LA-UR- 01-2160

Approved for public release; distribution is unlimited.

Title: Integration of Biaxally Aligned Conducting Oxides with Silicon Using Ion-Beam Assisted Deposited MgO Templates

Author(s): Luke A. Emmert, MST-STC
Bae Ho Park, MST-STC
J. Randy Groves, MST-STC
Raymond F. DePaula, MST-STC
Quanxi Jia, MST-STC
Paul N. Arendt, MST-STC

Submitted to: 2001 MRS Spring Meeting, San Francisco, CA, 4/14-22/01

Los Alamos
NATIONAL LABORATORY

Integration of Biaxially Aligned Conducting Oxides with Silicon using Ion-Beam Assisted Deposited MgO Templates

Luke A. Emmert, Bae-Ho Park, James R. Groves, Raymond F. DePaula, Q.X. Jia, and Paul N. Arendt
Superconductivity Technology Center, Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87544

ABSTRACT

Two conducting oxides, $La_{0.5}Sr_{0.5}CoO_3$(LSCO) and $SrRuO_3$, were deposited by pulsed laser ablation onto silicon substrates coated with biaxially textured MgO on an amorphous silicon nitride isolation layer. Comparison is made between templates using just 10 nm of ion-beam assisted deposited (IBAD) MgO and substrates with an additional 100 nm of homoepitaxial MgO. Both of these conducting oxide layers exhibited in-plane and out-of-plane texture, on the order of that obtained by the underlying MgO. The $SrRuO_3$ was c-axis oriented on both substrates, but exhibited a slightly sharper out-of-plane texture when the homoepitaxial MgO layer was included. On the other hand, the LSCO showed only (100) orientation when deposited directly on the IBAD-MgO templates, whereas a significant (110) peak was observed for films on the homoepitaxial MgO. A simple calculation of the distribution of grain boundary angles, assuming a normal distribution of grains, is also presented.

INTRODUCTION

Integration of oxides with silicon electronics has been a stumbling block for promising applications. One can deposit structures on oxidized silicon, but the resultant polycrystalline films dilute the properties that often depend on crystalline anisotropy. Moreover, the high-angle grain boundaries can adversely affect the properties by changing the defect chemistry. Alternatively, one can grow heteroepitaxial oxides directly on silicon, but thermochemistry limits one's choice of materials[1]. This method is best left to gate oxides where the silicon-oxide interface is crucial for the application.

Ramesh *et al.*[2] demonstrated that one can obtain fiber textured oxide films without using the crystallinity of the substrate by first depositing $Bi_4Ti_3O_{12}$ which has a tendency for uniaxial growth. Ion-beam assisted deposition is a useful way for controlling the texture of thin films during growth. Recently, Wang *et al.*[3] have shown that biaxial texture can be achieved by depositing MgO with ion-beam assistance(IBAD-MgO). We have applied this technique to superconducting tapes, where the elimination of high-angle grain boundaries improves the critical current density[4]. In this paper we demonstrate the growth of other conducting perovskites with biaxial texture in preparation for later studies on the effect of reducing the density of high-angle grain boundaries on electrical properties of other perovskites.

EXPERIMENTAL DETAILS

The IBAD-MgO films were deposited on as-received silicon wafers after coating with 20 nm of amorphous silicon nitride evaporated by electron-beam. MgO was evaporated by electron-beam evaporation while simultaneously exposing the substrate to a flux of argon ions accelerated to 750 eV. The ion current at the substrate was fixed at 100 $\mu A/cm^2$ as read by a nearby Faraday cup. The MgO vapor flux was set so that the ion-to-atom ratio was 0.7. Further details about the optimization of these parameters are provided in a previous publication[3]. The thickness of the IBAD-MgO layers was 10 nm. A second set of substrates was prepared with an additional 100 nm of MgO deposited without the ion beam while the substrate was held at 500°C. These latter substrates are referred to as having a homoepitaxial MgO layer.

