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Effect of deposition conditions on $YBa_2Cu_3O_{7-\delta}$ thin films by inverted cylindrical magnetron sputtering and substrate effects

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Abstract

The dependence of YBCO thin film properties on the deposition conditions was studied for different substrates. The deposition conditions were optimized for the epitaxial growth of high quality YBCO thin films of 1500 Å thickness onto single crystal (100-oriented) $SrTiO_3$ (STO), MgO and $LaAlO_3$ (LAO) substrates by DC Inverted Cylindrical Magnetron Sputtering (ICMS). The samples were investigated in detail by means of X-ray diffraction analysis (XRD), EDX, AFM, $\rho - T$, magnetic susceptibility and current–voltage ($I - V$) characterizations. The samples show strong diamagnetic behavior and sharp transition temperatures of 89–91 K with $\Delta T < 0.5$ K. XRD of the samples exhibited highly *c*-axis orientation. The full width at half maximum (FWHM) values of the rocking curves were ranging typically from 0.22 to 0.28°. The samples have smooth surfaces as shown from AFM micrographs. The surface roughness, R_a, changed between 5–7 nm. $I - V$ characteristics were obtained from the 20 μm-wide microbridges, which were patterned by a laser writing technique. The critical current densities (J_c, 1.06×10^6 for LAO-based YBCO, 1.39×10^6 for MgO-based YBCO, 1.67×10^6 A/cm^2 for STO based YBCO) of the microbridges were evaluated from $I - V$ curves at 77 K.

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1. Introduction

There are different methods of epitaxially growing $YBa_2Cu_3O_{7-\delta}$ thin films with smooth surfaces and optimal performance for device applications. In recent years, the researches in this topic have been increased enormously due to the requirement of high-T_c superconducting devices based on highly *c*-axis oriented YBCO thin films, such as microbridges, Josephson junctions and SQUID's [1–4]. In the process of epitaxial high-T_c superconducting thin film fabrication by DC sputtering, there are many parameters involved which all have to be taken into account. The YBCO thin film growth conditions of major importance are substrate preparation [5,6], target condition, target-substrate distance, gas mixture, growth temperature, deposition rate, oxygenation temperature and cooling rate. All these parameters should be optimized to obtain reproducible, homogeneous and epitaxial *c*-axis thin films on different substrates. Much effort recently has been devoted to search for the deposition conditions leading to epitaxial growth of YBCO thin films with superior superconducting properties. Degardin et al. [6] have reported that the substrate preparation is a major step for the elaboration of good quality superconducting films. Tsukamoto et al. [7] have reported that the film quality strongly depends on the target-substrate distance and the pressure. Taniwaki et al. [8] have reported that the crystalline structure and the crack formations in the YBCO thin films strongly depend on the substrate temperature. Zeng et al. [9] have reported that the film quality strongly depends on the sputtering pressure. Generally, the influences of these parameters on the YBCO thin film quality have been extensively studied for one type

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of substrate with varying deposition parameters [7–14]. Liu et al. [13] have commented that the self-consistent picture of the relationship between the deposition rate and the microstructure of YBCO thin films is lacking. To generalize this comment, in addition to the deposition rate, the other parameters should also be taken into account for obtaining YBCO thin films having superior superconducting properties on different substrates.

Epitaxial growth requires the controlled crystallographic orientation of the film with respect to the substrate. Generally this requires matching of the film and substrate lattice parameters, atomic position, crystallographic orientation, etc. The better the match of all these parameters, the more likely high quality epitaxial growth is to occur. The FWHM values of the rocking curves of the (005) peak characterized the degree of alignment of the *c*-axis with the normal to the substrate [13].

In this study, we systematically investigated the reproducibility of high quality YBCO thin film preparation on substrates of single crystal $SrTiO_3$, MgO and $LaAlO_3$ by using inverted cylindrical magnetron sputtering (ICMS) technique with fixed deposition parameters. The samples were studied by structural (XRD, EDX, AFM), magnetic ($\chi-T$) and electrical ($\rho-T$, current–voltage ($I-V$)) measurements. $I-V$ characteristics were measured on 20 μm-wide microbridges, which were patterned by laser writing technique [15]. This patterning technique was used to obtain narrow superconducting lines near the center of the YBCO films by directly applying the focused laser beam to the surface of the film without using any protection layer (mask or photoresist).

