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Dongqi Shi
University of Wollongong, dongqi@uow.edu.au

Mihail Ionescu
ANSTO, mionescu@uow.edu.au

Jeff McKinnon
University of Wollongong

Wu Ming Chen
University of Wollongong

S. X. Dou
University of Wollongong, shi@uow.edu.au

RELATIONSHIP BETWEEN ORIENTATION OF CeO_2 FILMS AND SURFACE MORPHOLOGY

D. Q. Shi, M. Ionescu, J. McKinnon, W.M. Chen, S. X. Dou

University of Wollongong, NSW 2522, Australia

ABSTRACT

Pure c-axis orientation CeO_2 films were deposited on YSZ<100> single crystal substrates by pulse laser deposition (PLD) in oxygen. The optimum epitaxial growth temperature for a high level of c-axis orientation is 790° C, and it is related to the morphology of CeO_2 grains. The more circular the shape of the grains is, the better the c-axis orientation and the smoothness of CeO_2 film surface are. This correlation appears to be an intrinsic feature of CeO_2 growth, independent of the type of substrate or the deposition method. Also it was found that the topography of the film surface grown by PLD reflects the morphology.

1. INTRODUCTION

Ceramic buffer layers play an important role in the YBCO superconducting film deposition. They are not only used on single crystal substrates for electronic applications, but also on flexible metallic substrates for YBCO coated conductor tapes. The commonly used, and therefore intensively studied, buffer layers include yttrium stabilised zirconia (YSZ), CeO_2, MgO, $SrTiO_3$ $PrBa_2Cu_3O_y$ (PBCO) et al. Among these materials, CeO_2 film is the most widely used buffer layer for YBCO film because it has the small lattice mismatch, similar thermal expansion coefficient and a good chemical compatibility with YBCO at the high deposition temperature [1,2,3]. As a buffer layer, the research of CeO_2 films commonly focused on the relationship between orientation and deposition conditions by x-ray diffractions, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) [4]. There are few works done to study the relationship between orientation and surface morphology of CeO_2 film [5]. Develos K. D. et al. [6,7] had studied the effect of deposition rate and film thickness on the surface morphology of CeO_2 films using AFM technique. In this study CeO_2 film was deposited on Al_2O_3 substrate by PLD. Wang A. et al. [8] deposited CeO_2 films by metal-organic chemical vapour deposition on YSZ substrates, and the surface morphology was analysed by using AFM.

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In this report, we focused on study the relationship between orientation and surface morphology of CeO_2 films grown on <100> YSZ by PLD. All CeO_2 films in our research are pure c-axis oriented, and difference of orientation between various samples was detected by X-ray rocking curve scan (ω-scan).

2. EXPERIMENT

CeO_2 films were deposited on <100> YSZ single crystal substrates by PLD, using an excimer laser system (Compex 301, 248nm from Lambda Physik). The particulars of the deposition system were: fixed laser beam; target-substrate distance 70 mm; target rotation 10 rpm; background pressure 1×10^{-6} Torr. The deposition conditions were: laser repetition rate 3 Hz; energy density on the target ~3.0 Jcm^{-2} (~600 mJ/pulse). The deposition pressure was 200 mTorr of high purity O_2. The YSZ substrates were mounted on the sample heater using silver paste.

Three CeO_2 films were produced at different temperatures, all having the same thickness of approximately 100nm: sample A at 775° C; sample B at 790° C; and sample C at 805° C. For all samples, after the deposition was completed, the heater was switched off, and then keeping oxygen pressure until the temperature of chamber was below 400° C.

The orientation of the films was characterised by X-ray θ-2θ and ω-scans. The surface morphology was examined by AFM (Digital Extended Multimode AFM) in contact mode, using a triangular gold-plated SiN tip, with a lever length of 200 nm. The measurements of roughness and size of outgrowths were carried out with height scan, and the images of morphology with deflection scan.

3. RESULTS AND DISCUSSIONS

The surface morphology of CeO_2 films consists of the following elements: a background structure of grains, outgrowths or island and droplets. The background structure is the area between the outgrowths

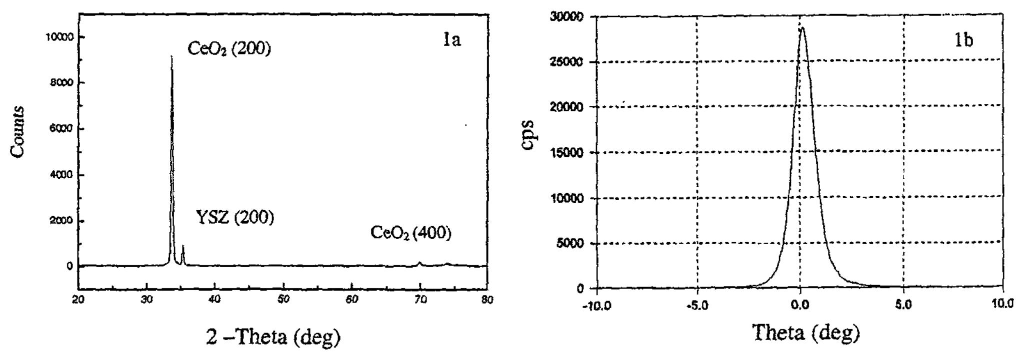


FIGURE 1a. X-ray θ-2θ scan for CeO_2 film of sample A, deposited at 790° C. **FIGURE** 1b. ω-scan for CeO_2 film of sample A, FWHM is 1.16°.

