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Modifying the critical current anisotropy of YBCO films via buffering layers on IBAD-MgO based templates

M. Z. Khan^{1,2}, E. Rivasto^{1,2}, Y. Wu³, Y. Zhao³, C. Chen⁴, J. Zhu⁴, H. Palonen¹, J. Tikkanen¹, H. Huhtinen¹ and P. Paturi¹

¹ Wihuri Physical Laboratory, Department of Physics and Astronomy, FI-20014 University of Turku, Finland

 2 University of Turku Graduate School (UTUGS), University of Turku, FI-20014 Turku, Finland

³ School of Electronic Information and Electrical Engineering, Shanghai Jiao Tong University, 200240 Shanghai, Peoples Republic of China

⁴ Shanghai Superconductor Technology Co. Ltd., 200240 Shanghai, Peoples Republic of China

E-mail: mukarram.z.khan@utu.fi

Abstract. The role of variations in the buffer layer structure of IBAD-MgO based templates on the critical current anisotropy has been investigated in undoped and $BaZrO_3$ (BZO) doped $YBa_2Cu_3O_{6+x}$ (YBCO) films. Not only do the natural defects grow distinct within the undoped YBCO lattice but also due to the different lengths of BZO induced nanorods within the YBCO matrix, the flux pinning properties are greatly affected by the underlying layers which in turn has a great impact on the angular dependent critical current density $J_{\rm c}(\theta)$. This has been verified by transport measurements where the shape of the $J_{\rm c}(\theta)$ varies in accordance with the substrates. Based on our results, the template having a cap layer with the minimum lattice mismatch and a good chemical compatibility with deposited YBCO is proven to be the best for growing both the undoped and BZO doped YBCO films. Furthermore, a model we can present based on the shapes of $J_{\rm c}(\theta)$ curves depicts how the formation of nanosized defects affects the flux pinning anisotropy.

1. Introduction

Superconductivity is concerned with the very act of cooling down the certain materials to their critical temperatures that brings about the exciting phenomenon of zero resistance. The discovery of high temperature superconductors (HTS) more than three decades ago opened up the gates to electrical power applications which are currently in frequent use [1, 2]. The potential of HTS realized in electrical power systems is arguably thrilling but one has to admit that HTS are complex, difficult to process and expensive to produce which all limit their usability to certain scales.

One of the main obstacles faced in solving these issues is that the structural growth dynamics of HTS restricts the range of suitable deposition substrates to certain flexible but complicated tapes manufactured by several methods [3]. From a laboratory to an industrial scale, the underlying substrate is not just the spectator but it generally plays a critical role on the growth of HTS film. On the other hand, the vortex motion in HTS under the magnetic fields (B)

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Figure 1. Schematics of IBAD-MgO based templates used for growing YBCO films (a)-(c). The three templates, CeO₂ (a), LMO-MgO (b), and LMO-Epi-MgO (c), differ from each other by the thicknesses and compositions of the buffer layer stacks grown on IBAD-MgO.

needs to be halted for zero resistance which leads to enhanced critical current density J_c and the usual approach for doing so is the introduction of artificial pinning sites within the lattice [2]. Moreover, besides other naturally created defects that appear during the growth process, the self-organized threading dislocations originating from the low-angle grain boundaries also act as strong vortex pinning sites. The out-of-plane threading dislocations are more pronounced when HTS films are grown on technical templates than with those deposited on single crystalline substrates due to larger lattice mismatch in the former [4, 5]. But the films on technical templates exhibit smaller J_c and they are more isotropic in comparison to the films grown on single crystalline substrates. However, thinking about their application, the HTS are required to be flexible so that they can easily be bent and wrapped. Due to these reasons, there is a motivation for developing technical templates that are less complicated, cost effective and sufficiently reliable for the growth of HTS films. Therefore, the fundamental understanding about this issue is a priority, should we want to improve the HTS growth from the substrate surface to the film top in pursuit of high critical currents (I_c) with small anisotropy.

