# Crystallographic Tilting of Oxide Films on Textured Metal Substrates Investigated Using X-Ray Microbeams

J. D. Budai,<sup>1</sup> W. Yang,<sup>1</sup> B. C. Larson,<sup>1</sup> G. E. Ice,<sup>1</sup> J. Z. Tischler,<sup>1</sup>

D. P. Norton,<sup>1</sup> J.-S. Chung,<sup>1</sup> C. Park,<sup>1</sup> W. Lowe<sup>2</sup>

<sup>1</sup> Oak Ridge National Laboratory, Oak Ridge, TN, U.S.A.

<sup>2</sup> Howard University, Washington, DC, U.S.A.

# Introduction

Although high-temperature superconducting materials have been studied extensively since their discovery in 1986,<sup>1</sup> practical applications have been hampered by difficulties in fabricating these brittle, oxide materials into long, flexible wires capable of carrying large critical currents ( $J_c$ ). In particular, high-angle grain boundaries in polycrystalline superconductors such as YBaCuO act as weak links, depressing  $J_c$  by orders of magnitude. Thus, significant efforts have focused on studying crystallographic alignment in superconducting wires.

One technique for establishing texture in oxide superconductors relies on the epitaxial growth of oxide films on oriented metal substrates.<sup>2</sup> Mechanical deformation and thermal recrystallization can be used to produce a continuous, highly-oriented metal tape. Ni(001) foils with mosaic spread  $\Delta \omega < 10^{\circ}$  are typically used in present prototypes. The subsequent deposition of epitaxial oxide films creates an oriented multilayer architecture known as rolling-assisted biaxially textured substrates (RABiTS).<sup>3</sup>

### **Methods and Materials**

Polychromatic x-ray microbeam diffraction has been used to investigate the crystallographic alignment (texture) and strain in each layer of RABiTS samples. The undulator beam was focussed to ~1 mm diameter using elliptical Kirkpatrick-Baez mirrors at the MHATT-CAT beamline 7-ID-4 Figure 1(a) shows an optical



FIG. 1. (a) Optical micrograph of typical RABiTS sample with ~50  $\mu$ m grain size; (b) Microbeam (001) Laue pattern from ~1  $\mu$ m area of a multilayer RABiTS sample.

micrograph of the grain structure of a RABiTS sample. Since substrate grains are typically ~50 mm in size and x-rays penetrate all layers, white microbeam diffraction simultaneously provides Laue patterns from each layer of the multilayer RABiTS samples. The resulting CCD detector image in Fig. 1(b) shows three superimposed Laue patterns corresponding to the Ni substrate, a CeO<sub>2</sub>/YSZ buffer layer, and a superconducting YBaCuO film. Analysis of these patterns yields the crystallographic orientations and deviatoric strain tensors with micron-spatial resolution.<sup>5</sup> The observation that (001) poles corresponding to different heteroepitaxial layers are not exactly superimposed illustrates the general result that crystallographic tilting occurs during film growth.

## **Results**

X-ray microbeam measurements were used to study the effect of the substrate temperature on the crystallographic orientation and strain of ~500-nm-thick CeO<sub>2</sub> oxide buffer films deposited on highly textured Ni foils by pulsed laser deposition (PLD). Temperature during film growth ranged from 450°C to 800°C. In general, high-temperature film growth (>600°C) showed sharp CeO<sub>2</sub> diffraction peaks and a crystallographic tilting of the oxide (001) poles towards the surface normal. In contrast, low-temperature growth (<500°C) showed more diffuse peaks and almost exact alignment of the (001) poles. These general trends are shown in Fig. 2 where the location of the Ni(001) and CeO<sub>2</sub>(001) poles are indicated for a large number of individual grains in samples grown at 450°C and 600°C.



FIG. 2. Location of Ni(001) and buffer layer  $CeO_2$  (001) poles in CCD detector images at two different growth temperatures.

The 600°C results show that the misorientation angle between the film and the substrate increases with the Ni(001) miscut angle for individual vicinal grains in the substrate. Quantitatively, the high-temperature oxide tilting can be described by a quadratic dependence on substrate miscut angle. The 450°C results show near alignment of the substrate and film (001) poles, with a slight bias (<0.5°) in a single direction.

#### Discussion

Heteroepitaxial growth on miscut substrates involves the deposition of lattice-mismatched material on a surface consisting of terraces separated by atomic ledges. Here, the high-temperature tilting behavior of  $CeO_2$  on Ni(001) can be described by a ledge growth model<sup>7</sup> which incorporates both elastic deformation at steps and interfacial misfit dislocations.<sup>8</sup> Note that the tilting towards the surface normal is desirable in applications since it increases the local texture.

At lower growth temperatures, the diffuse  $CeO_2$  peaks indicate limited film coherence (i.e., defects) at the submicron level.

Surface diffusion is suppressed and the lower kinetics gives rise to growth of aligned CeO<sub>2</sub> islands on the terraces. The small orientation bias in a single direction may be due to an off-axis energetic-ion effect during PLD growth, related to IBAD.<sup>9</sup>

This study of heteroepitaxial growth in RABiTS illustrates the ability of polychromatic x-ray microbeam diffraction to provide local structural information, enabling new classes of micronresolution experiments. Note that, in this study, x-ray microbeams enabled measurements from a large number of different vicinal grains grown under identical conditions on a single sample, i.e., a combinatorial approach.

#### Acknowledgments

Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract DE-AC05-00OR22725 with UT-Battelle, LLC. Experiments were performed on MHATT-CAT which is funded by the U.S. Department of Energy, Basic Energy Sciences.

Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

## References

<sup>1</sup> J.G. Bednorz and K.A. Mueller, Z. Phys. B 64, 189 (1986).

<sup>2</sup> J.D. Budai, R.T. Young, and B.S. Chao, Appl. Phys. Lett. **62**, 1836 (1993).

<sup>3</sup> D.P. Norton, A. Goyal, J.D. Budai, D. Christen, D. Kroeger, E.D. Specht, Q. He, B. Saffian, M. Paranthaman, C. Klabunde, D. Lee, B. Sales, and F. List, Science **274**, 755 (1996).

<sup>4</sup> G.E. Ice, J.S. Chung, J.Z. Tischler, A. Lunt, and L. Assoufid, Rev. Sci. Inst. **71**, 2635 (2000).

<sup>5</sup> G.E. Ice and B.C. Larson, Adv. Eng. Mat. 2, 643 (2000).

- <sup>6</sup> J.S. Chung and G.E. Ice, J. Appl. Phys. 86, 5249-5256 (1999).
- <sup>7</sup> H. Nagai, J. Appl. Phys. 45, 3789-3794 (1974).

<sup>8</sup> J.E. Ayers, S.K. Ghandhi, and L.J. Schowalter, J. Cryst. Growth **113**, 430-440 (1991).

<sup>9</sup> Y. Iijima, N. Tanabe, O. Kohno, and Y. Ikeno, Appl. Phys. Lett. **60**, 769 (1992).