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A method to fabricate biaxially textured MgO buffer layer for HTS coated conductor

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***Abstract*—In this study, a radio frequency magnetron sputtering system without any assisting ion source was employed to fabricate biaxially textured MgO films on the substrates of amorphous Y_2O_3/Al_2O_3/Hastelloy stacks. During the deposition process, the growing MgO film could be bombarded by off-normal energetic particle flux mainly composed of oxygen atoms and negative oxygen ions, which originated from the target surface. MgO could obtain biaxial texture due to such bombardment, thus this method was named as energetic particle self-assist deposition (EPSAD). The texture of EPSAD-MgO films was evaluated by XRD measurement of the post-deposited homo-epitaxial MgO layers. An out-of-plane orientation of MgO (111) and a 3-fold symmetric in-plane alignment were found. The MgO texture was optimal when the EPSAD-MgO thickness was about 10 nm. The establishment of biaxial texture during EPSAD process was verified by a control study, the influence of target inclined angle and target-substrate distance was also investigated. Compared with inclined substrate deposition (ISD) and ion beam assisted deposition (IBAD), the mechanism of EPSAD-MgO method was discussed. This study proposed a new method to fabricate biaxially textured MgO buffer layer for coated conductors, more optimization research will be conducted in our future study.**

***Index Terms*—MgO film, biaxial texture, in-plane orientation, magnetron sputtering.**

I. INTRODUCTION

Since the discovery of cuprate superconductors with T_c (critical temperature) over nitrogen boiling point (77 K) in the late 1980s, huge research efforts have been devoted to develop them into flexible wires. RE-123 superconducting materials ($REBa_2Cu_3O_{7-\delta}$, RE: rare earth elements) have excellent current property, but they have sensitive inter-granular weak-links and the critical current decreases sharply with increasing grain boundary angles [1], [2]. Therefore, the RE-123 superconducting films need high quality biaxial texture. The general way is to prepare proper buffer layers on a flexible Ni-based alloy tape that can serve as a biaxially textured template for the epitaxial growth of RE-123 layer [3], [4].

MgO thin film is widely regarded as a promising buffer layer, and two techniques have been proposed to develop biaxially textured MgO on non-textured substrates: inclined substrate deposition (ISD) [5], [6], [7] and ion beam assisted deposition (IBAD) [8], [9], [10]. ISD method, initially demonstrated by Sumitomo [5], can produce favorable biaxial texture using a vapor source at an inclined angle relative to the substrate normal, but the MgO thickness over 1 μm is required for epitaxial growth of superconducting layer [7]. IBAD can produce biaxial texture using the oblique assisting ion bombardment during the buffer layer deposition, and IBAD-MgO, initially developed at Stanford University in 1997 [9], has been demonstrated that a very small thickness (about 10 nm) is enough for favorable texture.

Magnetron sputtering deposition is a popular technique for buffer layer fabrication of coated conductors, such as the CeO_2 cap layer [11], on a textured template. During the magnetron sputtering deposition, the growing oxide film is usually simultaneously bombarded by energetic particles mainly composed of oxygen atoms and negative oxygen ions with energy of several hundred eV, which is named as re-sputtering effect [12], [13]. In our previous research [14], a well collimated energetic particle flux was reported, and its function similar to the ion beam in the IBAD process was proposed. In this work, we demonstrated a new method using the magnetron sputtering system to fabricate biaxially textured MgO buffer layer on non-textured substrates, named as energetic particle self-assist deposition (EPSAD). The sputtering parameters of deposition duration, target inclined angle and target-substrate distance were optimized, and the texture mechanism was discussed.

II. EXPERIMENTAL PROCEDURE

An off-axis radio-frequency unbalanced magnetron sputtering system with a magnesium metal target (99.99% pure, 50 mm in diameter and 4 mm thick) was used in this study [Fig. 1 (a)]. The sputtering atmosphere included oxygen and argon with a mole ratio of 1/7. The background pressure in the chamber was below 5×10^{-4} Pa, and the total sputtering pressure was fixed at 0.3 Pa. The details of the deposition parameters could also be found in our previous publication [14].