2. Experimental

$YBa_2Cu_3O_{7-\delta}$ thin films were deposited by in situ DC ICMS technique on (100-oriented and $10 \times 10 \times 0.5$ mm^3-sized) single crystal $SrTiO_3$, MgO and $LaAlO_3$ substrates. This sputtering method is an effective technique for the preparation of good quality films [16]. The sputtering device, as well as the YBCO target, was commercially purchased from Hitec-Materials [17]. The target-substrate distance was optimized at 35 mm. Besides the deposition conditions, the cleaning of the substrate surfaces within the appropriate solutions is a very important process for the epitaxial thin film growth. In this study, the substrates were cleaned with ultrasonic cleaner in acetone for 1 h and subsequently in isopropyl alcohol for 1 h. The substrates were attached to a Teflon holder with an angle of 45°. Then the ultrasonic cleaner was started by adjusting the temperature of the water inside to 30 °C. After cleaning, the substrate was glued to the substrate holder of sputter system with silver paste and the Ar partial pressure was adjusted to 0.5 mbar. The target was pre-sputtered in flowing Ar gas for 45 min. under 45 W power during which the temperature of the sample holder was increased up to the deposition temperature of 800 °C. This pre-sputtering is required for eliminating any contamination on the surface of the target. At 800 °C, the oxygen gas was introduced into the chamber to attain the partial pressure of oxygen of 0.1 mbar and finally the total pressure inside the chamber was 0.6 mbar. At these conditions thin film deposition was started with fixed deposition parameters; total pressure of 0.6 mbar, a substrate temperature of 800 °C, DC power of 45 W. The resulting deposition rate was about 1.3 nm/min. After the deposition, pure oxygen was introduced into the deposition chamber with the pressure of 1 mbar while the temperature was fixed at 800 °C for 10 min and then temperature was allowed to fall down to 600 °C at a rate of 20 °C/min. At the beginning of the cooling from 800 to 600 °C, oxygen pressure was increased up to 700 mbar and films were held at 600 °C for 20 min, then the temperature was allowed to fall down to room temperature at a rate of 30 °C/min.

We have performed resistivity-temperature ($\rho-T$) cryogenic measurements using a computer controlled Oxford Instruments Edwards Cryostat. These measurements were used to control the resistive behavior of the films depending on the temperature. For electrical measurements, gold (Au) pads were evaporated on the four points of the films and ohmic contacts were made using silver paste onto the gold pads. For $\rho-T$ measurements, the samples were cooled from room temperature to below T_c at a typical rate of 0.5 K/s adjusted by an ITC-502 temperature controller. Also, Keithley 220 Current Source and Keithley 6517 Electrometer were attached to the measurement system with standard GPIB interface and IEEE 488.2 card to achieve a computer controlled analyzing system. Current reversal technique was used to eliminate the effects of voltage

Table 1
Properties of the $YBa_2Cu_3O_{7-\delta}$ thin films deposited on different substrates

Sample	Substrate and crystal orientation	Thickness of the film (~Å)	Width of the bridge (~μm)	T_c ($R=0$) (K)	ΔT_c (K)	I_c (mA) (at 77 K)	J_c (A/cm^2) (at 77 K)	Mean roughness R_a (nm)	FWHM(°)
YBCO	$SrTiO_3$ (100)	1500	20	91	<0.5	50.16	1.67×10^6	4.6	0.22
YBCO	MgO (100)	1500	20	90	<0.5	41.63	1.39×10^6	6.2	0.26
YBCO	$LaAlO_3$ (100)	1500	20	89	<0.6	31.83	1.06×10^6	6.8	0.28

