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Epitaxial Growth of α -Fe₂O₃ and Cr₂O₃ Sputtered Films on Garnet

M. Gomi, H. Toyoshima and T. Yamada

Department of Materials Science, Japan Advanced Institute of Science and Technology, 1-1 Asahidai, Tatsunokuchi, Ishikawa 923-12, Japan

Abstract: Thin films of α -Fe₂O₃ and Cr₂O₃ have been prepared *in situ* on gadolinium gallium garnet (GGG) (111) substrates by rf sputtering. The α -Fe₂O₃ film showed a strong [0006] texture normal to the film plane at 500°C when deposited on an initial layer with 7 - 10 nm thick grown at ambient temperature, while the Cr₂O₃ films with a [0006] texture grew in pure Ar without the initial layer. X-ray pole figure analysis showed that nearly single crystalline (0001) films of α -Fe₂O₃ and Cr₂O₃ were grown on GGG (111) with an in-plane alignment of [2 $\bar{1}$ 10] parallel to GGG [1 $\bar{1}$ 0]. Thus, these films with the corundum crystal structure may be a promising buffer layer for achieving heteroepitaxy of various ferrite thin films onto a common substrate of GGG.

1. INTRODUCTION

Epitaxial film growth of ferrite such as yttrium iron garnet and Ba-hexaferrite onto a common substrate is of great importance for realizing microwave monolithic integrated circuit with high performance. It has been recently shown that Ba hexaferrite films can be epitaxially grown on basal plane sapphire with the corundum structure [1]. However, few studies on heteroepitaxy of the ferrite thin films have been reported so far. This is because there are few substrate crystals closely lattice matched to the ferrites with complicated crystal structures. Thus, buffer layer which provides a close structural fit with both of the substrate and the ferrite film is indispensable to achieve the heteroepitaxy.

We have recently reported that epitaxial thin films of α -Fe₂O₃ with the corundum structure can be fabricated by sputtering and subsequent annealing on gadolinium gallium garnet (GGG) which is a widely used substrate for the epitaxial growth of garnet films [2]. After that, we have successfully attained *in situ* growth of nearly single-crystalline films of α -Fe₂O₃ and Cr₂O₃ on GGG. Since these films have lattice constants close to that of the sapphire, they may be applied to a buffer layer which allows both of the Ba hexaferrite film and the iron garnet film to epitaxially grow onto a common substrate of GGG crystal. In this paper, we describe the conditions of *in situ* growth of their films and reveal crystallographic orientation relationships between the films and the substrate.

2. FILM GROWTH

The films were deposited on (111)-oriented GGG single crystal substrates by rf diode sputtering using sintered ceramic targets of α -Fe₂O₃ and Cr₂O₃. The rf power density was kept at 8 W/cm² which gives a deposition rate of 3 - 4 nm/min. The mixture gas of Ar and O₂ was used as a sputtered gas. The substrate was heated up to 500°C at maximum. When the film was deposited at ambient temperature, the substrate temperature T_s rose up to about 180°C due to plasma.

3. RESULTS AND DISCUSSION

Figure 1 shows typical X-ray diffraction spectra for the α -Fe₂O₃ films prepared under several substrate temperatures and gas pressures (partial pressure of O₂: 10 %). For deposition at high temperature, the spectra of the films show the diffraction peaks from (11 $\bar{2}$ 0), (0006) and (11 $\bar{2}$ 3) planes, while the films deposited at ambient temperature have a single peak from (11 $\bar{2}$ 0) plane for the gas pressure of 50 mTorr and (0006) plane for 10 mTorr. The peak seen around 37.6 deg in the spectrum at 50 mTorr is identified with that from (222) plane of γ -Fe₂O₃ which was made by insufficient oxidization of sputtered Fe ions on the substrate. These diffraction peaks are weak and broad, reflecting poor crystallinity of the films. No films with a strong texture were directly obtained under the sputtering conditions used.

Fig. 1 Typical X-ray diffraction spectra of α -Fe₂O₃ sputtered films.

Fig. 2 X-ray diffraction spectra of $\alpha\text{-Fe}_2\text{O}_3$ layers ($T_s=500^\circ\text{C}$) on the underlying layers with thickness t deposited at 10 m Torr and ambient temperature.

Fig. 3 X-ray pole figure taken from the $(11\bar{2}3)$ peak of $\alpha\text{-Fe}_2\text{O}_3$ film which was grown on the initial layer with 7 nm thick.

Fig. 4 X-ray pole figure of Cr_2O_3 film with a prominent $[0006]$ texture, which was taken from the $(11\bar{2}3)$ diffraction peak.