In EPSAD process, Hastelloy C276 alloy tapes (5 mm× 5 mm) coated with amorphous Y_2O_3/Al_2O_3 seed layers were used as substrate. The substrate was placed in the area directly facing the target center [the vicinity of point A as shown in Fig. 1 (a)], thus MgO deposition was accompanied with the bombardment of energetic particles. The angle α between the target normal and the substrate normal was 55°. The deposition was carried out at room temperature, with target power of 70

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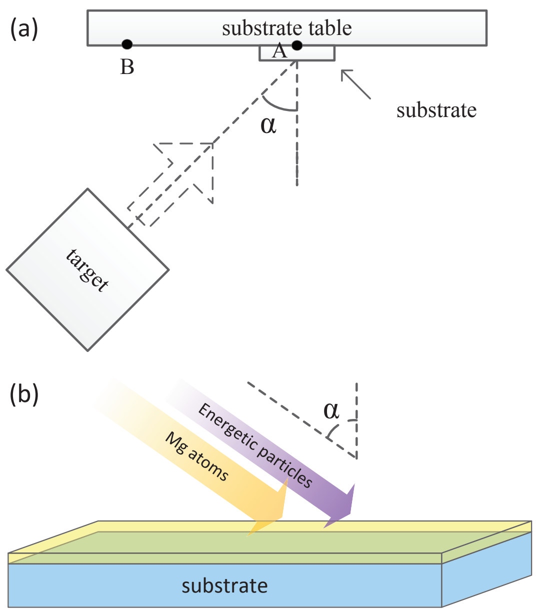

Fig. 1. Schematic of (a) the off-axis magnetron sputtering system and (b) energetic particle self-assist deposition (EPSAD).

W and target-substrate distance of 90 mm. EPSAD duration varied from 0.5 min to 20 min. According to our previous study [14], the MgO growth rate was about 2 nm/min.

In order to conduct X-ray Diffraction (XRD) characterization using Rigaku SmartLab for texture evaluation of the EPSAD-MgO film, a homo-epitaxial MgO layer about 100 nm thick was post-deposited. The homo-epitaxial deposition was carried out at a substrate temperature of 550°C in the area not bombarded by energetic particles, i.e. the vicinity of point B as shown in Fig. 1 (a).

III. Results

A. EPSAD duration optimization for biaxial texture

The XRD $\theta - 2\theta$ scan patterns for all homo-epi MgO/EPSAD-MgO films are shown in Fig. 2. For the sample with EPSAD duration of 0.5 min (about 1 nm thick), MgO peak was not found. For the samples with longer EPSAD duration (1∼20 min), MgO (111) peak could be observed, indicating that all these films were (111) out-of-plane oriented. MgO (100) ϕ scan patterns of the samples with EPSAD duration longer than 1 min are shown in Fig. 3, demonstrating in-plane alignment in these samples. In a word, the biaxial texture with (111) out-of-plane orientation and 3-fold symmetric in-plane orientation could be established in the MgO films with EPSAD duration longer than 1 min, i.e. with EPSAD-MgO layer thickness ranging from 2 nm to 40 nm.

Fig. 4 shows MgO (111) peak intensity and ϕ-scan FWHM (full width at half maximum) of all the films plotted against the EPSAD duration. When the EPSAD duration was longer than 1 min, biaxial texture began to develop and improved with increasing the EPSAD duration until it reached an optimum at about 5 min, and the optimal ϕ-scan FWHM ($\Delta\phi$) was about 18°. When the EPSAD duration was longer than 5 min, further

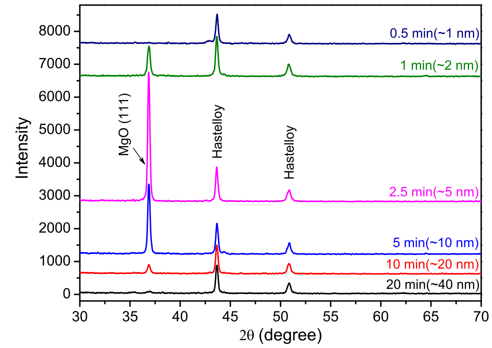

Fig. 2. XRD $\theta - 2\theta$ scan patterns of the homo-epi MgO/EPSAD-MgO/Y_2O_3/Al_2O_3/Hastelloy samples.