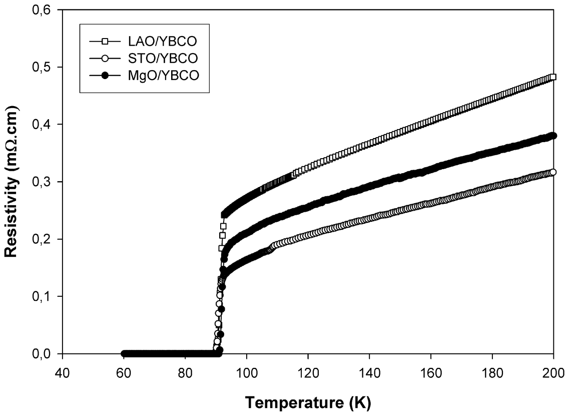

Fig. 1. Resistivity vs. temperature curves of the YBCO thin films on different substrates.

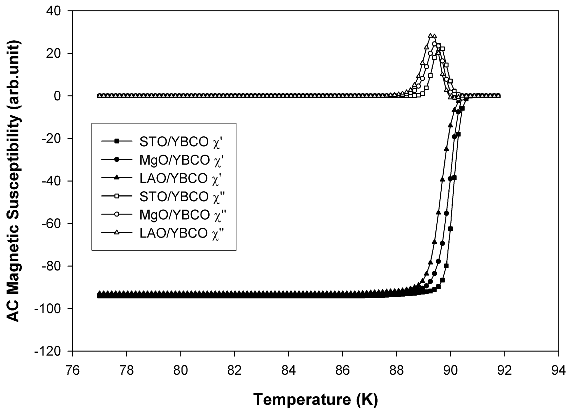

Fig. 2. Magnetic susceptibility vs. temperature curves of YBCO thin films on different substrates.

offsets. The AC complex susceptibilities ($\chi' - i\chi''$) of the samples were measured in a two-coil system using a two-phase SR530 model Lock-in amplifier to pick up in-phase (χ') and out-of-phase (χ'') signal of the secondary coil with AC magnetic field aligned perpendicular to the film surface. The excitation field within the primary coil was $h(t) = h_0 \sin 2\pi f t$ with $f = 1$ kHz and the amplitude h_0 was 18.5 A/m. For the structural analysis, XRD measurements were performed. Surface morphology and energy dispersive X-ray analysis were studied by AFM and EDX. Characterized films were patterned as microbridges by using a laser-writing technique. The commercial Solid State Laser System, has computer controlled $X-Y$ translation stage. The films were patterned with a focused laser beam resulting in about 20 μm-wide microbridges at the center of the samples. $I-V$ characteristics of the microbridges were measured at constant temperature of 77 K.

3. Results and discussion

The properties of our films are summarized in Table 1. Fig. 1 shows the temperature dependence of the resistivity. One observes a metallic behavior in the normal state between the room temperature and T_c having slope passing by very close to, but above the origin. The transition widths are about 0.5 K as given in Table 1. The normal state resistivity behavior of the films is already different depending on the substrates. In this study, the lowest normal state resistivity and highest T_c (91 K) were observed for the sample sputtered on $SrTiO_3$ but the highest normal state resistvity and the lowest T_c (89 K) were observed for the film sputtered on $LaAlO_3$. Fig. 2 shows real (χ') and imaginary (χ'') parts of the AC magnetic susceptibility responses of the samples for applied magnetic fields. The sharpness' of the transitions of these curves indicate the homogeneity of the films. In the reports [7,9], YBCO thin films were prepared with transition temperatures of about 85–88 K and transition widths (ΔT) greater than 1–2 K.

XRD patterns of the YBCO thin films grown at optimized growth parameters are shown in Fig. 3 where the c-axis orientation of the YBCO films can be seen clearly. The full width at half maximum (FWHM) values measured from rocking curves on the YBCO (005) peaks are 0.22° for $SrTiO_3$-based sample, 0.26° MgO-based sample and 0.28° for $LaAlO_3$-based sample. Good quality high-T_c films, determined by the figure of merit, such as critical current density, morphology, stability over time are those which grow epitaxially on the substrates of interest. The FWHM values of (005) diffraction peak of samples were employed to represent the thin films crystalline quality. In literature, the FWHM values of about 0.4° have been reported [7]. In our study, the FWHM values are in the range of 0.22–0.28°. These are the typical values for highly c-axis orientation of YBCO thin films. Some selected properties of the substrates that used in this study are listed in Table 2 [18].

