Microstructure and the grain boundaries evolution in sequential epitaxial buffer layers on RABiTS-Substrates

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Abstract. Epitaxial buffer layers of CeO₂ and Yttria-stabilized ZrO₂ (YSZ) have been deposited on biaxially textured nickel substrates using reel to reel thermal reactive evaporation, and rf sputtering. The degree of texture of the deposited buffer layers were analysed by X-ray pole figure, Out-of-plane ($\omega$-scan) and in-plane ($\Phi$-scan) texture. The Microstructures of oxide buffer layer was determined by electron backscattering diffraction (EBSD) partially and SEM in selected marked area, providing information on the propagation of the grain boundary network from the substrate to the buffer layers in the same expected area. The oxide layer sequence, YBCO/CeO₂/YSZ/CeO₂/NiW was the same in each sample, but different deposition techniques were used to deposit the YBa₂Cu₃O₇ films: MOCVD, PLD, and DC-sputtering. Transport measurements on the coated conductor samples were measured 1.2 MA/cm², and 1.0 MA/sm², 1.8 MA/cm² at 77 K (0 T) respectively.

1. Introduction

The RABiTS (rolling assisted biaxially textured substrate) technique is an important approach to obtaining YBa₂Cu₃O₇ (YBCO or Y123) coated conductors (CC’s) having high critical current densities in magnetic fields at 77 K. The technique is based on the epitaxial growth of Y123 on a buffered; highly in-plane textured metal tape obtained by recrystallization after heavy cold rolling. At present, the most challenging problems facing this technique are the scaling-up to long lengths and the understanding of the current limiting mechanism in the CC’s.

The two-dimensional grain boundary network (GBN) in the Y123 CC is the main feature responsible for the reduction in the overall critical current density, due to the exponential decrease in Jc with increasing grain boundary misorientation, as observed in bicrystal experiments. Secondary causes of the reduction in Jc can include GB grooving or Ni diffusion from the metal substrate into the Y123.
There are great efforts to grow sharp textured YBCO films with high critical current density on buffered Ni tapes. Many processes including evaporation, sputtering, pulsed laser deposition (PLD), and Metal organic chemical vapor deposition (MOCVD) have been used to produce oxide buffer layers and YBCO.

In this Work, we reported deposition of textured oxide buffer layers with the well known CeO$_2$/YSZ/CeO$_2$ multilayer on textured NiW tapes using reel-to-reel evaporation and rf sputtering. The oxide buffer layer sequence, CeO$_2$/YSZ/CeO$_2$/NiW, was the same in each sample, but different deposition techniques, DC-Sputtering, PLD and MOCVD, were used to deposit the YBa$_2$Cu$_3$O$_7$ films.

2. EXPERIMENT

2.1. Deposition of oxide buffer layers

A biaxially textured (100) oriented Ni-5 at.%W Tapes from IFW Dresden was used as substrate for Y123 CC’s. The NiW tape had a typical thickness of 80 µm and a sharp biaxial texture with an in-plane full width at half maximum (FWHM) of about 5°.

The Substrates were recrystallised in the HV chamber in forming gas (7% H$_2$ in Ar) at a total pressure of 1 mbar for 30 minutes at temperatures of 780 °C.

In our process metallic Ce has been reel-to-reel deposited from a thermal evaporator (tungsten) in water vapour atmosphere at 600 °C. H$_2$O and Ce react under the formation of CeO$_2$ and H$_2$, so during the reaction water was consumed and hydrogen was produced (typical hydrogen partial pressure 10$^{-5}$ mbar to 10$^{-5}$ mbar), which results in an atmosphere preventing the formation of NiO (111). The maximum substrate length for the evaporation process was 50 cm.

Initially, a 50-nm-thick CeO$_2$ was deposited, followed by a 200-nm-thick yttrium-stabilized zircon (YSZ) deposited by reel-to-reel rf sputtering at 700 °C in forming gas (the first 200nm in Ar/H$_2$ and 400nm in Ar/O$_2$) at a total pressure of 5.10$^{-3}$ mbar. For the last buffer layer, 70 nm CeO$_2$ were deposited by rf sputtering at 650°C. In all experiments the orientation of the first buffer was just transferred to the following layers.

