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Properties of Pr- and BZO-doped YBCO multilayers

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Abstract

Multilayers of 2 wt-% Pr- and 4 wt-% $BaZrO_3$-doped $YBa_2Cu_3O_{6+x}$ (YBCO) were deposited by pulsed laser ablation on $SrTiO_3$ and MgO single crystals. The structural and superconducting properties of the multilayers are presented. Multilayering was found to improve the critical temperature and structural quality of the samples on $SrTiO_3$, but it did not change the critical current density. On MgO, the critical temperature improved slightly, but the critical current density decreased.

Keywords: Keywords: YBCO, multilayers, $BaZrO_3$-doping, Pr-doping

1. Introduction

Widespread use of high-temperature superconductors in applications still requires the superconductors to have even higher critical current density, j_c, at high fields and temperatures. The j_c of the most studied high temperature superconductor, $YBa_2Cu_3O_{6+x}$ (YBCO), can be increased by adding artificial magnetic pinning sites, such as $BaZrO_3$ (BZO), into the superconducting matrix [1, 2]. Unfortunately, while adding BZO increases j_c and the isotropy of j_c [3], it also generally lowers the critical temperature, T_c, [1], which is undesirable as the T_c of YBCO is already quite close to the boiling point of nitrogen. The lowering of T_c can be attributed to strain and disorder in the lattice [4]. It has been shown recently [5] that doping the Y-site with Pr increases T_c and j_c in films with low angle grain boundaries. This effect is attributed to segregation of Pr to the grain boundaries and better oxygen stoichiometry at the grain boundaries, similarly to previously observed effect of Ca-doping [6]. Low angle grain boundaries are present in films deposited on textured metal substrates for coated conductors, but also in films on MgO [7].

In this work we have prepared multilayers of 4 wt-% BZO-doped and 2 wt-% Pr-doped YBCO films, in the hope that the BZO-induced drop of T_c can be remedied by alternately doping with Pr thus enhancing both j_c with BZO and T_c with Pr.

2. Experimental

The multilayers were prepared by pulsed laser deposition with a XeCl excimer laser (λ = 305 nm) at 0.3 Torr O_2 atmosphere. The multilayer structures were deposited *in situ* and changing the target between the layers took about 60 seconds. Fig. 1 shows the different multilayer structures. All the multilayer structures were grown on both single crystal $SrToO_3$ (100) (STO) and MgO (100) substrates. Substrate temperature of

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Table 1. The structural results, the onset T_c and transition width ΔT_c for all the films. c-axis is the measured c-axis length, Int(005)/(004) is the intensity ratio between peaks (005)/(004), r_c is the lattice coherence length, a is the amount of material with c-axis in the plane of the film and 45° is the amount of material turned 45° in plane of the film.

Sample	T_c (K)	ΔT_c (K)	c-axis (nm)	Int(005)/(004)	r_c (nm)	a (%)	45° (%)
STO 1	88.0	1.9	11.665	13.6	18.8	0.6	0
STO 2	88.7	1.7	11.670	13.8	20.5	0.6	0
STO 3	88.7	1.5	11.670	14.0	19.0	0.6	0
MgO 1	88.5	4.6	11.675	15.2	12.5	1.0	< 4
MgO 2	88.7	10.1	11.681	13.2	12.1	0.6	< 4
MgO 3	88.0	20.3	11.686	14.4	10.9	1.1	9.8

750°C was used for the STO samples and 725°C for the MgO samples. The lower deposition temperature for MgO films was chosen so that the formation of 45° turned grains in the MgO-films was minimized. After deposition the atmospheric pressure of oxygen was introduced in the deposition chamber at 700°C and the samples were cooled down to room temperature with 25°C/min.

The crystal structure of the films was measured using x-ray diffraction (XRD) with a Schultz texture goniometer. The investigated peaks were (102) in a- and c-axis perpendicular to the film surface and 45 ° in-plane rotated direction to study the orientation of the film, (122)/(212) peak set to investigate the twinning and low angle grain boundary structure, BZO (110) and (002) peaks to ascertain the orientation of the BZO rods, YBCO (005) peak and rocking curve to estimate the crystalline quality of the YBCO films and 2θ scan for c lattice parameter and oxygen content of the films.

