Inclined-Substrate Deposition of Biaxially Textured Magnesium Oxide Thin Films for YBCO Coated Conductors*

B. Ma, M. Li, Y. A. Jee, R. E. Koritala, B. L. Fisher, and U. Balachandran

Energy Technology Division, Argonne National Laboratory

Argonne, Illinois 60439

April 2001

(First revision 5/29/01)

For submittal to Physica C.

*This work supported by the U.S. Department of Energy (DOE), Energy Efficiency and Renewable Energy, as part of a DOE program to develop electric power technology, under Contract W-31-109-Eng-38

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Argonne National Laboratory, Argonne, IL 60439

ABSTRACT

Highly textured MgO films were grown by the inclined-substrate deposition (ISD) technique at a high deposition rate. A columnar grain with a roofing-tile-shaped surface was observed in these MgO films. X-ray pole figures and ϕ-scan and ω-scan were used to characterize in-plane and out-of-plane textures. MgO films deposited when the incline angle α was 55 and 30° exhibited the best in-plane and out-of-plane texture, respectively. High-quality YBCO films were epitaxially grown on ISD-MgO-buffered Hastelloy C substrates by pulsed laser deposition. T_c = 88 K, with sharp transition, and j_c values of $\approx 2 \times 10^5$ A/cm^2 at 77 K in zero field were observed on films 5 mm wide and 1 cm long. This work has demonstrated that biaxially textured ISD MgO buffer layers deposited on metal substrates are excellent candidates for fabrication of high-quality YBCO coated conductors.

PACS: 74.76.Bz
Keywords: YBCO thin film, Coated conductor, ISD, Biaxial-texture

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*Corresponding author. Address: 9700 South Cass Avenue, Argonne, IL 60439, USA; Tel: (630)252-9961; Fax: (630)252, 3604; e-mail: bma@anl.gov

INTRODUCTION

High-T_c thin film superconductors and coated conducting wires have many applications, including high-power transmission cables, high-field magnets, generators, magnetic shields, and large-scale microwave devices, if the materials can carry sufficient current [1-3]. Highly textured template films or buffer layers are necessary to successfully deposit biaxially aligned $YBa_2Cu_3O_{7-X}$ (YBCO) films on flexible metal substrates and thus to achieve high critical current density (j_c). Significant effort has been made in the last few years to develop fast, robust techniques for the fabrication of biaxially textured buffer layers on metallic substrates by vapor phase deposition (VPD) processes. One of the success stories has been the use of ion-beam-assisted deposition (IBAD) to fabricate biaxially textured yttria-stabilized zirconia (YSZ) films on polished metal substrates. Subsequently, YBCO films were deposited on these textured buffer layers by pulsed laser deposition (PLD), chemical vapor deposition (CVD), or electron-beam (e-beam) coevaporation methods [4-7]. By far, PLD is the most widely used and reliable method for growing YBCO films. The film growth rate in the IBAD YSZ process is slow, and it requires 500 to 1000 nm of material that is deposited at a rate of ≈0.1 nm/sec to achieve good in-plane texture with ϕ-scan full width at half maximum (FWHM) of ≈15°. A small FWHM is important for producing high-performance YBCO coated conductor films because large grain boundaries reduce j_c [8,9].

Magnesium oxide (MgO) thin films deposited on inclined substrates were first observed to have preferred orientation by Aboelfotoh in 1973 [10]. Recently, Bauer et al. reported on the use of the inclined-substrate deposition (ISD) technique to fabricate MgO buffer layers for coated conductor applications [11]. When compared with the IBAD YSZ process, the ISD MgO process has no need of ion beam assistance. Therefore, it is a much simpler approach and can be performed at a deposition rate of 300 nm/min, which is significantly faster than that in the IBAD YSZ process.

We grew biaxially textured MgO thin films on mechanically polished Hastelloy C (HC) substrates by ISD using an e-beam evaporation system. To decrease the surface roughness of the as-deposited ISD MgO films, an additional thin layer of MgO was deposited at an elevated temperature and a flat angle. YBCO films were subsequently deposited on these ISD-MgO-buffered substrates with an Excimer laser system. Surface morphology of the films was investigated by scanning electron microscopy (SEM). X-ray pole figures, and ϕ-scan and ω-scan were used to analyze texture. In this paper, we report the growth mechanism, microstructure, dependence of the biaxial alignment of the ISD MgO thin films on substrate incline angles, and superconducting properties of YBCO films deposited on the highly textured ISD MgO buffer layers fabricated on polished HC substrates.

