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Control of Step and Terrace Structures of LSMO Thin Films by IBS Deposition

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LSMO thin films were fabricated using ion beam sputtering (IBS) technique on LAO ($LaAlO_3$) (100) and MgO (100) substrates at different substrate temperatures of 550-750°C and oxygen partial pressures of 0.5-1.5 mTorr. Oxygen was supplied either in molecular or plasma form. At higher temperatures the plasma supply improves the crystallinity. Mosaicity of the films on LAO substrate is always better than on MgO substrate. The films on LAO substrate show two-dimensional step and terrace type growth. The terrace is very long as 2.45μm for T_S = 550°C. The step height and terrace length decrease at the higher temperature.
Key words: LSMO thin films, LAO and MgO substrates, Crystallinity, Mosaicity, Step and terrace

1. INTRODUCTION

The discovery of colossal magnetoresistance (CMR) effect in perovskite manganite compounds with general formula $R_{1-x}A_xMnO_3$ (where, R: trivalent rare earth and A: divalent alkaline earth) has renewed the interest in this class of materials [1,2]. Because of its chemical stability as well as the most common lattice spacing, CMR manganites are suitable for fabrication of ultrathin film or artificial supperlattice structure with different compositions. CMR thin films are of special interest due to their advantage in practical applications. The magnetoresistance value could be strongly affected by A-site doping and oxygen content.

However, for a given chemical composition and oxygen stoichiometry, CMR material grown epitaxially on different substrate can have very different resistivity (ρ), metal-insulator transition temperature (T_p) and ferromagnetic transition temperature (T_c). This feature was attributed to the different types of strain caused by different substrates. In epitaxial films, biaxial strain has strong effect on the changes in T_c and T_p [3-9]. On the other hand, there are many reports mentioning the variation of T_c and T_p with film thickness [6-10]. However, strain and film thickness are not the only factors, but microstructure also determines the magnetotransport properties of the films [11]. The x-ray diffraction technique measures lattice constants over a large length scale and can characterize the strain as uniform. However, in submicron length scale the film may contain internal boundaries, discontinuities and steps, which will affect the magnetotransport properties of the film. These intriguing features of the thin films motivate the researchers to explore the film fabrication at various conditions using different techniques like PLD [12-14], MBE [15-17], IBS [18-23] etc.

In the present article, we report the microstructure analysis of the $La_{1-x}Sr_xMnO_3$ thin films grown on LAO and MgO substrates using ion beam sputtering (IBS) technique. We discuss the film fabrication at different substrate temperatures (T_S) and oxygen partial pressures (P_O), and characterizations. We have seen the different growth characteristics of the film on LAO and MgO; which are mainly associated with the lattice mismatch between the substrate and film. The magnetotransport properties of these films will be published elsewhere.

2. EXPERIMENTAL

LSMO thin films were fabricated on both LAO (100) and MgO (100) substrates simultaneously using IBS technique. These substrates have a size of 5 mm× 10 mm, and surface roughness around 0.1-0.5 nm. A sintered disk-shaped target of $La_{0.9}Sr_{0.4}MnO_3$ was sputtered by 4 keV Ar^+ ion. The sputtered particles were deposited on heated substrates. Substrate temperature (T_S) was varied in the range 550-750°C. During the film growth oxygen gas was supplied through a plasma source either in molecular (ML) or plasma (PL) form at various partial pressures P_O. In the case of plasma supply a high AC voltage of 60 Hz was applied to the plasma source to produce oxygen plasma. The oxygen pressure P_O was varied in the range 0.5-1.5 mTorr.

The crystalline structure of the films was characterized by x-ray diffraction (XRD) (Philips X'pert, Cu K_α) measurements. The crystallinity was estimated from the average half-width (Av.Δ_θ) of the Bragg's peaks. From the average half-width of the rocking curve (Av.Δ_ω) we have checked the mosaicity of the films. The surface morphology was examined and surface roughness (R_{sf}) was determined using atomic force microscopy (AFM). The film thickness is around 50-100 nm.

3. RESULTS

In Fig.1, we show the representative x-ray diffraction patterns of the LSMO films on both LAO and MgO substrates atdifferent substrate temperatures (T_S) at 1 mTorr of P_0. The absence of any impurity peak in the patterns indicates that the films are single phase. The labels L and M in the figures indicate the LAO and MgO substrates peaks, respectively. Figure 2 shows the variation of crystallinity (Av.Δ_θ), mosaicity (Av.Δ_ω) and surface roughness (R_{sf}) of the films as a function of T_S. We observed that crystallinity is degraded with the increase in T_S on both the substrates for ML supply (Fig. 2(a)). However, at higher T_S, the crystallinity could be improved by PL supply on MgO at 700°C, and on LAO at 700°C and 750°C as indicated by arrows in Fig.2(a). At 750°C mosaicity of the film is little improved by ML supply on LAO, whereas at 700°C the PL supply for MgO substrate considerably improves the mosaicity (Fig. 2(b)). One should notice the large difference in mosaicity of the films on LAO and MgO substrates. The films on LAO substrate have much smaller crystal plane distribution compared to MgO substrate. Figure 2(c) shows that the R_{sf} on LAO substrate is minimum (1.44 nm) at 550°C for ML supplied film, it increases with the increase in temperature up to 700°C followed by decrease above 700°C. This behavior can be considered as the change in growth characteristic as 2-dimensional (D)→3D→2D as indicated by arrows in Fig. 2(c). Though R_{sf} on MgO substrate does not show significant changes below 700°C, on further increase in T_S it decreases (Fig. 2(c)). It is interesting to observe that PL supply at higher T_S improves R_{sf} of the films on both LAO and MgO substrates (Fig. 2(c)).