The LSCO and $SrRuO_3$ films were deposited in a separate vacuum system using pulsed laser ablation of ceramic targets using a XeCl excimer laser. The laser fluence was 2 J/cm^2. Prior to deposition, the substrates were first annealed at 800°C for 10 minutes at the base pressure (10^{-5} Torr) in order to remove any adsorbed water on the MgO surface. Afterwards, the substrates were set to the deposition temperature – 750°C for LSCO and 775°C for $SrRuO_3$ – and the system pressure was set to 200 mTorr with flow of molecular oxygen. The deposition started with the laser set to a 2Hz pulse rate for 2 minutes followed by 10 minutes at 5 Hz. Afterwards, the substrates were allowed to cool in a background of pure oxygen near 400 Torr.

DISCUSSION

As shown in Figure 1, x-ray diffraction demonstrates that the films grow with a single out-of-plane orientation. The lone exception is the LSCO deposited onto a substrate with the thick homoepitaxial layer of MgO. Our previous experience has been that this additional layer improves the template for subsequent film growth while sharpening the diffraction spots in our *in situ* RHEED system. However, it appears from this evidence that this layer can be a source of problems possibly because it is deposited at such a low temperature compared to typical oxide growth.

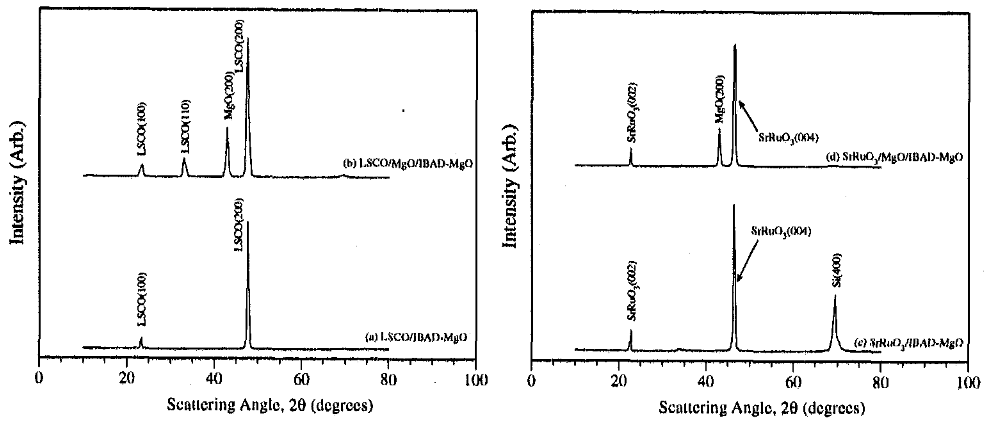

Figure 1: X-ray normal scans of the four cases. These wide angle scans show that in all cases except LSCO on homoepitaxial MgO, the films grow with their [001] axis oriented near the surface normal.

Figure 2 shows rocking curves of the dominant peaks of LSCO, $SrRuO_3$, and MgO. In this case the x-ray source and detector are locked to a fixed angle specified by the peaks in the Figure 1 while the substrate is rocked through the Bragg condition. This shows the distribution of the grains oriented about the normal to the substrate. Both conducting

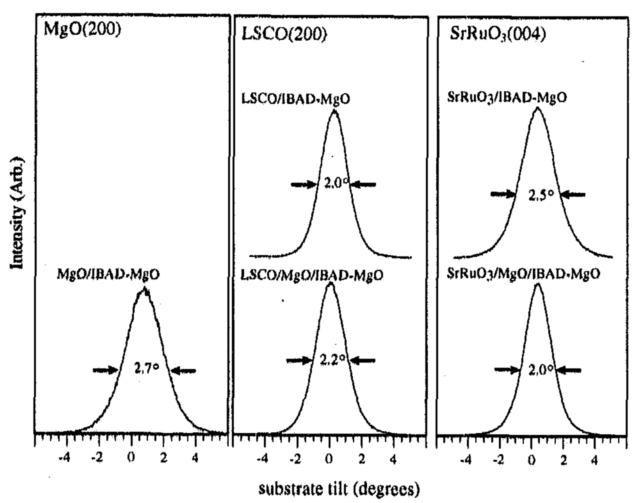

Figure 2: Rocking curves of out-of-plane texture for the films and planes as labeled. There is no rocking curve of the IBAD-MgO alone as there is not enough diffracted intensity from a 10nm film to make a good comparison.

oxides improve upon the full-width at half-maximum(FWHM) for the homoepitaxial MgO.