To *in situ* grow the films with a strong $[0001]$ texture normal to the film plane, a two-step deposition process, which involved an initial growth of $\alpha\text{-Fe}_2\text{O}_3$ at ambient temperature followed by growth at 500°C , was tried. Both growths were done under a gas pressure of 10 m Torr. Figure 2 shows relationship between the crystal orientation of the overlying layer and the thickness t of the underlying layer. At $t < 3.5$ nm and $t > 14$ nm, the films are polycrystalline with no preferred crystal orientation, as well as the film without underlying layer, while, when $t = 7$ nm and 10.5 nm, the films showed a strong $[0006]$ texture. Figure 3 shows pole figure taken from the $(11\bar{2}3)$ peak of the $\alpha\text{-Fe}_2\text{O}_3$ film which was grown on the initial layer with 7 nm thick. It exhibits six pole densities every 60° for ϕ scans at $\phi = 61^\circ$, similar to that for the epitaxial film obtained by heat treatment previously [2]. The width of the pole density in ϕ scans are rather broad, indicating substantial twist misalignment of the grains. These make an angle of about 30° in ϕ scans with the pole densities of the substrate taken from the (440) diffraction peak. Other narrow spots with the strongest intensity simultaneously observed, are due to reflections from the substrate. The results obtained from pole-figure analysis indicate that a nearly single-crystalline film of (0001) -oriented $\alpha\text{-Fe}_2\text{O}_3$ was *in situ* grown on GGG (111) with an in-plane alignment of $\alpha\text{-Fe}_2\text{O}_3$ $[2\bar{1}\bar{1}0]$ parallel to GGG $[1\bar{1}0]$. These results are explained by assuming that, when T_s rose above 500°C , the thin initial layer having an appropriate thickness is recrystallized and the in-plane alignment in the layer traces the surface potential of the substrate to reduce the film-substrate interface energy: Too thin initial layer which incompletely covers the surface of the substrate, or too thick initial layer in which in-plane crystalline reorientation would not occur sufficiently because T_s of 500°C is rather low results in growth of polycrystalline film.

Films of Cr_2O_3 having a lattice constant a little bit smaller than that of $\alpha\text{-Fe}_2\text{O}_3$ were grown onto GGG (111) as well. The films sputtered in atmospheres containing oxygen showed X-ray diffraction spectra similar to those of $\alpha\text{-Fe}_2\text{O}_3$ films, except that the film at ambient temperature was amorphous, while the film sputtered below 400°C in pure Ar exhibited a strong $[0006]$ texture. However, the latter film was considerably oxygen deficient. Figure 4 shows pole figure of the Cr_2O_3 film with prominent (0006) -orientation, taken from the $(11\bar{2}3)$ diffraction peak. It exhibits a pair of spotlike pole densities, which is clearly separated, every 60° in ϕ scans as well as the case of $\alpha\text{-Fe}_2\text{O}_3$. It is apparent from pole-figure analysis that the orientation relationship between the film and the substrate is the same as that for $\alpha\text{-Fe}_2\text{O}_3$ film. These results indicate that nearly single crystalline Cr_2O_3 (0001) film can be grown *in situ* onto GGG (111) without an underlying layer, in contrast with the $\alpha\text{-Fe}_2\text{O}_3$ film.

The above orientation relationship between the films with the corundum structure and the GGG substrate is explained in terms of the close structural fit between the cation sites as reported previously [2]: The arrangement of Fe ion or Cr ion in triangular symmetry in the corundum structure roughly fits into the Gd site which is the largest cation site in the garnet structure. In this case, the $[2\bar{1}\bar{1}0]$ direction of the corundum structure lies parallel to the $[1\bar{1}0]$ direction of the substrate, consistent with the experimental results. However, the in-plane lattice arrangement of the dodecahedrally coordinated Gd and tetrahedrally coordinated Ga in the GGG (111) plane is locally distorted, leading to considerable deviation from the ideal arrangement with triangular symmetry. This gives flexibility to the in-plane orientation of the film, which results in substantial twist misalignment of the grains observed at the $\alpha\text{-Fe}_2\text{O}_3$ film. The difference in the shape of pole density and epitaxial behavior between the $\alpha\text{-Fe}_2\text{O}_3$ film and the Cr_2O_3 film may be ascribed to a different lattice constant. Further studies on it are required.

4. CONCLUSION

We have epitaxially grown $\alpha\text{-Fe}_2\text{O}_3$ (0001) films and Cr_2O_3 (0001) films with the corundum crystal structure *in situ* on GGG (111) by rf sputtering. It was achieved for $\alpha\text{-Fe}_2\text{O}_3$ films by a two-step deposition process, which involved an initial growth of $\alpha\text{-Fe}_2\text{O}_3$ at ambient temperature followed by growth at 500°C , while, for Cr_2O_3 films, by direct deposition. These films were grown with an in-plane alignment of $[2\bar{1}\bar{1}0]$ parallel to GGG $[1\bar{1}0]$.

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