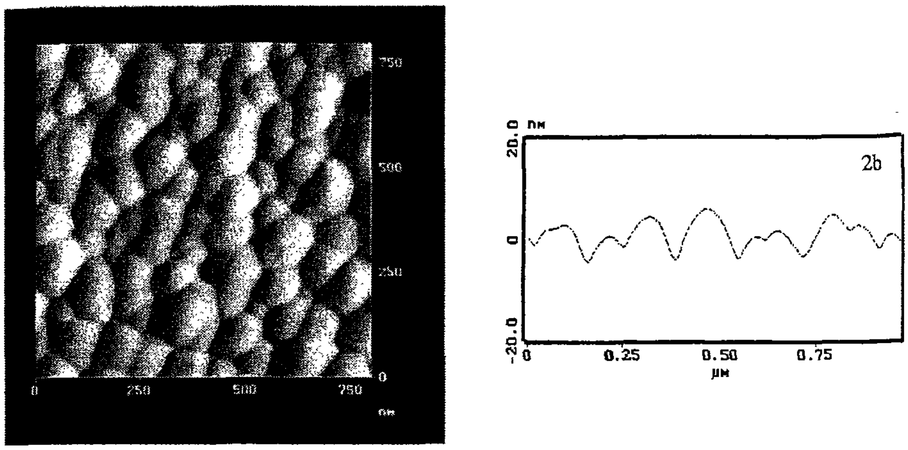


FIGURE 2a. AFM image of background structure of CeO_2 film of sample B. **FIGURE** 2b. Section profile of image in FIG 2a.

In some instances when the density of outgrowths is high, it is difficult to distinguish the outgrowths from the background structure. The formation of droplets, usually associated with the laser ablation process, was reduced in our case by using a long target-substrates distance.

Pure c-axis oriented CeO_2 films were obtained for all three samples. The X-ray θ-2θ scan of sample B is presented in FIGURE 1a, and the X-ray ω-scan of the (200) reflection for the same sample is presented in FIGURE 1b. In the case of samples A and C, the X-ray θ-2θ and the ω-scan of the (200) reflection are similar to these presented of sample B. However, the departure from an ideal c-axis orientation is larger for samples A and C, as compared to sample B. This departure was assessed using the full width at half maximum (FWHM) for the (200) reflection obtained in the ω-scan, and summarised in TABLE 1. The dependence of the FWHM value on the deposition temperature shows a minima of 0.16°

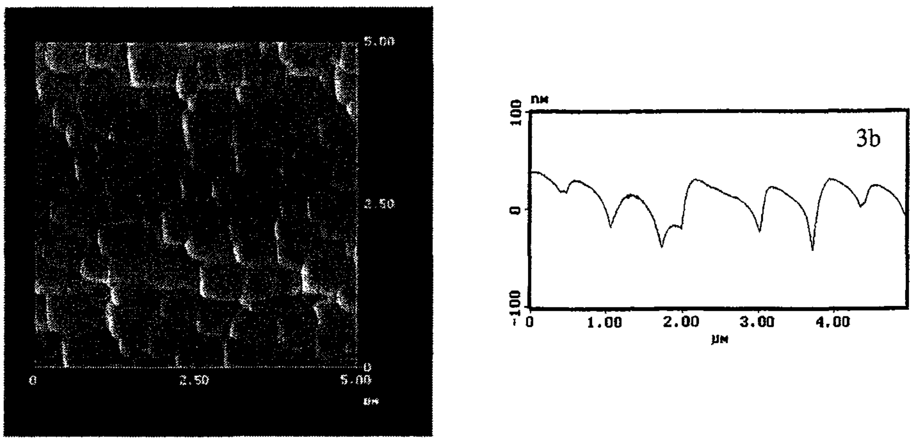
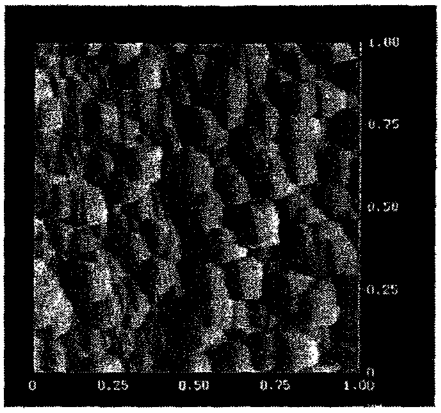


FIGURE 3a. AFM image of background structure of CeO_2 film of sample A. **FIGURE** 3b. Section profile of AFM image in FIG 3a.