Herein we present the superconducting properties of YBCO films grown on three different ion beam assisted deposition—magnesium oxide (IBAD-MgO) based templates. We investigated the properties of both undoped and artificially BZO doped YBCO films and observed the relative differences in the I_c anisotropies with respect to varying substrate buffer layers architectures. Furthermore, a schematic model of the growth of the natural and artificial defects within the YBCO lattice when grown on these distinct substrates is proposed.

2. Experimental details

Undoped YBCO films and YBCO films doped with 4 % BZO by weight were deposited on three variably structured $5 \times 5 \text{ mm}^2$ IBAD-MgO based templates by the pulsed laser deposition (PLD) method. The schematics of templates are shown in Fig. 1 where the differences in top buffer layers can be seen. From here onwards, the structures in the subfigures (a), (b) and (c) in Fig. 1

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Template	YBCO lattice	$\Delta 2\theta$	$\Delta \phi$	I(005)/		
	c (Å)	(004) (°)	(102) (°)	I(004)		
undoped YBCO						
CeO_2	11.69	0.12	3.40	14.0		
LMO-MgO	11.70	0.15	4.36	13.7		
LMO-Epi-MgO	11.70	0.14	3.77	13.5		
YBCO + BZO						
CeO_2	11.74	0.19	3.37	13.9		
LMO-MgO	11.75	0.21	4.42	12.5		
LMO-Epi-MgO	11.73	0.20	4.75	12.9		

Table 1. Structural parameters for undoped and 4 % BZO doped YBCO thin films grown on three different IBAD-MgO based templates.

will be referred as CeO₂ for CeO₂ cap layer, LMO-MgO for LaMnO₃ cap layer and LMO-Epi-MgO for LaMnO₃-Epi-MgO, templates, respectively. Firstly, three sets of undoped YBCO films were deposited on these templates at temperatures from 625 °C to 850 °C with 25 °C step size to optimize the growth temperature (T_g). The optimized T_g emerged as 775 °C and all the films were deposited at this T_g . A XeCl laser (wavelength $\lambda = 308$ nm, pulse duration 25 ns and pulse repetition rate 5 Hz with 1.3 Jcm⁻² laser fluence) was used as a source for cleaving off the target material and depositing it on the substrate. The applied 1600 laser pulses with a growth rate of ≈ 0.1 nm/pulse gave us the films' thicknesses as ≈ 160 nm.

The structural properties were characterized using a Panalytical Empyrean X-ray diffractometer (XRD) and performing (00*l*) directional θ -2 θ and ϕ scans to observe the phase purity and in-plane crystallographic texturing of all the films, respectively. A Quantum Design physical property measurement system (PPMS) was utilized for magnetic measurements, where ac magnetization curves were measured from 10 K to 100 K at 0.1 mT ac field for determining the critical temperatures (T_c).

For transport measurements, all the films were patterned by wet chemical etching to obtain 200 μ m wide current stripes. The electrical contacts from Quantum Design dc pucks to sample pads were made by using TPT HBO5 Wire bonder with 33 μ m thick Al wire. Then the angular dependent J_c anisotropies were measured using a horizontal rotator available in the PPMS. All the measurements were done at the temperature of 40 K under 0.5 T, 1 T, 2 T, 4 T, 6 T and 8 T magnetic fields and 0° to 360° angular range. 40 K was chosen as the measurement temperature as it is far below the T_c and is also a reasonable intermediate temperature when thinking about the low (10 K) and high temperature (77 K) applications.

3. Results and discussion

3.1. Crystallographic and superconducting properties

The most important structural parameters of the grown films are presented in Table 1. The *c*-axis lengths calculated by the Nelson-Riley method [6] reveal that undoped films have the *c*-parameters very close to the nominal value (11.68 Å) of YBCO [7] in comparison to all the doped films where relatively long *c*-axis is observed. This depicts the BZO as a cause of lattice perturbation during the growth process. The $\Delta 2\theta$ shows the undoped films as a winning competitor in terms of better structural growth of YBCO compared to the BZO doped films. Here the notation $\Delta 2\theta$ is the full width at half maximum of the peak. We observe that the films grown on CeO₂ template have the least variation along *c*-axis among the other templates. For both the undoped and BZO doped films, the $\Delta 2\theta$ values are way smaller than our previously investigated templates [4, 8, 9, 10] showing signs of texture improvement in our current substrates. In addition, the $\Delta \phi$ values for all the films are varying with respect to the

