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# An approach to (103) oriented $YBa_2Cu_3O_{7-\delta}$ thin films epitaxially grown on (100) MgO substrates

J. Chen,[a] X.D. Su,[a,*] D.L. Zhu,[a] J. Fan,[a] H. Yang,[a] Y. Chen[b,*] and V.G. Harris[b]

[a]*Jiangsu Key Laboratory of Thin Films, Department of Physics, Soochow University, Suzhou 215006, People's Republic of China*
[b]*Department of Electrical and Computer Engineering, and the Center for Microwave Magnetic Materials and Integrated Circuits, Northeastern University, Boston, MA 02115, USA*

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The preferred growth orientations of $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films on (100) MgO substrates produced by pulsed laser deposition are reported. In particular, (103) perpendicular oriented films were successfully demonstrated. The crystallographic properties of the oriented films were examined by transmission electron microscopy and X-ray diffraction pole figure analyses, indicating a 45° tilt of the superconducting $CuO_2$ planes relative to the substrate surface. This work provides a unique pathway to realizing the growth of (103) oriented YBCO films on highly valued electronic and microwave (100) MgO substrates.


*Keywords:* YBCO thin films; (103) Orientation; Pulsed laser deposition; Pole figures of thin films; Domain epitaxy

Epitaxial $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films have demonstrated excellent high temperature superconducting (HTS) properties, such as critical temperatures $T_c$ greater than 90 K, critical current densities $J_c$ (77 K and 0 T) greater than $10^6$ A $cm^{-2}$ and microwave surface resistances $R_s$ (77 K and $f = 10$ GHz) less than 500 μΩ ($R_s(T,f) \propto f^2$) [1–3]. These unique properties have created significant opportunities for HTS materials in modern microwave communication electronics and in highly sensitive magnetic field sensors, among other applications [4–6]. Because HTS materials have demonstrated substantial performance advantages over conventional materials systems they offer an, as yet, largely untapped potential.

In the past two decades the fabrication of YBCO films has received considerable attention. Among these studies, the oriented growth and physical properties of YBCO films have been of particular interest, resulting in numerous engineering applications [2]. For example, perpendicularly *c*-axis oriented films are suitable for applications in superconducting devices and tapes which require large currents densities along the film/tape surface [7,8], *a*-axis oriented films are often used in multilayered structures such as those containing Josephson junctions [9,10] and (103) oriented films enable the measurement of the intrinsic anisotropic properties of YBCO due to a coherence length $\xi_{(103)}$ that is longer than that along the *c*-axis $\xi_c$ [11].

The oriented growth of films can be realized using physical vapor deposition techniques (e.g. pulsed laser ablation, molecular beam epitaxy, rf and dc sputtering, etc.) in which the operator can control adatom mobility by adjusting the deposition conditions. This, together with a substrate that is appropriately matched in terms of lattice constant and thermal expansion coefficient, such as MgO, $SrTiO_3$, $LaAlO_3$, etc., allows epitaxial and textured growth [12]. Previous work has indicated that (103) oriented YBCO films can be realized on the (110) $SrTiO_3$ or (110) $LaAlO_3$ substrates, whereupon the (110) plane functions as a template for (103) epitaxial growth of YBCO films. The resulting films have an intrinsic roughness of ~40 nm due to the (110) cut of the substrates inducing two different growth directions, (103) and ($\bar{1}$03) [13,14]. Tafuri et al. reported the growth of YBCO junctions with artificial grain boundaries at the interface of the *c*-axis and (103) oriented films on a (110) $SrTiO_3$ substrate [14]. Such oriented films feature a very low normal state resistivity, combined with substantial in-plane anisotropy in both the normal and superconducting states [15]. Hence, (103) oriented YBCO grown on MgO may offer further opportunities to develop new HTS devices.

* Corresponding authors; e-mail addresses: xdsu@suda.edu.cn; y.chen@neu.edu

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In this paper, we report a novel approach to realizing and stabilizing high quality growth of perpendicularly oriented (103) YBCO films on (100) MgO substrates. (100) MgO single crystals are widely used in electronic and microwave applications which, when combined with the textured growth of (103), *c*-axial and *a*-axial oriented YBCO films, could lead to efficient and flexible lateral junction structures that may find application in a broad range of microwave and spintronic applications [16].

Specifically, YBCO films were deposited on (100) MgO single crystal substrates using pulsed laser deposition (PLD). PLD used a 248 nm KrF Excimer laser (Lambda Physik Compex 205). The deposition conditions, such as laser energy density, pulse repetition frequency, and oxygen pressure (P), were systematically varied to explore energetically favorable conditions for epitaxy. Previous work on this system indicated that low deposition temperatures, i.e. below 720 °C, failed to stabilize oriented (103) YBCO films on a (100) MgO substrate (Su et al. unpublished). Here we demonstrate the growth of YBCO films at higher deposition temperatures, ranging from 720 °C to 840 °C over a broad range of oxygen pressures varying from 0.02 to 5 Pa (see Table 1). The optimized pulse repetition rate and laser energy density were $\sim$2 J cm$^{-2}$ and 3 Hz, respectively. After deposition all films, having a typical thickness of $\sim$200 nm, underwent in situ annealing at an oxygen pressure of 500 Torr and a temperature of 500 °C for 1 h. This was done in order to achieve a stoichiometric oxygen content.

