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Working around HTS Thickness Limitations – towards 1000+ A/cm – Class Coated Conductors

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Abstract

Increasing the HTS film thickness would be the straightforward route to enhance the current transport capacity of coated conductors. Usually, however, growth defects lead to a strong deterioration of the critical current density with thickness. The unique growth mode of HTS films on graded and tilted MgO buffer layers grown by inclined substrate deposition (ISD) paves the way to overcome this limit. This contribution presents micro-structural and performance data of coated conductors with monolithic REBCO-films up to 7.5 μm thickness exhibiting perfect crystallinity and critical currents in excess of 1000 A/cm-width.

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1. Introduction

The engineering current density of coated conductors is the key performance parameter for all technical applications. Hence, a lot of research has been directed towards pushing the intrinsic performance limits of HTS films e.g. by means of nano-engineering. However, since the functional HTS layer contributes just a few percent to the entire coated conductor cross section the most straightforward and economic approach is increasing the effective HTS film thickness. Unfortunately, however, epitaxial growth of thick films constitutes a major challenge. The thickness of HTS films e.g. grown by metal-organic deposition (MOD) and re-crystallization of amorphous precursors, seems to be fundamentally

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limited due to the underlying nucleation process. But also epitaxy by physical vapor deposition (PVD) is hampered by the accumulation of growth defects and secondary phases which result in a strong degradation of the critical current density with increasing film thickness [1]. Eventually, this leads to a dead surface layer with a high degree of disorder and usually limits the useful HTS thickness to 2-3 μm .

In order to suppress or retain the current density of thin films or to avoid the growth of defects two successful approaches have been reported. One is to introduce 30 nm thin CeO_2 intermediate layers after every 500 nm of YBCO growth [2]. In such multi-layers it was possible to retain extremely high current densities up to 3.5 μm thickness and to achieve critical currents of 1400 A/cm (@ 75 K). However, the successive switching between two components constitutes a major complication and cost factor for production. Another successful approach was made by maintaining a precise growth temperature throughout the process by means of a hot-wall radiation heater [3]. In this way up to 6 μm thick monolithic GdBCO layers were demonstrated with 1040 A/cm (@ 77 K). However, this heating concept is restricted to pulsed laser deposition within a relatively small deposition area and will be difficult to transfer to large area deposition by other techniques, which is the ultimate goal for scaled commercial production.

Nucleation and the epitaxial growth mode depend strongly on the substrate surface conditions. Inclined substrate deposition (ISD) produces a uniquely tilted and terraced buffer surface [4-5]. Hence, such ISD-buffers are fundamentally different from those plain surfaces which are commonly produced (e.g. by RABiTS, IBAD) in coated conductor fabrication. This study is investigating the impact of the unique ISD surface structure on the growth of thick DyBCO films.

2. Experimental

All coated conductor samples for this survey were prepared by e-beam evaporation on electro-polished Hastelloy C 276 substrates (100 mm long, 10 mm wide, 90 μm thick). Details of this process have been reported previously [6-7]. In general, 2.5 μm thick MgO buffer layers were deposited at room temperature by inclined substrate deposition, where the substrate was tilted with respect to the e-beam evaporator. The samples originate from an ISD deposition series where the tilt angle was varied between 30-38° and the MgO deposition rate was 4-5 nm/s. The buffer was completed by a 300 nm thin MgO cap layer grown at 720°C under normal incidence. The characteristic features of the ISD-MgO buffer are depicted in Fig. 1. Fig. 1a) shows the unique surface morphology of the MgO surface, composed of cubic crystals tilted by 25° with respect to the substrate normal and forming characteristic terraces of 50 - 150 nm width.

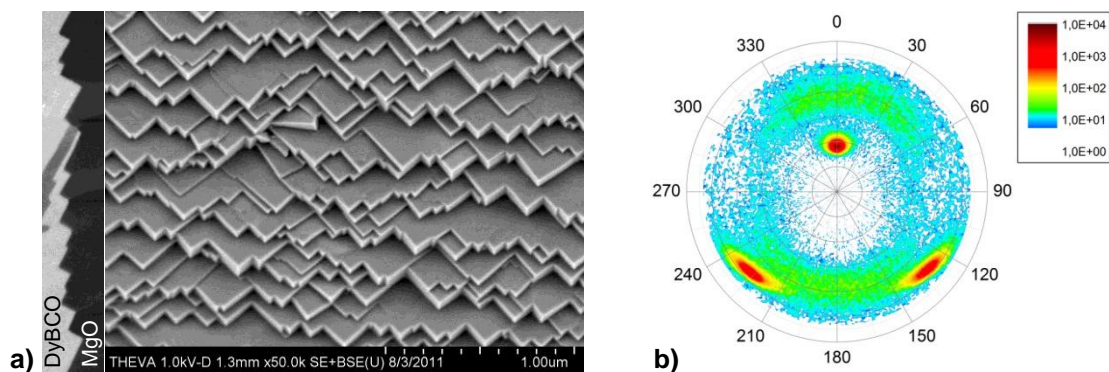


Fig. 1. a) MgO buffer surface and cross sectional view (left) of the MgO/DyBCO interface; b) Pole figure of the MgO (200) reflex.

