Microcrack-free and high $J_c$ thick YBCO films on vicinal $r$-cut sapphire buffered with CeO$_2$

J. C. Nie $^{1,2}$, H. Yamasaki $^2$, Y. Nakagawa $^2$, K. Develos-Bagarinao $^2$, M. Murugesan $^2$, H. Obara $^2$, and Y. Mawatari $^2$

$^1$Department of Physics, Beijing Normal University, Beijing 100875, China
$^2$Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki, 305-8568, Japan

E-mail: jcnie@bnu.edu.cn

Abstract. By growing CeO$_2$ buffer layers on deliberately miscut Al$_2$O$_3$ (1102) surfaces, we have successfully fabricated stoichiometric YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thick films by pulsed laser deposition. Atomic force microscopy and scanning electron microscopy observations demonstrated that YBCO films with thickness up to $t \sim 1.0$ $\mu$m were microcrack-free. Characterization of the films revealed a porous morphology, consisting of interconnected islands and deep holes (pores). This feature is considered as one of the sources contributing to the strain-relieving mechanism responsible for the increase in film thickness without microcracking. Microcrack-free thick YBCO films revealed $T_c = 90.5 \pm 0.5$ K, $J_c(77.3$ K, 0 T ) $= 2.0\text{--}3.0 \times 10^6$ A/cm$^2$, and a substantial enhancement of $J_c \times t$ (77.3 K, 0 T ) up to 246 A/cm.

1. Introduction

YBCO thin films deposited on sapphire substrates have been proposed as ideal base materials for resistive fault current limiter applications [1,2]. Sapphire is the most ideal substrate due to its good thermal conductivity, good mechanical strength, as well as its availability in large areas at relatively low cost compared to other single crystal substrates. In particular, as switching elements for resistive fault current limiters, thick YBCO films are necessary to increase the critical current per unit width (i.e., $J_c \times t$ product, here $J_c$ is the critical current density and $t$ is the film thickness) and thereby enhance the nominal power. However, it is well-known that microcracking occurs in YBCO films deposited on CeO$_2$-buffered sapphire when the thickness exceeds a critical value of $\sim 300$ nm [3]. The reason for cracking is attributed to the thermal strain during cooling (from the deposition temperature) since the expansion coefficient of YBCO is about two times larger than that of sapphire. Microcracking is a big problem towards the use of YBCO films in practical applications. The presence of microcracks in YBCO films leads to a drastic reduction of $J_c$ and a serious degradation of the microwave characteristics. Efforts have been made by shifting the film composition to Y-rich to prevent the formation of microcracks [4,5]. However, together with the suppression of microcracking a deviation in composition from Y-123 phase and a decrease of a factor of 2--3 in $J_c$ were observed.

Moreover, the mismatch between [1120] axis of sapphire (4.76 $\text{Å}$) and [100] axis of CeO$_2$ (5.41 $\text{Å}$) is remarkable (about 12%). This means that lattice matching needs in principle one misfit for every 8 lattices of [1120] sapphire. In this case mismatch between 7 lattices of CeO$_2$ [100] and 8 lattices of sapphire [1120] is 0.55%. If there is a single step of sapphire [1102] between the two adjacent (1102)
planes, the mismatch will be greatly released. This will result in a vicinal surface with tilt angle of 5.22°. More advantages of miscut Al₂O₃ (1102) are: (i) a vicinal surface could provide a large number of steps for initial nucleation and lead to a rapid step-flow growth, which has been found to suppress the formation of precipitates [6] and to enhance $J_c$ of YBCO films; (ii) step-flow growth often forms step-bunches and there are many pores between the step-terraced grains [7]. Our strategy is to make use of such pores to release the thermal strain during the cooling. In addition, the $a$-$b$ plane of YBCO on the vicinal substrate has a free termination at the surface, which could further enable the strain relaxation of the YBCO films. In this paper, we report the successful preparation of microcrack-free (up to 1.0 μm thick) and high $J_c$ YBCO films on the deliberately miscut Al₂O₃ (1102) (5.22° off toward [1120]) substrates buffered with CeO₂.

2. Experimental

CeO₂ buffer layers were first deposited by pulsed laser deposition (PLD) utilizing a KrF excimer laser source (248 nm wavelength, LPX305xi) operated at 300 mJ. The oxygen pressure was 300 mTorr and the substrate temperature $T_s$ was held at 810°C. The deposition rate was about 0.028 nm/pulse at a laser repetition rate of 1 Hz and the thickness of the CeO₂ layers was about 30 nm. Immediately after preparation of the CeO₂ layer, the substrate temperature was changed to $T_s = 760°C$ for the preparation of YBCO. The laser fluence of ~ 4.5 J/cm² (600 mJ) on a YBCO target at 5 Hz laser repetition rate was used for the YBCO deposition. Deposition was carried out in 300 mTorr of O₂ ambient at a rate of 0.133 nm/pulse. After deposition the samples were annealed in 400 Torr oxygen at 430°C for 1 hour and then cooled down to room temperature within 40 min.
Structural properties of the films were measured by using a so-called “in-plane” X-ray diffractometer (ATX-G, Rigaku Co.) [8,9]. The surface morphology of the films was examined by atomic force microscopy (AFM) and scanning electron microscopy (SEM). The microscopic composition of the films was analyzed by energy dispersive X-ray spectroscopy (EDS).