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Figure 2. Magnetization curves for the undoped and BZO doped films grown on various templates.

templates, being only half to those reported earlier for YBCO films on NiW substrates [8]. This in-plane $\Delta \phi$ (102) lattice variation is crucial in flux pinning as it correlates positively with the severity of the low-angle grain boundaries which channel the YBCO growth along its *c*-axis and finally leads to the pass through of threading dislocations from the interface to the film surface. Conclusively, the oxygen content for all the films is well in the acceptable range, that is, $\delta < 0.1$ [11].

Fig. 2 displays the temperature dependences of magnetization (deriving the corresponding T_c values) for all the films. The undoped films have higher T_c and sharper transitions than the doped ones which is attributed to the local strain field arising due to the lattice mismatch (~8%) between YBCO and BZO and also the possible local oxygen deficiency that can occur around BZO nanocolumns [9, 12, 13]. Comparing the templates, we see that the undoped YBCO grown on CeO₂ has the highest T_c which can be due to less strain caused by the least lattice mismatch between the CeO₂ and YBCO layers. Among the doped films, YBCO deposited on CeO₂ and LMO-Epi-MgO follows the similar T_c curves but LMO-MgO has the lowest T_c and the broadest transition. Our goal is to get as high a J_c as possible at high B so we believe that the importance of the T_c values should be assessed on a per-application basis.

3.2. Angular dependent J_c and anisotropy

In order to compare the $J_c(\theta)$, we shifted the data separately for each constant value of B in such a way that the lowest point for every curve falls at the same level. Moreover, the c-peak always occur at 0°, whereas the ab-peaks falls at \pm 90°. In Fig. 3(a), we display the $J_c(\theta)$ curves for undoped films measured at 0.5 T, 1 T and 2 T. It is observed that, at 0.5 T, both the films grown on CeO₂ and LMO-MgO are almost completely isotropic with the exception of very slight but broad ab-peaks. The $J_c(\theta)$ for film on LMO-Epi-MgO is more anisotropic where it shows the wide and pronounced ab-peaks. The occurrence of ab-peaks in the latter can be due to the more pronounced vortex pinning across the stacking faults. With increasing B from 0.5 T up to 2 T, we see that the shape of $J_c(\theta)$ curves for CeO₂ and LMO-MgO starts to be anisotropic, that is, the appearance of ab-peaks and occurrence of c-peak for film on CeO₂ is clearly visible. The cpeak for film on CeO₂ template should be attributed to the vortex pinning within the threading dislocations formed by the low-angle grain boundaries [14]. It is notable that with increasing B, ab-peaks become narrower and sharper for film on LMO-Epi-MgO template. This can be related to the presence of the greater number of pinning paths along ab-direction which pin



Fig. 3. $J_{c}(\theta)$ curves for undoped (a)–(b) and BZO doped (c)–(d) YBCO films grown on CeO₂, LMO-MgO and LMO-Epi-MgO templates measured at different *B* and 40 K.

the increasing number of vortices with respect to higher B [15]. This means that the template governs the YBCO growth and is responsible for the overall $J_{\rm c}(\theta)$.

Fig. 3(b) displaying the $J_{\rm c}(\theta)$ graphs for all the undoped films measured at 4 T, 6 T and 8 T reveals the remarkable differences in the shape of the curves when comparing with those measured at 0.5 T-2 T. Firstly, the c-peak for film on CeO_2 template is almost indiscernible at 4 T. As can be seen, that the curves are still somehow isotropic at 4 T for films on CeO₂ and LMO-MgO templates, we observe the subtle and strange behaviour of *ab*-peaks for film on LMO-Epi-MgO. Earlier we have also reported such a behaviour for the YBCO films grown on single crystalline $SrTiO_3$ (STO) [4]. With increasing B, the ab-peaks are observed to be intense along with the pronounced shoulderings. This means that the film grown on LMO-Epi-MgO template has more pinning paths along *ab*-direction. Generally, the undoped films have more pinning paths along ab-direction than the c-axis [15]. The shoulders seen beside the ab-peaks can be due to the interplay of in-plane and out-of-plane pinning [16]. Back to the film on CeO_2 template, it is clear that the $J_{\rm c}(\theta)$ starts to follow anisotropic behaviour from 6 T where relative broad *ab*-peaks with a wide drop in J_c curve along 0° measured at 8 T can be observed. Finally, in the case of film on LMO-MgO template, the relative isotropic $J_{\rm c}(\theta)$ is observed even at high B. These results reveal that not only do the natural pinning defects vary in number but they also differ in sizes with respect to the underlying template as can be deduced from an analysis of the differences in the shapes of $J_{\rm c}(\theta)$ curves (Figs. 3(a) and (b)). Especially, the film on LMO-Epi-MgO template behaves exceptionally in terms of anisotropic $J_{\rm c}(\theta)$ as compared with

