Critical Current Density of YBa$_2$Cu$_3$O$_{7-x}$ Films with BaZrO$_3$ Inclusions On SrTiO$_3$ and Al$_2$O$_3$ Substrates

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Abstract. Recently, many efforts have been dedicated to the development of a reliable technology for the introduction of artificial pinning sites in YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) films with the aim of improving the in-field $J_c$ performances. One of the most effective technique resulted to be the inclusion of BaZrO$_3$ (BZO) second phase embedded in the YBCO films. In this contribution we present $J_c$ measurements on BZO-added YBCO films deposited on SrTiO$_3$ (STO) and CeO$_2$-buffered-Al$_2$O$_3$ (ALO) substrates. Samples were deposited by pulsed laser ablation technique using a composite YBCO + 5mol.% BZO target at the optimum conditions for fully oxygenated $c$-axis oriented YBCO films. Despite of a slight $T_c$ reduction, BZO addition in YBCO-STO films resulted in an improvement of in-field performances with the appearance of a $J_c$ plateau in the low field region which extends up to about 2.5 Tesla irrespective of the temperature at least in the investigated range (down to 65K). On the other hand, samples deposited on ALO did not exhibit any remarkable difference neither in the $J_c$ value nor in the magnetic field dependences as compared with pure YBCO. The presence of $\theta^\circ$ (magnetic field parallel to the $c$-axis) peaks in the $J_c$. angular behaviour revealed a $c$-axis correlated character of the pinning forces in BZO added YBCO films grown on both STO and ALO substrates. X-ray diffraction measurements and AFM investigations were carried out in order to determine the influence of BZO addition on films crystalline quality and microstructure.

1. Introduction

Recently the addition of second phase nanometric sized particles in ReBaCuO (Re = Rare Earth) superconducting films revealed to be an effective technique for the enhancement of in field transport properties [1]. In YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) coated conductors the best results in terms of critical current density and irreversibility field values have been obtained by the introduction of BaZrO$_3$ (BZO) inclusions in the film matrix [2] though this effect is not fully understood yet. Many possible mechanisms have to be taken into account in order to understand the role of BZO in determining pinning properties of YBCO: nanosized BZO particles are non-superconducting regions with

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dimensions which are comparable to the superconducting coherence length acting as isotropic pinning sources; BZO inclusions could induce a strain field at the interface with YBCO matrix resulting in dislocation appearance [2]; the possible formation of self organized columnar structures of BZO particles acting as correlated pinning sites as suggested by TEM analyses [3]; the presence of BZO could decrease the YBCO mean grain size increasing the number of grain boundaries containing dislocations [4]. Understanding which of these effects take place in BZO added YBCO films and determining the relative influence on pinning properties would be of crucial importance to obtain further progresses in enhancing the in-field performances.

In this work we report the study on the effect of BZO addition in YBCO thin films grown by pulsed laser deposition method (PLD) on two different substrates, (001)-SrTiO$_3$ single crystal and CeO$_2$ buffered $r$-cut Al$_2$O$_3$ single crystal substrate. D.C. transport properties measurements performed on both sample sets revealed that while a remarkable improvement of in field critical current density values can be observed for samples grown on SrTiO$_3$, almost no effect of BZO introduction in films grown on sapphire can be noticed. X-ray diffraction analyses and atomic force microscope (AFM) investigations were carried out in order to determine if the observed effects can be ascribed to any change in microstructural and morphological features induced by BZO addition.