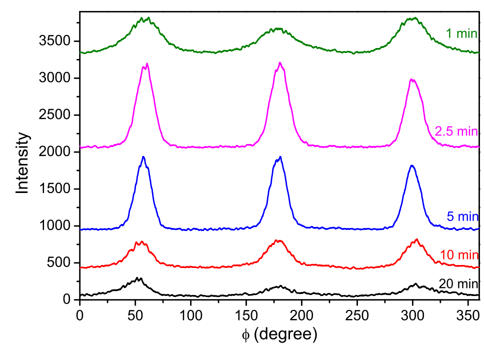

Fig. 3. MgO (100) ϕ scan patterns of the homo-epi MgO/EPSAD-MgO/Y_2O_3/Al_2O_3/Hastelloy samples.

increasing the EPSAD duration would cause degradation in the texture.

The rocking curves for homo-epi MgO/EPSAD-MgO samples are shown in Fig. 5, representing the out-of-plane texture. The behaviors of the rocking curve FWHM and the tilt angle of MgO <111> axis are illustrated in Fig. 6. The rocking curve FWHM was optimal at the EPSAD duration of about 2.5 or 5 min, which was similar to the ϕ scan result.

The above texture variation with the deposition duration was similar to that reported for IBAD-MgO [15], [16], [17]. In IBAD-MgO process, when the thickness of IBAD-MgO layer was greater than the optimal value, increasing the thickness (i.e. the IBAD duration) would cause texture degradation and tilting of MgO <001> axis.

B. Control study to verify the texture establishment by EPSAD

As demonstrated above, biaxial texture could be detected in most of the homo-epi MgO/EPSAD-MgO/Y_2O_3/Al_2O_3/Hastelloy samples. The XRD measurement of homo-epitaxial MgO for texture evaluation was a traditional and indirect method in IBAD studies [9], [15], [18]. However, *in situ* characterization of texture evaluation using reflection high-energy electron diffraction (RHEED) was not available for us. Considering the fact that the square substrate was 4-fold symmetric and the measured in-plane texture was

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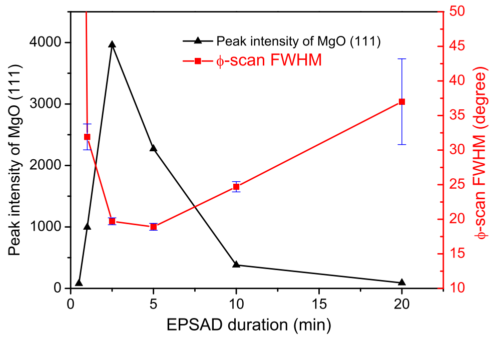

Fig. 4. The MgO (111) peak intensity and in-plane texture variation with EPSAD duration.

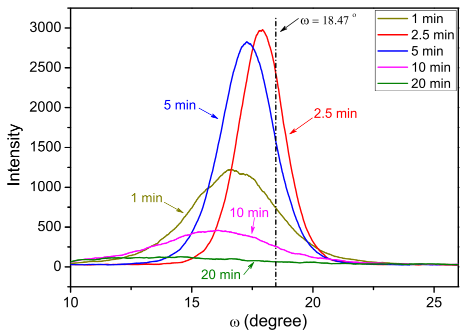

Fig. 5. The MgO (111) ω-scan patterns (rocking curve) of the homo-epi MgO/EPSAD-MgO/Y_2O_3/Al_2O_3/Hastelloy samples (2θ=36.94°, θ=18.47°) for different EPSAD duration.

3-fold symmetric, a control study was designed to verify the establishment of MgO biaxial texture during EPSAD process, not during the homo-epitaxial deposition process.

