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Characterization of the Structure of Y-Ba-Cu-O Coated Conductors

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Abstract-- Transmission electron microscopy (TEM) has been applied to the microstructural investigation of $YBa_2 Cu_3 O_{7-\delta}$ (YBCO) thick films deposited on polycrystalline Ni-based metal substrates by pulsed laser deposition. The films were found to be strongly textured with c-axis oriented grains aligned perpendicular to the substrates. Despite the large average in-plane misorientation ($\theta \sim 14°$), as was estimated from selected area electron diffraction and x-ray diffraction, TEM inspection reveals colonies of submicron-sized grains with low angle ($\theta \leq 7°$) tilt grain boundaries. The linkage of the colony structures may provide a continuous percolation pathway for the supercurrent transport in YBCO, which may provide the mechanism for the higher than expected critical current density J_c. Periodic arrays of grain boundary dislocations were observed, which may serve as effective flux pinners.

I. INTRODUCTION

Future applications of high temperature superconductors (HTS) will require long lengths of superconducting materials with current density exceeding ~ 10^6 A/cm^2 at the operating temperatures and fields. It has been shown that the grain boundaries in polycrystalline conductors can severely degrade the transport properties, and the presence of magnetic fields further degrades the performance. The benefit of in-plane grain alignment has been documented in YBCO [1] and Tl (1223) thin film bicrystals [2], in which high critical current density (Jc) has been observed across low angle grain boundaries, and deteriorates exponentially with grain boundary misorientation angles beyond 7°. The in-plane alignment can be obtained via the formation of a strongly textured template. The two leading techniques are: ion beam assisted deposition (IBAD) of a buffer layer [3], and the rolling-assisted-biaxially-textured-substrates (RABiTS) method [4]. Critical current densities exceeding 10^6 A/cm^2 at 77K, zero field, have been reported in these textured YBCO films. These findings illustrate the potential and importance of controlling in-plane grain alignment for approaching the performance required for large-scale HTS applications.

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The recent progress at Los Alamos National Laboratory (LANL) in achieving high supercurrent density in one-meter-long YBCO superconducting tapes can be attributed to the success in forming highly textured superconducting thick films on oxide templates grown by the IBAD process. The best result to date is a tape with a current density of 1x10^6 A/cm^2 at 75K for a one-meter length [5]. In this report, a TEM investigation of the degree of texture and grain boundary structure in one micron thick YBCO films grown on polycrystalline metal substrates will be discussed to elucidate the mechanisms of supercurrent transport in the one-meter-long YBCO coated conductor tapes.

II. EXPERIMENTAL PROCEDURES

The deposition of YBCO coatings was conducted by first depositing an IBAD yttria-stabilized-zirconia (YSZ) layer with a thickness of 0.5 µm - 1 µm on Inconel 625 alloy substrates. This was followed by the deposition of a 120 nm YSZ layer, a 20 nm - 30 nm Y_2O_3 layer, and then the final coating of a 1-µm-thick YBCO layer by pulsed laser deposition (PLD). Fig. 1 shows the schematic of the layered geometry of the coated YBCO film. A detailed description of the facility, deposition parameters and superconducting properties characterization has been reported previously [6].

For plan-view TEM examination, 3mm discs were first punched out of the tape, and mechanically thinned from the substrate side to ~ 15 µm. Low angle Ar ion milling was then carried out to further thin the sample from the substrate side until perforation and electron transparency. A final ion polishing/cleaning from both sides was conducted to remove potential surface contamination during the ion thinning. TEM characterization was performed on a Philips CM30 analytical TEM, and a JEOL 3000F high resolution TEM, both were operated at 300 kV.

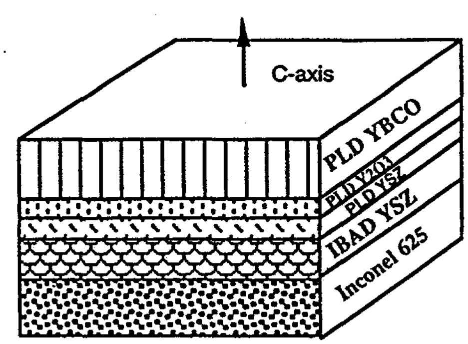

Fig. 1. Schematic diagram showing the layered structure of the coated YBCO thick film on Inconel substrate.