EXPERIMENTAL PROCEDURES

Mechanically polished HC pieces measuring ≈0.1 mm thick, 1 cm long, and ≈5 mm wide were used as substrates for ISD MgO and subsequent YBCO deposition. The experimental setup we used for ISD is illustrated in Fig. 1. MgO thin films were grown from a magnesium oxide source by e-beam evaporation. Fused lumps of MgO (Alfa Aesar, 99.95% metals basis, 3-12 mm pieces) were used as target material. The substrates were mounted onto a tiltable sample stage above the e-beam evaporator. The substrate incline angle α, substrate normal with respect to evaporation direction, was varied between 10 and 70°. Oxygen flow was introduced into the system during film deposition. The base pressure of the vacuum system was 1×10^{-7} torr, which rose to an operation pressure of 2×10^{-5} torr during deposition. A quartz crystal monitor was mounted beside the sample stage to monitor the deposition rate. High deposition rates of 120-300 nm/min were used and the substrate temperature was maintained between room temperature and 50°C during deposition. After the deposition of ISD films, a thin layer of MgO was deposited at a zero degree incline angle at elevated temperatures to improve the surface roughness of the buffer layer with respect to the epitaxial YBCO growth.

YBCO films were deposited by the PLD method with a Lambda Physik COMPex 201 Excimer laser with a Kr-F_2 gas premixture as lasing medium. Commercial YBCO targets, of 1-inch in diameter and 0.25-inch thick, were used. Substrates were attached to a heatable sample stage with silver paste and heated to 760°C during deposition. The laser spot focused at the rotating target was ≈ 2 mm^2 which resulted in an energy density of ~2 J/cm^2. The distance between the target and the substrates was 7.5 cm. The desired oxygen partial pressure (in the range of 100-300 mtorr) was obtained by flowing ultra-high-purity oxygen through the chamber. A pulse repeat rate of 8 Hz produced a growth rate of ≈ 15 nm/min.

The superconducting critical transition temperature (T_c) and critical current density (j_c) were determined by the inductive method and confirmed by the transport method at 77 K in liquid nitrogen. The inductive test was used as a standard characterization tool to measure the superconducting properties of our YBCO films. Thin-film superconductor samples were placed between a primary and secondary coil pair with inner diameter of 1 mm and outer diameter of 5 mm. Alternating current was introduced to the primary coil and detected from the secondary coil by a lock-in amplifier (Stanford Research Systems SR830 DSP). Samples used for transport measurements were first coated with 2-μm-thick silver by e-beam evaporation and then annealed in flowing high-purity oxygen at 400°C for 2 h. Typical samples used for four-probe transport measurement were 3-5 mm wide and 1 cm long.

Biaxial-texture analysis was conducted by X-ray diffraction pole figure analysis with Cu-K_α radiation. In-plane texture was characterized by the FWHM of ϕ-scans for the MgO (002) reflection ($2\theta = 42.9°$), and out-of-plane texture was characterized by the FWHM of ω-scan at the MgO [001] pole for the same reflection. Plane-view and fracture cross-sectional SEM (Hitachi S-4700-II) were used to study the morphology of the MgO films.

RESULTS AND DISCUSSION

1. X-ray Texture Analysis

The biaxial texture of the ISD MgO films was characterized by X-ray diffraction pole figure analysis. Typical pole figures of an ISD MgO film deposited at an incline angle $\alpha = 55°$, film thickness of 2 μm, are shown in Fig. 2. Unlike the YSZ films prepared by inclined-substrate PLD [7], where the (001) planes are parallel to the substrate surface, the [001] axis of the

ISD MgO buffer layer is tilted away from the substrate normal. The asymmetric distribution of the pole peaks reveals that the MgO (001) planes have a tilt angle β toward the deposition direction, as indicated by an arrow in the pole figures. These ISD MgO films exhibit good texture; distinct in-plane alignment can be seen as well-defined poles for not only the (001) axis but also the [010] and [100] axes in Fig. 2. The tilt angle for the (001) plane was determined from the chi angle value of the [001] reflection in the MgO (002) pole figure. Out-of-plane alignment was characterized by ω-scan; data were taken at the [001] pole.

Table. 1. Tilt angle and FWHMs* of ϕ-scan and ω-scan for ISD MgO films deposited at various incline angles.

Incline Angle α (°)	Tilt Angle β (°)	ϕ-FWHM (°)	ω-FWHM (°)
10	16	37.8	9.5
15	16	47.2	11.3
20	17	20.2	5.9
25	17	23.4	6.8
30	22	14.4	5.6
35	18	19.4	6.4
40	16	35.5	10.5
45	20	39.3	12.5
50	24	27.4	11.3
55	32	12.2	6.3
60	28	14.1	6.6
65	32	12.8	6.9
70	37	13.8	7.8

*As determined for MgO (002).

Figure 3 shows typical ϕ-scan and ω-scan patterns for the ISD MgO film deposited when α = 55°. The FWHMs of ϕ-scan and ω-scan and the tilt angles for the ISD MgO films made at various incline angles are listed in Table 1. The smallest MgO (002) ϕ-scan FWHM (12.2°) was observed for samples deposited when α = 55°, and the smallest MgO (002) ω-scan FWHM (5.6°) was observed for samples deposited when α = 30°, with slightly larger FWHM values at higher or lower α angles. The FWHMs of ϕ-scan and ω-scan for MgO (002), plotted in Fig. 4 as a function of substrate incline angle, clearly show minimum values at incline angles of ≈30 and 50°, with MgO (002) tilt angles β = 22 and 32°, respectively.

