X-ray microdiffraction study of epitaxial oxide films on textured nickel substrates

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Introduction

Fabrication of high-current superconducting wires using high-T_c oxide materials such as YBCO requires that intrinsic materials limitations such as grain-boundary conductivity and brittle mechanical properties must be overcome. Studies have revealed that high-angle grain boundaries act as weak links, depressing the critical current density by orders of magnitude. Thus, a practical superconducting wire should be both crystallographically aligned and flexible over long lengths.

One approach to achieving these properties involves crystallographic alignment induced by the heteroepitaxial growth of oxide layers on rolling-assisted biaxially textured substrates (RABiTS) [1]. In this approach, nickel foils are rolled and recrystallized to produce [001]<100> cube texture with mosaic spread <10° full width half maximum (FWHM). Epitaxial oxide films are then deposited by pulsed laser ablation or other techniques. Because of chemical interactions and thermal cracking, the YBCO layer cannot be deposited directly on the Ni foil. Instead, suitable epitaxial buffer layers, such as CeO_2 and yttria-stabilized zirconia (YSZ), are typically used. Short (~1 cm) samples with remarkable current densities (J_c > 10^6 A/cm^2) have demonstrated the feasibility of this approach. However, reliably controlling the local microstructure in order to scale up to kilometer lengths is a significant challenge [2]. Understanding the details of processes, such as epitaxial growth on vicinal surfaces and stress cracking under a variety of growth conditions, is essential for the fabrication of practical superconducting wires.

Methods and Materials

Since substrate grains are typically ~50 µm in diameter and x-rays penetrate all epitaxial layers, x-ray microbeam diffraction is ideally suited for studying the local structure, orientation, and strain of each epitaxial layer. Electron techniques [e.g. electron backscatter patterns (EBSP)] do not probe underlying layers and do not provide high-resolution (~10^-3) strain tensor measurements. As described in the activity report by G.E. Ice et al., conventional microbeam diffraction using monochromatic x-rays and sample rotations has limited spatial resolution and data collection speed when considering polycrystalline samples. We are interested in quantitatively studying both vicinal growth on a grain-by-grain basis as well as microstructural variations within individual grains.

We have developed a white-microbeam Laue method based on focused undulator radiation and a CCD area detector. The broad-bandpass x-rays are focused to a beam diameter of ≤1 µm diameter FWHM by a pair of elliptical Kirkpatrick-Baez mirrors [3]. As shown schematically in Figure 1, a complete Laue diffraction pattern is generated from each layer of the sample and is recorded by a 1242 x 1152 pixel CCD detector.

![Figure 1: Schematic diagram of ~1 µm diameter focused, white x-ray beam incident on a multilayer sample.](image1)

Results

Figure 2 shows a typical microdiffraction result consisting of the superposition of white-beam Laue patterns from the different heteroepitaxial layers.

![Figure 2: CCD image showing (001) Laue patterns from each layer of a multilayer RABiTS sample.](image2)
It reveals that the successive layers are not exactly crystallographically aligned. Instead, the [001] poles of the YSZ/CeO$_2$ buffer layers are tilted towards the surface normal when compared with the substrate Ni[001]. Similarly, the YBCO (001) planes are tilted further in the same direction. We find this form of crystallographic tilting to be a general result for RABiTS samples prepared under usual growth conditions.

To obtain quantitative results, the positions of the x-ray peaks were fit and the images were indexed using computer programs capable of handling overlapping Laue patterns [4, 5]. In most samples, the tilt of the epitaxial film away from the substrate increases monotonically with the substrate grain misorientation away from the sample normal. Systematic measurements at different growth temperatures have been obtained for the initial CeO$_2$/Ni interface. Here, the film tilt increases approximately linearly as a function of substrate vicinal angle for samples grown under the usual high-temperature (600°–800° C) conditions. However, the slope is temperature dependent, and deviations from linearity are observed. Further, measurements on a sample grown at a relatively low temperature (450° C) reveal small tilts that are nearly independent of substrate grain orientation.

In addition to orientation information, analysis of the Laue patterns provides the local three-dimensional distortional strain tensor [4, 5]. Results from the heteroepitaxial CeO$_2$ films reveal biaxial strain with an expanded lattice spacing normal to the film (c/a ~ 1.007).

**Discussion**

Examples of crystallographic tilting of heteroepitaxial films on vicinal surfaces have been reported previously for semiconductor and oxide systems [6–9]. Films are often observed to grow tilted with respect to miscut single-crystal substrates. Although many of our observations are in qualitative agreement with a geometrical model based on elastic deformation at surface steps proposed by Nagai [6] we find that the quantitative value for the slope in the linear relation can depend on processing conditions and that deviations from a linear relation can occur. These observations suggest that other mechanisms, such as the preferential insertion of particular misfit dislocations, are needed to understand the process of heteroepitaxial growth in RABiTS samples.

Discovery of these effects has been made possible by the x-ray microbeam. To our knowledge, all previous investigations used large single-crystal substrates; that is, they required that a new sample be grown for each miscut angle. Thus, studies were limited to a small number of observations and were susceptible to errors introduced by changes in processing conditions. The use of the x-ray microbeam and textured substrates eliminates these limitations. Films grown under identical conditions on a large number of grains with a distribution of miscut angles can easily be measured from a single sample. In effect, the x-ray microbeam enables a combinatorial approach with greatly improved statistics to the measurement of heteroepitaxial growth.

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**References**