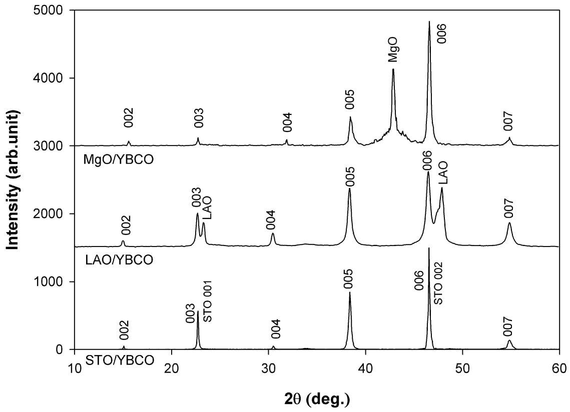

Fig. 3. XRD analysis of YBCO thin films on different substrates.

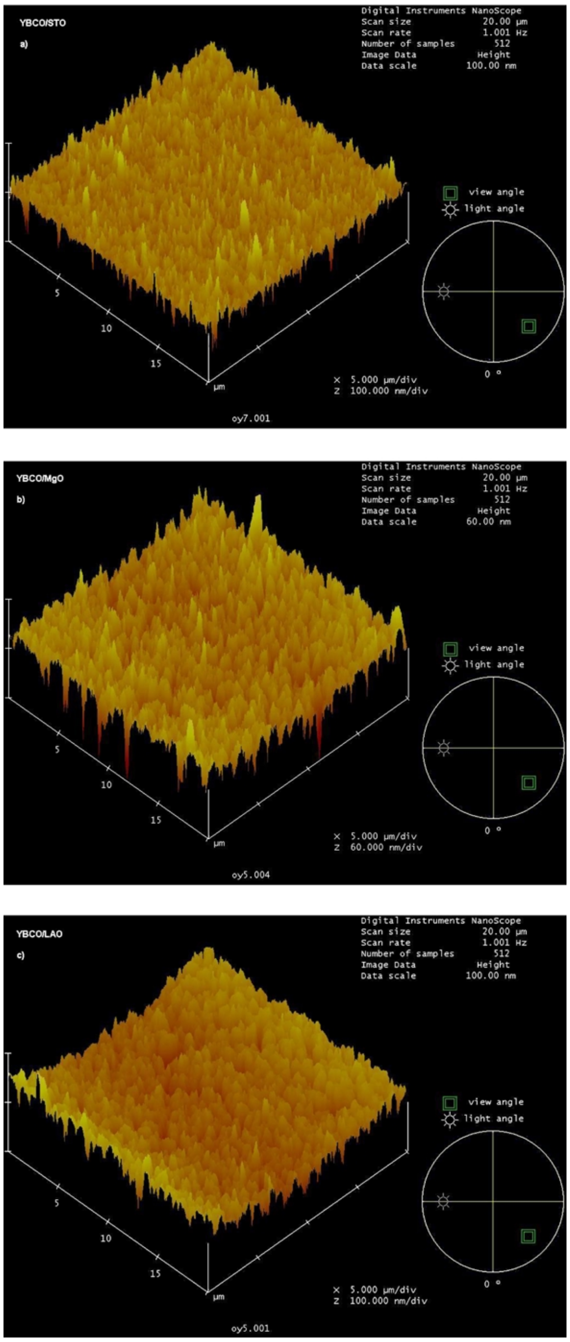

Fig. 4. AFM images of YBCO thin films: (a) YBCO on STO, (b) YBCO on MgO, (c) YBCO on LAO.

