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Inclined Substrate Deposition by Evaporation of Magnesium Oxide for Coated Conductors

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ABSTRACT

Biaxially textured MgO buffer layers were deposited on metal substrates using "inclined substrate deposition" (ISD). The influence of the substrate inclination angle, deposition rate, and film thickness on the texture is shown. Scanning electron microscopy reveals columnar growth. We developed a growth model to explain the texturing. To test this model we have carried out 3D Monte-Carlo simulations. We find that the preferred orientation arises from mutual shadowing of the columns and directional surface diffusion due to their initial momentum.

$YBa_2Cu_3O_7$ (YBCO) films deposited on the ISD buffer layers are highly textured. The ab-planes of the YBCO are tilted with respect to the surface by typically 25° towards the direction of MgO vapor incidence. Therefore, the critical current density j_c is anisotropic with up to 8×10^5 MA/cm² in one direction and 4×10^5 MA/cm² in the other. For tape coating the MgO deposition direction can be chosen so that the high j_c is along the tape.

INTRODUCTION

Thin films of YBCO must be highly textured in order to have good superconducting properties. One way to achieve this is to grow textured buffer layers on arbitrary polycrystalline substrates by ISD. This was first proposed by Hasegawa et al. [1] using pulsed laser deposition of yttria-stabilized ZrO_2 (YSZ). We use evaporation techniques to make the ISD process scalable to large areas and high production rates. The buffer material is MgO. It was already reported that high critical current densities were achieved on MgO ISD buffer layers [2]. This shows that ISD-MgO is a very promising candidate for the successful realization of YBCO coated conductors.

In this paper we will give an overview of the experimental results. In order to further optimize the buffer layer deposition it is important to understand the mechanism of texturing. We present a model that can explain the growth of textured ISD films. Monte-Carlo simulations based on this model were also carried out. The results will be compared with the experimental findings.

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EXPERIMENTAL DETAILS

The experimental setup for ISD is shown in figure 1. We used an e-gun with MgO or Mg target as evaporation source. Oxygen was supplied locally at the substrate. The deposition rate, which was measured by a quartz crystal monitor, was varied between 0.1 nm/s and 8 nm/s. The substrate was positioned above the source and inclined by an angle α as shown in figure 1. The substrates were mechanically or electropolished Hastelloy C276 or stainless steel pieces (10 × 10 mm²) with a thickness between 0.05 mm and 0.5 mm. For experiments, where no YBCO was deposited on the ISD buffer layer, thermally oxidized Silicon substrates were used as well. Further details of the deposition setup were described earlier [2].

YBCO was deposited by reactive thermal co-evaporation that was described in detail elsewhere [3]. Briefly, the metals are evaporated from three different resistively heated boats. At a temperature of 680 °C the rotating substrate alternately moves through a deposition zone and an oxidation zone with increased oxygen pressure. A MgO single crystal substrate is deposited together with the ISD substrates as a monitor of the YBCO deposition process. The YBCO films on these monitor substrates had critical temperatures between 86 K and 90 K indicating high quality YBCO films.

The critical current density j_c was determined inductively and resistively at 77 K in liquid nitrogen. The inductive measurement with a 3rd harmonic method was used as a standard characterization tool as it is a nondestructive method. In order to measure the anisotropy of the critical current density, the films were patterned with a cross like bridge for resistive measurements. The width of the bridges was 0.6 mm and the length 3.6 mm. A criterion of 1 µV/mm was used.

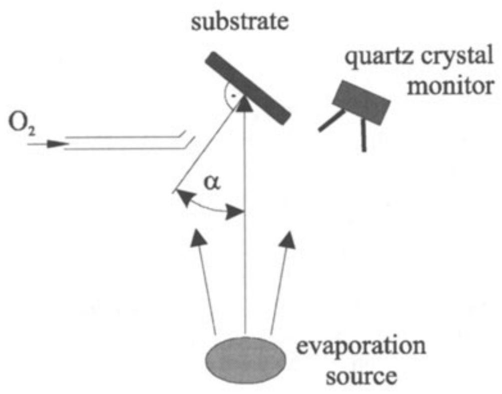

Figure 1. *Experimental setup for inclined substrate deposition. The evaporation source was an e-gun with MgO or Mg target.*

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RESULTS AND DISCUSSION

ISD buffer layer

To investigate the structure and texture of the MgO films, the deposition parameters like substrate inclination, deposition rate and film thickness were varied. The exemplary results, which are shown below, are from MgO deposition experiments if not mentioned otherwise. Similar results were obtained in the case of reactive evaporation of Mg. The results of these investigations are important to understand the properties of the YBCO films and will be used for the motivation of a growth model for ISD that can explain the texturing.

