Journal of Alloys and Compounds, 195 (1993) 97–100
JALCOM 7423

Surface Morphology of $YBa_2Cu_3O_{7-\delta}$ Thin Films on $SrTiO_3(100)$ Studied by Scanning Tunneling Microscopy

H. P. Lang, R. Sum, H. Haefke, and H.-J. Güntherodt

Institute of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland*

Abstract

Thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) have been prepared by pulsed laser deposition on single-crystalline $SrTiO_3(100)$ substrates at different substrate temperatures T_s. Depending on T_s, different orientations of the films are revealed by X-ray diffraction: (001) orientation between 660 to 800 °C, (100) orientation between 610 and 660 °C, and no YBCO reflections for films prepared at lower T_s. In-plane orientation (Φ-scan) and out-of-plane orientation data (mosaic spread) of the grains are compared to scanning tunneling microscopy (STM) images. The images show rectangular-shaped growth hills. Their growth steps are aligned along the [100] and [010] directions of YBCO and are one c-axis distance in height (1.2 nm). Occasionally screw dislocations are observed. Highest critical transition temperatures are reached at about T_s = 750 °C. The relationship between film thickness and surface morphology has been studied using YBCO films with a thickness gradient prepared by a half-shadow technique. Thus, films of different thicknesses can be obtained simultaneously under identical conditions. STM images give evidence of different growth stages such as nucleation and coalescence of two-dimensional islands, their growth and, finally, the formation of growth hills.

1. Introduction

The growth of superconducting thin films of high-temperature superconductors (HTSC) on single-crystalline substrates without further post-annealing by pulsed laser deposition (PLD), first described by Dijkamp et al. [1], is a rapid and facile way to prepare samples of high quality. Their electrical properties – especially the critical current density – surpass those of single crystals, polycrystalline bulk material or melt-textured samples by powers of ten. The structural properties – as determined by X-ray diffraction (XRD) – of epitaxial HTSC thin films do not differ very much from those of HTSC single crystals. One of the most intensively studied HTSC is $YBa_2Cu_3O_{7-\delta}$ (YBCO), especially its growth on $SrTiO_3(100)$ substrate crystals, which have lattice parameters very well adapted to that of YBCO.

Recent efforts in investigating HTSC thin films by scanning tunneling microscopy (STM) [2-4], which is a local method to image the surface structure from the micrometer to the atomic scale, have revealed that the surface morphology of thin films differs clearly from that of single crystals [5]. Whereas single crystals exhibit growth terraces several micrometers in width, the surface of thin films is dominated by spiral growth hills (typical diameter: 100-200 nm [2-4]).

* Work supported by Swiss National Science Foundation and Kommission zur Förderung der Wissenschaftlichen Forschung.

In this study, the PLD growth of YBCO on $SrTiO_3(100)$ at different substrate temperatures is characterized by XRD and STM. The electrical and structural properties of the YBCO films have been optimized. A special half-shadow technique [6,7] is applied to prepare films with thickness gradients to study different growth stages by STM.

2. Experimental

2.1. Thin-film growth

Thin films of YBCO are prepared by standard PLD [1] in a high vacuum chamber. For good reproducibility, a 1 inch $SrTiO_3(100)$ wafer is cleaved to obtain many substrates of identical quality. After pre-heating the substrate at 820 °C in the evacuated preparation chamber (10^{-4} Pa), the substrate temperature is decreased to the value chosen for YBCO deposition. After establishing an oxygen pressure of 27 Pa, the film (thickness 220 nm, nominal growth rate 0.6 nm/s) is deposited using a KrF excimer laser focused onto a stoichiometric YBCO target (2.5 J/cm^2). Following deposition, oxygen is let in to a pressure of 4×10^4 Pa and the sample is cooled to 600 °C at a rate of 4 K/min, followed by oxygen annealing at 500 °C for 30 min and cooling to room temperature. All temperatures given are determined by an optical pyrometer. A chromel-alumel thermocouple embedded in the heating block is used as a sensor for the computer-controlled feedback loop of the heating system.

0925-8388/93/$06.00

2.2. Thickness-gradient films

YBCO films of variable thickness (thickness-gradient films) are prepared using a shutter between ablation target and substrate [6,7]. Thus the substrate is partially shaded. This leads to YBCO deposition at different rates resulting in a varying film thickness.