2. Experimental details

Samples were obtained by PLD technique from YBCO-BZO composite targets with 5 mol.% BZO nominal content. Further details on targets manufacturing can be found elsewhere [5]. The films were grown on (001)-SrTiO$_3$ (STO) and $r$-cut Al$_2$O$_3$ (ALO) single crystal substrates using a 308 nm XeCl excimer laser (Lambda Physik 110i cc) adjusting the laser energy at $\sim$110 mJ and with a repetition rate of 10 Hz. The number of laser shots was 7000, corresponding to a film thickness ranging from 150 to 190 nm on STO and from 110 to 130 nm on ALO substrate. The chosen deposition conditions resulted from the achievement of optimal conditions for fully oxygenated YBCO films. The substrate-target distance was 4.8 cm and the oxygen partial pressure was 300 mTorr. During the deposition process the substrate temperature was monitored by a thermocouple inserted in the substrate holder and was 850 °C and 750 °C for deposition on STO and ALO respectively. After the deposition the temperature was slowly cooled down in 570 mTorr oxygen atmosphere whit a 15 minute dwell at 450 °C. In the case of deposition on ALO prior to the YBCO film a 20 nm CeO$_2$ film was deposited in 250 mTorr oxygen pressure and at 750 °C. Samples were patterned using standard U.V photolithographic process in order to obtain 1 mm long and 30 µm wide stripes, mounted on a one-angle variable sample-holder with an accuracy greater than 0.05° and loaded in a He gas flow cryostat provided with a 12 T superconducting magnet. The magnetic field direction was kept always normal to the bias current direction in order to guarantee the maximum Lorenz force configuration. I-V characteristics were recorded using four point method at different magnetic field and temperature values. Critical current values were extracted from I-V with standard 1 µV cm-1 voltage criterion. YBCO films grew epitaxially on the (001) STO substrates and on epitaxial CeO$_2$-Al$_2$O$_3$ structure as shown by X-ray diffraction spectra performed in $\theta$-2$\theta$ configuration. (001)-oriented YBCO film with (005) rocking curve FWHM of about 0.16° and 0.70° are typically obtained on STO and ALO. The BZO crystallites are (h00) oriented, indicating single epitaxial relationship between BZO and YBCO. AFM analyses were performed in air with a Veeco - Digital Instruments Dimension 3100 Microscope, equipped with an optical deflection system in combination with silicon cantilever and tips, working in tapping mode. Topographic images have been recorded over scanned areas ranging from 400 x 400 nm$^2$, up to 30 x 30 µm$^2$, each with a resolution of 512 x 512 data points.

3. Results and discussion

In figure 1 the resistivity versus temperature curves collected for BZO added YBCO films grown on both ALO and STO substrates are shown. As expected, films grown on both substrates exhibit a reduction in the zero resistance critical temperature ($T_c$) as compared with standard pure YBCO. The
measured $T_c$ values (86.6 K for film on ALO and 87.2 K for film on STO) and room temperature resistivity values $\rho_{RT}$, are within ranges typically reported for good quality 5 at.% BZO-YBCO films [6].

Figure 1. Resistivity versus temperature curves for BZO added YBCO films grown on both ALO and STO substrates. In the inset the transition region has been magnified. Lines represent the best linear fit of the normal state resistivity.

The comparison of the magnetic field dependence of the critical current densities, $J_c(B)$, exhibited by BZO-added and pure YBCO for both substrates films is shown in the left panel of figure 2. While the BZO addition in ALO samples does not affect the $J_c(B)$ trend except for a irreversibility field decrease due to lower $T_c$ values, STO samples show a completely different scenario. In BZO-added samples a low field plateau of $J_c$ values and higher irreversibility field value are evidences of better in field performances which can be estimated trough the $J_c(B)/J_{c0}$ ratio at 77 K (where $J_{c0}$ is the self field critical current density value) for $B=1$ and 3T. While in pure YBCO a ratio of 0.14 and 0.02 is achieved, in BZO samples the corresponding values are 0.33 and 0.15.

Figure 2. Magnetic field dependence of critical current density (left panel) and pinning force density as a function of normalized field (right panel) at 77K for pure and 5 at.% BZO added YBCO films grown on both ALO (circles) and STO (squares) substrates.

The analysis of the pinning force densities, $F_p=J_cB$, as a function of the normalized field, $b=B/B^*$ ($B^*$ is the magnetic field where the pinning force density is lower than 1% of its maximum) as shown in the right panel of figure 2 confirms the different effect of BZO introduction depending on the substrate. In
samples grown on ALO, the lower $T_c$ value determined by BZO addition results in a decrease in $F_{p\text{max}}$ and $B^*$ absolute values while the shape of the $F_p(b)$ curve is not changed. On the contrary, samples on STO with BZO inclusions exhibit a remarkable increase of the maximum pinning force density (9.9 GN/m$^3$ against 1.7 GN/m$^3$ exhibited by pure YBCO) and a shift of the $F_{p\text{max}}$ towards higher normalized field values. These features, coupled with the shift of the irreversibility field towards higher magnetic fields, suggests a better pinning sites lattice efficiency. Taking into account that the $J_c(B)$ dependence is improved in the whole magnetic field range inspected, it can be deduced that BZO inclusions introduce strong pinning sources inside STO samples. As suggested by the analysis of the angular properties showed in figure 3 these sources exhibit a $c$-axis correlated character. Apart from the intrinsic peak due to the layered structure of YBCO which is always detectable, the appearance of a broad peak centered at 0° (magnetic field direction parallel to the $c$-axis) in BZO films on STO evidences the presence of extended defects along the $c$-axis direction. As already reported in literature [2], the correlated defects can be identified with dislocations arising at BZO-YBCO interfaces as a consequence of the lattice mismatch. Actually, a small 0° contribution can be measured also for ALO films even though it can not be ascribed to BZO introduction since pure YBCO on ALO shows the same angular behavior.

