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ScienceDirect
Journal of the European Ceramic Society 30 (2010) 481–484
www.elsevier.com/locate/jeurceramsoc

Characterization of the biaxial textures of MgO thin films grown by E-beam evaporation

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Available online 8 July 2009

Abstract

The biaxial textures of MgO thin films with thicknesses 4500–5500 Å deposited by electron beam evaporation on glass substrates have been characterized. The surface normal to the amorphous soda-lime glass substrate was placed parallel to the MgO vapor flux, i.e., at zero degree with respect to the MgO vapor source. The MgO thin films showed biaxial texture regardless of the deposition parameters. XRD and SEM have been used to characterize the crystal structure and thin film surface morphology. In this study we devised a novel method for the characterization of the biaxial texture of MgO thin films with thicknesses ≤5000 Å, for which the X-ray pole figure method cannot be used due to the low scattering intensity from the MgO film containing only low atomic X-ray scattering ions. We report the biaxial texture development in MgO thin films grown by E-beam evaporation on the amorphous glass substrate inclined at zero degree with respect to the MgO vapor source.

Keywords: Films; X-ray methods; Non-destructive evaluation; MgO; Functional applications

1. Introduction

MgO thin films have been widely used as the dielectric layer in PDPs (plasma display panels), functioning as the protecting and secondary electron emission layer,[1,2] as well as the buffer layers for the epitaxial growth of thin films, such as superconducting YBCO and ferroelectric perovskites on silicon and metal substrates.[3–6] In order to use MgO films to prepare buffer layers and the PDP dielectric, the crystallographic orientation, surface morphology, and stoichiometry of the film are very important factors. For instance the buffer layer application requires the film to be highly oriented in the [1 0 0] direction. As the dielectric protective layer, the preferred orientations of MgO thin films have been reported to vary from (1 0 0) to (1 1 1). In some cases (2 2 0) has also been reported, depending on the film deposition methods and conditions such as deposition rate, atmospheric gas, temperature, film thickness and the type of substrate.[1,7,8]

Normally, polycrystalline thin films show some fiber texture, in which a certain crystallographic direction is aligned with the surface normal to the substrate (out-of-plane texture). However, in these polycrystalline films the in-plane texture is nearly random.[6] For growing epitaxial films of YBCO, ferroelectrics, and Co_3O_4, the biaxially textured MgO buffer layer must be laid on the substrate before the thin film deposition. Several methods[9–12] such as ion beam assisted deposition (IBAD), rolling-assisted biaxially textured substrates (RABiTS),[1,6] inclined-substrate deposition (ISD)[7,8] and pulsed laser deposition (PLD)[5,9] had been applied for the biaxial growth of MgO thin films, e.g., (1 0 0)-oriented MgO on Si(1 0 0) substrates using a Mg metal target[5] and (1 1 1)-textured MgO on Si(1 0 0) substrates using a MgO target.[9]

For the analysis of the preferred orientation of MgO thin films, XRD and TEM have been used. Most of the previous reports determined the degree of the preferred orientation simply by comparing the relative peak intensities of (1 1 1), (2 0 0), (2 2 0) and (2 2 2) in the XRD patterns of the MgO films. Comparing the relative intensities of the diffraction peaks in the XRD patterns only reveals the fibrous texture of the MgO grains grown on the substrates. When the MgO thin films have some in-plane texture, the previous texture analysis method, carried out by simply comparing the peak intensities, results in a fundamentally erroneous conclusion on the degree of preferred orientation. Actually in many previous reports the preferred orientation of the MgO films varies from the stated (2 0 0), (1 1 1) or (2 2 0) and

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doi:10.1016/j.jeurceramsoc.2009.06.015

the crystallinity also varies with minor changes of preparation conditions.[5,7,9,12]

In this study we report a novel method for determining the in-plane as well as the out-of-plane texture of MgO thin films using XRD diffraction. By this method the in-plane and out-of-plane preferred orientations can be clearly identified. The conventional X-ray pole figure method could not be applied for our sample due to the low X-ray diffraction intensities from the MgO thin film, consisting as it does of low atomic X-ray scattering factor ions and a relatively small film thickness of <6000 Å. The X-ray pole figure measurement has been reported only for MgO thin films with a thickness of about 20,000 Å[3] and 25,000 Å,[7] which are thick enough to significantly scatter the X-rays.