2.3. X-ray diffraction

Standard Cu K_α XRD in Bragg-Brentano geometry is used to determine the structural bulk properties of the films. The samples are mounted on the XRD stage and oriented using the YBCO(005) reflection. The azimuth angle is varied to obtain maximum intensity (Φ-scan, in-plane orientation of the grains). Thereafter, the sample stage rock (Θ) is varied while leaving the detector at the 2Θ position of the YBCO(005) reflection (mosaic spread, out-of-plane orientation of the grains). The analysis is completed by a $\Theta - 2\Theta$ scan.

2.4. Scanning tunneling microscopy

The surface morphology of YBCO thin films is imaged by STM applying tunneling currents below 0.3 nA and sample bias voltages below -800 mV. The constant current mode is used to obtain topographical information. Several mechanically prepared $Pt_{90}Ir_{10}$ tips are used for each sample to exclude tip-geometry induced artifacts. The roughness of the surface imaged is indicated by the maximum height difference and the standard deviation of the data points measured.

2.5. Electrical properties

The critical transition temperature T_c of the films has been determined by a standard four-probe DC technique and compared to the values obtained by an inductive method. Critical current densities j_c are determined at 77 K by use of a microbridge patterned by laser ablation.

3. Results and discussion

The structural and superconducting properties of YBCO thin films grown on $SrTiO_3(100)$ substrates at different substrate temperatures T_s, ranging from 550 to 800 °C, are discussed.

3.1. Structure of a sample prepared at T_s = 740 °C

As an example, the structural properties of a thin film sample prepared at T_s = 740 °C as obtained by XRD are compared to STM results. XRD ($\Theta - 2\Theta$) reveals that the film is (001) oriented. The determination of the mosaic spread (fig. 1(a)) shows that the rock of the grains is less than 0.26° (full width at half maximum (FWHM) of the YBCO (005) reflection).

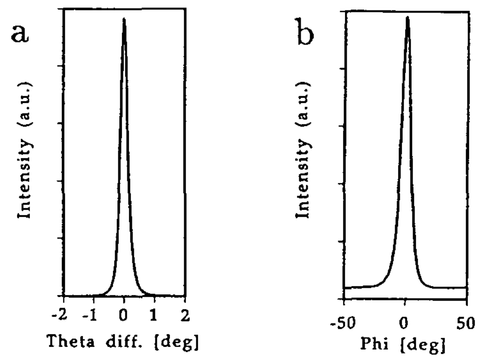

Figure 1. YBCO thin film prepared at 740 °C. (a) Mosaic spread of YBCO(005) reflection (FWHM = 0.26°). (b) Φ-scan of YBCO(005) (FWHM = 6°).

The in-plane misorientation relative to the substrate (fig. 1(b)) of the YBCO grains is within 6° (FWHM of Φ-scan of YBCO(005) reflection). Figure 2 shows a STM image (a) and a line section of the surface (b). The surface morphology is dominated by growth hills. Their steps are aligned in two main directions (indicated), corresponding to the crystallographic [100] and [010] directions of YBCO as determined by XRD (Φ-scan). Only occasionally, screw dislocations are observed. The height of the growth steps (1.2 nm) corresponds to the height of a unit cell in the YBCO[001]

