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Author(s):

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Materials Science Division, Los Alamos National Laboratory
Los Alamos, NM, 87545

ABSTRACT

We have studied the growth of magnesium oxide using ion-beam assisted deposition (IBAD) to achieve (100) oriented, bi-axially textured films with low mosaic spread, for film thicknesses of 10 nm on silicon substrates. We have refined the process by using reflected high-energy electron diffraction (RHEED) to monitor the growth of IBAD MgO films and found that the diffracted intensity can be used to determine (and ultimately control) final in-plane texture of the film. Here we present results on our work to develop the use of real-time RHEED monitoring to deposit well-oriented IBAD MgO films. The results have been corroborated with extensive grazing-incidence X-ray diffraction (GID). Results of these analyses have allowed us to deposit films on metallic substrates with in-plane mosaic spread less than 7°.

INTRODUCTION

There is great interest in the high temperature superconductor (HTS) coated conductor community to develop economically scalable processes for fabricating bi-axially textured templates on which high quality $YBa_2Cu_3O_{7-\delta}$ (YBCO) can be heteroepitaxially deposited. In order to achieve good superconducting properties, YBCO grains require good alignment between each other to achieve high (>1 MA/cm^2) critical current densities (J_c)[1]. The two competitive processes to produce the bi-axial texture required by YBCO have been Roll-Assisted BI-axial Texturing of Substrates (RABiTS)[2] and Ion-Beam Assisted Deposition (IBAD)[3]. Our group has focused on the latter technique and expended much effort to deposit yttria-stabilized zirconia (YSZ) with IBAD for coating lengths on technically important metal substrates. This effort has resulted in the development of a process, coupled with pulsed laser deposited (PLD) YBCO, that has produced meter lengths of second generation superconducting wire with critical current densities over 1 MA/cm^2 and critical currents over 100 A[4].

One of the criticisms of the IBAD YSZ process has been that the length of time required to deposit the material with sufficient in-plane texture for high quality YBCO is too long. In order to develop texture, YSZ requires a thickness between 0.5 and 1 μm to achieve a $\Delta\phi$ (or full-width at half-maximum of the ϕ-scan peak) better than 12°. This translates to an IBAD deposition time of ~20 hours per meter of tape[5]. The viability of this process is questionable for cost effective, industrial fabrication.

Wang et al. showed that magnesium oxide (MgO) could be deposited with the IBAD process and produce a film with in-plane texture comparable to YSZ that was only 10 nm[6]. This translates to a process which is ~100 times faster than IBAD YSZ. We have continued to develop this process for use with HTS coated conductors[7]. To date, we have produced short length samples (< 4 cm long) using IBAD MgO templates that have achieved J_c's over 1 MA/cm^2 and have the potential for use in our meter length coating efforts[8].

For this type of rapid processing, it is necessary to determine the in-plane texture in real time. Reflected high-energy electron diffraction (RHEED) has been used to gather qualitative real-time data during IBAD MgO deposition. A method has been proposed to use the RHEED data as a process monitor to control the final in-plane texture of IBAD MgO[9]. In this method, the spot intensity versus time (I vs. t curve) is monitored as the film is grown. This paper will address the use of this data as a potential real-time process monitoring technique during the growth of IBAD MgO.

EXPERIMENTAL DETAILS

The substrates used were as-received polished silicon wafers. The native oxide was not removed. A 5 nm thick amorphous silicon nitride layer was deposited on the silicon surface using electron beam deposition with a stoichiometric source. A background partial pressure of nitrogen (6.7×10^{-3}Pa) was introduced into the deposition chamber during the silicon nitride coating process. A subsequent layer of MgO was deposited on the amorphous SiN_x layer using IBAD. Argon ions were accelerated to 750 eV with a current density of 100 mA/cm^2 using a 22 cm × 2.5 cm Kaufman ion source. The ion source incidence angle is at 45° relative to the substrate which corresponds to the MgO <110>. Concurrently, an electron beam evaporator provided the MgO vapor flux of 0.15 nm/s during the IBAD growth. The ion to atom ratio was kept constant at 0.7, which reduced the effective deposition rate by 30% to 0.1 nm/s due to resputtering. The vapor flux and the ion fluence were monitored with a quartz crystal microbalance (QCM) and a Faraday cup, respectively. All IBAD depositions were performed at room temperature. The process is described in greater detail elsewhere[7]. Silicon substrates were chosen as they provide a consistently smooth surface for deposition and would deconvolute any surface roughness effects.