its other two counterparts where $J_{\rm c}(\theta)$ is seen to be more isotropic.

Figs. 3(c) and (d) display the $J_{\rm c}(\theta)$ of BZO doped YBCO films on the set of studied templates. In Fig. 3(c), the measured curves at 0.5 T-2 T reveal that $J_{\rm c}(\theta)$ changes shape with respect to B. At 0.5 T, the shape of $J_{\rm c}(\theta)$ for BZO doped films on CeO₂ and LMO-MgO templates is quite similar to those of undoped films measured at 2 T. This is related to less c-axis flux pinning at this B and the pinning mostly occurs in-plane which in turn gives us broad ab-peaks. Unlike in the undoped case, more isotropic $J_{\rm c}(\theta)$ is seen at this B for film on LMO-Epi-MgO template which is due to the pinning of vortices by the dopant thus decreasing the anisotropy. As the BZO grows in the form of nanocolumns along the *c*-axis in both the single crystalline and buffered metallic templates, it is obvious that they are pinning the vortices effectively [12, 17]. The $J_{\rm c}(\theta)$ measured at 1 T already reveals the vortex trapping by the induced nanocolumns as we see the c-axis peaks in both the films on CeO_2 and LMO-MgO templates, whereas the film on LMO-Epi-MgO template has not shown any c-peak. This behaviour is related to the lengthened nanorods in films on CeO_2 and LMO-MgO, whereas the shorter nanocolumns in film on LMO-Epi-MgO template are weaker in vortex pinning. The broad and intense c-peaks along with very intense ab-peaks are visible at 2 T for films grown on CeO₂ and LMO-MgO templates. However, the total vortex pinning is not only attributed to the BZO nanocolumns but also to the other naturally created defects during the growth process. This is due to the fact that ab-peaks were seen to become more pronounced with increasing B from 0.5 T-2 T in all the films.

Fig. 3(d) depicting the $J_{\rm c}(\theta)$ at high B (4 T) indicates a strong flux pinning accross YBCO c-axis for all the films. At 4 T, the film on CeO₂ template again beats its counterparts in terms of isotropic $J_{\rm c}(\theta)$. A very strong c-peak in this case predicts the nice and elongated growth of BZO nanocolumns within YBCO which are both similar in shape and size to that of vortices [4]. Due to the occurrence of a wide c-peak via enhanced c-axis flux pinning, the ab-peaks tend to be narrow and less intense [16]. A similar tendency is seen for the film grown on LMO-MgO template, whereas with intuitive argument, the c-peak starts to appear in LMO-Epi-MgO substrate due to the growth of shortened and randomly distributed nanocolumns within the film. This result reveals the effect of the substrate on both the YBCO and the dopant induced within it. At 6 T, the c-axis flux pinning gets more pronounced for film on CeO_2 template with the appearance of a very broad and intense c-peak thus telling us that most of the vortices get pinned at this B with maximum pinning force $F_{\rm p}$, whereas the c-peaks starts to slightly decrease for the rest of the films. To an end, the film on CeO_2 template still shows a very strong c-peak even at 8 T with pronounced ab-peaks thus revealing the enhanced pinning along both the *c*-axis and *ab*-planes. On the other hand, the *c*-peaks are barely visible for the films on both LMO-MgO and LMO-Epi-MgO templates.