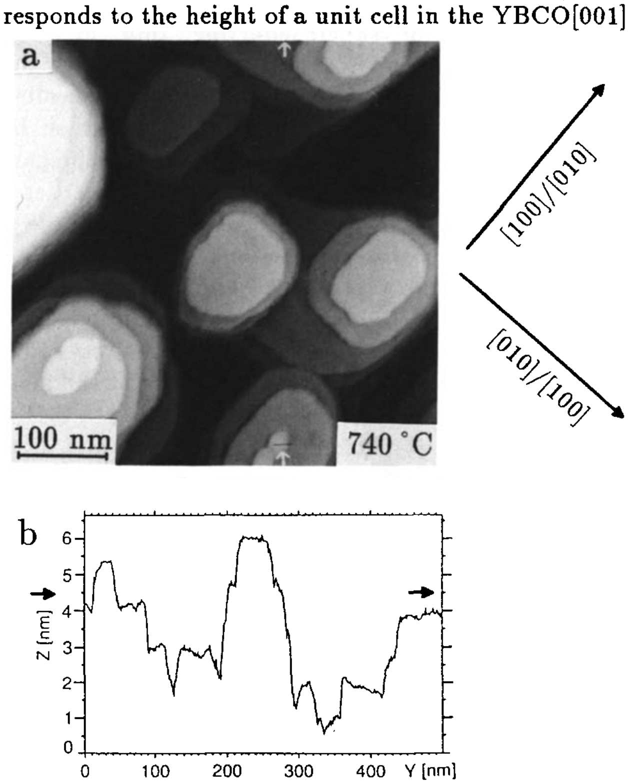

Figure 2. Surface of a sample prepared at 740 °C. (a) STM topview showing growth hills with steps aligned to [100] and [010] of YBCO (indicated) and one c-axis length in height. A line section is indicated by small arrows in (a) and displayed in (b).

