Comparison of CSD-YBCO growth on different single crystal substrates

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Abstract: 2G HTS Coated Conductors properties can be improved by comparing different raw materials, precursor production routes and coating and annealing conditions. To suppress the influence of varying substrate tapes and buffer layer qualities on the HTS layers, a standard substrate is needed to improve the Jc values. In this work various pure single crystal substrates (SrTiO3 [STO], (LaAlO 3)0.3(Sr2AlTaO6)0.7 [LSAT], LaAlO 3 [LAO], NdGaO 3 [NdGaO]) are investigated to find the material which is best in terms of price, texture and morphological layout and instantaneous availability. YBCO films deposited onto these substrates via chemical solution deposition (CSD) are analysed using XRD texture analysis, surface morphology analysis (high resolution AFM) and inductive measurement of the critical current density.

1. Introduction:
The production of 2G HTS Coated Conductors by an all chemical solution deposition (CSD) process is considered as one of the most promising technologies concerning the cost / performance ratio of long lengths. A prototype of a continuous all solution coating device for tape lengths of up to 250 m has been successfully installed. Measurements on the first samples on single crystals show auspicious Jc values > 2.5 MA/cm². The conductor architecture is based on a biaxial textured metal tape - Ni or Ni-alloys - as substrate [1], a multilayer LZO (Lanthaniumzirconate) / CeO2 (Ceriumoxide) buffer system or multi-STO (Strontiumtitanate) buffer layers and superconducting YBCO-layers. Both, the buffer and YBCO layers are deposited by a low-cost CSD process with a throughput of up to 7 m/h for buffer layers and 15 m/h for the YBCO thin-film.

The YBCO layer quality defined by the maximum current density strongly depends not only on the processing conditions and precursor materials for this film itself, but also on the quality (texture and surface morphology) of the under laying substrate and the buffer layers. As each of them are under constant improvement, the application of inert and easily available reference material with suitable lattice parameters enables us to separate the substrates influence from the superconductors process conditions. The aim of this work is to find a suitable reference material for biaxial YBCO growth via CSD with reasonable current densities under conditions close to the coated conductors processing parameters. No crystal growth kinetics or stress parameters during cooling of the final YBCO film were taken into account, because all those details will change in any case for YBCO on CSD-grown buffers.
YBa$_2$Cu$_3$O$_{7-x}$ is orthorhombic for high oxygen contents ($x < 0.6$) and has lattice parameters of $a = 3.817 \text{ Å}$, $b = 3.883 \text{ Å}$ and $c = 11.633 \text{ Å}$. A biaxial texture is necessary to avoid weak grain links which could block partially or totally the current flux in the a-b-planes between the grains.

Many crystalline substrates have suitable lattice parameters for epitaxial growth of YBCO, but not all materials are sold in appropriate quality. STO is the most common material, but delivery times are growing and the material’s quality is sometimes poor, so some alternative materials are needed.

2. Experiments:

Substrates under investigation are SrTiO$_3$ [STO], (LaAlO$_3$)$_{0.3}$,(Sr$_2$AlTaO$_6$)$_{0.7}$ [LSAT], LaAlO$_3$ [LAO] and NdGaO$_3$ [NdGaO]. The lattice mismatch of all these substrates compared to YBCO is not higher as 1.5% (see Table 1). All of these substrates are chemically inert to YBCO TFA reaction and had a well polished surface in the orientation given in table 1.

| Material   | Crystal structure | Orientation | Lattice constant $[\text{nm}]$ | Plane Space $d$ $[\text{nm}]$ | Misfit to (00)YBCO $[\%]$ |
|------------|-------------------|-------------|--------------------------------|-------------------------------|-----------------------------|
| YBCO       | Orthorhombic      | 001         | $a = 0.382$  $b = 0.388$  $c = 1.163$ | $a+b)/2 = 0.385$ossed          | ($+/-$) 0                   |
| LSAT       | cubic perovskite  | 100         | 0.387 $b = 0.387$          | 0.379 $c = 0.379$            | ($+/-$) 0.47                |
| LAO        | rhombohedral      | 100         | 0.379 $b = 0.379$ $a = 0.543$ | 0.391 $c = 0.391$            | ($+/-$) 1.51                |
| STO        | cubic perovskite  | 100         | 0.391 $b = 0.391$ $a = 0.543$ | 0.391 $c = 0.391$            | ($+/-$) 1.43                |
| NdGaO$_3$  | orthorhombic      | 110         | 0.543 $c = 0.543$ $a = 0.543$ | 0.386 $c = 0.386$            | ($+/-$) 0.36                |

Small single crystals (10 mm x 10 mm x 1 mm) cannot be treated in a continuous coating process like long tapes with respect to reel-to-reel cleaning, coating, and drying routines. So preparation modes were slightly adjusted. The different crystals were rinsed with acetone and isopropanol for fifteen minutes in an ultrasonic bath respectively to cleanse the substrate of possible fouling. The single crystal substrates were not thermally treated after the cleaning. The crystals were coated with a 0.25 M (0.25 M refers to the yttrium ion concentration) TFA-YBCO-precursor solution by dip coating process with a withdrawal velocity of 0.3 cm/s and a dipping angle of 90°. The coating direction was the diagonal axes of the substrate’s surface. All coatings were carried out with the same parameters, so the sample’s thickness is comparable. It is generally between 260 and 270 nm. The crystals were then transferred without any extra drying step to the tube furnace. In a pyrolysis under wet oxygen atmosphere (20°C dew point) the organic compounds are decomposed and then they are forming oxyfluorides. During the next stage of the heat treatment up to 800°C under wet nitrogen atmosphere with 100 ppm oxygen [3]. Finally the films are kept at 450 °C and oxygen atmosphere for 90 minutes to increase the oxygen content. The conditions used are described elsewhere [4, 5, 6].