To measure the $J_c$ of the vicinal YBCO films, 2-mm-long and 20-μm-wide bridges were etched in the YBCO films by pulsed laser etching, in $L$ and $T$ directions, respectively. The current-voltage curves of these bridges were measured by a four-probe transport method using pulse currents. A voltage criterion of 1 μV/mm was used to define $J_c$. We specify $\theta$, the tilt angle of $H$ with respect to the vertical direction of the film plane; the field is always perpendicular to the direction of the current through the bridge. The $\theta$ was varied from 0 to 360°.

3. Results and Discussion

Figure 1 shows 10×10 μm AFM images and corresponding profiles of eight typical YBCO films with thicknesses of 200, 600, 1000, 1200, 1400, 1600, and 2400 nm, respectively, on vicinal Al$_2$O$_3$ (1102) buffered with 30-nm-thick CeO$_2$. Line scan profiles show that the root mean square (rms) surface roughness was in a range of 15–26 nm and increased monotonically with increase of thickness. The thin (e.g., 200 nm) YBCO film showed small islands with nanoscale terraces separated by parallel steps along Al$_2$O$_3$ [1101] (by small-scale AFM checking, data not shown), suggesting that the YBCO films grown by a step-flow growth mechanism. In the following, the directions which are longitudinal and transverse to these step edges will be called $L$ (i.e., along Al$_2$O$_3$ [1101]) and $T$ (i.e., along Al$_2$O$_3$ [1120]), respectively (see Fig. 1a). The islands were frequently interrupted in the $L$ direction at ragged steps, forming irregular-shaped nanoholes (pores) between the islands. The discontinuity of the YBCO terraces might be related to strain fields in the vicinity of grown-related defects. The existence of substrate surface defects resulting from a small additional miscut in $L$ direction can be responsible for the observed local discontinuity of the terraces. With increase of thickness, YBCO films grown on the vicinal substrates consisted of interconnected islands, showing also the presence of larger and deeper pores between the islands. The characteristic slope of the step-terraced islands was frequently observed to be ~ 4° away from the sample surface in $T$ direction, this is well consistent with the XRD observations which showed that the CuO planes of YBCO was tilted 4.06° from the substrate surface toward Al$_2$O$_3$ [1120]. In addition, the combination of AFM and SEM observations demonstrated that YBCO films with thickness up to $t \sim 1.0$ μm were microcrack-free and stoichiometric (by EDS). Over 1.2 μm, short-microcracks were observed on the vicinal YBCO films [10]. The porous morphology and the inclination or termination of CuO planes are considered to contribute to the strain-relieving mechanism responsible for the increase in film thickness without microcracking.

YBCO films grown on the CeO$_2$-buffered miscut sapphire exhibited only one epitaxial orientation with YBCO(001) || CeO$_2$(001) || Al$_2$O$_3$(1102) and YBCO[110] || CeO$_2$(100) || Al$_2$O$_3$(1120). After YBCO film (600 nm) deposition the lattice parameters of the CeO$_2$ layer were distorted to $b = 0.54224$ nm and $c = 0.53924$ nm, while the corresponding lattice parameters of a 30-nm-thick CeO$_2$ single layer are $b = 0.5416$ nm and $c = 0.54054$ nm [9]. Moreover, although the full width at half-maximum (FWHM) of CeO$_2$(202) $\phi$-scan, $\Delta \phi$ (e.g., ~ 1.68°), was almost unchanged after YBCO deposition, the FWHM of CeO$_2$(002) rocking curve, $\Delta \omega$ (typically ~ 0.27°), was about 4 times larger than that of the single CeO$_2$ layer (about 0.07°) [8]. Since the mean thermal expansion coefficient of CeO$_2$ is between that of YBCO and sapphire [3], this distortion could be an effect of the different thermal expansion coefficients of Al$_2$O$_3$, CeO$_2$ and YBCO, which leads to extending the strain within the YBCO to the CeO$_2$ layer during cool down from deposition temperature to room temperature.

Fig. 2(a) shows the magnetic field dependence of $J_c$ at different temperatures towards $L$ and $T$ direction for a 800-nm-thick YBCO film. $J_c$(77.3 K, 0 T) = 2.68×10^6 A/cm$^2$ and 2.46×10^6 A/cm$^2$, $J_c$(77.3 K, 8 T) = 3.48×10^4 A/cm$^2$ and 1.66×10^4 A/cm$^2$, towards the $T$ and $L$ direction, respectively. Evidently, the film showed superior $J_c$ at all the measured temperatures towards the $T$ direction than towards the $L$ direction. Fig. 2(b) shows the thickness dependence of $J_c,T$ and $J_c,T\times T$ for different films
at zero field and 77.3 K. For the 1000-nm-thick microcrack-free YBCO film, a $J_{c,T}$ value of $\sim 2.46 \times 10^6$ A/cm$^2$ and a $J_{c,T,u,t}$ value of $\sim 246$ A/cm were obtained. With further increase of the YBCO film thickness, however, $J_c$ as well as $J_{c,T,u,t}$ drastically decreased, probably due to the formation of short-microcracks observed by SEM.

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