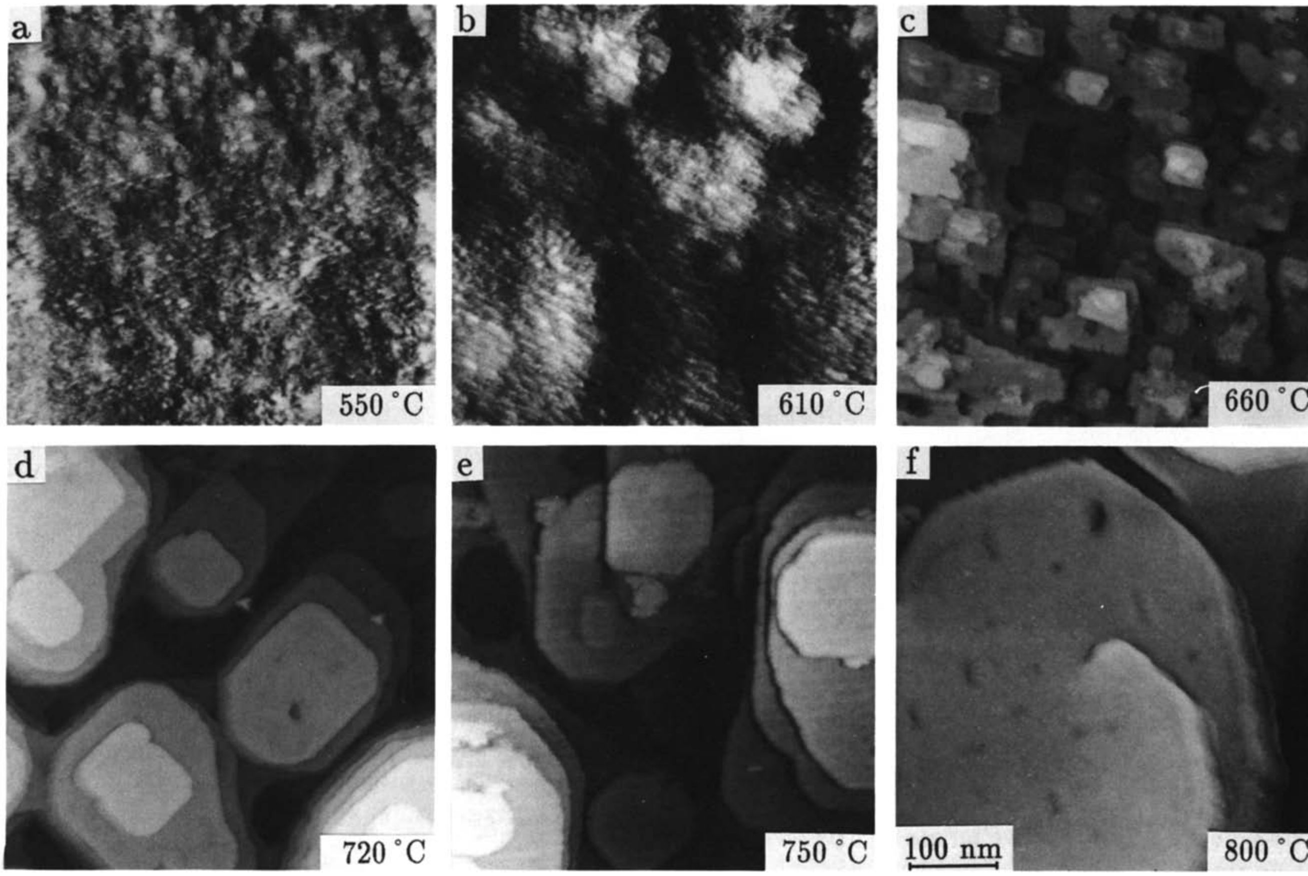

Figure 3. STM topviews of YBCO films on $SrTiO_3(100)$ prepared at different substrate temperatures: (a) 550 °C (film orientation: amorphous), (b) 610 °C (100), (c) 660 °C (001), (d) 720 °C (001), (e) 750 °C (001) and (f) 800 °C (001).

direction (1.17 nm) as can be seen from the line section (fig. 2(b)). The rock of YBCO grains is too small (0.26°) to be seen in the STM image.

3.2. Film orientation depending on T_s

Table 1 summarizes the XRD data and the surface roughness values determined by STM for the samples prepared at different T_s. XRD reveals that the samples prepared above 660 °C are (001) oriented, and the samples prepared below 660 °C are (100) oriented or show no YBCO reflections at all.

Table 1. Structural properties of YBCO films on $SrTiO_3(100)$ prepared at different T_s.

T_s	$2\Theta^a$ [deg]	Mosaic^a [deg]	Φ-scan^a [deg]	Roughness^b [nm]
550 °C	–^{c}	–^{c}	–^{c}	7±1
610 °C	0.12^d	0.24^d	6.5^d	14±2
660 °C	0.48	0.90	17	16±2
700 °C	0.23	0.42	13	21±3
740 °C	0.19	0.26	6	28±4
750 °C	0.19	0.24	6	20±4
800 °C	0.20	0.53	7	10±1

[a] XRD: FWHM of YBCO(005) reflection.
[b] STM: maximum height difference and standard deviation of data points measured.
[c] no YBCO reflections observed.
[d] YBCO(100) reflection.

3.3. Film morphology as a function of T_s

The surface morphologies of samples prepared at different T_s are compared. Figure 3 shows STM images of YBCO films on $SrTiO_3(100)$ prepared at 550, 610, 660, 720, 750 and 800 °C, respectively. Films prepared at 550 °C exhibit a grainy, disordered surface (fig. 3(a)), those prepared at T_s=610 °C show *a*-axis growth steps with a height of 0.4 nm (fig. 3(b)). The films prepared at $T_s \geq 660$ °C exhibit growth hills with unit cell steps in the YBCO[001] direction (figs. 3(c) - (f)). With increasing T_s the growth hill diameters increase, too. This is caused by the higher T_s applied and the prolonged cooling process to 600 °C, leading to increased thermal diffusion.

3.4. Electrical properties

The transition temperatures T_c to superconductivity, the transition widths ΔT_c and the ratios of the resistivity at 300 K to the resistivity at 100 K (R_{300K}/R_{100K}) of the samples prepared at different T_s are compiled in Table 2.

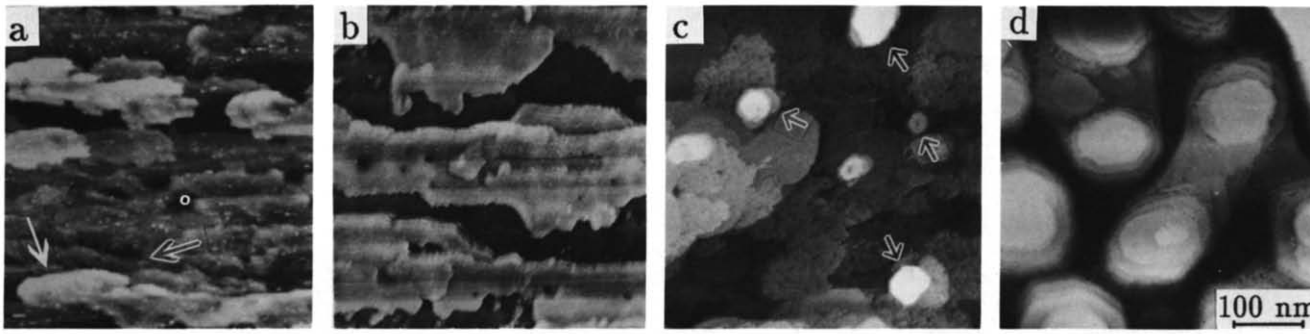

Figure 4. STM topviews of a YBCO thickness-gradient film grown on $SrTiO_3(100)$ at 740 °C: (a) Nucleation of two-dimensional YBCO islands (bold arrow) near a substrate step (open arrow), (b) coalescence of two-dimensional YBCO islands, (c) formation of growth hills at preferred sites (see arrows), (d) thick film with growth hills.