In cross-section (outermost left insert) the interface between MgO and the DyBCO layer resembles a staircase. Fig. 1b) shows a typical XRD result for such a highly oriented MgO buffer layer. In the pole figure the MgO (200) peak is tilted by 25° from the center and the orientation deduced from the FWHM in ϕ - and χ -sections is $\Delta\phi < 10^\circ$ in-plane and $\Delta\chi < 4^\circ$ out-of-plane. Since XRD is averaging over the film thickness, the orientation of the topmost surface layer is usually better reflected in the subsequent DyBCO orientation.

On top of the ISD-MgO buffer layers a series of up to $7.5 \mu\text{m}$ thick DyBCO layers was deposited by reactive e-beam deposition of DyBCO powder. We intentionally employed a Ba-deficient powder and the composition of the resulting films as analyzed by inductively coupled plasma (ICP) analysis was Dy:Ba:Cu = 17:29:54. Compared to the ideal 123-stoichiometry this means a massive Cu- and Dy-excess of 24% and 17%, respectively.

Structural properties were characterized by XRD (Θ - 2Θ -scans and pole figures) and electron microscopy (plan-view and cross sections). All DyBCO films exhibited excellent in-plane ($\Delta\phi < 6^\circ$) and out-of-plane ($\Delta\chi < 2.5^\circ$) orientation. For electric transport measurements, each sample was coated with a 200 nm silver contact layer and several bridges (1.1 mm wide and 3 mm long) were patterned by wet-etching. Measurements were carried out in liquid nitrogen applying a $1 \mu\text{V}/\text{cm}$ – criterion.

3. HTS growth on terraced ISD buffers

The most common defect structures observed in HTS films are a-axis oriented grains and secondary phases due to deviations from the ideal 123-stoichiometry. A-axis grains are easily generated because the c-axis length is practically three times the a-axis lattice constant and hence both orientations satisfy epitaxial conditions. Once generated, due to the strong anisotropy of the growth speed in both crystallographic directions $v_{ab} \gg v_c$, a-axis grains form platelets growing in size and height much faster than the surrounding c-axis matrix of the film. In a similar way secondary phases once emerged from the HTS film surface are hardly overgrown and gain in size. Eventually, these processes lead to an accumulation of defects and failure of epitaxy.

This prospect changes significantly when considering growth on a staircase as in the case of ISD-buffers. The implications of the stepped surface are sketched in Fig. 2. First, the terrace width sets a natural limit to the lateral growth of a-axis grains. Second, on a staircase there is always a higher step from which an a-axis grain can be overgrown laterally by the HTS layer (arrows in Fig 2a). In a similar way the HTS layer can overgrow secondary phases (arrows in Fig. 2b) thus pushing them to the side until they encounter a grain boundary (GB). These growth mechanisms lead to a termination of such defects, so that their density cannot become critical.

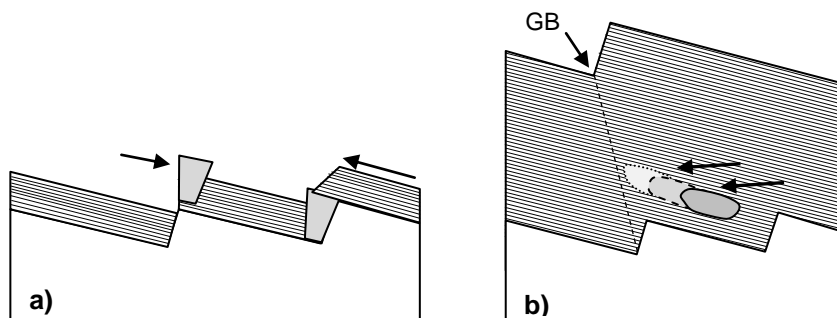


Fig. 2. Proposed growth mode of HTS films with a-axis grains (a) and secondary phases (b) on a stepped surface

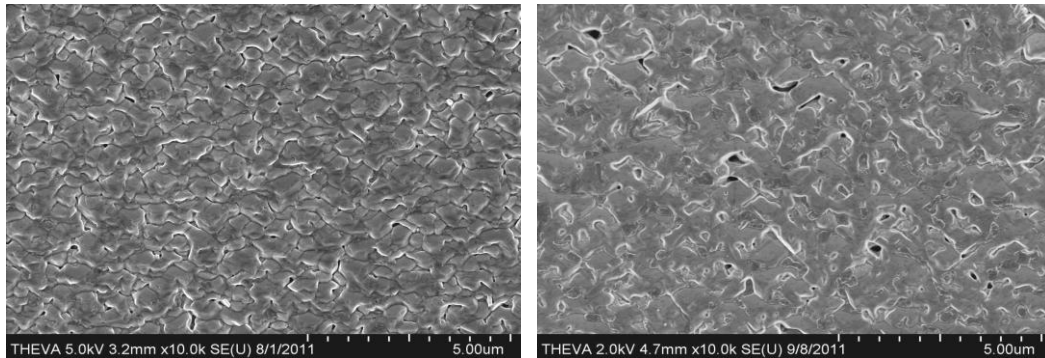


Fig. 3. Surface structure of 1 μm (left) and 5 μm thick (right) DyBCO coated conductors on ISD buffers.