The film tilt angle increases with increasing substrate incline angle, as shown in Table1. This finding implies that β cannot be independently adjusted without affecting the texture of the films. The best in-plane texture can be obtained at α = 55°. MgO films deposited when α = 55° were used for buffer layers and subsequent deposition of YBCO films by PLD. An interesting observation is that we obtained the smallest ϕ-FWHM and ω-FWHM at fairly large β when compared with the adjacent values listed in the table.

To improve the surface roughness, an additional thin layer of MgO was deposited on the ISD MgO films at elevated temperatures (600-800°C) by e-beam evaporation at zero-degree α angle (flat substrate). Table 2 lists the FWHMs for several ISD MgO films before and after flat substrate deposition of an additional 1 μm MgO at 700°C. We had observed ≈2 and

≈1° improvements in FWHMs from the MgO (002) ϕ-scan and ω-scan, respectively.

Table 2. FWHMs of ϕ-scan and ω-scan for ISD MgO films before and after deposition of an additional 1 μm MgO at 700°C.

Sample#	ϕ-scan MgO (002) FWHM (°)			ω-scan MgO (002) FWHM (°)		
	Before	After	Difference	Before	After	Difference
1	11.8	9.7	2.1	6.6	5.6	1.0
2	11.6	9.8	1.8	7.0	5.8	1.2
3	12.7	10.9	1.8	6.5	5.6	0.9
4	13.2	9.2	4.0	6.8	5.2	1.6

2. SEM Morphology

Figure 5 shows the top plane-view and fracture cross-sectional view of an ≈2-μm-thick ISD MgO thin film grown at room temperature. The fracture cross-sectional view revealed that MgO films grow with a distinct columnar structure. The top of the MgO columns terminated with (002) planes [12] and formed a roofing-tile like rough surface, as shown by the top plane-view SEM image.

Figure 6 shows SEM images of the top plane-view and fracture cross-sectional view of a sample made of 1.5-μm-thick ISD MgO layer deposited at room temperature with α = 55°, followed by deposition of a 0.5-μm-thick additional MgO layer at 700°C with $\alpha = 0°$. The surface smoothness was significantly improved, and the in-plane and out-of-plane textures were also improved. Plate-shaped grains were formed during the flat substrate deposition at 700°C, in contrast to columnar grains during ISD deposition at room temperature.

3. YBCO Grown on ISD MgO

Biaxially aligned YBCO films were successfully deposited on ISD-MgO-buffered HC substrates by PLD. Figure 7 shows ϕ-scans for the MgO (220) and YBCO (103) grown on the MgO buffered HC substrate; it reveals epitaxial growth, with the usual cubic-to-cubic biaxial alignment: YBCO [001] // MgO [001] and YBCO [100] // MgO [100] (or MgO [010]). The FWHMs of the YBCO films were generally 1-2° smaller than those of the MgO films underneath; the FWHMs of MgO buffer layers on which YBCO films had been deposited were also slightly smaller. This finding may represent a bulk value due to larger penetration depth of the X-ray; a similar finding was reported by Bauer et al. [11].

To improve lattice mismatch and therefore enhance the superconducting properties of YBCO films, various buffer layers were explored. These buffer layers were put between MgO and YBCO layers by either e-beam evaporation or the PLD method. Details of buffer layer architecture will be reported elsewhere [13]. We obtained $T_c = 88$ K for a 0.5-μm-thick YBCO film deposited on an MgO-buffered HC substrate. From inductive measurements, Fig. 8 shows that the superconducting transition completed at 85 K. The critical current density for this sample is ~2 x 10^5 A/cm^2 at 77 K with zero external field.

CONCLUSIONS

Biaxially textured MgO films were successfully grown by the ISD method. This method is much more time-efficient for fabrication of buffer layers when compared with the IBAD deposition of YSZ. ISD MgO films grow columnar grains with (002) planes that are truncated at the surface. Plane-view SEM revealed a roofing-tile-shaped structure. An MgO (002) ϕ-scan with FWHM = 12.2° was observed for 2-μm-thick MgO thin films deposited with $\alpha = 55°$, and an MgO (002) ω-scan with FWHM = 5.6° was observed for MgO thin films deposited with $\alpha = 30°$. Tilt angles $\beta = 22$ and 32° were observed for films deposited with $\alpha = 55°$ and 30°, respectively. The surface roughness and biaxial texture of ISD MgO thin films were significantly improved by flat substrate deposition of an additional thin layer of MgO at elevated temperature.

YBCO films epitaxially grown on ISD-MgO-buffered HC substrates by pulsed laser deposition at 760°C in a 250-mtorr high-purity oxygen flowing environment exhibited good superconducting properties. T_c = 88 K with sharp transition, and $j_c \approx 2 \times 10^5$ A/cm^2 at 77 K in zero field were observed. This work has demonstrated that biaxially textured ISD MgO buffer layers deposited on metal substrates are excellent candidates for fabrication of high-quality YBCO coated conductors.