Figure 2 shows the scanning electron microscope (SEM) image of the fracture cross section of an ISD buffer layer. It reveals a pronounced columnar structure. In this image the columns are tilted slightly away from the vapor direction which is indicated by an arrow. In general the columns are always oriented about parallel to the surface normal. SEM images also reveal that the general shape of the columns is not round but faceted. This means that the tip is a plane which is tilted towards the deposition direction, as it can be seen in figure 2, and also the sides of the columns are planes.

The pole figure of an ISD buffer layer is shown in figure 3. In contrast to the common case the [001]-axis is not parallel to the substrate normal but tilted by the texture angle β towards the deposition direction which is indicated in the pole figure, too. Due to the tilt, the poles of the (100) and (010) planes are clearly visible and indicate a high degree of texture. The in-plane alignment of the film, measured by φ-scans of the (220) peak, was 8° FWHM. If we compare the pole figure and the SEM image, we find that the facets of the growth columns are {001} planes. This tendency of MgO, namely to generate crystals with {001} planes, is well known and can be explained by the high mobility of MgO molecules on these planes [4].

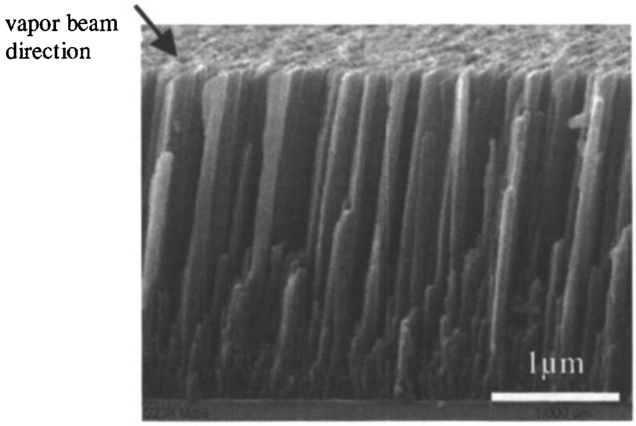

Figure 2. *Fracture cross section of an ISD film deposited by reactive evaporation of Mg (SEM image)*

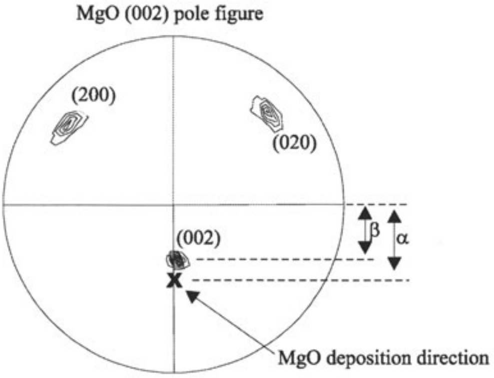

Figure 3. (002)-pole figure of an ISD MgO film. α is the angle between the substrate normal and the deposition direction indicated by the **x***. β is the angle between the substrate normal and the (002)-pole.*

The influence of the substrate inclination on the texture is shown in figure 4. The texture angle increases with increasing substrate inclination angle. The FWHM of the in-plane alignment varies more drastically. At $\alpha < 20°$ no alignment is observed (FWHM = 90°). With increasing substrate inclination the alignment improves drastically and the smallest FWHM is observed at $\alpha = 50°$. These results show that β and the in-plane alignment cannot be adjusted independently. A good in-plane alignment would imply a rather high tilt angle of more than 25° whereas larger FWHM values have to be accepted if small tilt angles are needed.