This perception is impressively confirmed in the micrographs of such films. Fig. 3 shows the surfaces of a 1 μm and a 5 μm thick DyBCO film. Apart from a slightly coarser grain structure, there is little difference and neither a-axis grains nor segregations are detectable on the surface. The absence of a-axis growth in the bulk of the films was also confirmed by XRD and high resolution transmission electron microscopy (TEM) as reported separately [7-8].

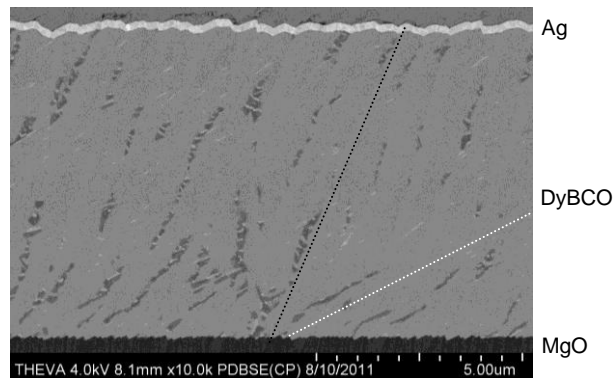


Fig. 4. Cross section of 7.5 μm thick DyBCO film on ISD-MgO. The dotted white line indicates the terrace tilt angle, the black line the prevailing direction of grain boundaries within the DyBCO layer.

Fig. 4 shows a cross section of a 7.5 μm thick DyBCO film. Starting from the MgO interface the DyBCO grain size and surface corrugation increase to 0.5-1 μm . The massive surplus of Cu and Dy leads to obviously visible secondary phases. By EDX microprobe the dark inclusions could be identified as Cu-rich particles, whereas the bright inclusions depict a Dy-rich phase. In accord with the concept presented in Fig. 2b a lot of the Cu-rich segregations, especially in the lower part of the film where terraces are yet small, are growing along these terraces (direction of white line) and pushed aside until encountering a grain boundary. However, there is also another growth mode observable. DyBCO grain boundaries are usually related to the kinks and corners in the stepped growth front. For energetic reasons these corners are also preferred nucleation sites for secondary phases. Hence, these tend to pile up at there following the grain boundaries in the film (direction of black line). Such inclusions consist of alternating Cu- and Dy-rich segregations. In the course of film growth most of them are also overgrown and terminated. Their density remains practically constant throughout the film and is naturally given by the excess of the related metal species. Hence, their volume can be controlled by adjusting the stoichiometry of the HTS film.

4. Results

Fig.5 shows the critical current measured as a function of film thickness. Due to the different ISD buffers used for this study there is considerable scatter in the data. However, overall there is a practically linear increase with the thickness and samples with 6-8 μm exceed 800 A/cm. The best sample with 5.9 μm thickness exhibited 1018 A/cm. For these very Cu-rich films the average current density was 1.3 MA/cm². Lately, using DyBCO powder closer to the ideal 123-composition, we were able to increase the current density beyond 2 MA/cm² in films up to 2 μm and are currently preparing another thick film coating series. Therefore we expect that films with optimized ISD buffer and DyBCO composition will easily exceed the 1000 A/cm-width level beyond 6 μm thickness.

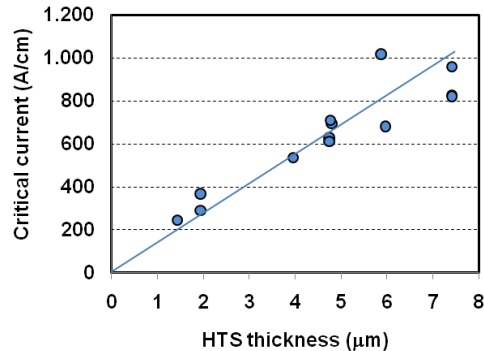


Fig. 5. Critical current of ISD-based coated conductors versus HTS thickness

5. Conclusion

In summary, the unique HTS growth mode on ISD buffer layers leads to a suppression of a-axis growth and termination of common defects. This allows deposition of thick monolithic HTS films without strong degradation of the critical current density. Thus, coated conductors with extremely high critical transport currents beyond 1000+ A/cm can be realized by simply extending the HTS deposition.

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