IBAD film growth was monitored in situ using Reflected High-Energy Electron Diffraction (RHEED) by collecting a spot intensity versus time (I vs. t) curve that used the reflections corresponding to the (002) and (022) planes. Images were captured using kSA400 software (k-Space Associates, Ann Arbor Michigan). All patterns were taken at a beam energy of 30 keV.

The IBAD MgO-coated silicon substrates were then heated to approximately 500°C and coated by e-beam evaporation with an additional homoepitaxial layer of MgO (~100 nm). The additional layer was necessary to provide sufficient MgO for standard X-ray diffraction (XRD) of the samples to characterize their in-plane texture. The films were then characterized using XRD to determine in-plane mosaic spread (ϕ-scan).

DISCUSSION

The RHEED spot intensity has been collected as a function of time for a growing IBAD MgO film. In figure 1, progressive snapshots have been assembled from 0 seconds to 75 seconds during the deposition. From 0 seconds (figure 1a) to approximately 30 seconds no spot pattern is observed; only the amorphous background pattern from the substrate is seen. At 35 seconds (figure 1b), a faint spot pattern begins to appear. A distinct pattern is observed and the intensity begins to increase significantly at 40 seconds (figure 1c). The maximum intensity value is

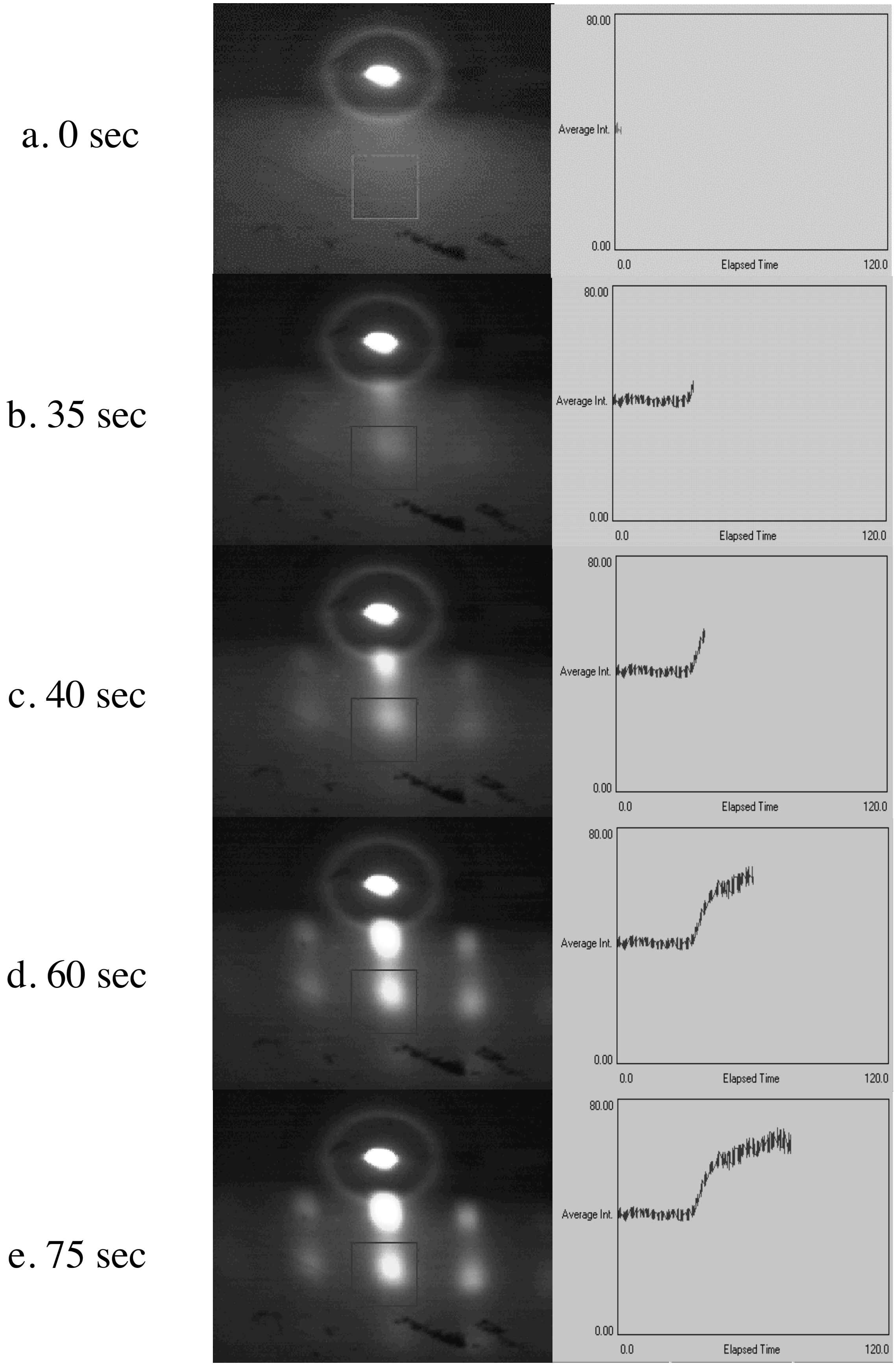

Figure 1. Snapshots of IBAD MgO at sequential times during deposition using RHEED pattern imaging and spot intensity plotting functions.