ACKNOWLEDGMENT

SEM analysis was performed in the Electron Microscopy Center for Materials Research at Argonne National Laboratory. This work was supported by the U.S. Department of Energy (DOE), Energy Efficiency and Renewable Energy, as part of a DOE program to develop electric power technology, under Contract W-31-109-Eng-38.

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FIGURE CAPTIONS

Fig. 1. Schematic illustration of experimental setup for inclined substrate deposition.

Fig. 2. Pole figures of ISD MgO film deposited with $\alpha = 55°$, (a) MgO (002) and (b) MgO (220). The direction of MgO flux, deposition direction, is indicated by arrow.

Fig. 3. (a) ϕ-scan and (b) ω-scan patterns of ISD MgO film deposited when $\alpha = 55°$.

Fig. 4. FWHMs of MgO (002) ϕ-scan and ω-scan for IBAD MgO films deposited at various substrate incline angles.

Fig. 5. (a) Top plane-view and (b) fracture cross-sectional-view SEM images of ISD MgO film deposited with $\alpha = 55°$.

Fig. 6. (a) Top plane-view and (b) fracture cross-sectional-view SEM images of MgO film made of a layer of ISD MgO deposited at room temperature when $\alpha = 55°$, followed by an additional layer of MgO e-beam evaporated at 700°C with $\alpha = 0°$.

Fig. 7. X-ray ϕ-scan patterns of YBCO (103) and MgO (220), showing epitaxial growth.

Fig. 8. Critical-temperature transition curve for YBCO film deposited on ISD-MgO-buffered Hastelloy C substrate.

Figures:

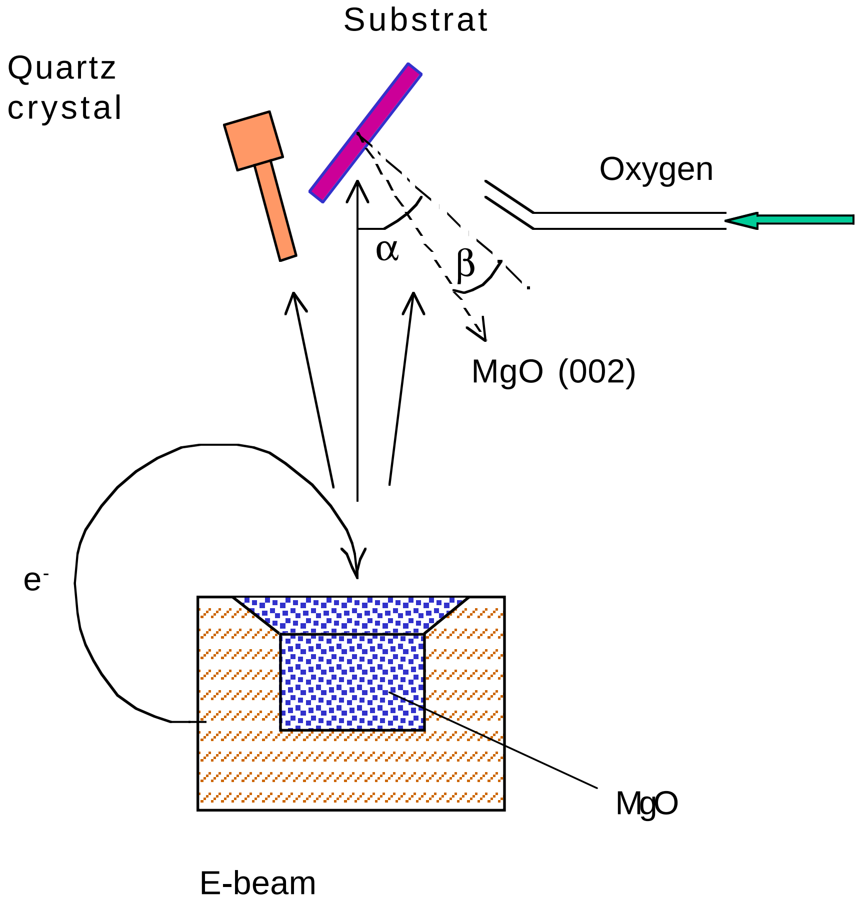


Fig. 1.