All the $J_{\rm c}(\theta)$ curves presented in Figs. 3(c) and (d) are asymmetric in the vicinity of 0° which is the typical behaviour for all the films grown on IBAD-MgO based templates being caused by the inclined buffer layer growth during the IBAD process [12, 19, 20]. Furthermore, the low lattice mismatch between the CeO₂ cap layer and the YBCO [21] makes the CeO₂ template the best for growing both undoped and BZO doped YBCO as least anisotropy of $J_{\rm c}(\theta)$ is revealed when the films were grown on it. To bring forth a comprehensive understanding of this explanation we present a schematic model, in the next section, which illustrates the growth of the defects in both the undoped and doped YBCO films when grown on all of our three templates.

For the detailed anisotropy analysis, the anisotropy of undoped YBCO is best described via the Blatter anisotropy parameter γ , which ultimately represents the electronic mass anisotropy of YBCO [22]. This parameter can be experimentally estimated by plotting the $J_c(\theta, B)$ values (shown in Fig. 3) in the regions where they collapse (between 90° and 180°) as a function of effective magnetic field $B_{\text{eff}} = B\varepsilon(\theta, \gamma)$, where $\varepsilon(\theta, \gamma) = (\cos^2(\theta) + \gamma^{-2}\sin(\theta))^{1/2}$, and finding a value of γ that produces linear B_{eff} - J_c plot [18, 23]. Fig. 4 presents the calculated B_{eff} - I_c curves

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Figure 4. The I_c as a function of effective magnitic field B_{eff} along the *c*-axis of YBCO for all the undoped films. The calculated γ parameters are also given.

and corresponding γ -values which respectively produce the linearity observed in the curves. Our films have the γ values in the range of 2 to 3, which are much lower compared with other YBCO films where γ is always found to be above 5 [18, 24, 25]. In our previous work [24], we calculated the γ values of YBCO films on STO where the films grown by micro- and nanocrystalline sized targets have $\gamma = 5$ and 1.8, respectively. This observed difference is explained by the increased number of correlated pinning sites in n-YBCO (grown from nanocrystalline sized target) that on the other hand, has a lowering effect on γ value. Similarly, Xu et.al. [26] compared the anisotropy of two YBCO films, one with Y2O3 precipitates and other with stacking faults, where the calculated γ value for the film with Y₂O₃ precipitates is much lower than for the film with stacking faults due to the effective pinning properties of Y_2O_3 precipitates in a wide region around YBCO *c*-axis. This suggests that in our case, the decreased anisotropy especially in CeO₂ and LMO-Epi-MgO films is due to the increased number of weak point like defects, precipitates or voids. This scaling procedure has not been applied to BZO doped films since the highly correlated BZO nanorods perturb the $J_c(\theta)$ curves in such a way that they do not collapse to a line properly at low temperature and B [24], thus making the anisotropy analysis difficult.

3.3. TEM analysis

Based on our transport properties shown in Fig. 3, we chose the best film (in terms of enhanced isotropy of $J_c(\theta)$) for the TEM analysis. Fig. 5 displays the TEM image of one of the doped films, that is, BZO doped YBCO grown on CeO₂ template. It can be seen that the BZO nanocolumns grow elongated along the *c*-direction with the presence of short stacking faults (mainly next to the CeO₂/YBCO interface) in between them. Our analysis shows that the BZO nanocolumns are just slightly tilted and are randomly distributed. The lengthened nanorods increase the flux pinning along *c*-axis, thus reducing the J_c anisotropy, whereas the short stacking faults are the source of pinning the vortices along *ab*-direction. Moreover, while connecting the microstructure with the angular dependent results (Fig. 3), it can be concluded that the nanorods are very effective in flux pinning as revealed by the most pronounced *c*-peak of doped film grown on CeO₂ template, especially at high fields from 4 T to 8 T.



Figure 5. TEM image of BZO doped film grown on CeO_2 template. The yellow boundaries delimit the BZO nanocolumns, whereas the presence of a stacking fault next to the interface is marked by sky blue area.