Finally, the films were characterized for their in-plane mosaic spread by means of a phi scan. In this case the diffractometer is set to reflect off of planes not in the zone axis defined by the film's out-of-plane orientation. In the case of MgO and LSCO, the planes used belonged to the {202} and {101} families, respectively. The [001] axis of the films is a four-fold rotation axis as reflected in the scans (a), (b), and (c) in figure 3.
For the $SrRuO_3$ films, the planes chosen were the {132} family. There are four such planes with non-degenerate orientations which exhibit the 2-fold rotational symmetry of the orthorhombic structure(figure 3d and 3e). There are eight peaks in the trace because the $SrRuO_3$ films can have two domains related by a 90° rotation. The orientations are characterized by $SrRuO_3$[100]||MgO[100] and $SrRuO_3$[100]||MgO[010]. This is the result of putting a two-fold structure on a four-fold substrate

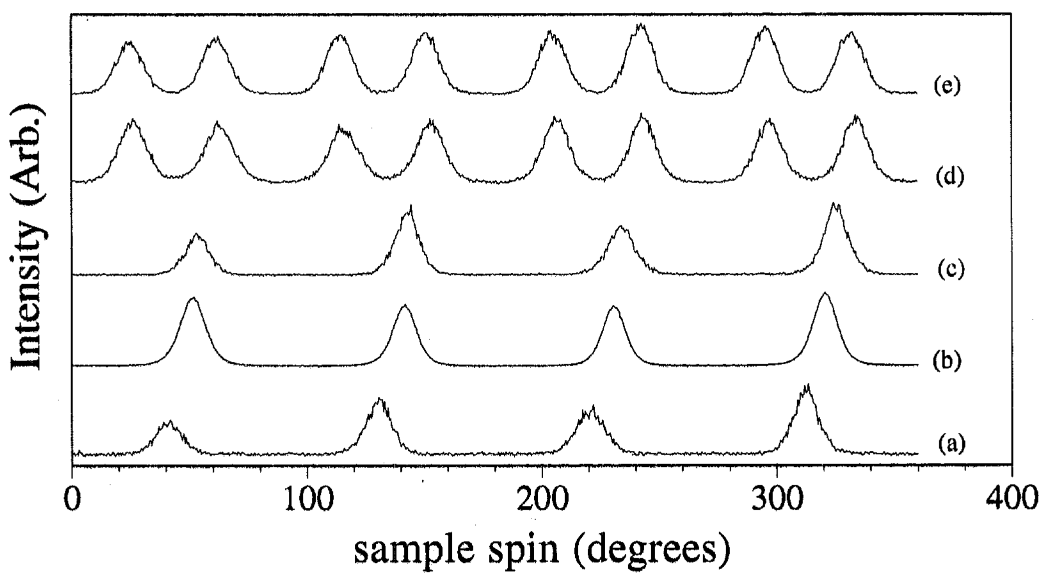

Figure 3: Collected phi scans in the following order: (a) {202} planes of homoepitaxial MgO; (b) {101} planes of LSCO on IBAD-MgO only; (c) {101} planes of LSCO on homoepitaxial MgO; (d) $SrRuO_3$ on IBAD-MgO only; and (e) $SrRuO_3$ on homoepitaxial MgO.

Table 1: Compilation of full-width at half-maximum values.

film	out-of-plane; rocking curve	in-plane phi scan
MgO/IBAD-MgO	2.7°	13°
(La,Sr)CoO_3/MgO/IBAD-MgO	2.2°	13°
(La,Sr)CoO_3/IBAD-MgO	2.0°	13°
$SrRuO_3$/MgO/IBAD-MgO	2.0°	14°
$SrRuO_3$/IBAD-MgO	2.5°	15°

For comparison, the values of the full-width at half-maximum for the rocking curves and phi scans are compiled in Table 1 for all four combinations of perovskite and substrates. The value for the homoepitaxial MgO is also included. One can see that the distribution of grains in the plane of the film is roughly the same for all combinations.