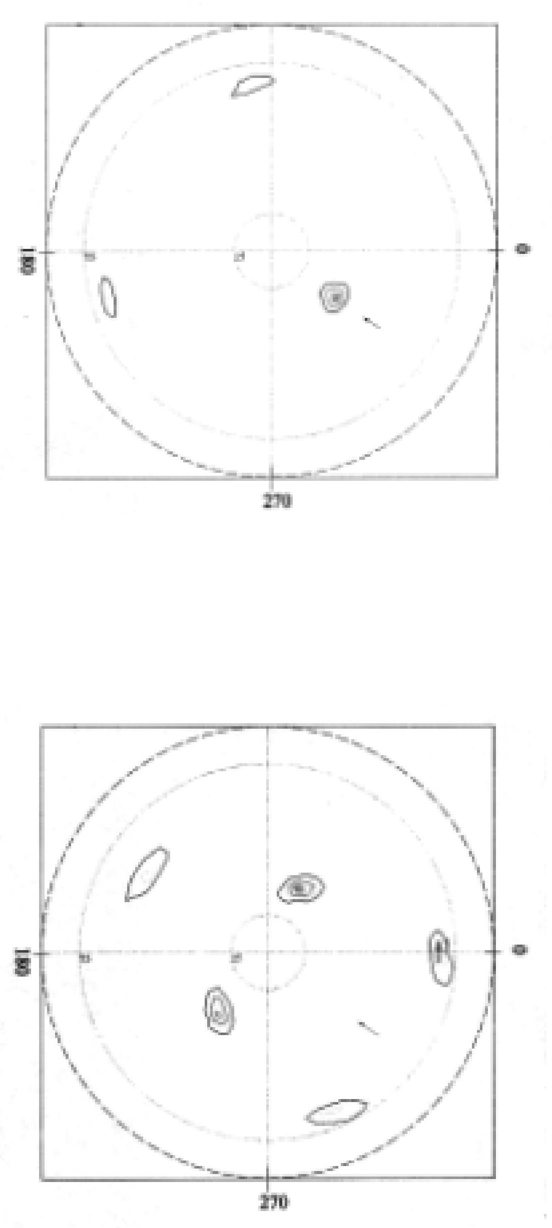
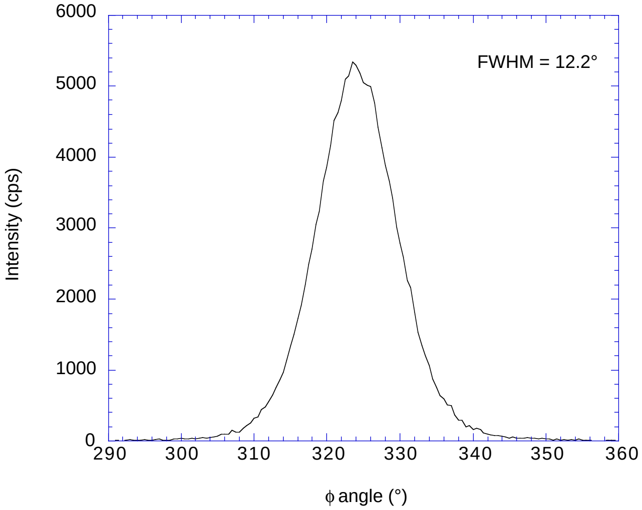
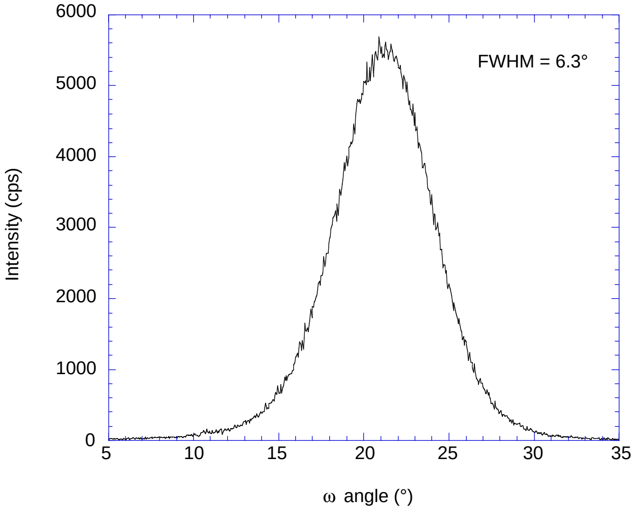


Fig. 2.


6000
5000
4000
3000
2000
1000
0
Intensity (cps)
FWHM = 12.2°
290
300
310
320
330
340
350
360
ϕ angle (°)