In Fig. 7, the sample arrangement of three deposition routes are sketched. The sample prepared via route A was the control sample. In route B, the only difference from A was the 90° rotation in the epitaxial growth stage, and the obtained sample had almost the same in-plane ϕ scan pattern as the control sample. In route C, the only difference from A was the 90°

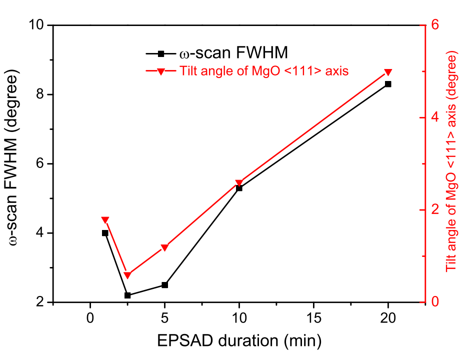

Fig. 6. The behaviors of rocking curve FWHM and tilt angle of MgO <111> axis with different EPSAD duration.

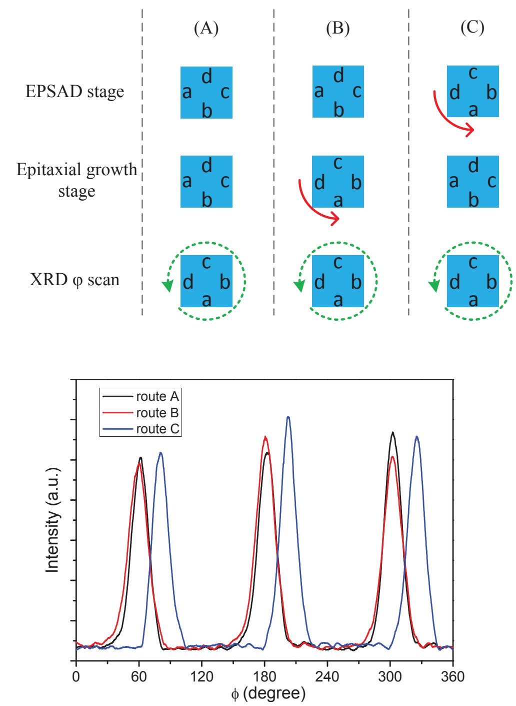

Fig. 7. The sample arrangement of three deposition routes designed as a control study, and the in-plane texture patterns of the obtained samples.

rotation in the EPSAD stage, and the peaks in ϕ scan pattern of the obtained sample shifted 90° to the left (i.e. 30° to the right), compared with that of the control sample. Therefore, it could be verified that the biaxial texture originated from the EPSAD-MgO layer.

C. Influence of target inclined angle and target-substrate distance

In order to study the influence of target inclined angle α on the texture, α was changed from 55° to 45° or 35°. At each angle, the in-plane texture was optimized. The MgO (100) ϕ scan patterns of the optimal samples are shown in Fig. 8. The intensity of the peak decreased when decreasing the inclined angle, but the ϕ-scan FWHM is almost the same and the position of the peak remained unchanged. The change of α, i.e. the incidence angle of the energetic particle flux, did not change the in-plane alignment of MgO films. Summarizing all these cases, the projection of the incidence direction of energetic particle flux onto the substrate surface, indicated in the inset image of Fig. 8, is always perpendicular to MgO <101> axis.

The target-substrate distance (d_{s-t}) was also changed from 5 cm to 15 cm, and the in-plane texture was also optimized for each d_{s-t}. The ϕ scan FWHM of the optimal samples are plotted against d_{s-t} in Fig. 9. When d_{s-t} ranged from 9 cm to 15 cm, FWHM was almost stable, about 18°, while decreasing of d_{s-t} to 5 cm would result in a degraded texture.

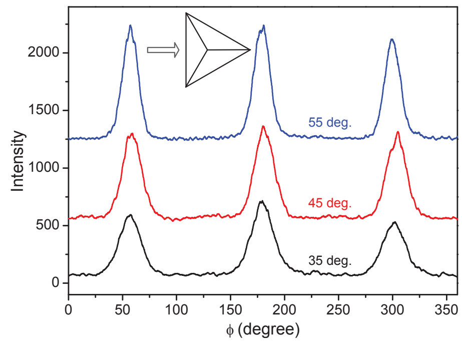

Fig. 8. MgO (100) ϕ scan patterns of the homo-epi MgO/EPSAD-MgO/Y_2O_3/Al_2O_3/Hastelloy samples optimized at three target inclined angle. Figure inset: schematic view from the top of a (111) out-of-plane oriented MgO cell limited by {100}-facets and the energetic particle flux indicated by the arrow.