3.4. Growth model of the defects

According to our schematic shown in Fig. 6 (drawn based on the angular dependent measurements, XRD results and TEM image), one realizes relative differences not only in the number but also in the sizes of both the naturally and artificially induced defects within the films. Comparing the undoped films, we concluded that the growth of long threading dislocations throughout the thickness of the film on CeO_2 template is the source of effective vortex pinning which in turn reveals the c-peak (Fig. 3(a)). This is in line with our structural results where the low-angle grain boundaries ($\Delta \phi$) channel the YBCO growth which results in threading dislocations. For LMO-MgO template, the intermediate sized dislocations are still perturbing the vortex motion which increases the isotropic behaviour of the $J_{\rm c}(\theta)$ especially at low B (Fig. 3(a) and (b)). On the other hand, the same sized stacking faults gave us the similar shapes of $J_{\rm c}(\theta)$ ab-peaks in both the CeO₂ and LMO-MgO templates, whereas more pronounced abpeaks under all the B for film on CeO_2 template is related to the greater number of stacking faults compared with the LMO-MgO template. Thinking about the similarity of *ab*-peaks for film on LMO-Epi-MgO template with that of YBCO grown on single crystalline substrate [4], we see that the growth of long stacking faults makes it energetically preferable to pin a huge number of vortices. Principally, the film on LMO-Epi-MgO template beats the other two in terms of both the number and size of stacking faults and thus has the highest J_c along the ab-direction. Moreover, the small-sized threading dislocations in this case are also the source of vortex pinning but do not affect the shape of the *c*-axis peak.

The induced defects in BZO doped films as revealed by Fig. 5 and schematically drawn in Fig. 6, we find in our analysis that the BZO grown nanocolumns are slightly unidirectionally splayed in all the films as this is the cause of anistropic c-peaks (Fig. 3(c) and (d)). The main difference among all the films is the length of these nanorods. The nanorods are the longest when film is grown on the CeO_2 template and this is the most effective for strongly enhanced flux pinning thus producing very broad and intense *c*-peak in this case. The length of the

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Figure 6. Schematic of some of the induced defects within the (a)-(c) undoped and (d)-(f) BZO doped films grown on different buffered substrates. The red horizontal lines present the stacking faults, whereas the vertically oriented blue threads depict the low-angle grain boundary based threading dislocations. The green filled pipes correspond to the BZO induced nanocolumns.

nanorods is smaller in the film on LMO-MgO template which affects the flux pinning especially at high *B* as revealed in Fig. 3(d)). The very small sized nanorods grown in the film on LMO-Epi-MgO template cause weakened vortex pinning as a slight *c*-peak was observed in this case (Fig. 3(c) and (d)). The difference in the shapes of the $J_c(\theta)$ curves of the undoped films from the doped ones can be explained by the Paulius model [27] where J_c is reduced when the inclination angle of the applied field is increased with respect to the *c*-axis thus decreasing the length of trapped vortex portion. Unlike our undoped films where vortices are only pinned by the natural defects and they can become free and move around at sufficient magnetic field angles, the doped films have the significant edge of firstly pinning the vortices by the nanocoloumns and at certain angles when vortices get untrapped by them the proximate natural defects like threading dislocations and stacking faults can be the cause of their trapping. This effect improves the $J_c(\theta)$ and thus the creation of enhanced natural defects on our templates, especially for film on CeO₂, can be used to help advance fundamental understanding and applied for electrical power applications in wide temperature, field and angular ranges.

4. Conclusion

Three variously buffered IBAD-MgO based templates were used to grow both undoped and BZO doped YBCO films. The analysis of our results showed that the buffer architecture is crucial for growing superconducting films with desired properties. We succeeded in optimizing one of the substrates to achieve the goal of improving the isotropic behaviour of $J_{\rm c}(\theta)$ on technical

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substrates. The optimized CeO₂ template is suitable for the creation of highly effective natural and decorated defects within YBCO films which enhances the pinning of the vortices. Both isotropic and high-valued $J_c(\theta)$ curves are achieved on this template in comparison to other two counterparts. Furthermore, our growth model of the defects gives a basic understanding about the induced defects within all the films deposited on our differently buffered templates. Thus, our results can be utilized in a variety of electrical power applications.

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