III. RESULTS AND DISCUSSION

Fig. 2(a) shows a plan-view bright field image of the YBCO film viewed along the c axis. The film is seen to consist of grains with near equi-axed shape, and an average grain size of ~0.5 μm. The presence of the {110} twins in the YBCO grains provides an internally calibrated axis and orientation marker, which can be used to estimate the misorientation between neighboring grains. The grain boundary misorientation angles, as estimated from the twin contrast, were marked on Fig. 2(a). It can be clearly seen that most of the grain boundaries are low angle boundaries, with misorientation angles $\theta \leq 7°$. Some of the boundaries have such small in-plane rotation angles that periodic arrays of dislocations can be distinguished, as marked by arrows, on the image. Fig. 2(b) is the corresponding selected area diffraction (SAD) pattern that reveals the strongly textured in-plane orientation relationship. The arc of the diffraction spots represents the distribution of the a-axis (or b-axis) in all the grains included by the SAD aperture (approximately 15 grains). The angular distribution of 14°, as measured from Fig. 2(b), gives an estimated "average" in-plane misorientation. This is consistent with the x-ray ϕ scan result, which shows a 12° FWHM in-plane misorientation.

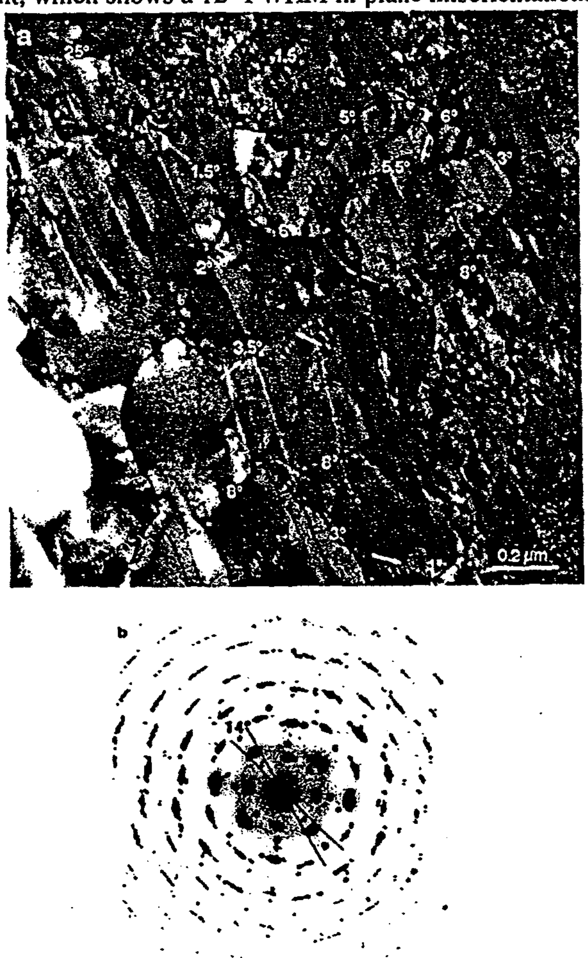

Fig. 2(a) Plan-view TEM image showing the grain structure of the strongly textured YBCO film. The misorientation angles across the boundaries were estimated from the twin contrast and marked on the images. (b) Corresponding SAD pattern showing the average in-plane misorientations.

A high resolution TEM (HRTEM) view of one of the low angle boundaries in Fig. 2(a) is shown in Fig. 3(a), which shows the lattice image of a [001] low angle tilt grain boundary. It can be seen that the grain boundary plane lies very close to the (010) plane of the two grains neighboring the boundary, and the two grains are related by a small rotation about the c axis. The strain contrast from the periodic dislocation array is clearly visible from the lattice image. A computer generated FFT (Fast Fourier Transform) pattern taken from the region marked in Fig. 3(a), is shown in the inset. The pattern clearly shows the superposition of two slightly misoriented [001] patterns from the two grains. The misorientation angle can be estimated to be ~ 7±0.5°. Image reconstruction by filtering out all but the (010) reflections resulted in the inverse-FFT image as shown in Fig. 3(b). The presence of the periodic array of grain boundary dislocations (GBDs) can be clearly visualized as indicated by arrows in Fig. 3(b). By using the {100} sets of lattice fringes as a reference, the dislocation spacing can be estimated to be 31± 2 Å.

