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Substrate decoration for improvement of current-carrying capabilities of $YBa_2Cu_3O_x$ thin films

Alexey V. Khoryushin [a,*], Peter B. Mozhaev [a,1], Julia E. Mozhaeva [a,1], Igor K. Bdikin [b], Yue Zhao [c], Niels H. Andersen [a], Claus S. Jacobsen [a], Jørn Bindslev Hansen [a]

[a] Department of Physics, Technical University of Denmark, DTU Building 309, Kgs. Lyngby DK-2800, Denmark
[b] Department of Mechanical Engineering, Centre for Mechanical Technology & Automation, University of Aveiro, 3810-193 Aveiro, Portugal
[c] Department of Energy Conversion and Storage, Technical University of Denmark, DK-4000 Roskilde, Denmark

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ABSTRACT

The effects of substrate decoration with yttria and $Y:ZrO_2$ on the structural and electrical properties of the $YBa_2Cu_3O_x$ (YBCO) thin films are studied. The films were deposited on $(LaAlO_3)_3$–$(Sr_2AlTaO_8)_7$ substrates by pulsed laser deposition. Two different structures of decoration layer were applied, a template layer of nanoparticles and an uniform ultra-thin layer. Significant improvement of current-carrying capabilities was observed, especially at high external magnetic fields. Structural studies of these films reveal the presence of extended linear defects in the YBCO matrix. The formation of these structures is attributed to seeding of randomly oriented YBCO grains due to suppression of epitaxy in the very beginning of the deposition. The films of both kinds of decoration layers show nearly the same improvement of j_C over the reference film at 77 and 50 K: j_C (5T and 50 K) reaches 0.92 and 0.97 MA/cm^2 for uniform and template decoration layers. At 5 and 20 K the effect of template decoration layers is more beneficial: j_C (5T and 20 K) values are 3.5 and 4.1 MA/cm^2, j_C (5T and 5 K) values are 6.4 and 7.9 MA/cm^2, for uniform and template decoration layers, respectively.

1. Introduction

Modern high temperature superconducting wires (so-called "coated conductors") have a high potential in power applications. For some applications like DC lines, fault current limiters and persistent magnets the current-carrying capabilities of existing coated conductors are satisfactory, for others like motors or generators a stronger pinning in magnetic fields of 3–5 T than the natural pinning sites can provide is required. Dense and efficient artificial pinning centers (APCs) which do not deteriorate the superconducting properties of the surrounding material are essential to meet this demand [1]. One of the promising approaches is decoration of the substrate surface by yttria nanoparticles, which may result in formation of extended linear defects (dislocations, stacking faults, etc.) in the YBCO matrix overgrowing the yttria nanoparticle [2–4]. Such structures show improved current-carrying capabilities in an external magnetic field with arbitrary orientation (of a certain cone of angles around the c-orientation) [1], which is important for motors and generators. The mechanisms of extended defects formation over yttria nanoparticles remains unclear, and even the reasons for this are under discussion.

Nanoparticle decorations are usually fabricated using pulsed laser deposition (PLD) techniques: cooling of a dense plume of ablated material may result in condensation into nanoparticles on the way from target to substrate [5–8]. The size and density of the nanoparticles may be tuned by changing deposition conditions, mainly pressure, composition of the working atmosphere, and the laser beam energy density on the target surface. Usually the ablated material is not completely consumed by nanoparticles, and some part of it is coming to the substrate surface as atoms and clusters. As a result, the nanoparticles are partially embedded into a thin uniform film of the same material [5]. The effect of this uniform layer on the superconducting film was not studied.

In this paper we present results of our studies of YBCO films deposited by PLD on decorated substrates. Different decoration materials and techniques are applied and compared. The structural and electrical properties of the grown films are reported.