We also report that MgO thin films with biaxial texture can be grown by E-beam deposition on a soda-lime glass substrate placed vertically with respect to the MgO vapor flux, i.e., at zero degree of inclination of the substrate with respect to the MgO vapor source. In a previous report it was reported that MgO thin films grown at normal incidence (substrate inclination angle $\alpha = 0$) have only an out-of-plane texture, i.e., fiber texture but not in-plane texture.[7,8] Only MgO films with thickness >10,000 Å have been reported to show a biaxial texture.[7,8]

2. Experimental

Soda-lime glass substrates (2.8-mm thick), commercially available for PDP panel applications, were used for MgO thin film deposition by an electron beam evaporator. The glass substrate was washed with DI water and IPA (isopropyl alcohol) before deposition. The MgO thin film deposition conditions were $\sim 3 \times 10^{-6}$ Torr vacuum, temperature 100–180 °C, evaporation source was a polycrystalline MgO pellet, and the deposition rate was 2–6 Å/s. The soda-lime glass substrates were placed vertically in the MgO vapor flux, i.e., at zero degree with respect to the MgO vapor source. The resulting MgO film thickness was 4500–5500 Å.

The surface morphology of the MgO thin films was characterized using FE-SEM (field emission scanning electron microscopy). For the biaxial texture analysis we used an X-ray diffractometer (Cu-Kα target) fitted with a thin film measurement goniometer allowing the sample to rotate in-plane. Generally, during the XRD measurement, the thin film sample inside the sample holder either remains stationary or rotates in-plane. However, when the MgO thin film sample has some degree of in-plane texture, neither of these two scans can reveal the true texture. Consequently, the analysis results can be false and misleading.

Therefore, in this study, in-plane texture was analyzed as follows. At first, the sample is placed on the thin film goniometer and 2θ-scanned with a fixed incident angle (θ). After completion of the 2θ-scanning over the desired angle range (35–65°), the sample in the goniometer was rotated by 22.5° and the 2θ-scan repeated over the same angle range. By repeating this procedures 16 times, a full 360° rotation for the sample can be achieved. Actually the 360° rotation of the sample inside the holder corresponds to a phi (ϕ) scan in the X-ray pole figure method. This analysis procedure is schematically represented in Section 3.

3. Results and discussions

The surface morphology of the MgO thin films deposited at 150 °C and 100 °C are shown in Figs. 1 and 2, respectively. The MgO grain size is larger in the thin film grown at higher temperature (150 °C) than at 100 °C. The characteristic feature of the MgO grain shape is the appearance of triangular facets at the grain corners, which are aligned along a diagonal

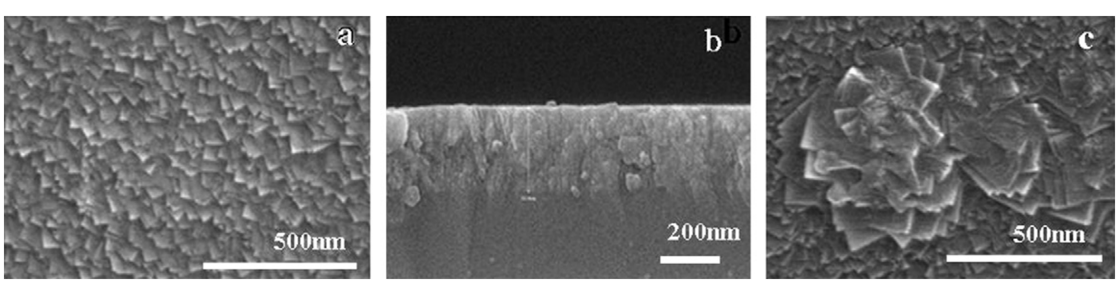

Fig. 1. FE-SEM images of MgO thin film surfaces deposited at 150 °C on a glass substrate.