The distribution of grain boundaries is a function of the texture that has been measured. For the grains so nearly aligned as these here, the intergrain misorientation has both a tilt and twist component related to the texture as shown in figure 4. To first order, the magnitude of the grain boundary misalignment is dominated by the in-plane distribution because it is so much larger than the out-of-plane spread. Thus one can describe the grains by a distribution function $n(\phi)$ which gives the fraction of the grains in the range from ϕ to $\phi + d\phi$. We can then define a distribution function for the grain boundaries which is a function of the grain boundary tilt $\Delta\phi$ in similar fashion. This new function is just the convolution of the grain boundary distribution function with itself.

$$n(\Delta\phi) = \int_{-\infty}^{+\infty} n(\phi)n(\phi + \Delta\phi)d\phi$$

The limits are set to infinity to simplify the integration. If we take $n(\phi)$ to be a normal

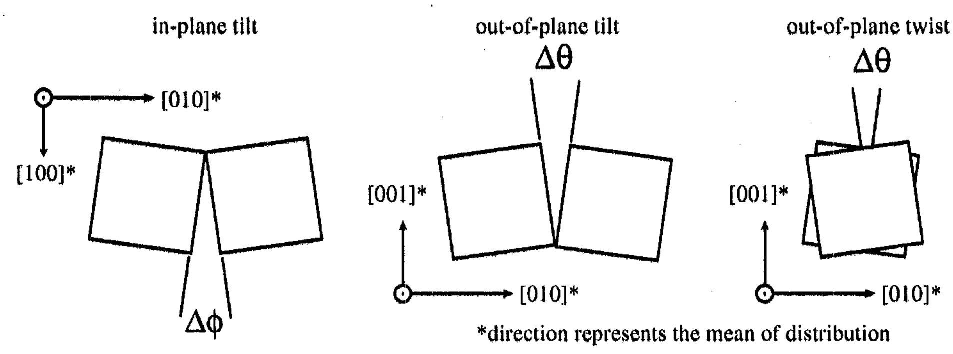

Figure 4: Relationship between grain orientation and grain boundary type. The in-plane difference creates a tilt boundary characterized by $\Delta\phi$. The out-of-plane distribution leads to grain boundaries with both tilt and twist character.

distribution characterized by its standard deviation σ, then the integration is simple and leads to another normal distribution with standard deviation $\sqrt{2}\sigma$. Therefore, under the assumptions of this model, we can expect the grain boundaries to have a distribution 40% broader that those measured in the phi scans. Considering the LSCO films, we can expect that most of the grain boundaries would be below 20°.

CONCLUSIONS

We've demonstrated integration of perovskites with silicon, having both out-of-plane texture below 3° and in-plane texture from 13° to 15°. While a homoepitaxial layer of MgO might provide a small benefit in the $SrRuO_3$ films, the LSCO film deposited on a substrate with homoepitaxial MgO showed a second out-of-plane orientation. Therefore, IBAD-MgO alone can provide an improvement in the texture of oxide films grown onto silicon without requiring any additional heating steps. In addition, the silicon nitride layer isolates the underlying silicon devices from while providing the amorphous starting point for the nucleation of the IBAD-MgO template.

BIBLIOGRAPHY

1. K.J. Hubbard and D.G. Schlom, J. Mater. Res. **11**, 2757-2776 (1996).
2. R. Ramesh, J. Lee, T. Sands, V.G. Keramidas, and O. Auciello, Appl. Phys. Lett. **64**, 2511-2513 (1994).
3. C.P. Wang, K.B. Do, M.R. Beasley, T.H. Geballe, and R.H. Hammond, Appl. Phys. Lett. **71**, 2955-2957 (1997).
4. J.R. Groves, P.N. Arendt, S.R. Foltyn, R.F. DePaula, E.J. Peterson, T.G. Holesinger, J.Y. Coulter, R.W. Springer, C.P. Wang, and R.H. Hammond, Appl. Supercond. **9**, 1964-1966 (1999).