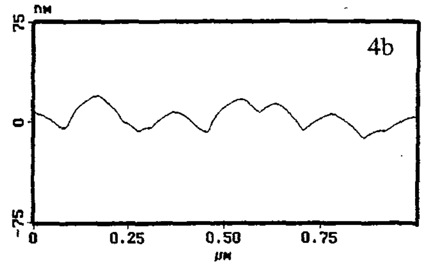


FIGURE 4a. AFM image of background structure of CeO_2 film of sample C. **FIGURE** 4b. Section profile of AFM image in FIG 4a.

around 790° C. This value of FWHM of CeO_2 film is comparable to the corresponding value of the YSZ substrate, of 0.15°. From this result it is inferred that the highest degree of c-axis orientation of CeO_2 film can be obtained at a deposition temperature of 790° C.

The morphology of the background structure was investigated by AFM for all three samples. The result for sample B which has the highest degree of c-axis orientation, is presented in FIGURE 2a. For this deposition temperature, the shape of the CeO_2 grains is approximately round, with an average size of approximately 100nm. The average roughness of this film determined on a scan area of (5x5) μm^2, was 3.62nm Rms. From cross section profile analysis, shown in FIGURE 2b, it appears that the top of the grains is located in the same layer.

Identical AFM measurements were performed on the samples A and C, and the AFM surface images are presented in FIGURE 3a and FIGURE 4a respectively. For the sample A deposited at the lower range of the deposition temperature, the shape of the grains is approximately square, and the average grain size is approximately 600 nm. The average roughness of this sample is 26.42 nm Rms, and the cross section profile analyses shown in FIGURE 3b suggests that the top of the grains is located in different layers.

In the case of sample C deposited at the higher end of the deposition temperature, the shape of the grains appears to be also square-like, but the average grain size is approximately 150 nm. The average roughness of this sample is 16.27nm Rms, and the cross section profile analyses shown in FIGURE 4b suggests again that the top of the grains is located in different layers.

A summary of the data obtained for samples A, B and C is presented in TABLE 1.

Develos K. D. et al studied the surface morphology of CeO_2 films deposited by PLD on Al_2O_3 single crystal substrate [6,7]. In this reference, the film deposited under optimum conditions has also round-shaped grains, and as the quality of the c-axis orientation is deteriorating, the shape of the grains which form the background structure is changing as well.

TABLE 1: Summary

Sample	Deposition Temperature [° C]	ω-FWHM [deg]	Average Roughness [nm Rms]	Average Grain Size [nm]	Grain Shape
A	775	0.19	26.42	600	Square-like
B	790	0.16	3.62	100	Round-like
C	805	0.18	16.27	150	Square-like

Wang A. et al studied the surface morphology of CeO_2 film deposited by metal-organic chemical vapour deposition (MOCVD) on YSZ single crystal substrates [8]. The result of this investigation shows again a correlation between the optimum c-axis orientation of CeO_2 film and the round shape of the grains, as well as a small value of surface roughness.

The results cited above, together with the results of our investigation, suggests that the c-axis orientation of CeO_2 film is related to the morphology of the grains, and independent of the substrates and deposition method. The more circular the shape of the grains is, the better the c-axis orientation and the smoothness of CeO_2 film surface are. It appears that this is an intrinsic feature of the CeO_2 films, and is related to its growth mode. A high degree of c-axis orientation is the result of a 2-D layer-by-layer growth. Any tendency not to restrict this growth mode results in a simultaneous growth on different layers (3-D), and a decrease in the c-axis orientation. This change in the growth mode is apparently independent of the initial roughness of the substrates, which in our samples was approximately 12nm Rms. It is apparent that the topography of the film surface grown by PLD reflects the morphology.

4. CONCLUSION

This paper presented a study of c-axis orientation of CeO_2 films grown on YSZ<100> single crystal substrates by PLD. It was found that the optimum epitaxial growth temperature in oxygen for a high level of c-axis orientation is 790° C. The degree of c-axis orientation is related to the morphology of CeO_2 grains. The more circular the shape of the grains is, the better the c-axis orientation and the smoothness of CeO_2 film surface are. This correlation appears to be an intrinsic feature of CeO_2 growth, independent of the type of substrate or the deposition method. A high degree of c-axis orientation is the result of a 2-D layer-by-layer growth. Any tendency not to restrict this growth mode results in a simultaneous growth on different layers (3-D), and a decrease in the c-axis orientation. Also it was found that the topography of the film surface grown by PLD reflects the morphology.

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