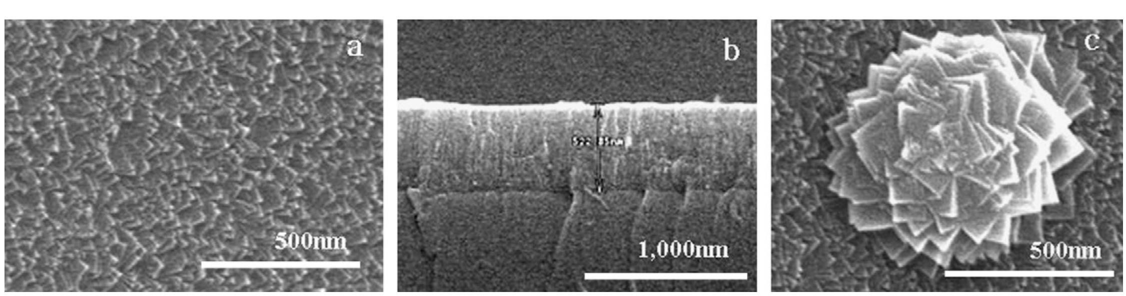

Fig. 2. FE-SEM images of MgO thin film surfaces deposited at 100 °C on a glass substrate.

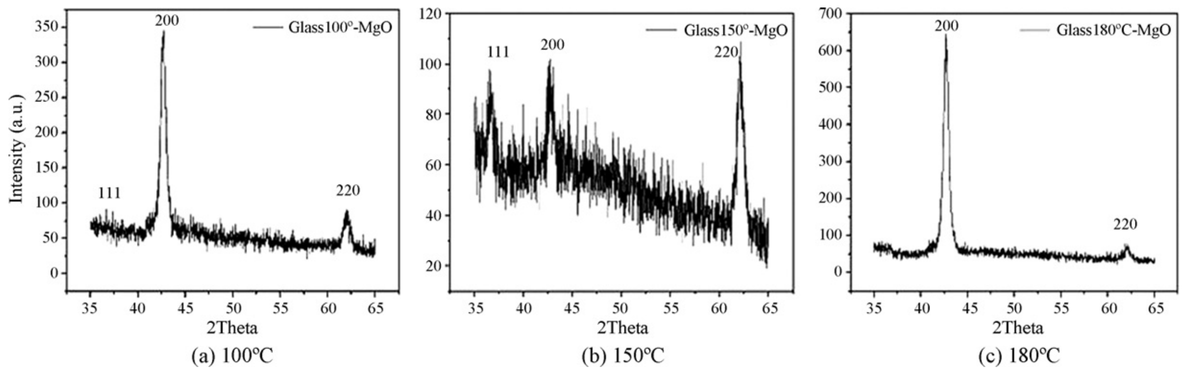

Fig. 3. XRD patterns measured by a conventional 2θ-scan with a fixed incident angle (θ) for MgO thin films deposited at (a) 100 °C, (b) 150 °C and (c) 180 °C on the glass substrate.

direction in both figures. The alignment of the triangular facets along a particular direction has also been observed in the biaxial MgO films grown using the inclined-substrate deposition (ISD) method.[7,8] Figs. 1(b) and 2(b) show cross-sectional images of the same samples, in which columnar alignment of the grains is observed. Figs. 1(c) and 2(c) show some of the abnormal MgO grain islands having a flower petal shape about 6000 Å diameter on the surface of the MgO thin films. In this floral island the triangular facets are clearly delineated and lie in the plane of the substrate. The corners of the triangular facets are directed towards the radial directions in the plane, which implies that the MgO crystal grains are aligned parallel to the substrate (out-of-plane). Hence our preliminary conclusion from the SEM images in Figs. 1 and 2 is that the MgO thin film has considerable out-of-plane and in-plane texture.