2.2. YBCO Deposition

YBCO films were deposited on the CeO$_2$/YSZ/CeO$_2$ buffered substrates by three deposition techniques: PLD$^1$, DC-Sputtering$^2$ and MOCVD$^3$. The thickness of the YBCO layer was about 300 nm, 450 nm and 320 nm respectively.

All layers were characterised by XRD analysis ($\theta$-2$\theta$, $\omega$- and $\phi$-scans) using Cu k$_\alpha$ radiation. Philips X’Pert four-circle diffractometer was used to determine pole figure, out-of-plane, and in-plane orientation of the film. The out-of-plane alignment (omega scan) was measured by scanning of (200) plane of the film. The in-plane alignment (phi scan) was determined by measuring of (111) planes. The buffer layers (111) pole figure was collected to determine whether the film had a single cube texture.

A simple method has been devised for examining selected areas of surfaces using Diamond pyramid of Vickers micro-Hardness Tester, this was employed to create a set of markers so that the exact area can be refound in the subsequent EBSD and SEM Measurment.

The selected area is identified by fine replica about 2 µm on the sample surface. A typical pattern is reproduced in Fig 4 and 6. In the selected marked area the GBN behavior of oxide buffer layer was determined by electron backscattering diffraction (EBSD) partially optical micrograph and SEM, providing information on the propagation of the grain boundary network from the substrate to the buffer layers in the same expected area.
3. RESULTS AND DISCUSSIONS

3.1. NiW substrates and buffer layer

The substrate and buffer layers were analysed by XRD. Fig.1 shows a θ-2θ-scan after each buffer layers deposition on a NiW substrate. Single-phased (100)-orientation of CeO₂ and YSZ can be seen.

![Fig. 1. XRD theta/2theta (a) and Phi scan(b) of CeO₂/YSZ/CeO₂ on NiW.](image)

ω- and Φ-scans revealed high degree of alignment in out-of-plane and in-plane texture of the buffers as can be seen in Fig.2. The FWHM(200) out-of-plane alignment value was measured to be 5.4°, 3.8° and 3.4° for Nickel, CeO₂ and YSZ respectively. Once again a significant decrease in the out-of-plane FWHM after each buffer layer can be seen. The FWHM (111) in-plane alignment was measured to be 6.3° in the Nickel substrate and around 6.5° in the buffer layer (Fig.3 and 4).

![Fig. 2. XRD (200) omega scan of NiW substrat and CeO₂ and YSZ buffer layers](image)

Fig. 3 shows AFM height images of NiW-substrate(a),CeO₂ (b) and YSZ (c) buffer layers. The root mean-square (RMS) surface roughness value was measured 17.6 nm, 21.6 nm, 33.7 nm and 15.3 nm for NiW substrate, CeO₂/NiW, YSZ/CeO₂/NiW and CeO₂/YSZ/CeO₂/NiW respectively.
The rough aspect of the surface is caused by the grooves that develop at the grain boundaries. As observed by AFM and SEM, grooves are related to the boundaries misoriented grains.

![Fig. 3. AFM height images of NiW-substrate(a), CeO$_2$ (b) and YSZ (c) buffer layers](image)

More detailed texture measurements were carried out on the NiW substrate and CeO$_2$ buffer layer. The marked area was then investigated by EBSD. Fig 4 and 5 present the EBSD Measurement of the interesting area. The results of the EBSD measurement are presented in form of maps and histograms, indicating the crystallographic orientation of each individual point. Automated EBSD scans were performed with a step size of 40 nm. The measurement show the grain boundaries with misorientation angles (in-plane Fig.4 (a) and out-of-plane Fig. 4 (b) relative to the ideal cube texture). The encircled area in Fig. 4 (a) was one of the three markers. Fig. 4 (c) map is a so called image quality (IQ)-map, which gives an information about the Kikuchi pattern quality to each measured data point.