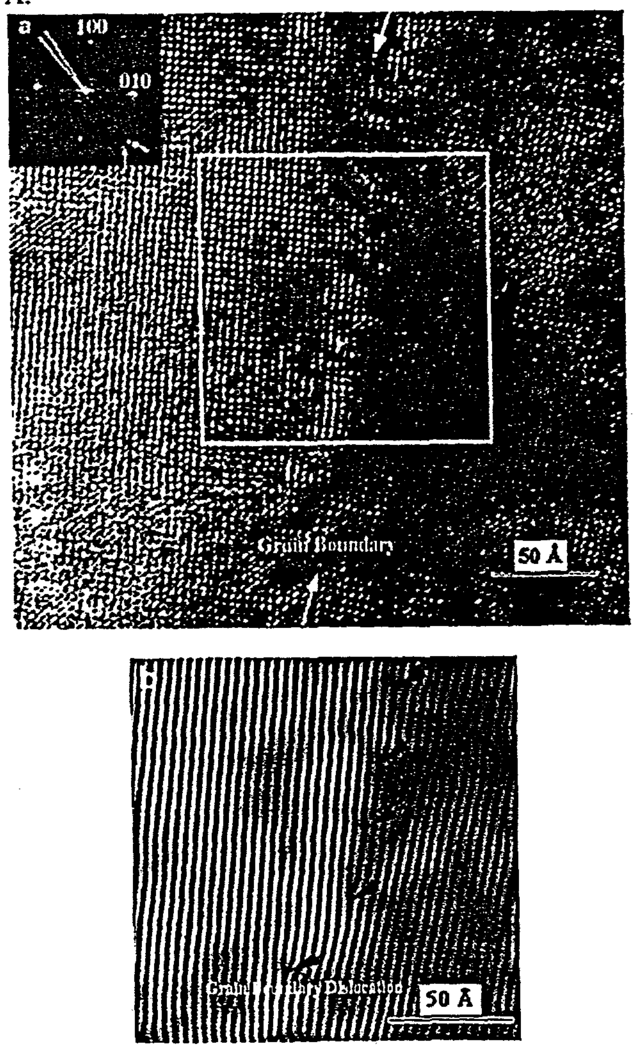

Fig. 3(a) HRTEM image of a low angle tilt grain boundary. FFT pattern (inset) obtained from the center region shows the misorientation of the two neighboring grains is ~ 7°. (b) Reconstructed (inverse FFT) image formed by including reflections from the (010) planes. An array of grain boundary dislocations (as marked by arrows) is visible on the image.

From a simple geometrical consideration, for a low angle grain boundary, the spacing (d) of grain boundary dislocations (GBDs) can be estimated from the ratio of the magnitude of the shortest lattice translation vector (Burgers vector, b) and the misorientation angle (θ), as described by Frank's formula:

$$d \approx \frac{|b|}{\theta} \qquad (1)$$

For a dislocation spacing of 31 Å and a misorientation angle of 7°, the magnitude of the GBD Burgers vector is estimated to be ~3.8± 0.1 Å. This corresponds, as expected, to the unit lattice translation vector of the b axis (3.88Å), i.e. [010], of orthorhombic YBCO.

The edge type GBDs with line direction along the c-axis of YBCO have been postulated [7] to be the origin of strong flux pinning due to the localized nature and spacing of these line defects. Recently, a study by Diaz et al. [8] demonstrated that the critical current density, Jc, across a 4° tilt grain boundary displays a maximum for magnetic fields close to the c axis in fields up to 7 tesla. The observation is consistent with vortex pinning by a dense planar distribution of line pins. From Figs. 2 and 3, it can be seen that the polycrystalline YBCO films consist of strongly textured grains with a majority of them at small angles- in the range of 2° – 7°. This will result in GBDs with a range of spacing from 110 Å to 30 Å. The numerous, well-aligned, edge type grain boundary dislocations are spaced at the appropriate length to potentially serve as effective flux pinners up to high magnetic fields.

The grain boundary structure has been characterized extensively for YBCO thin film bicrystals [9, 10]. The present study is the first systematic study carried out on YBCO polycrystalline coated conductors grown on metal substrates. Despite the different synthesis routes, one common feature found in the coated conductors as well as the bicrystal thin films is the presence of the periodic array of GBDs in low angle grain boundaries and the relatively undisturbed channels between the dislocations. The burgers vectors of GBDs in both cases are determined to be of the unit lattice translation vector: a[100} or b[010] for boundaries with {100} type of grain boundary plane. However, the grain boundaries in the polycrystalline coated conductors are found to be wavier and form fewer facets than were reported for thin film bicrystals. This could be related to the submicron grain size and the large radius of curvature of the polycrystalline grains.

It is worth noting that despite the large (14°) average in-plane angular distribution, the misorientation between grains is small enough in most cases to reduce the overall weak link effect. This suggests the possibility that percolation pathways for supercurrent can be formed in regions consisting of grains with very low-angle (less than 7 degrees) grain boundaries. Indeed, in a low magnification view of the microstructure, as shown in Fig. 4, one can identify colonies consisting of regions of well aligned grains that are continuously connected across a region of ~ 20 μm in length. The linkage of the colony structures may provide a continuous percolation pathway for the supercurrent transport in YBCO, which may provide the mechanism for the higher-than-expected critical current density observed in many samples. For example, the 100-A I_c of the sample shown in Figs. 2-4 is much greater than anticipated, considering the YBCO in-plane misorientation of 12°, as determined by x-ray φ scan.