achieved at 60 seconds (figure 1d). As the deposition is allowed to progress beyond this maximum, a decrease in the spot intensity is observed in the final snapshot at 75 seconds (figure 1e).

Further investigation into this intensity profile during the growth of IBAD MgO has led to some interesting observations. A series of films were deposited at thicknesses that corresponded to particular times along the I vs. t curve. This was done to determine any correlation between the I vs. t curve and the in-plane texture. The films were overcoated with a homoepitaxial layer of MgO and then analyzed using XRD ϕ-scan. Although there is some inherent difference between the IBAD film in-plane texture and that of an overcoated IBAD film, we have shown, in a previous work, that the trend from sample to sample is preserved[10]. The XRD Δϕ (ϕ-scan full-width at half-maximum) results are plotted along with the I vs. t curve at the corresponding times that their individual depositions were stopped in Figure 2. At successive points along the curve the films' Δϕ measured from 15° at the onset of growth to a minimum value of 8° at the spot intensity maximum and then increased to 18° at ~15 seconds past the time at which the maximum spot intensity was observed. From the results of these experiments, it appears that the best texture is achieved at this maximum intensity and that the in-plane texture drastically degrades if the IBAD MgO film is allowed to progress past this maximum intensity value. A thickness of 10 nm has been found to correspond to this maximum intensity.

If we allow the films to grow beyond the maximum in the I vs. t curve, we find that some other surprising effects occur. In the present system configuration, the RHEED beam is parallel to the ion source. If, after the deposition is completed, the samples are rotated 90° with respect to the ion beam direction, the spot pattern tilted away from the ion source as the film continued to grow beyond 10 nm. The tilting progresses from ~0° at 10 nm to ~5° at 20 nm and to 12° at 30 nm. The effect was first observed by Wang and repeated by our group[11]. Figure 3 shows results of tilting of these (100) planes that were observed at both Stanford and here at LANL. As one continues to deposit IBAD MgO beyond 30 nm, the tilt continues to increase until it appears to approach an asymptotic value of ~30° near a thickness of 100 nm. We hypothesized that as the film grows beyond 10 nm and the planes begin to tilt away from the ion beam, the ions do

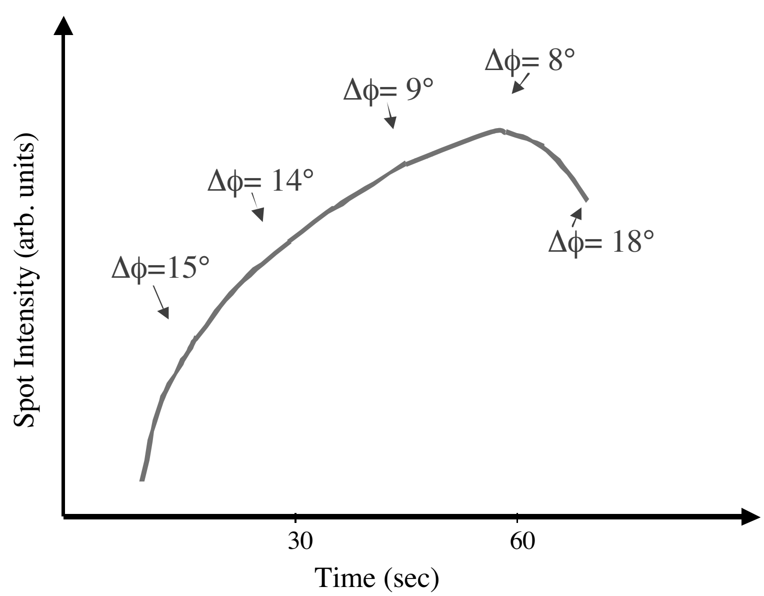

Figure 2. Graphical representation of RHEED spot intensity as a function of time. Included are XRD Δϕ values for films with IBAD MgO thicknesses which correspond to depositions stopped at times along the curve as shown by arrows.