The influence of film thickness and deposition rate on the texture is shown in figure 5. Although neither the film thickness nor the deposition rate have an effect on the texture angle, the in-plane FWHM in clearly influenced. With increasing thickness the

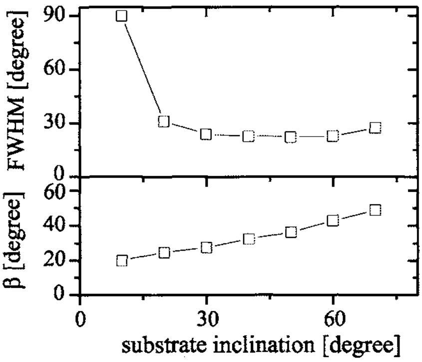

Figure 4. Texture angle β and FWHM of the in-plane texture versus the substrate inclination angle a.

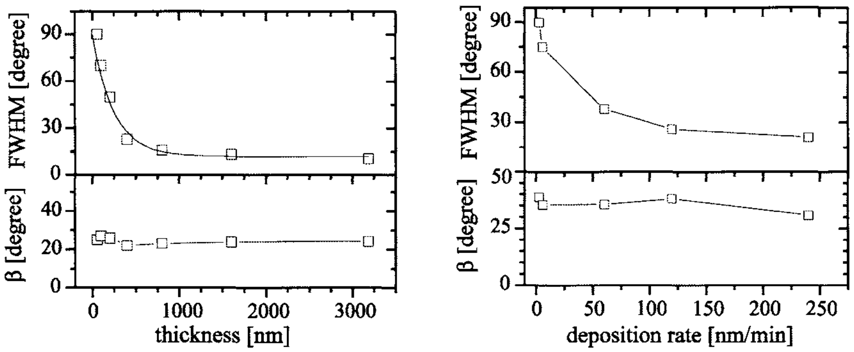

Figure 5. *Texture angle β and FWHM of the in-plane texture versus the film thickness (left side) and the deposition rate (right side). The line on the upper left side is an exponential decay function fitted to the data.*

FWHM decreases exponentially. A similar dependency was observed in biaxially textured YSZ films deposited by ion beam assisted deposition [5]. It can be explained by an evolutionary selection process where misoriented grains are slowly overgrown by grains with the 'correct' orientation. Also the deposition rate strongly influences the in-plane texture in the way that high deposition rates give highly textured films. As the deposition rate is limited by the evaporation source in our experiments, even higher deposition rates seem to be possible. To achieve a good in-plane alignment, a more than 1 μm thick film must be deposited with high deposition rates.

Growth Model and Monte-Carlo Simulation

Based on the experimental results the growth of the ISD MgO films can be explained as shown in figure 6. First randomly oriented grains nucleate on the substrate surface (figure 6(a)). Once the substrate is covered by MgO, new material is added by local epitaxial growth on the already existing MgO grains and growth columns are formed (figure 6(b)). Due to the inclined deposition, the columns tend to shade each other. High columns gather more material whereas low columns tend to stop growing as they are shaded by their neighbors. Because MgO tends to form (100) surfaces, the shape and with it the height of the columns is linked to their crystalline orientation. The crystalline orientation, which gives the highest columns, will survive during the further film growth (figure 6(c)). This way, the alignment of the surface of the film improves with increasing film thickness.

As shown in figure 2, the columns grow in a direction nearly perpendicular to the substrate surface. This is unusual for inclined deposition because one would expect them to be inclined towards the deposition direction [6]. It was proposed earlier that the momentum of the molecules or atoms hitting the substrate leads to a directional diffusion as it is shown in figure 7(a) [7]. This effect causes a mass transport on the flat planes of the columns so that every new atomic layer is shifted away from the deposition source and the growth direction changes (figure 7(b)). In our case a directional diffusion is very

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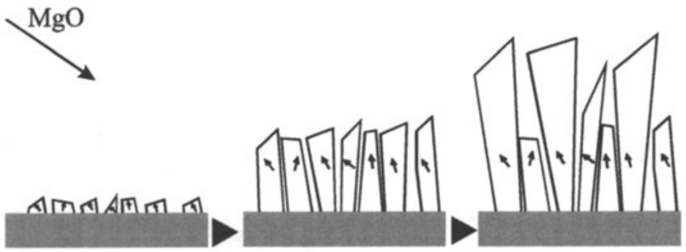

Figure 6. Growth model for inclined substrate deposition of MgO. The small arrows indicate schematically the crystalline orientation.