Some clues to the different effect of BZO on STO and ALO can be obtained by X-ray diffraction investigations. $\theta$-2$\theta$ spectra performed on BZO-added YBCO films grown on both STO and ALO templates evidence the formation of BZO crystallites with $(h00)$ orientation as suggested by the presence of the BZO (200) peak centered in the range 43.5°-44.0° (see figure 4). From the analysis of the (005) YBCO peak some parameters concerning the structural properties of the samples can be deduced. The full width half maximum (FWHM) of the (005) rocking curve (RC) can be related both to the film mosaic spread on the $ab$-planes and to the x-ray coherence length on the planes, $l_{ab}$ [7]. As shown in table 1, where x-ray parameter has been summarized, the FWHM exhibited by ALO films is quite large. The RC broadening is mainly due to a large mosaic spread as expected for films grown on the CeO$_2$ template. Therefore, $l_{ab}$ values obtained for ALO films are probably undervalued and any quantitative consideration on the $ab$-planes coherence length can be meaningful. However, only a small relative change can be observed in BZO films suggesting that BZO introduction does not lead to any remarkable effect on films structural properties. On the contrary, the small RC broadening exhibited by STO films can be strictly related to $l_{ab}$ since the shape of the RC peak is well described by a simply Lorentzian function (mosaic spread induces Gaussian shaped RC, as observed in ALO films, since the statistical distribution of the spread is uncorrelated in space). In this case BZO introduction induces a large $l_{ab}$ increase evidencing a positive effect of BZO crystallites on films structural properties. Moreover, neglecting the instrumental broadening, the inverse of the (005) YBCO reflection integral width in the $\theta$-2$\theta$ pattern can be related to the $c$-axis coherence length, $l_c$ through the Scherrer formula.

![Figure 3. Critical current density dependence from the angle between magnetic field direction and films $c$-axis measured for STO samples (squares) and ALO samples (circles) at 77K and 1T.](image-url)
Also the analysis of the $l_c$ parameter suggests that BZO films on STO are characterized by an improved film crystallinity while no remarkable effect of BZO inclusion can be observed in ALO films.

**Table 1.** FWHM of YBCO (005) reflection and X-ray coherence length on the $ab$-planes and on the $c$-axis obtained from the analysis of YBCO (005) reflection for both pure and BZO-YBCO films on ALO and STO.

| Substrate | BZO | FWHM | $\lambda$ (nm) | $l_c$ (nm) |
|-----------|-----|------|----------------|------------|
| ALO       | 0 % | 0.70°| 5.9            | 44.8       |
| ALO       | 5 % | 0.84°| 5.2            | 43.0       |
| STO       | 0 % | 0.16°| 21.8           | 38.0       |
| STO       | 5 % | 0.09°| 49.6           | 73.0       |

Since we are interested in evaluate the influence of BZO on the growth of YBCO, a comparison was carried out between samples by AFM investigations on pure and BZO-added YBCO films grown on both templates. The most representative images for these samples categories are reported in figure 5.

**Figure 5.** AFM images collected on pure YBCO films (left panel) and BZO-added YBCO films (right panel) on both STO (upper images) and ALO (lower images) substrates. In the images maximum brightness corresponds to the higher heights.