XRD patterns were measured by a conventional 2θ-scan with 2θ = 35–65 with a fixed incident angle (θ) for MgO thin films deposited at 100 °C, 150 °C and 180 °C and are shown in Fig. 3. The reflection peaks observed in the samples are (1 1 1), (2 0 0), and (2 2 0). The MgO thin films deposited at 100 °C and 180 °C appear to have a highly oriented grain along the (2 0 0) direction. However, the thin film deposited at 150 °C appears to be poorly crystalline and has little preferred orientation, the diffraction intensities being similar in the (1 1 1), (2 0 0), and (2 2 0) directions, in contrast to the highly oriented alignment of the MgO grains observed in Fig. 1. Furthermore, when the same sample was repeatedly measured by XRD, different patterns were produced in respect to the relative diffraction intensities and crystallinity, data not shown in this paper. We ascribe this inconsistency as due to in-plane texture. X-ray pole figure measurements had been attempted for the MgO thin film samples but were not successful; the pole figure pattern could not be obtained due to the low diffraction intensity from the sample, e.g. the (2 0 0) reflection could not be located.

Instead of X-ray pole figure measurements we devised a novel method as described in Section 2 for the in-plane texture measurement. Fig. 4 shows the total of 16 XRD-patterns for the MgO sample deposited at 180 °C, obtained using the method described in Section 2. Each of the 16 patterns in Fig. 4 shows a conventional 2θ-scan (2θ range = 35–65°) with a fixed incident angle. The 16 scans were obtained from one sample by rotating the sample inside the holder by 22.5° 16 times, thus rotating the sample from 0° to 337.5° inside the sample holder.

The 16 patterns clearly indicate that the MgO thin film has in-plane texture, in which the [2 0 0] direction is aligned to a substantial degree along a particular direction on the substrate. The in-plane alignment of the triangular facets of the MgO

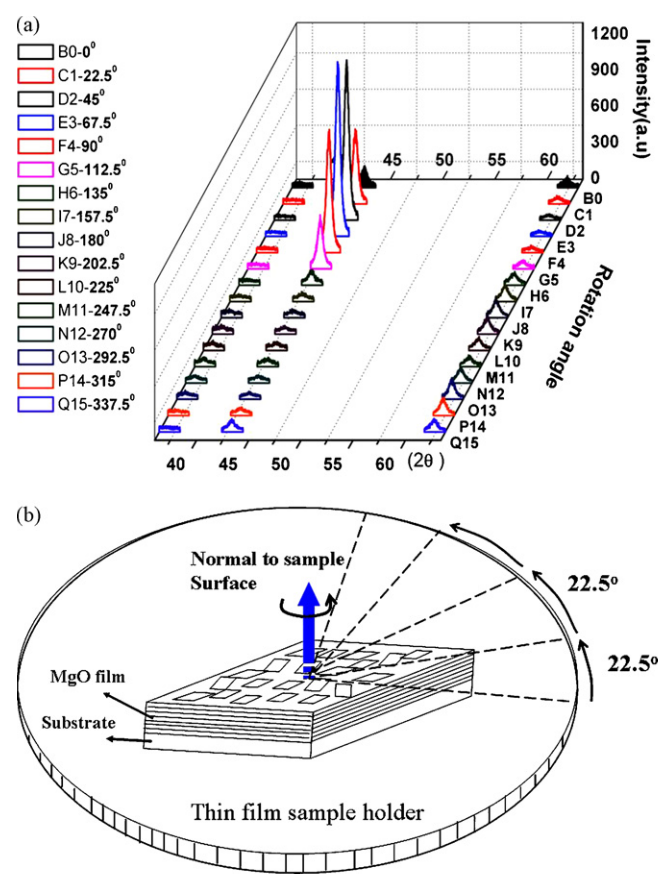

Fig. 4. (a) XRD patterns measured by a conventional 2θ-scan (2θ range = 35–65°) with a fixed incident angle on the resulting MgO thin film deposited at 180 °C on the glass substrate. For one sample a total of 16 scans were measured, between each of which the sample was rotated by 22.5° from 0° to 337.5° inside the sample holder. (b) Schematic representation for the biaxial texture analysis procedure described in (a).