(a)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3000
2000
1000
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Intensity (cps)
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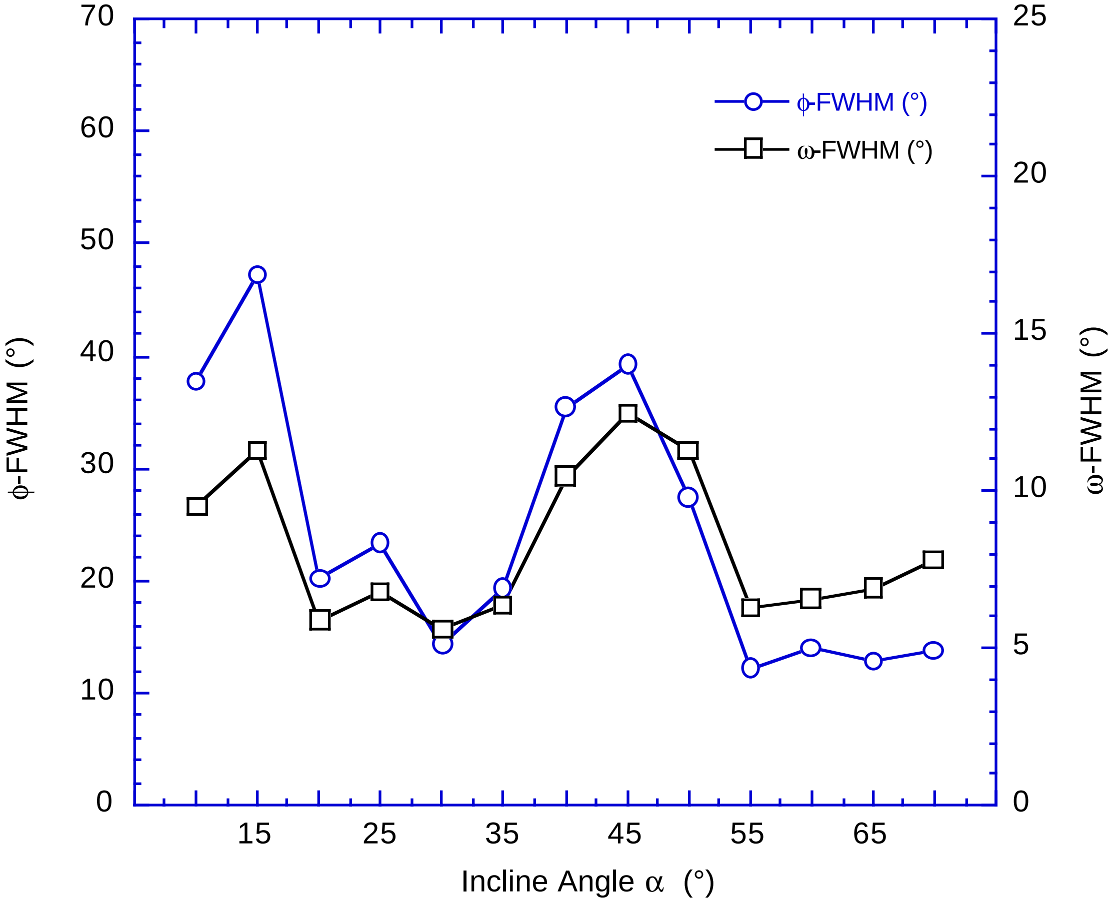
10
15
20
25
30
35
ω angle (°)

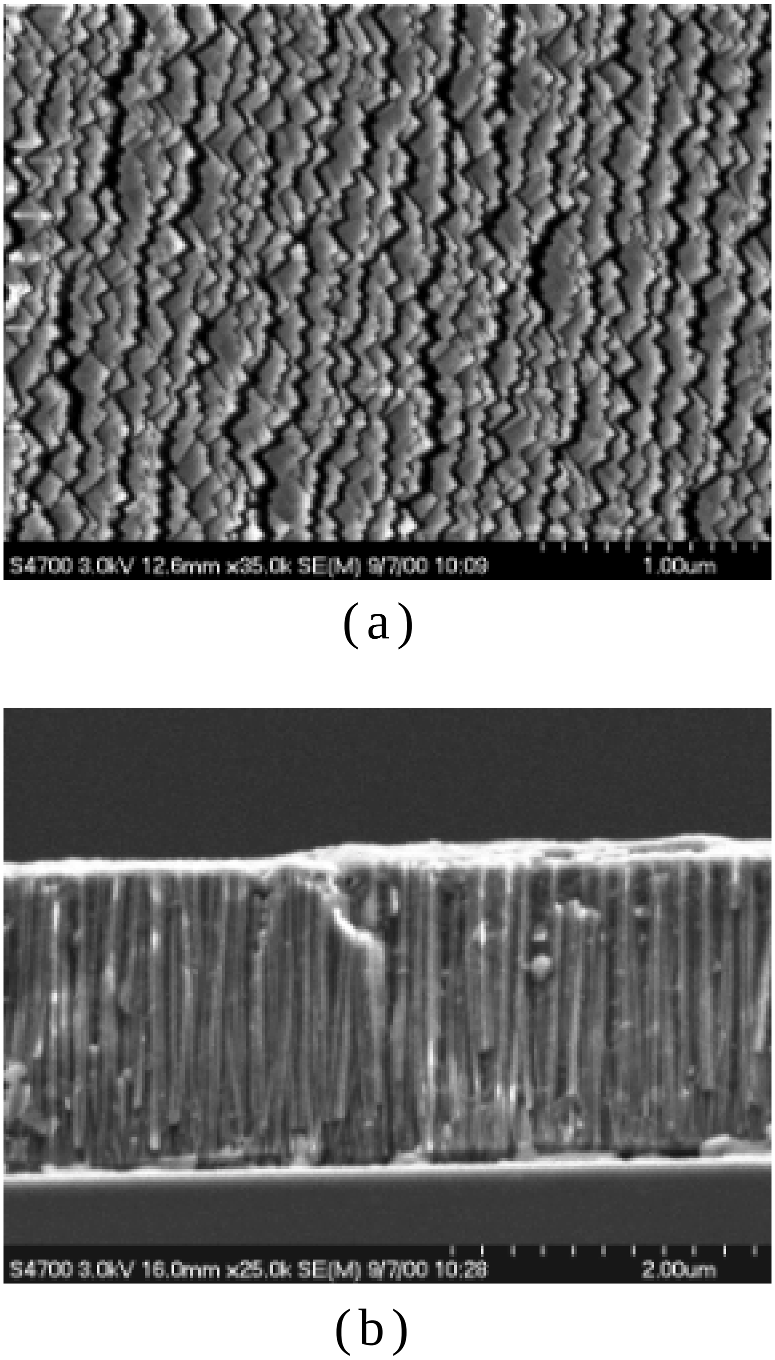
(b)