The AFM images were also taken to provide a clearer insight into the surface morphology. As shown in Fig. 4, the surfaces of the films were found to be quite smooth. The mean surface roughness, R_a, of the samples were measured with scanning areas of $20 \times 20\ \mu m^2$. R_a values are; 4.6 nm for STO based YBCO thin film, 6.2 nm for MgO based sample and 6.8 nm for LAO based sample. These values are typical and in well agreement with the SEM and XRD results about the smoothness and microstructural properties of the samples. For all measurements, the film sputtered on $SrTiO_3$ substrate showed many superior values compared to the other substrates, which means that this substrate has the best lattice match with the YBCO thin film. The energy dispersive X-ray analyses, EDX, results are given in Fig. 5. From these analyses, we found that there is no detectable impurity phases in the YBCO structure.

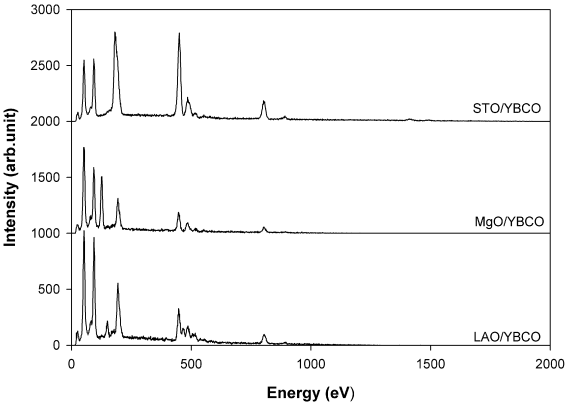

Fig. 5. EDX analysis of YBCO thin films on different substrates.

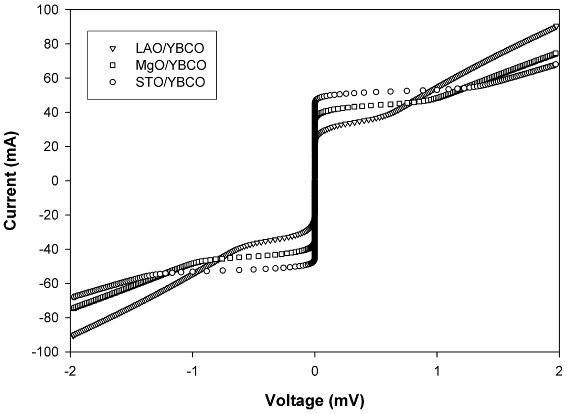

Fig. 6. Current–voltage characteristics of the YBCO microbridges on different substrates at 77 K.

The current-voltage ($I-V$) characteristics of the microbridges were measured at the constant temperature of 77 K (Fig. 6). The critical currents of the samples were resistively measured at 77 K using 1 µV/cm criteria and critical current densities of microbridges were calculated as, 1.67×10^6 A/cm^2 for STO based YBCO, 1.39×10^6 A/cm^2 for MgO based YBCO and 1.06×10^6 A/cm^2 for LAO based YBCO at 77 K. For all samples, the critical current densities are typically in the order of 10^6 A/cm^2.

Table 2
Selected properties of $SrTiO_3$, MgO and $LaAlO_3$ substrates

Substrate	Crystal system	Lattice constants (Å)	Chemical stability wrt. YBCO	Twining	γ (10^{-6}/K)	Melting point (K)
$SrTiO_3$	Cubic	3.91	Good	No	10.4	2353
$LaAlO_3$	Rhombohedral	3.79 $\alpha = 90°5'$	Good	Yes	11	2453
MgO	Cubic	4.20	Good	No	10.5	3100

4. Conclusion

We have grown YBCO thin films on MgO, LAO and STO substrates by ICMS technique. Highly *c*-axis oriented YBCO thin films with superior superconducting properties were obtained on different substrates by optimizing their deposition conditions. The samples were investigated by structural, magnetic and electrical measurements. In the literature, different deposition conditions have been used for obtaining YBCO thin films by taking a single type of substrate [7–14]. In our study, we fixed the deposition conditions for all three different substrates and this has produced YBCO thin films having high quality microstructure and the superior superconducting properties.

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