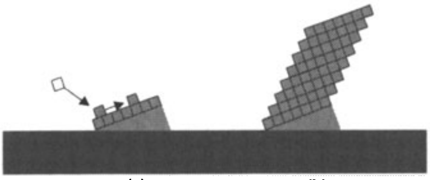

Figure 7. Schematic sketch of the directional diffusion due to the momentum of the molecules or atoms in the vapor (a) and changing of the column growth direction (b)

likely because of the flat surfaces and the high mobility of MgO. Unfortunately, to our knowledge no theoretical work was published on directional diffusion, so that a more precise theoretical investigation is not possible yet.

We performed 3D Monte-Carlo simulations of the thin film growth of a mono-atomic cubic material to substantiate our growth model. Inclined deposition as well as surface diffusion were taken into account. Additionally, also the influence of directional diffusion was investigated by increasing the probability of an atom to move parallel to the direction of the vapor beam during the first diffusion steps. To simplify the calculations, only one crystalline orientation, e.g. $\beta = 30°$, was considered in each calculation. This is different from the real film growth where grains with various crystalline orientations nucleate and grow next to each other. The idea is to calculate simulations with differently chosen crystalline orientations and to compare the results. The simulation starts with a flat surface. Then an atom is generated at an arbitrary point above the surface and moved towards it with the inclination angle $\alpha = 40°$. After hitting the surface, it can diffuse along <001> and <011> directions until it finds a position with more than one nearest neighbor. At this position it is fixed permanently to the surface and the next atom is generated. Typically 5 x 10^6 Atoms are deposited for one simulation.

The interesting question now is whether the experimentally observed crystalline orientation and the one, which gives the highest columns in the Monte-Carlo simulations, are the same. It is useful to discuss the results of three exemplary calculations first. We start with a simulation where no directional diffusion is taken into account and the

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orientation of a real MgO film (figure 3) is chosen for the simulation, too. This means that the texture angle is 30° and the projections of the [011] axis and the vapor beam direction on the substrate surface plane are parallel. The cross section of this simulation is shown in figure 8 (a). The crystalline orientation and the vapor beam direction are indicated in the image. Growth columns form which are tilted towards the vapor direction. In figure 8 (b) the same crystalline orientation of the film is employed but in this case also directional diffusion is taken into account. As expected, this changes the growth direction of the columns and perpendicular columns develop. Because also the tips of the columns are inclined planes, the structure of the film is similar to the real one (fig. 2). This proves that our simulations can reproduce the real film growth. The film structure depends on the crystalline orientation, too. For example, if the texture angle is increased from 30° in figure 8 (b) to 50° in figure 8 (c), the growth columns become much more tilted and hardly observable. Comparing figure 8 (b) and (c), β = 30° gives higher columns and will therefore overgrow columns with β = 50°.

Up to now only two different orientations were shown. The next step is to vary the film orientation in a broader range and to compare the column heights. They were determined by calculating the mean film height and normalizing this value to the height of a dense layer without voids. In figure 9 (a) the dependency on the texture angle β is shown. The highest columns grow at a texture angle of 30° (cross section shown in figure 8 (b)), which is equivalent to the real film orientation. It is noticeable that the height decreases drastically with smaller texture angles and becomes nearly 1 at β = 0°. At this orientation no steps are built on the (001) surface plane so that the atoms can diffuse over long distances without being captured by a monoatomic step. This way, every hole is filled before a new layer is build and a dense films grows. The influence of the in-plane orientation φ, which is the angle between the projection of [011]-plane and the projection of the vapor direction on the substrate plane, is shown in figure 9 (b). At φ = 0°, which again corresponds to the experimentally found orientation, the highest columns are found.