Fig. 3.



ϕ-FWHM (°)
ω-FWHM (°)
70
60
50
40
30
20
10
0
25
20
15
10
5
0
15
25
35
45
55
65
Incline Angle α (°)

Fig. 4.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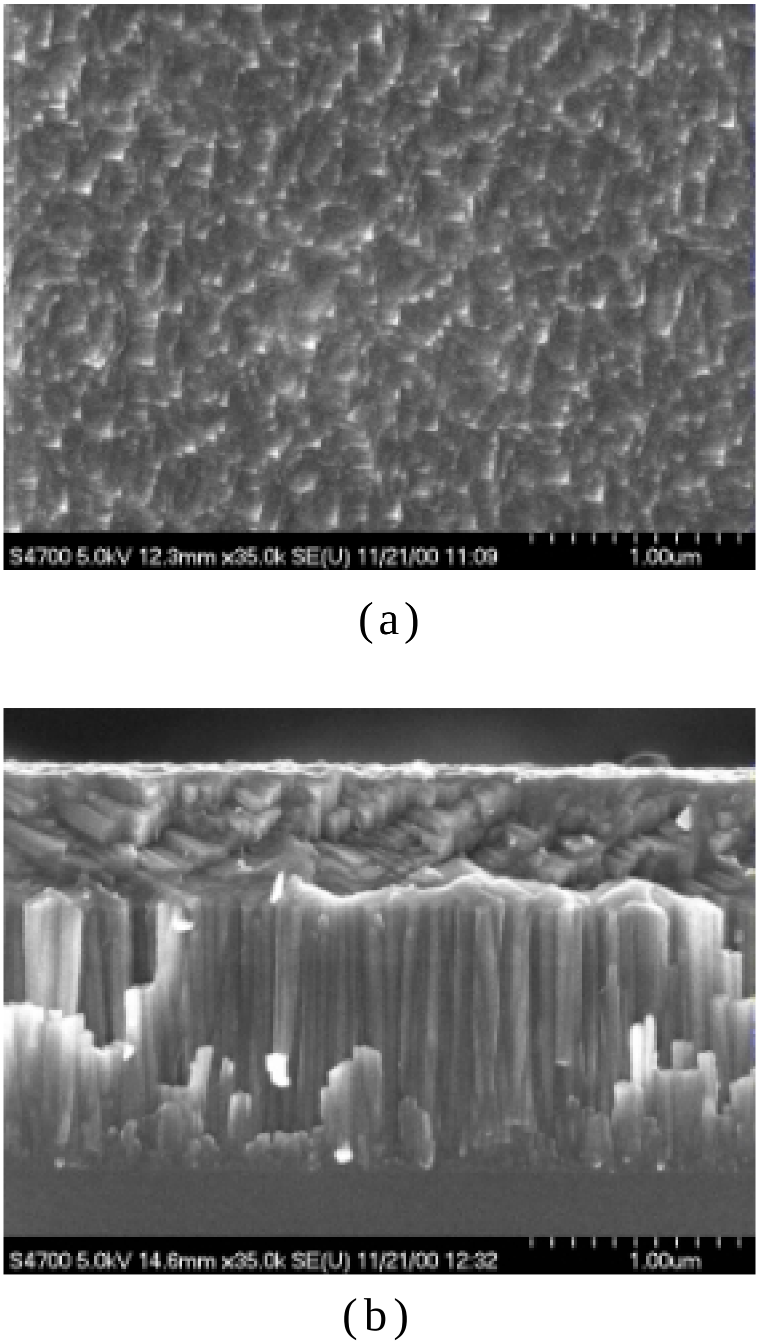
2.00um

(a)

(b)

Fig. 5.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(a)
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(b)

Fig. 6.

