO buffer was not so smooth. In the latter case, the crystallinity of YBCO film becomes worse due to the rough surface of BSO, especially in grooved area between BSO grains. The degraded crystalline areas became the nucleation sites of rectangular and disk-shaped grains [6]. Nucleation and growth of these grains made the surface of YBCO rough, leading to a reduction of  $J_c$  of the YBCO film. Surface roughness of buffer layer is one of the key factors in order to fabricate the better buffer layers for YBCO coated conductor using MOD method.

## **IV. CONCLUSION**

We have investigated the deposition of BZO and BSO buffer layers by MOD method and have successfully fabricated *c*-axis and in-plain oriented buffer layers on STO single-crystals. Subsequently YBCO films were deposited by PLD on both BZO buffer layer annealed at 700 °C and BSO buffer layer annealed at 900 °C. A  $J_c$  value of 0.29 MA/cm<sup>2</sup> was obtained for YBCO/BZO/STO sample. On the other hand,  $J_c$ of YBCO/BSO/STO sample was lower. The rectangular and disk-shaped grains were frequently observed on YBCO films deposited on BSO buffers. These grains were originated from the rough surface of BSO buffer layers and  $J_c$  of the films were degraded. Control of the roughness of buffer layers is one of the key factors for YBCO coated conductor fabrication by MOD method.

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