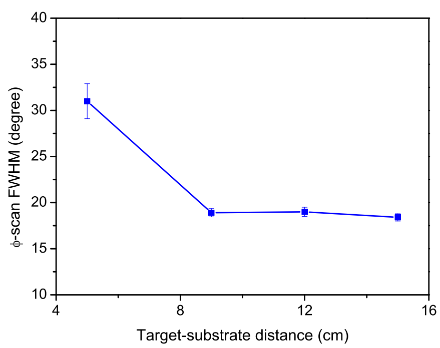

Fig. 9. Influence of target-substrate distance on the in-plane texture.

IV. Discussion

As demonstrated above, biaxial texture could be established in the EPSAD-MgO process. In this study, Mg atoms arrived at the substrate from an off-normal direction, similar to ISD-MgO process. The oblique incidence of Mg atoms might induce biaxial texture through the ISD mechanism. However, the competition growth mechanism in ISD process requires the MgO thickness of about 1 μm to form biaxial texture [5], [6], [7], while the thickness of about 10 nm was optimal for EPSAD-MgO layer in this study. Therefore, we supposed the texture mechanism of EPSAD-MgO is similar to that of IBAD-MgO, which also requires the thickness of about 10 nm [9], [19], [20]. Moreover, the energetic particles in EPSAD are well collimated and have energies of several hundred eV, according to our previous study [14]. Such characteristics are similar to the assisting Ar ions in IBAD process. Thus the energetic particles might play an important role in the nucleation stage to cause the biaxial texture of MgO films.

As mentioned in section III C and indicated in Fig. 8, the energetic particle flux could only control the in-plane alignment of MgO films. This phenomenon could also support the mechanism similarity between EPSAD and IBAD. According to the IBAD-MgO report of Wang [15] and the simulation of ion sputtering yield of MgO by Dong et al. [21], [22], the assisting ions in the IBAD-MgO process could help to establish the in-plane texture, but not determine the out-of-plane orientation. In this study, as shown in Fig. 8, the direction of energetic particle flux was perpendicular to MgO $<101>$ axis. Such a fact might be due to the sputtering yield anisotropy of MgO, which was proposed to determine the in-plane texture of MgO [21], [22] as the out-of-plane orientation was determined by other factors.

Although the establishment of biaxially textured MgO by EPSAD was verified in this study, there are still two issues in the EPSAD-MgO fabrication method. First, the in-plane texture is 3-fold symmetric. Although there was also 3-fold symmetric in-plane texture established in the IBAD-MgO process [23], the practical IBAD-MgO buffer layers utilized in the coated conductors are generally 4-fold symmetric. Second, the FWHM value of ϕ-scan patterns in this work (about 18°) is larger than that of IBAD-MgO (typically about 5° [18]). These issues might cost the feasibility of EPSAD-MgO film to serve as a good buffer layer for the coated conductor. More optimization work for other parameters of the EPSAD-MgO process, such as gas content and substrate temperature, will be conducted in our future research in order to overcome the issues.

V. Conclusion

In this study, energetic particle self-assist deposition (EPSAD), was demonstrated as a new method to fabricate biaxially textured MgO buffer layers on non-textured substrates. It was verified that the biaxial texture was induced by the EPSAD process. The EPSAD-MgO process without any ion source was supposed to have a mechanism of texture establishment similar to IBAD-MgO process. XRD characterization of the post-deposited homo-epitaxial MgO layer exhibited that the MgO layer was (111) out-of-plane oriented with 3-fold symmetric in-plane orientation. The optimal FWHM of MgO ϕ scan was about 18°. Further improvement of the biaxial texture by EPSAD method will be carried out for subsequent epitaxial growth of YBCO superconducting layer.

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