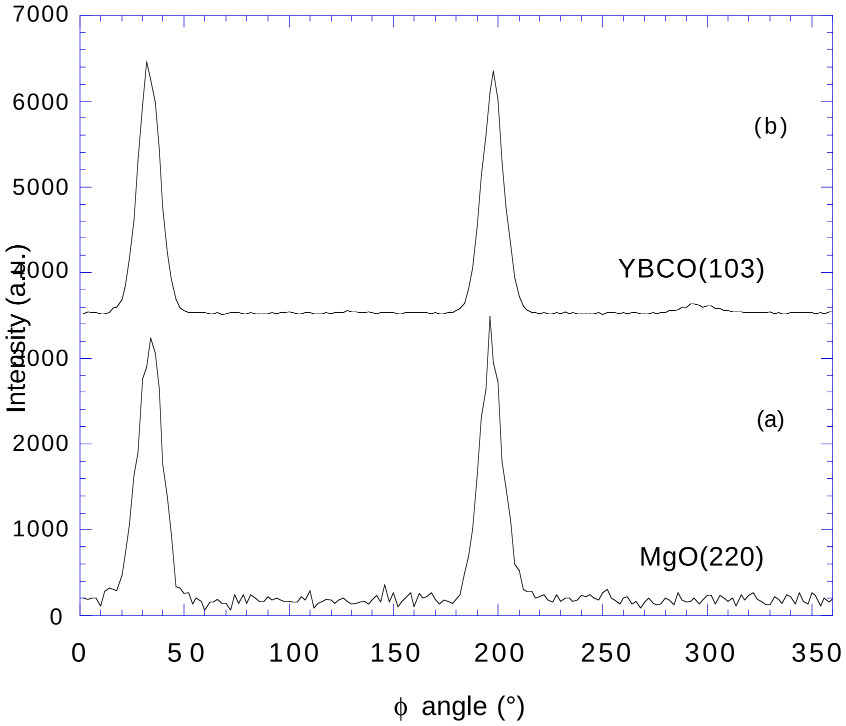
7000
6000
5000
4000
3000
2000
1000
0
0
50
100
150
200
250
300
350
Intensity (a.u.)
φ angle (°)
(b)
YBCO(103)
(a)
MgO(220)

Fig. 7.

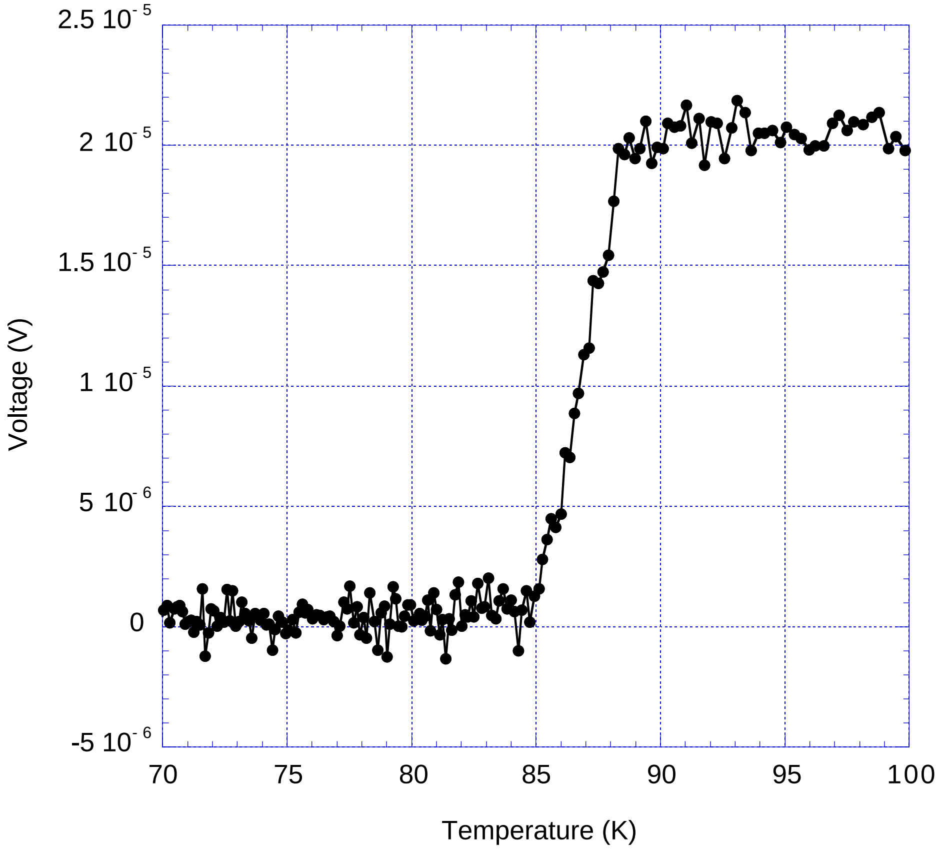
2.5 10- 5
2 10- 5
1.5 10- 5
1 10- 5
5 10- 6
0
-5 10- 6
Voltage (V)
70
75
80
85
90
95
100
Temperature (K)

Fig. 8.