

Deposition of in-plane textured MgO on amorphous Si₃N₄ substrates by ion-beam-assisted deposition and comparisons with ion-beam-assisted deposited yttria-stabilized-zirconia

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We report the growth of in-plane textured (100) MgO on amorphous Si₃N₄ substrates by ion beam assisted deposition (IBAD). The textured MgO can be used as a structural template for subsequent epitaxial thin film deposition. The results are compared with IBAD of (100) and (111) yttria-stabilized-zirconia (YSZ). Based on transmission electron microscopy (TEM) and *in situ* reflection high energy-electron diffraction (RHEED), we find that MgO texturing is a nucleation-controlled process and the alignment is a function of nuclei size and density. This differs greatly from the evolutionary-type texturing process observed for IBAD (100) YSZ. Consequently, we are able to make 100 Å thick MgO films with 7° in-plane alignment, whereas IBAD (100) YSZ films need to be thicker than 5000 Å to achieve in-plane alignment better than 13°. This has important implications for the economical application of IBAD induced alignment in real manufacturing processes, including high *T_c* superconductor (i.e., YBCO) coated tapes, photovoltaics, magnetic thin films, and semiconductor devices. © 1997 American Institute of Physics. [S0003-6951(97)03346-9]

Ion beam assisted deposition (IBAD) has shown promise as a mean to produce textured films on nonepitaxial substrates.^{1–3} These textured films can then be used as structural templates for the deposition of other films, thus eliminating the requirement of single crystal substrates. The template material most commonly used in IBAD is (100)-oriented yttria-stabilized-zirconia (YSZ).^{4–8} However, the texturing of (100) YSZ evolves slowly, a thick film (>0.5 μm) is required to achieve good in-plane alignment (~13°).^{6,7} This limits the application of IBAD YSZ due to high processing time and cost.⁹ This letter reports a study of IBAD MgO deposited on amorphous Si₃N₄ substrates in which we have obtained 7° in-plane alignment in films as thin as 100 Å. The texture originates at the nucleation stage, which is similar to the result of the IBAD (111) YSZ, but differs greatly from that of (100) YSZ. Combining the results, we suggest that the preferred out-of-plane alignment contributes to the differences.

The MgO films were deposited using an *e*-beam evaporation system. A Kaufman 3 cm ion source was used to bombard the surface during deposition. The diffraction pattern of the film surface was monitored in real time using reflection high energy electron diffraction (RHEED). A Faraday cup and a quartz crystal monitor were used to measure the ion flux and the evaporation rate of MgO, respectively. In this work, a 700 eV Ar⁺ ion energy was used with fluxes ranging from 110–120 μA/cm². The angle between the ion beam and the substrate normal was 45°, which is an open or channel direction for (100) MgO. The evaporation rate of MgO was selected by monitoring the appearance of the MgO diffraction patterns in RHEED. In-plane textured MgO films have been obtained from room temperature (RT) to 300 °C. For a RT deposition, the best alignment was obtained with a 1.5 Å/s evaporation rate.

Figure 1 shows a typical texturing sequence observed with the *in situ* RHEED. When the proper deposition conditions are set, the RHEED pattern changes from a diffuse-scattering pattern (from the amorphous Si₃N₄ substrate) to a crystalline pattern of MgO at about 20 Å [Fig. 1(a)], which is the limiting diffraction thickness of the RHEED. The pattern suggests that the MgO molecules form well-aligned small 3D crystallites right at the inception of the deposition, i.e., right at the nucleation stage, with (100) planes aligning out-of-plane and the [110] direction facing the ion source. The pattern gradually improves [Fig. 1(b)], and is optimized at about 100 Å [Fig. 1(c)]. At the optimal texture, the ion beam