The crystallographic properties of all films grown under different deposition conditions were examined by X-ray diffractometry (XRD) (Rigaku D-Max diffractometer, Cu$K_\alpha$ radiation) and transmission electron microscopy (TEM). The resulting relationships between the out-of-plane orientation and deposition conditions are listed in Table 1. It should be noted that there are principally three regions representing different crystallographic orientations in the PLD YBCO films: in region I (bold in Table 1) the high temperature and low pressure is favorable to (103) oriented growth; in region II (underlined in Table 1) the moderate temperature and pressure result in preferred growth of perpendicular *c*-axis oriented films; in region III (not bold or underlined in Table 1) the low temperature and high pressure correspond to a perpendicular *a*-axis orientation. There are regions of mixed crystallographic orientation: specifically, films within the transitional regions between regions I and II and regions II and III exist with a mixture of both *a*- and *c*-axis orientations. As a result, the (103) oriented films correspond to only a very narrow region of deposition conditions, i.e. high temperature (>810 °C) and low pressure (<0.2 Pa). In particular, highly oriented growth of the (103) plane is achieved at a substrate temperature of 810 °C and oxygen pressure of 0.02 Pa. Interestingly, an overheated substrate results in discontinuous crystal growth, resulting in misalignment of the crystallographic axes.

Several representative XRD patterns of YBCO films grown under different conditions and illustrating different crystallographic orientations are presented on the left-hand side of Figure 1. The results show dramatic variations in film orientation with substrate temperature and oxygen working gas pressure. The XRD spectra in Figure 1a–c have been indexed to single phase YBCO structures and verified as having a single crystallographic orientation, corresponding to *c*-axis, *a*-axis and (103) orientations, respectively. It should be pointed out that either *c*-axis or *a*-axis growth is controlled by the oxygen pressure at a common substrate temperature. Clearly, the oxygen pressure is a key growth parameter in determining the preferred crystallographic orientation due to the adatom motilities of the constituent elements of YBCO [17]. At higher growth temperatures, i.e. 810–840 °C, the (103) diffraction features broaden and (001) and (002) lines appear, as depicted in Figure 1d.

The surface morphology of the YBCO films grown under different deposition conditions are displayed on the right-hand side of Figure 1. The morphologies of the films as seen by SEM are fully consistent with the XRD analysis. For instance, the *c*-axis oriented film

**Table 1.** The out-of-plane orientations of YBCO films prepared at different temperatures (T) and oxygen pressures (P).

| T (°C) | P (Pa) | | | |
|---|---|---|---|---|
| | 0.02 | 0.2 | 2 | 5 |
| 720 | *c*[I] | *a*(+*c*)[a] | *a*(+*c*) | *a*[II] |
| 750 | *c* | *c* | *c*(+*a*)[b] | *a*(+*c*) |
| 780 | *c* | *c* | *c* | *c*(+*a*) |
| 810 | **(103)**[III] | *c* | *c* | *c* |
| 840 | **(103)** | **(103)** | *c*(+103)[c] | *a*(+103)[d,IV] |

[I–IV] Corresponding XRD and SEM results are presented in Fig. 1a–d, respectively.
[a] A primary *a*-axis orientation with a minor *c*-axis orientation.
[b] A primary *c*-axis orientation with a minor *a*-axis orientation.
[c] A primary c-axis orientation with a minor (103) orientation.
[d] A primary a-axis orientation with a minor (103) orientation.

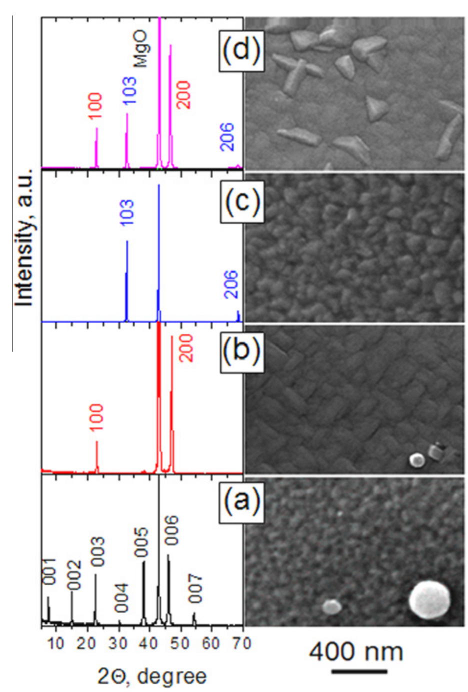


**Figure 1.** X-ray diffraction patterns (left) and SEM morphologies (right) for the YBCO films with different orientations: (a) *c*-axis; (b) *a*-axis; (c) (103); (d) *a*(+103).