2. Experimental

The films were grown on $(100)(LaAlO_3)_{0.3}$–$(Sr_2AlTaO_8)_{0.7}$ (LSAT) perovskite substrates by PLD (KrF excimer laser, $\lambda = 248$ nm). The LSAT substrates provide fine conditions for YBCO formation: a good

* Corresponding author. Tel.: +45 45253243; fax: +45 45931669.
E-mail address: khoryushin@ya.ru (A.V. Khoryushin).
[1] On leave from Institute of Physics and Technology RAS, Nakhimovsky Ave. 34, 117218 Moscow, Russian Federation.

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lattice match (3.867 Å, less than 0.3% mismatch with the (*ab*) plane of YBCO), a small difference of thermal expansion coefficients and no twinning or lattice phase transitions in the range from deposition to room temperature.

Two different kinds of yttria-decorated substrates were fabricated by changing conditions of the PLD process: a "template" layer of Y_2O_3 nanoparticles and a "uniform" YO_x layer with uniform thickness and smooth surface.

The YBCO thin films (~240 nm) were deposited simultaneously from the stoichiometric target on two LSAT samples with different yttria decoration layers and on a reference bare LSAT substrate (HLP25, HLP29, HLP33). The deposition conditions were optimized for formation of high crystal quality and high critical temperature thin YBCO films on $LaAlO_3$, $SrTiO_3$ perovskite substrates (stoichiometric target $YBa_2Cu_3O_7$, substrate temperature 770 °C, 0.8 mbar total pressure, Ar/O_2 flow ratio 8/2 sccm, laser energy density on target 1.7 J/cm^2, repetition rate 2 Hz, deposition rate 0.165 nm/s, post-deposition oxygenation in 500 mbar O_2 at 450 °C for 1 h), other details of the deposition routine can be found elsewhere [10]. Additional samples were fabricated with a lower oxygen partial pressure of 0.12 mbar (HLP48, HLP52 and HLP56).

The surface morphology of the films was observed with atomic force microscopy (AFM), the structural parameters were determined by X-ray diffraction (XRD) measurements. The lattice constants *a*, *c* and the variations $\delta d/d$ along the substrate normal were determined using all available diffraction peaks of $\theta/2\theta$-scans (see, e.g., [11]). The in-plane spread of the orientation of the grains $\Delta\varphi_{\text{in-plane}}$ was determined using the measured rocking curve width $\Delta\omega_{(005)}$ and width of the peaks of the φ-scan for the (104) peak of YBCO:

$$\Delta\varphi^2_{\text{in-plane}} = (\Delta\varphi^2_{(104)} - \Delta\omega^2_{(005)}\cos^2\chi)/\sin^2\chi,$$

where $\chi = 52.66^\circ$ is the tilt angle for the (104) reflection.

The cross-sectional study of the YBCO film deposited on template yttria layer was performed on a transmission electron microscope (Tecnai T20 G2 from FEI, the sample was prepared by in situ lift-out method by Quanta 200 3D SEM-FIB from FEI, resulting in the sample thickness less than 150 nm).

The superconducting properties of the samples were examined by magnetic non-destructive methods. The accuracy of the T_c determination by the AC-susceptibility technique is expected to be better than 0.1 K. The width of superconducting transition was determined as full width at half maximum of the out-of-phase component of the AC signal. The magnetization measurements were done using a Cryogenic Ltd. vibrating sample magnetometer (VSM) instrument. Hysteresis loops *m* vs. *B* were recorded at 77, 50, 20 and 5 K (*m* is the magnetic moment of the sample and *B* is the applied magnetic field). For estimation of the critical current density j_C the critical state Bean model was employed for a geometry of thin films of rectangular shape: $j_C(B) = \frac{6\cdot 2m(B)}{t\cdot a^3(3b/a-1)}$, where *t* is the film thickness, and *a* and *b* denote the dimensions of the film ($b > a$). At a given field the gap of the hysteresis loop ($2m$) was utilized in order to minimize the effect of any possible paramagnetic background signal, specific for the setup and the sample holder.