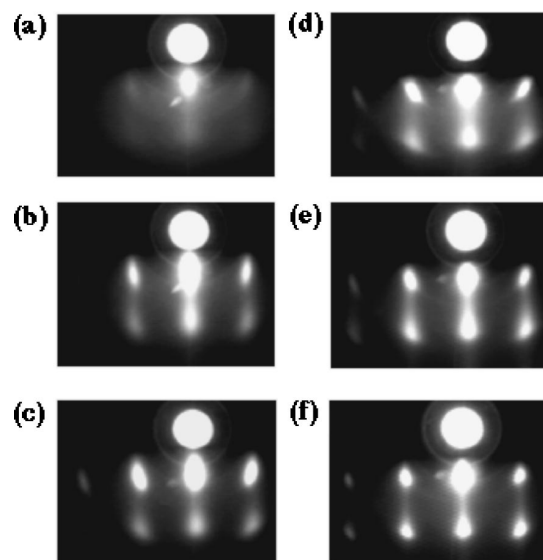


FIG. 1. Real time RHEED observation of an IBAD (deposited at room temperature) and homo-epi MgO film (deposited at 600 °C), in which (a) to (c) are IBAD MgO at 20, 50, and 100 Å, respectively, and (d) to (f) are homo-epi MgO at 100, 400, and 800 Å, respectively.

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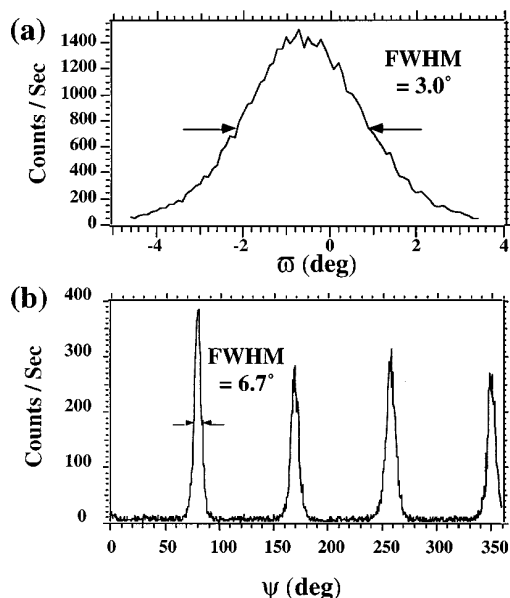


FIG. 2. X-ray diffraction of the MgO film shown in Fig. 1 (100 Å IBAD and 800 Å homo-epi film): (a) ω -scan of the MgO (200) peak and (b) ϕ -scan of the MgO (220) peak.

is turned off, the substrate temperature is raised to 600°C and subsequently an 800 Å of homo-epi MgO is deposited so that the film is thick enough for x-ray diffraction. Based on the RHEED, no obvious additional features are introduced during the homo-epi process [Figs. 1(d)–1(f)]. X-ray diffraction of the sample (shown in Fig. 2) indicates a 3° out-of-plane alignment full width at half-maximum (FWHM) of the ω -scan and a 7° in-plane alignment (FWHM of the ϕ -scan).

The cross-sectional TEM (X-TEM) image in Fig. 3 reveals the texture development of the IBAD MgO pictorially, suggesting that the MgO growth is pseudocolumnar with each column consisting of small 3D crystallites, and that the alignment of these crystallites starts at the MgO/SiN inter-

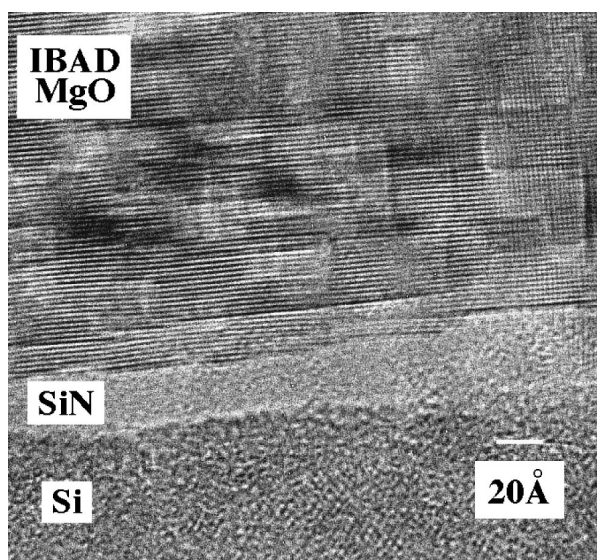


FIG. 3. X-TEM of a IBAD MgO (deposited at 300°C), in which the ion beam was incident at roughly 45° with respect to the normals of the substrate and image planes.