Table 2. Electrical properties of YBCO films on $SrTiO_3(100)$ prepared at different T_s.

T_s	T_c [a]	ΔT_c [a]	T_c [b]	R_{300K}/R_{100K}
550 °C	– [c]	– [c]	– [d]	0.3
610 °C	– [c]	– [c]	– [e]	1.3
660 °C	71 K	7 K	89 K	1.5
700 °C	86 K	3 K	87.5 K	2.0
740 °C	88 K	4 K	91 K	1.9
750 °C	87.6 K	2.5 K	92 K	2.4
800 °C	87.5 K	2.5 K	90 K	3.3

[a] determined by an inductive method (onset).
[b] determined by a resistive DC method (onset).
[c] no transition observed.
[d] semiconducting.
[e] metallic.

Best resistively determined T_c's are obtained at a T_s of 740 to 750 °C. The critical current density of a sample prepared at 740 °C amounts to 3×10^6 A/cm^2 at 77 K. Such films are also optimized with respect to their structural properties (Table 1). STM images of these films reveal a surface morphology dominated by well-developed growth hills. Films having a smoother surface morphology (e.g. samples prepared at 660 °C) exhibit inferior structural and electrical properties compared to the 740 °C sample.

3.5. *Thickness-gradient films*

In order to study the relationship between film thickness, i.e. different growth stage, and film morphology, thickness-gradient films have been investigated. Figure 4 shows four STM images of the surface morphology of a thickness-gradient film prepared at 740 °C. The thinnest region (fig. 4(a)) is characterized by nucleation of two-dimensional islands (bold arrow) at substrate steps (open arrow). At a film thickness of a few monolayers, coalesced two-dimensional islands are observed (fig. 4(b)). At a thickness of about 50 nm, sites for the preferred formation of growth hills are imaged (arrows in fig. 4(c)). Finally, fig. 4(d) shows that the thick region of the YBCO film (220 nm) consists of growth hills.

4. Conclusion

Superconducting YBCO thin films have been prepared at different substrate temperatures on $SrTiO_3(100)$. They have been characterized by XRD and STM, and their electrical properties have been determined. Films with optimized structural and electrical properties exhibit a surface morphology dominated by growth hills, whereas films optimized for a smooth surface morphology have inferior structural and electrical properties. Preparing films by a thickness gradient technique, the various growth stages of YBCO on $SrTiO_3(100)$ have been documented by STM. A more detailed study of YBCO growth on $SrTiO_3(100)$ is in preparation [8].

References

1 D. Dijkamp, T. Venkatesan, X.D. Wu, S.A Shaheen, N. Jisrawi, Y.H. Min-Lee, W.L. McLean, and M. Croft, Appl. Phys. Lett. 51 (1987) 619.
2 C. Gerber, D. Anselmetti, J.G. Bednorz, J. Mannhart, and D. G. Schlom, Nature, 350 (1991) 279.
3 M. Hawley, I.D. Raistrick, J.G. Beery, and R.J. Houlton, Science, 251 (1991) 1587.
4 H.P. Lang, T. Frey, and H.-J. Güntherodt, Europhys. Lett. 15 (1991) 667.
5 H.P. Lang, J.P. Ramseyer, D. Brodbeck, T. Frey, J. Karpinski, E. Kaldis, and T. Wolf, Ultramicroscopy 42-44 (1992) 715.
6 H. Haefke, H.P. Lang, G. Leemann, and H.-J. Güntherodt, Appl. Phys. Lett. 60 (1992) 3054.
7 H.P. Lang, H. Haefke, G. Leemann, and H.-J. Güntherodt, Physica C 194 (1992) 81.
8 H.P. Lang et al., to be published.