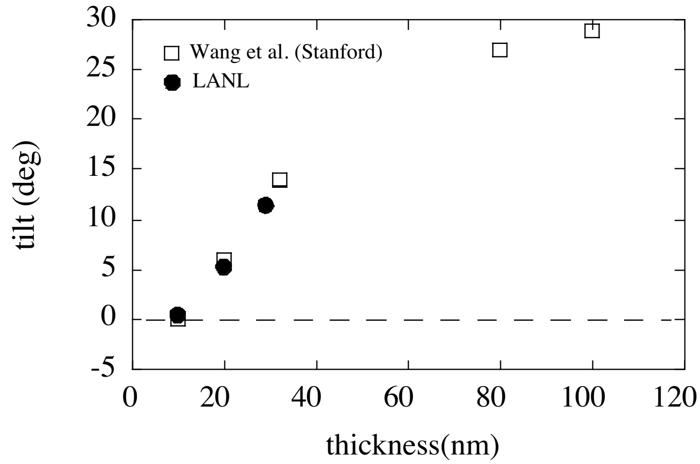

Figure 3. Plot of MgO (100) plane tilt as a function of thickness.

not channel as well along the <110> direction and induce a greater amount of damage in the surrounding film thereby increasing the misorientation between grains.

If we allow the films to grow beyond the optimum thickness of 100 nm, the film orientation begins to tilt away from the ion beam as observed by Wang[11]. That is, the MgO initially grows with the [001] axis nominally parallel to the substrate normal. Eventually, the crystal axis tilts away from the ion beam in the plane defined by this axis and the ion beam itself. The tilting is observed after deposition by rotating the sample 90° and observing the RHEED pattern. It should be unchanged, as the (001) surface has four-fold symmetry, but instead the spots appear tilted to the left or right depending on which direction the substrate was rotated. We have confirmed the Stanford results for the initial tilting as shown in figure 3. At first, the growing surface tilts rapidly past the first 10 nm and eventually approaches a rotation of about 30° at 100nm. We hypothesize that as the film tilts out of the channeling condition the ions produce more damage in the film that is reflected in the worsening in-plane texture.

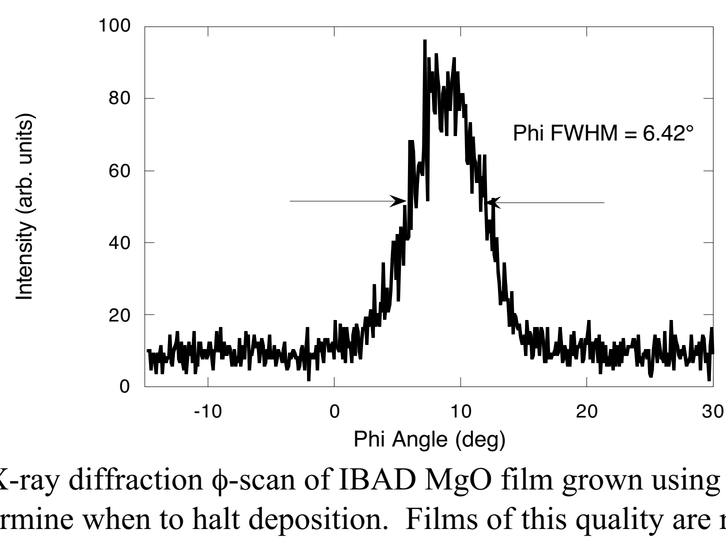

Figure 4. X-ray diffraction ϕ-scan of IBAD MgO film grown using its RHEED I vs. t curve to determine when to halt deposition. Films of this quality are routinely deposited using this technique.

CONCLUSION

We have demonstrated that the RHEED I vs. t curve can be used as an effective in situ process-monitoring scheme for IBAD MgO deposition. We observed that the I vs. t curve reaches a maximum spot intensity and then decreases off sharply. Subsequent XRD analysis of films grown at various thicknesses has shown that the minimum $\Delta\phi$ value corresponds to the maximum intensity observed in the curve and to an IBAD MgO thickness of ~10 nm. As film thickness is increased beyond 10 nm, the films' channeling planes begin to tilt away from the ion beam impingement direction. We hypothesize that as the film grows beyond 10 nm and the planes begin to tilt away from the ion beam, the ions do not channel as well along the <110> direction and induce a greater amount of damage in the surrounding film thereby increasing the misorientation between grains. Use of in situ RHEED monitoring has allowed for optimization of the IBAD MgO process and has resulted in our ability to routinely produce IBAD MgO films with $\Delta\phi$ values between 6° and 8°.

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