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# Liquid phase epitaxy of $YBa_2Cu_3O_{7-\delta}$ film on single crystalline MgO and NdGaO<sub>3</sub> substrate

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Abstract. YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (YBCO) films were deposited by a liquid phase epitaxy (LPE) method on MgO single crystalline substrates that were covered by thin film of pulsed-laser-deposited (PLD) YBCO as a seed layer. The 990°C-deposited LPE film showed c-axis orientation perpendicular to the substrate surface and in-plain two -dimensional alignment. The LPE film contained twin structure with good crystalline perfection and the PLD film showed broad in-plain alignment, meaning lattice constant distribution or lattice distortion.

### 1. Introduction

Because of the high transition temperature, a film of oxide superconductor of  $YBa_2Cu_3O_{7-\delta}$  (YBCO) has been expected to apply to superconducting electronic devices. To construct multilayer structure and draw patterns on the film, a highly orientated defect free film, that is, a single crystalline film, is indispensable. For that purpose YBCO film deposition by a liquid phase epitaxy (LPE) method was performed because films with high crystalline perfection was easily obtained due to its near equilibrium growth process.

NdGaO<sub>3</sub> (NGO) substrates were often used for YBCO hetero-epitaxial deposition by the LPE method. However, the grown films contain cracks due to a difference of the thermal expansion between the substrate and the film [1]. Yamada *et al.* reported that YBCO films grown on MgO substrates have no cracks after complete oxygenation [2]. To prevent the reaction between MgO and the solution used in the LPE process and achieve epitaxial growth, a YBCO thin film as a seed layer was deposited by a pulsed laser deposition (PLD) method on MgO substrate. This paper describes crystallographical quality of the LPE grown YBCO film and the PLD seed layer of YBCO.

# 2. Experimental

YBCO deposition was performed by a temperature gradient solution growth method [3]. Raw materials of  $Y_2O_3$ , BaCO<sub>3</sub> and CuO were put into an yttria crucible and heated to 990°C to make solution in air. After keeping a proper period the solution consisting of the solute of YBCO on the bottom of the crucible and the solvent of Ba-Cu-O was obtained. The solvent composition was selected to be BaO:CuO=3:5 to grow YBCO as a primary phase. MgO (100) substrate covered by YBCO thin film formed by the PLD method was used for the substrate of the LPE growth. The MgO substrate was top-seeded horizontally on the solution surface and rotated at 30 rpm for 30 min.

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temperature difference between the solution surface and the bottom was about 5°C, making driving force of the growth.

#### 3. Results and discussion

Figure 1 shows surface morphology of an LPE grown YBCO film on an MgO substrate observed by optical microscopy. Surface morphology of an LPE grown YBCO film on a NGO substrate is also shown as a comparison. The obtained YBCO film on the MgO substrate have no cracks, on the other hand cracks are observed on the NGO substrate. It is confirmed that MgO substrate is suitable for YBCO film formation. Surface observation by optical microscopy also showed square shaped macrosteps, which is a typical surface morphology appearing on the (001) facet face of a YBCO crystal. This indicates that the LPE film is c-axis perpendicular to the surface. Some amounts of droplets of the solvent were inhered on the surface. Therefore, surface polishing is needed for the usage of substrate for the electronic devices.



Figure 1 Surface morphology of LPE grown YBCO film (a) on MgO and (b) NGO substrate.

 $\theta/2\theta$  X-ray diffraction (XRD) patterns are shown in Figure 2(a) for the LPE film and Figure 2(b) for the PLD seed layer. They indicated that both the films were c-axis oriented, indicating that the LPE film grows epitaxialy on the PLD film. No diffraction from MgO substrate can be seen on the LPE film. This means that the LPE film was much thicker than the PLD seed layer.

Lattice constants of the c-axis of the LPE grown film and the PLD seed layer were estimated to be 0.1690 nm and 0.1672 nm, respectively. Peak width of the diffraction peaks of the PLD layer is broader than the LPE film, meaning that lattice constant in c-axis of the PLD YBCO has distribution.



Figure 2  $\theta/2\theta$  X-ray diffraction (XRD) pattern of (a) LPE film and (b) PLD seed layer on MgO substrate

 $\phi$ -scan XRD of the LPE film, which was measured using YBCO(104) diffraction showed four-fold symmetry due to c-axis orientation as shown in Figure 3(a). However, each diffraction has two peaks with angle interval of about 2°, as shown in Figure 3(b). The peak splitting indicates that there was an in-plain miss-orientation alignment with the angle of about 2°. This miss-orientation is an obvious evidence of twin formation arising from tetragonal to orthorhombic transition during cooling process after the growth.



Figure 3  $\phi$ -scan XRD of the film (a) showing four-fold symmetry and (b) in-plain miss-orientation

On the other hand,  $\phi$ -scan of the PLD seed layer shows a broad peak with no peak split as shown in Figure 4. The width of the peak is broader than that of the LPE film. If orthorhombic a- and b-axis lattice parameters are the same as those of the LPE film, the same peak split should be observed. No peak split and single broader peak indicate that a-/b-axis lattice parameters are widely distributed. The c-axis lattice parameter of the PLD film is shorter than that of the LPE film, meaning that oxygen content is sufficient to form twin structure. Therefore, oxygen deficiency in the film is not the reason for the lattice parameter distribution.



Figure 4  $\phi$ -scan of the PLD seed layer.

The PLD film was deposited on an MgO substrate hetero-epitaxialy. The lattice mismatch is more than 8% and the YBCO lattice should receive a tensile stress. Possible reason of the single broader inplain alignment is that the PLD YBCO lattice is distorted by the stress and the distortion is inhomogeneous. In-plain a-/b-axis lattice distortion should affect c-axis lattice. Therefore, This

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understanding is consistent with the broader width of  $\theta/2\theta$  XRD line of the PLD film than the LPE film. Consequently, crystalline perfection of the PLD film is inferior to the LPE film.

# 4. Summary

Crack free YBCO film was formed by the LPE method on the PLD seed layer on MgO substrate. The film was oriented c-axis perpendicular to the surface.  $\theta/2\theta$  XRD shows that crystalline lattice perfection is higher in the LPE film than in the PLD layer.  $\phi$ -scan XRD indicated there are in-plain miss-orientation with angle of about 2° in the LPE YBCO film, however, the PLD seed YBCO has no apparent miss-orientation but shows broader distribution. Therefore, it is concluded that the LPE grown film had twin structure with high crystalline perfection and the PLD film had distributed distortion, causing less crystalline perfection.

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