The critical temperatures, T_c, of the films were determined from ac-susceptibility measured with a Quantum Design Physical Property Measurement System (PPMS) ACMS-option with 0.1 mT ac-magnetic field at 113 Hz. The T_c's were determined as the onset of the transition and and the width of the transition, ΔT_c, as the temperature difference between 10 % and 90 % of the full transition. The critical current densities, j_c, were determined from hysteresis loops measured between -8 – 8 T at 10 – 80 K. j_c was determined using the Bean critical state model for rectangular films $J_c = 2\Delta m/[a(1 - a/3b)V]$, where a and b ($b \geq a$) are the side lenghts of the film, Δm is the opening of the hysteresis loop and V is the volume of the sample [8]. The accommodation field, B^*, was determined as the field where the critical current has decreased to 90 % of the zero field value. The j_c decrease exponent α was determined by fitting B^α to the $j_c(B)$ curve above B^*.

3. Results and discussion

3.1. Structural properties

The 2θ scans showed that all the films contained only YBCO and BZO and that the YBCO was c-axis oriented. We calculated the c-lattice parameter from the (004), (005) and (007) peaks (shown in table 1). In the both sample sets, c increases slightly when the layer thickness in the multilayer decreases. From the intensity ratio of the (005) and (004) peaks, we estimated that the oxygen content [9] of all the films is optimal, i.e. the observed intensity ratio is below 20. The lattice coherence length $r_c = 1/\pi \cdot d/l\Delta\omega$ was calculated from the width of the (005) rocking curve [10]. Here d is the c lattice parameter, l is the order of the Bragg reflection and $\Delta\omega$ is the full width at half maximum (FWHM) of the rocking curve. In the films on STO, r_c is around 19 nm, which is closer to the value of 19–22 nm on undoped YBCO [11] than the observed values of around 14 nm for 4 wt-% BZO-doped films. This means that the Pr-doped layers improve the structural alignment of the lattice. On MgO, r_c is clearly lower, around 12 nm, which has been observed also earlier for films on MgO [5]. One can see that the multilayering can not improve the lattice coherence above the value determined by the substrate.

The amount of grains with their c-axis in the plane of the film (a-orientation) was determined from the geometry corrected intensities of the (102) peaks for both a- and c-orientations. All the samples had less than 1.5 vol-% of material in a-orientation and as this is very close to the detection limit, we can conclude that the films were purely c-axis oriented. In films on MgO, if the deposition temperature is too high, some

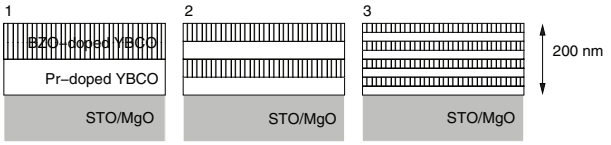

Fig. 1. Schematic of the deposited structures. The substrate is either MgO or STO. Films 1 have one 100 nm thick Pr-doped and one similar BZO-layer, films 2 have two layers each with 50 nm thickness and films 3 have 4 layers each with 25 nm thickness.

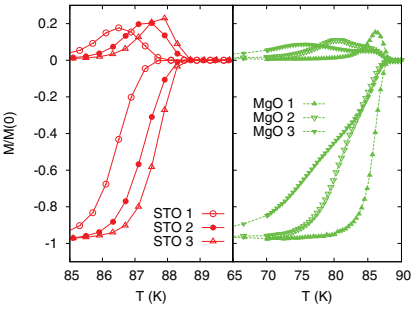

Fig. 2. $M(T)$ curves of the films. Note that the temperature scale is different for films on STO and MgO.

of the grains frequently turn 45° in the plane of the film. Since these grains form weak links, they greatly decrease the superconducting properties of the films. The amount of 45° material was measured also from the (102) peaks and was below 4 % for the MgO 1 and 2 films, which is around the detection limit, since the overlapping tail from a substrate peak complicates the determination. The MgO 3 film had clearly more 45° material, around 10 %, and this affected the superconducting properties. Changing the deposition parameters only resulted in worse films.

All the films on STO showed the four peaks normal to undoped YBCO in (122)/(212) scans. This indicates normal twin formation and smaller lattice distortion than is usually observed in BZO-doped films [1]. On the other hand, the peaks were not separable in any the samples on MgO, indicating abundance of low angle grain boundaries, as are usually observed in films on MgO [7]. Therefore, we can conclude that multilayering with Pr improves the crystalline structure on BZO-doped films on STO, but does not make it better than is dictated by the substrate on MgO.