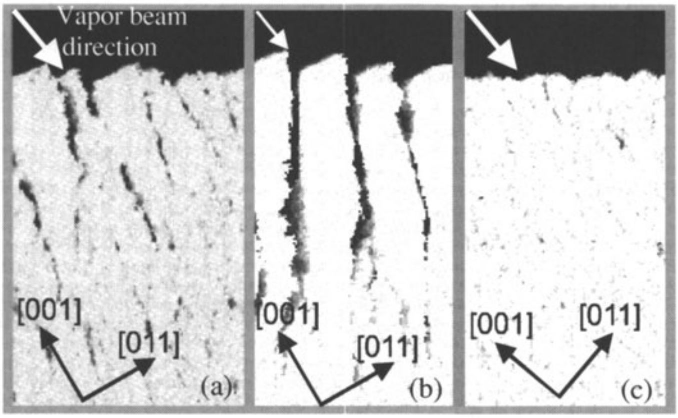

Figure 8. *Cross sections of Monte-Carlo simulations: 5 x 10^6 Atoms deposited, $\alpha = 40°$; (a) $\beta = 30°$, no directional diffusion; (b) $\beta = 30°$, with directional diffusion; (c) $\beta = 50°$, with directional diffusion.*

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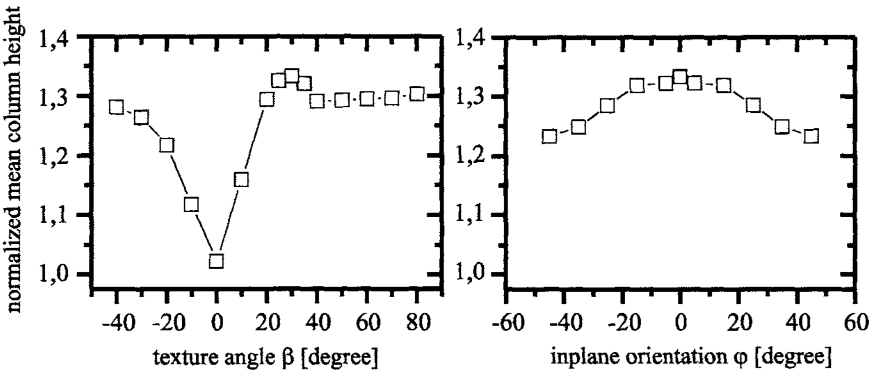

Figure 9. *The mean column height of Monte-Carlo simulations normalized to the height of a ideally dense layer in dependence of the crystalline orientation*

The simulations therefore confirm that the biaxial alignment can be explained by a evolutionary growth process, where only the highest columns survive.

YBCO Films on ISD Buffer Layers

Fig. 10 shows the (103) pole figure of a YBCO film grown on biaxially textured MgO. It reveals epitaxial growth with the usual cube on cube orientation: YBCO [001] || MgO [001] and YBCO [100] || MgO [100] (or [010]). Correspondingly, the c-axis is tilted by the texture angle and not parallel to the substrate normal. The in-plane texture of the YBCO is in general up to 6° smaller than the FWHM of the MgO film underneath. As YBCO is sensitive only to the texture directly at the surface of the MgO buffer layer, the smaller FWHM indicates that the biaxial texture of the MgO surface is better than the value measured by XRD. As the latter represents a bulk value due to the large penetration depth of the X-rays (> 1 μm), this corresponds to the decrease of the MgO in-plane FWHM with increasing film thickness (figure 5(a)).

The critical current density versus the in-plane texture, as it was measured by an inductive method, is shown in figure 11. It rises nearly exponentially with decreasing FWHM which is due to the decrease of high angle grain boundaries. The highest value is 0.57 MA/cm² at 7.5° FWHM. The texture angle is indicated by the error bars in figure 11, too. YBCO films with higher tilt angles have reduced inductive critical current densities compared to films with the same FWHM but smaller tilt angles. This can be explained by an anisotropy of the critical current density in the substrate surface plane. If the current flows parallel to the projection of the MgO vapor beam i.e. the tilt direction of c-axis of YBCO ($j_{c||}$), it has a lower value than if it is applied perpendicular to it ($j_{c\perp}$). This anisotropy is due to the tilt of the ab-planes and the intrinsic anisotropy of the critical current density of YBCO [2]. As the inductive j_c measurement applies a circular current (perpendicular as well as parallel directions) only the smaller $j_{c||}$ is measured. Films with high texture angles have a more pronounced anisotropy so that the inductive j_c decreases.