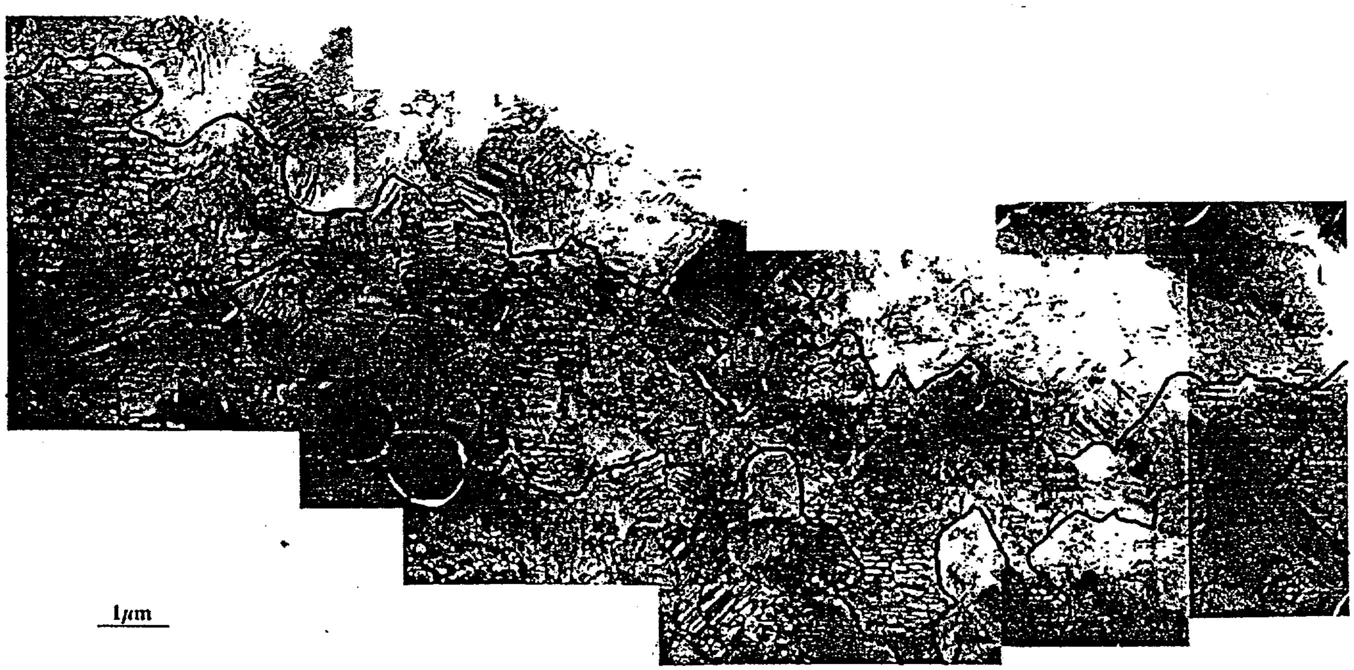

Fig. 4 Low magnification montage view of the colony structures consisting of well-aligned grains. The linkage of the colonies, as shown by the outlined regions, provides a percolation pathway for the transport of supercurrent.

One very intriguing feature as reflected in Figs. 2 and 4 is the fact that only one set of twins was seen to be present in the samples, despite the fact that there are two twin variants: [110] and [1 $\bar{1}$ 0] in the orthorhombic YBCO structure. This is especially prominent when viewed in the low-magnification montage view, where a region of 20 μm in length consists of grains with twins almost completely oriented along the same direction. This could be due to the poor sampling statistics of the TEM technique. However, the intrinsic surface stress, related to the growth geometry, may result in a preference for forming one set of twins over the other. A more in-depth study will be carried out to elucidate the relationship between the residual stress and twin structures in the YBCO coated conductors.

IV. CONSLUSIONS

In summary, the microstructure and grain boundary structure of YBCO thick films deposited on polycrystalline Ni-based metal substrates was characterized by TEM. The films were found to be strongly textured with c-axis oriented grains aligned perpendicular to the substrates. Despite the large average in-plane misorientation (θ ~ 14°), a major portion of the grain boundaries are low angle tilt boundaries with θ in the range of 2° to 7°. As a result, edge type of GBDs with spacing ranging between 11 and 3 nm, and line direction parallel to the c axis are found in these low angle grain boundaries. The presence of the dense array of line defects can serve as effective flux pinners. TEM inspection also reveals colonies consisting of strongly textured submicron-sized grains. The linkage of the colony structures may provide a continuous percolation pathway for the supercurrent transport in YBCO, which is a possible mechanism for the higher than expected critical current density.

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