3.2. Magnetic properties

The ac-magnetization curves for all the samples are shown in Fig. 2 and the both midpoint T_c and ΔT_c are listed in table 1 for all samples. $T_c \approx$ 88 K is quite normal for BZO-doped films [1]. For the films on STO, T_c increases and the transition width decreases slightly with multilayering, whereas on MgO the T_c increases from sample 1 to 2, but decreases for sample 3. The transition width increases from 4 K to 20 K from sample 1 to 3. The sample 3 has clearly 2 phases, which can be due to the observed 45° turned grains and the weak links formed, although that does not explain the increase of transition width in sample 2. The improvement of T_c and transition width in samples on STO can be explained by the healing of the lattice with Pr-doping, which also evident from the XRD results as increase of r_c, decrease of peak widths and clear separation of twins.

j_c as a function of the magnetic field at 10 K and 70 K are presented for all the samples in Fig. 3. It is clear that on STO the multilayering does not appreciably change the values or the shape of $j_c(B)$. The $j_c(0) = 5$ MA/cm^2 value at 70 K is quite high even for BZO-doped film [3]. On MgO j_c decreases from sample 1 to 3, i.e. the multilayering makes the sample worse. Samples on MgO are worse than the samples on STO at all temperatures and magnetic fields. The MgO 3 sample, which is worst in all other respects, actually surpasses the MgO 2 sample in high fields at all temperatures. This is probably due to large amount of pinning sites resulting from the bad structural film quality.

The temperature dependences of B^* and α are shown in Fig. 4. The MgO multilayers have clearly higher B^*, indicating a larger amount of pinning sites, which can be interpreted to be due to the dislocations in the low angle grain boundaries. Inside the film sets, the B^* decreases in the STO set and from MgO 1 to 2, whereas the B^* in MgO 3 is higher than in the others. The temperature dependence of B^* is normal for all the films. The decrease of B^* in the STO films can be understood from decrease in the amount of pinning sites as the lattice heals with thinner layers. In MgO samples the same works for samples 1 and 2, but the clearly worse quality of sample MgO 3 makes its B^* higher.

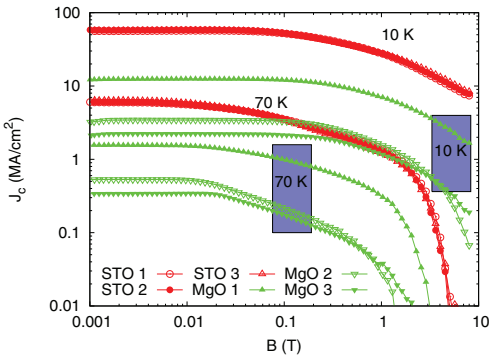

Fig. 3. $j_c(B)$ for all the multilayers at 10 K and 70 K.

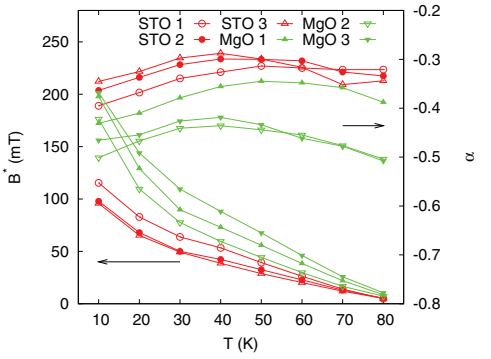

Fig. 4. Determined B^* and α values as function of temperature for all the films.

The α-values for the STO multilayers are close to those usually observed in BZO-doped films [1] and all the samples give similar α-values. In MgO multilayers $\alpha \approx -(0.4 - 0.5)$ is slightly lower than in the STO samples. In samples with BZO nanorods α is generally less than -0.3, but decreases if the rods are shorter [12] and is around -0.4 for nanoparticles [13], which have diameter of a few nanometers. Therefore, we can suggest that in the MgO samples BZO forms shorter nanoparticles instead of long nanorods. This is supported by the fact that α decreases as the layer thickness decreases. The temperature dependence of α is negligible for all the samples, which is quite usual [14].

4. Conclusions

Multilayers of 2 wt-% Pr- and 4 wt-% $BaZrO_3$-doped $YBa_2Cu_3O_{6+x}$ (YBCO) were deposited by pulsed laser ablation on STO and MgO single crystals. Multilayering was found to improve slightly the critical temperature and structural quality of the samples on STO, but it did not affect the critical current density appreciably. On MgO, however, the onset critical temperature improved slightly with 50 nm thick layers, but the critical current density decreased with all multilayerings. Therefore multilayering is not the solution for increasing the performance of BZO-doped YBCO films.

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