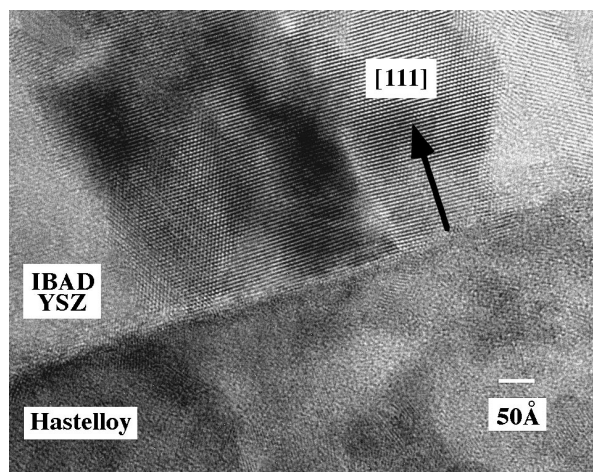


FIG. 4. X-TEM of an in-plane textured (111) YSZ deposited at 600°C by IBAD, in which the ion beam was incident roughly from a 55° angle with respect to the normals of the substrate and image planes.

face. Combining the observations from both TEM and RHEED, a consistent picture emerges in which the MgO texture develops at the beginning of the growth, i.e., the nucleation stage. The optimal thickness of the growth probably corresponds to the point at which the surface of the substrate is fully covered by textured MgO crystallites. After that, the texture improves only slightly and eventually starts to degrade. We also observed that both the alignment and coverage of the textured area improved at a lower substrate temperature, suggesting that the texturing process is facilitated by the denser and smaller nuclei obtained at lower temperature. This is consistent with the nucleation-texturing mechanism suggested for MgO.

The MgO texturing mechanism and alignment results are very different from those of IBAD (100) YSZ. One of the major differences between the two materials is the preferred out-of-plane alignment. At high temperature, the YSZ tends to grow with a (111) fiber texture (i.e., the (111) planes aligned out-of-plane while randomly oriented in-plane), and this out-of-plane alignment cannot be changed by the ion beam.^{10,11} Thus, the best chance to obtain (100) YSZ growth is at low temperature. At low temperature, however, the YSZ molecules lack mobility, resulting in kinetically limited growth. Without IBAD, either a poorly crystallized film results^{6,10} or several out-of-plane alignments coexist.¹¹ With IBAD, the ratio of the different out-of-plane orientations depends on the ion energy, deposition methods, etc., and favors (111) when sufficient molecular mobility is obtained,¹¹ resulting in a complicated texturing process. Conversely, the (100) direction is the preferred out-of-plane alignment for MgO (a rock-salt structure). Thus, the ion beam only needs to align the in-plane orientation, simplifying the texturing process and allowing the texture to appear at an early stage.

We have deposited in-plane textured (111) YSZ at 600°C by IBAD.¹⁰ The X-TEM image of the film is shown in Fig. 4 which clearly illustrates the columnar growth. Based on the high resolution TEM image and Fourier transform analysis of the image, the texture originates at an early stage in some of the columns and several of the misaligned

columns are observed to be shadowed and outgrown by the aligned columns.

Similar film structure has been reported for IBAD (100) YSZ^{12,13} with an additional polycrystalline region near the interface. The poly-region, is a result of the polycrystalline nature of low-temperature deposited YSZ. It is observed that the best alignment of YSZ is achieved when the angle of the ion beam to the normal of a (100) film is 55°. In the poly-region, however, the relative incident angle of the ions to the grains is random so poor alignment results. It has been suggested that the ion beam induces the aligned grains and to overgrow the misaligned grains and the texture develops gradually through a prolonged competition.^{6,13}

Together, the results on IBAD (100) MgO, (111) YSZ, and (100) YSZ suggest that when the film has only one type of out-of-plane alignment, the texturing process is simple and alignment starts from the beginning of the growth. However, YSZ films show a remarkable tendency to align to the ion beam.¹² Thus, the YSZ is able to turn into a textured film gradually even when the initial layer is poorly textured. This tendency makes YSZ more tolerant to the substrate condition. In contrast, the texture of IBAD MgO is totally determined at the initial stage, where the initial texture, substrate condition, and film/substrate interaction become important factors.

In summary, we have made well in-plane textured MgO films on amorphous Si₃N₄ substrates, which are promising structural templates for the deposition of many materials without the need for single crystal substrates. This has important economic implications for a number of potential products. The texturing process for MgO is a nucleation process and the alignment is a function of nuclei size and density. Hence it is very different from the evolutionary-

texturing process for IBAD (100) YSZ but similar to (111) YSZ. The dissimilar behaviors in the out-of-plane alignment of the two materials contribute to this difference.

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