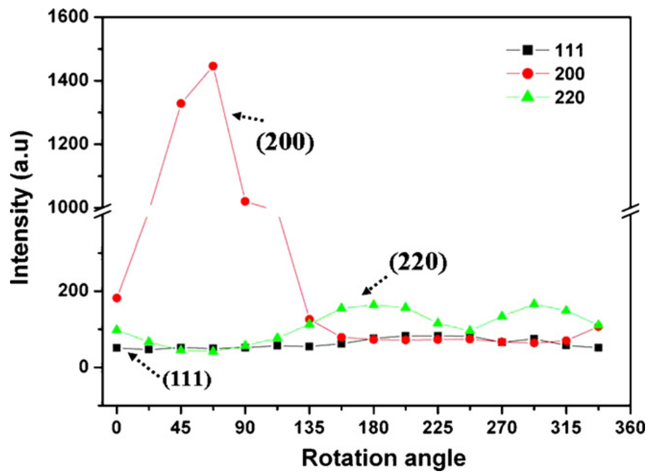

Fig. 5. Maximum intensities of the three peaks (1 1 1), (2 0 0), and (2 2 0) as a function of the rotation angle of the sample from 0° to 337.5° with the 16 scans from Fig. 4. (Vertical axis: maximum peak intensity, Horizontal axis: step-wise rotation angle, 22.5° for each step.)

grains observed in Figs. 1 and 2 is analytically confirmed by our method.

The maximum peak intensities of the three peaks (1 1 1), (2 0 0), and (2 2 0) in Fig. 4 are re-plotted along with the step-wise rotation angle from 0° to 337.5° in Fig. 5. The strong (2 0 0) intensity in the figure indicates that the MgO thin film has a highly preferred orientation of [2 0 0] grown vertically to the substrate (out-of-plane texture). Meanwhile, the dependence of the (2 0 0) intensity on the step-wise rotation angle implies that the [2 0 0] direction is also preferentially oriented along a particular direction in the substrate (e.g., at 67.5° in Fig. 5), which is an in-plane texture developed parallel to the substrate. The FWHM of the (2 0 0) curve, corresponding to the phi(ϕ) FWHM, is about 45°, as estimated from Fig. 5. The in-plane texture in not as perfect as the MgO thin film with thickness 20,000 Å, as grown by the ISD technique,[3] at an inclination of 60° in which the phi(ϕ) FWHM is about 14°, but comparable to the phi(ϕ) FWHM of the MgO film of 20,000 Å at an inclination angle (α) = 15°. The relative peak intensity of (2 2 0) decreases to a minimum at the rotation angle corresponding to the maximum of (2 0 0) and increases to its maximum value at the angle of minimum of (2 0 0), as shown in Fig. 5. The (1 1 1) curve also shows a slight change in the intensities in a similar manner as the (2 2 0) curve.

4. Conclusion

The preferred orientation and crystal morphology of MgO thin films grown by electron beam evaporation at a substrate inclination angle $\alpha = 0$ are characterized and found to have a biaxial texture. The [2 0 0] in-plane texture was substantially developed in a MgO film of thickness <6000 Å as well as the (2 0 0) out-of-plane (fiber) texture. A simple and novel method for the analysis of the biaxial texture has been devised in this study and was found to be suitable for very thin films comprising weak X-ray scattering species. The crystal morphologies of the MgO grains were shown to have a columnar structure growing vertically from the substrate and also to have triangular facets strongly aligned in a particular direction parallel to the substrate. The texture analysis by the analytical method using XRD conforms to the SEM morphological characteristics of MgO thin films.

Acknowledgements

This work was supported by grant no. RTI04-01-02 from the Regional Technology Innovation Program of the Ministry of Commerce, Industry and Energy (MOCIE).

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