YBCO films on STO substrates reveal some features that can be explained in terms of a mixed 2D-3D growth mechanisms. According to the Stranski-Krastanov model, once the critical thickness for a pure 2D growth is reached the stress is released promoting island growth mechanism. As it can be seen in figure 5a, the film morphology shows a flat and uniform background where 3-D islands not regularly
shaped are observed. They clearly grow around screw dislocation vectors as revealed by the typical spiral shape. The height between two steps can be estimated by section analysis, in the range of 8-18 lattice parameters of YBCO. The morphology of the BZO films deposited on STO substrate presents remarkable changes (see figure 5b). The inclusion of BZO seems to induce a decrease in the spiral features density (0.15 µm\(^{-2}\) against 1 µm\(^{-2}\) exhibited by YBCO/STO films) as well as a different morphology of the background. In fact, the morphology reveals details that could be associated to almost bi-dimensional growth, producing terraces (mean size 500 nm) oriented along (110) direction, coherently with the morphology of the STO annealed substrate [9]. On the contrary, the YBCO on ALO template appears as a granular surface with a uniform coverage of randomly disposed grains (see figure 5c). Flat grains of 100-200 nm in dimension are connected by deep trenches. The spiral features were not observed for these samples. The inclusion of BZO, does not change significantly the morphology of the surface (see figure 5c), resulting in a decrease of droplets density, which become bigger in size.

The increased density of grains for YBCO on ALO compared to STO naturally leads to the extension of the boundary regions where structural defects (such as edge dislocations) are expected to arise during the grains coalescence. In this way the stress is released at the grain boundaries preventing the nucleation of screw dislocation with the related spiral growth mechanisms observed on STO.

The presence of c-axis correlated defects together with the improved structural quality of BZO-added YBCO films grown on STO can be related to the observed change in the surface morphology assuming that an effective stress release mechanism is provided by edge dislocations at YBCO/BZO interfaces which allows the growth of an unstrained YBCO film inhibiting the spiral growth mechanism.

4. Conclusions
Transport properties analysis have been carried out on pure and 5 at.% BZO-added YBCO films grown on SrTiO\(_3\) and CeO\(_2\) buffered Al\(_2\)O\(_3\) substrates. In films grown on STO a remarkable effect of BZO introduction on the in-field performances and on the pinning force density values can be noticed while the same effect has not been observed for ALO films. X-ray analyses confirm the different influence of BZO introduction depending on the substrate evidencing a significant improvement of the film crystallinity for BZO films as compared with pure films only in the case of STO substrate. AFM investigations have revealed that this difference could be ascribed to a change in the film growth mechanism induced by the presence of BZO crystallites embedded in the YBCO matrix grown on STO.

References
[1] Yamada Y, Takahashi K, Kobayashi H, Konishi M, Watanabe T, Ibi A, Muroga T, Miyata S, Kato T, Hirayama T and Shiohara Y 2005 Appl. Phys. Lett. 87 132502
Matsumoto K, Horiede T, Ichinose A, Horii, Yoshida Y and Mukaida M 2005 J. J. Appl. Phys. 44 246
[2] Mac-Manus D, Foltyn S R, Jia Q X, Wang H, Serquis A, Civale L, Maiorov B, Hawley M P and Peterson D E 2004 Nat. Mat. 3 439
Gutierrez J, Llordes A, Gazquez J, Gibert M, Roma N, Ricart S, Pomar A, Sandiumenge F, Mestres N, Puig T and Obradors X 2007 Nat. Mat. 6 367
[3] Goyal A, Kang S, Leonard K J, Martin P M, Gapud A A, Varela M, Paranthaman M, Ijaduola A O, Specht E D, Thompson J R, Christen, Pennycook S J and List F A 2005 Supercond. Sci. Technol. 18 1533
[4] Dam B, Huijbregts M and Rector H 2002 Phys. Rev. B 65 064528
[5] Galluzzi V, Augieri A, Ciontea L, Celentano G, Fabbri F, Gambardella U, Mancini A, Petrisor T, Pompeo N, Rufoloni A, Silva E and Vannonzi A 2006 IEEE Trans. Appl. Supercond. 17 3628
[6] see for example Traito K, Peurla M, Huhtinen H, Stepanov Y P, Safonchik M, Tse Y Y, Paturi P and Laiho R 2006 Phys. Rev. B 73 224522
[7] Gauzzi A and Pavuna D 1995 Appl. Phys. Lett. 66 1836
[8] Guiner A 1994 X-ray diffraction: In crystals, imperfect crystals and amorphous bodies (Dover)
[9] Fabbri F, Padeletti G, Petrisor T, Celentano G and Boffa V 2000 Supercond. Sci. Technol 13 1492