3. Results and discussion

3.1. Yttria-decorated substrates

The parameters of nanoparticles deposition, argon pressure (0.2 mbar) and laser beam energy density on target (1.1 J/cm^2) were optimized to provide the smallest size of the nanoparticles together with almost no formation of uniform film. The yttria nanoparticles seem to be liquid when deposited on the substrate, as one can suppose from their shape (height smaller than diameter). Deposition at low substrate temperature results in non-oriented particles, while at temperatures above 600 °C a clear set of (00*l*) peaks can be seen on X-ray $\theta/2\theta$-scans, proving that crystallization of the nanoparticles takes part under influence of the substrate lattice.

The employed "template" yttria layers consisted of Y_2O_3 nanoparticles 20–40 nm in diameter and 10–25 nm high of $\sim 1.5\times10^{10}$ cm^{-2} density, with addition of less dense and bigger particles (Fig. 1a). The nature of these bigger particles (50–200 nm in diameter) is clearly different from the condensation on the way from target to substrate – presumably, it is droplets formation.

The uniform layers were fabricated from the same target after surface modification with the laser beam. The ablation of a metallic shiny YO_x surface provides no nanoparticles, only a smooth (roughness R_a less than 1 nm, Fig. 1b) uniform (001) orientated film for all studied deposition conditions and thicknesses.

The reason for such a change in the film features is metallization of the target surface: the ablation threshold of yttrium is much smaller than that of yttria (0.9 and 1.3 J/cm^2, [9]), so for the same energy density the temperature of the material ablated from metallized target is much higher and when it cools down to the condensation point the density of the plume is insufficient to result in nanoparticles, or the distance to substrate is insufficient for cooling below the condensation point.

The lattice constants of yttria measured along the substrate normal were 11.608–11.618 Å, close to that of bulk yttria (11.605 Å). The (001) yttria orientation provides good matching for *c*-oriented YBCO films growth, so we deposited all yttria layers at high deposition temperatures, 650–800 °C. Even for depositions at these temperatures the resulting nanoparticles can be easily moved along the substrate surface by the AFM tip in the contact mode.

The overall amount of deposited yttria is the same for both layers (average thickness ~1 nm). Detailed description of deposition procedures and dependences of yttria layers properties on deposition parameters will be published elsewhere. For comparison we prepared an $Y{:}ZrO_2$ layer in the same deposition conditions and of the same nominal thickness as yttria layers (Fig. 1c). This material has a lattice structure (fluorite) similar to that of yttria (defective fluorite), with close lattice constants (5.15 Å and 10.605/2 = 5.302 Å for $Y{:}ZrO_2$ and Y_2O_3, respectively). The fabricated $Y{:}ZrO_2$ layer is even more smooth than the uniform yttria layer, and is (001)-oriented as well.

3.2. YBCO films

The XRD $\theta/2\theta$-scans showed only peaks of YBCO for all samples on bare and yttria-decorated substrates (Fig. 2). No signs of yttria peaks or other oxides could be detected. The quality of crystal structure of the *c*-oriented grains is higher for the films on yttria layers: the peaks on the XRD $\theta/2\theta$-scans are smeared for the YBCO film on a bare LSAT substrate, while a clear $K_{\alpha1}/K_{\alpha2}$ splitting can be seen on other curves (Fig. 3 left). The strain evaluation explains this observation: the films on yttria layers have $\delta c/c = 0.3\%$, while on a bare substrate the strain increases to 0.5% (Table 1). The *c* lattice constant shows clear anti-correlation with the variation $\delta c/c$, with longest $c = 11.674$ Å corresponding to the smallest variation. The rocking curve width, as well as width of the (104) peaks on the φ-scan, is increasing with a decrease of the variation $\delta c/c$. All observed set of features reveal weaker epitaxial bonding of the YBCO film to the substrate when a decoration layer is applied: the spread of grains orientations increases both in and out-of the substrate plane. As a result of this increased spread, the substrate-induced strain in the film is partially relaxed, with corresponding decrease of variation $\delta c/c$.