![Fig. 4 EBSD measurements of RABiTS tape. Grain boundaries with misorientation angles relative to the ideal cube texture : (a) In-Plane (b) Out-of-plane (c) Image quality](image)
(a) 
(b) 
(c) 

Fig. 5 Misorientation profile of the RABiTS_Substrates.

(a) 
(b) 
(c) 

Fig. 5 EBSD measurements of CeO₂/NiW. Grain boundaries with misorientation angles relative to the ideal cube texture (a) In-Plane (b) Out-of-plane (c) Image quality.
Already the CeO2 (Fig. 6 a) films provide good coverage on the nickel surface. Most of the grain boundary grooves on the nickel surface were found to be well covered more and more by the buffer layers with very few instances of micro-cracking of the film near the nickel boundaries.

![Fig. 6 A comparison of SEM micrographs taken in the same marked area of the film after each buffer layers deposition. CeO2/NiW, YSZ/CeO2/NiW and CeO2/YSZ/CeO2/NiW (d).](image)

3.2. YBCO Deposition

For this deposition two buffer layers architectures were expected: YBCO on single CeO2 buffered substrate (YBCO/CeO2/NiW) and YBCO on the standard YBCO/CeO2/YSZ/CeO2/NiW. The thickness of the YBCO layer was about 320 nm.

Fig. 7 present results of pole figures analysis of the samples were done after the deposition of YBCO. This were collected from (111), (200), and (220) planes (Fig. 4 (a)). A four-fold symmetry is clearly seen, which is an indication of cube-on-cub textured growth. Sample with only CeO2 buffer layer show low YBCO intensities. Cracking of the buffer layers, which occurs in the highly oxidising conditions of YBCO growth, was found to be the main obstacle for the achievement of high critical current densities on metallic tapes. To avoid the cracking multilayer buffer layers are required.
3.3. YBCO

3.3.1. PLD

By this deposition two buffer layers architectures were expected: YBCO on single CeO$_2$ buffered substrate (YBCO/CeO$_2$/NiW) and YBCO on the standard YBCO/CeO$_2$/YSZ/CeO$_2$/NiW. The thickness of the YBCO layer was about 320 nm. Details on the deposition procedure can be found in [1]. Fig. 7 shows results of pole figures analysis of the samples were done after the deposition of YBCO. This were collected from (111), (200), and (220) planes.

The first sample Fig. 7 (a) shows bad Texture and low YBCO intensities. Cracking of the buffer layers, which occurs in the highly oxidising conditions of YBCO growth, was found to be the main obstacle for the achievement of CeO$_2$ single buffer layer.

However, the Sample with YBCO/CeO$_2$/YSZ/CeO$_2$/NiW architecture shows a perfect alignment of the buffers and the YBCO. A four-fold symmetry is clearly seen, which is an indication of cube- oncubtextured growth.

The FWHM for the in plane texture of the YBCO (103) (Fig.7) and the out of plane texture of the YBCO (005) was measured to be 7-8° and 4.1° respectively. The critical current density (inductive measurement) was measured to be 1.2 MA/cm$^2$ at 77 K.
Textured buffer layer architectures with high quality YBCO films have been performed on biaxially textured Ni.5%W tapes using evaporation and sputtering techniques. The texture of the substrate and buffer layers had strong 4-fold symmetry and [111] ND. The FWHM(200) out-of-plane alignment value was measured to be 5.4°, 3.8° and 3.4° for Nickel, CeO2 and YSZ respectively. Most of the grain boundary grooves on the nickel surface were found to be cumulated covered by the buffer layers. A method has been devised for examining selected areas of surfaces using Diamond pyramid of Vickers micro-Hardness Tester, this was employed to create a set of markers so that the exact area can be refunded in the subsequent EBSD and SEM Measurement.

The deposition of YBCO directly on single CeO2 buffer layer show bad growth of the YBCO film, associated with destruction of the CeO2 (100) texture.

The deposition of YBCO on CeO2/YSZ/CeO2/NiW by PLD, MOCVD and DC-Sputtering show a perfect alignment of the buffers and the YBCO with high current density.
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