The atomic force microscope analysis of the YBCO film on the single crystals was progressed with a VEEO CP-II microscope. The measurement area was 10 µm$^2$ and the frequency value was 1 Hz. The roughness was assigned with the software called WSxM 4.0 Develop 11.0 [7].The XRD analysis was made with a 2-circle-diffractionometer (Model: Bruker axs D4 Endeavor), which works with copper $K_\alpha$ X-ray radiation (wavelength $\lambda = 1.54$ Å).

The critical current at 77 K was inductively measured with a CryoScan from Theva.
3. Results and Discussion:

Based on the lattice constant, we expect very good results for the YBCO-films grown onto the different substrates, especially on NdGaO and LSAT (mismatch < 0.4 %). Figure 3.1 shows the YBCO-film onto the LSAT substrate after pyrolysis and annealing. It shows a thick, dense and epitaxial c-axis orientated grown YBCO film.

Figure 3.1 AFM of YBCO on LSAT substrate shows high P-V value (277.1 nm) and good epitaxial growth. Measured roughness RMS= 63.9 nm.

Figure 3.2 shows the YBCO film on the NdGaO substrate. Many needles in 45° direction can be observed on the films surface which are equal to a (X00)-oriented (a-axis) growth. The displayed area is not exactly parallel to the surface borders. Because of the (110)-orientation of the NdGaO substrate, the YBCO a-axis (100) or (010) grows in 45° direction to the border of the (110) cut substrate.

Figure 3.2 AFM of YBCO on NdGaO substrate shows strong unwanted growth of YBCO crystals in 100-direction. Measured roughness RMS= 48.8 and P-V = 212.8 nm.

The YBCO film on the STO substrate is shown at figure 3.3, where a strong epitaxial c-axis growth with a fine arrangement of the peaks can be seen.

Figure 3.3 AFM of YBCO on STO substrate shows a fine, dense surface. Measured roughness RMS= 40.7 nm and P-V= 175.7 nm.
The fourth tested substrate was LAO. The AFM (figure 3.4) shows characteristic cross-needles which represent an unwanted a-axis growth. The needles grow parallel to the edge of the substrate.

Figure 3.4 AFM of YBCO on LAO substrate shows partial crossed needles (a-axis growth). Measured roughness RMS= 41.4 nm and P-V= 182.62 nm.

Figure 3.5 shows a window of the 0-20 XRD of the films. Reflexes for YBCO 006 are visible in every spectrum except in the STO scan, because in this case YBCO (006) reflex coincident with the shoulder of the STO peak. The (006) on LAO and NdGaO₃ is higher than the one on LSAT. YBCO (200) is seen in none of the scans. It is just not present in the STO and LSAT scan and it coincident with the substrate peaks in the LAO and NdGaO₃ cases. This 2nd coincidence directly gives the explanation for the intense a-axis growth. The desired (00l)-YBCO growth on NdGaO₃ is calculated for two YBCO unit cells growing on the diagonal axes of one NdGaO₃ unit cell (Table 1). But on the other hand doubling the a-axis of NdGaO₃ leads nearly to the lattice constant of the c-axis of YBCO. That is not only why the YBCO (200) peak and the NdGaO₃ substrate peak coincide in Fig 3.5, More severe is the fact, that this also triggers a growth of YBCO with the c axes parallel to the substrate and the (00l) axes perpendicular, which is a axis growth. This also explains why the needles in Figure 3.2 occur in 45° direction, which is the 100 direction of the (100) cut substrate.

Figure 3.5: XRD of the single crystals under investigation with YBCO films deposited onto them. The high double peaks refer to the substrates, and the positions for YBCO (006) and YBCO (200) are indicated.
Misorientation reduces the critical current because of the anisotropy of the superconducting effect. This is obvious on the NAGaO$_3$ sample, which has the highest (00l)-YBCO peaks, but the current density is rather low due to YBCO (200) crystals blocking the current (Table 2). The current densities of the other films are all above 2 MA/cm$^2$, the values on LSAT and STO substrates are practically the same.

| Substrate | (006) Peak height | (200) YBCO growth | Current density [MA/cm$^2$] at 77K |
|-----------|------------------|-------------------|---------------------------------|
| LSAT      | +                | no                | 2.23                            |
| NdGaO3    | ++               | strong            | 1.44                            |
| STO       | Under substrate reflex | no              | 2.18                            |
| LAO       | +                | present           | 2.02                            |

4. Conclusion

Four different single crystals were coated with a 0.25 M TFA-YBCO-Precursor solution via chemical solution deposition by dip coating process.

A good epitaxial c-axis growth ((00X) orientation) was indicated onto STO and LSAT substrates with critical current measurements of 2.18 MA/cm$^2$ and 2.23 MA/cm$^2$ at 77 K. An unwanted a-axis growth was detected onto LAO and especially NdGaO$_3$ substrates. So STO and LSAT are the more suited crystals as YBCO reference substrates. LAO is slightly worse and NdGaO$_3$ is unsuited due to massive a-axes growth of YBCO crystals.

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6. References

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