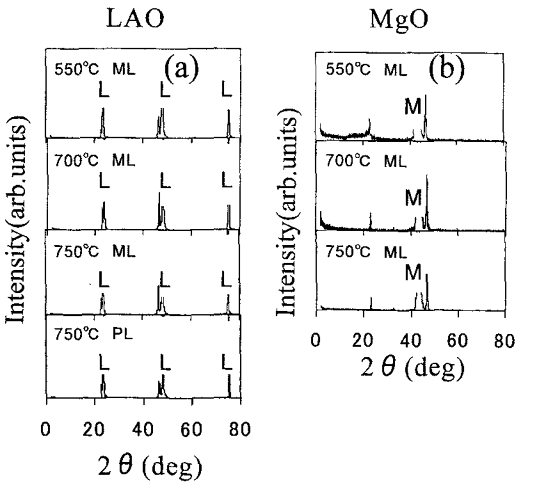

Fig. 1. XRD patterns of the LSMO films grown at different temperatures and 1 m Torr of P_0 on (a) LAO and (b) MgO substrates.

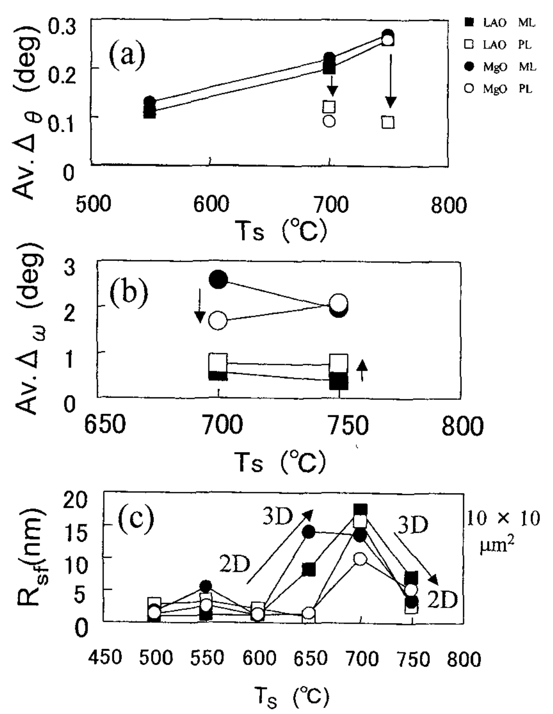

Fig. 2. Ts dependence of (a) Av. Δ_θ, (b) Av. Δ_ω and (c) R_{sf} of the films grown at 1 mTorr of P_0 on both LAO and MgO substrates. Solid and open symbols correspond to ML and PL supply, respectively. The films on MgO at 500°C are amorphous.

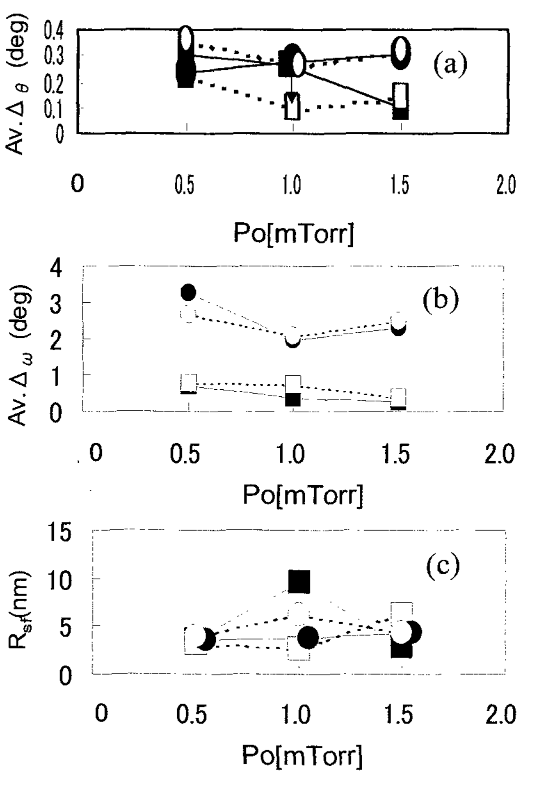

Fig. 3. Po dependence of (a) Av.Δ_θ, (b) Av.Δ_ω and (c) R_{sf} of the films grown at 750℃ on both LAO and MgO substrates. Solid and open symbols correspond to ML and PL supply, respectively.

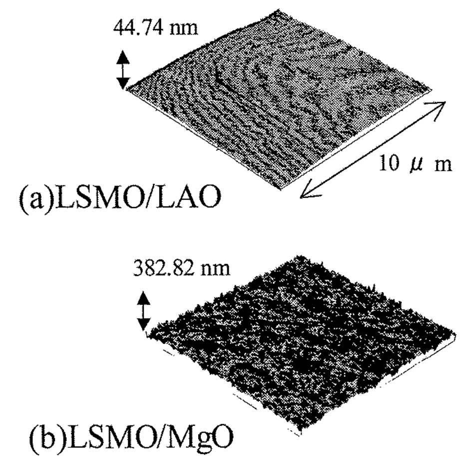

Fig.4. AFM images of LSMO films grown at 550℃ on (a) LAO and (b) MgO substrates. (a) shows the step and terrace type growth of the film on LAO substrates.

In Fig. 3, we plot Av.Δ_θ, Av.Δ_ω and R_{sf} of the films grown at 750°C on both LAO and MgO substrates as a function of P_0. Using PL supply at 1 mTorr we are able to considerably improve the crystallinity of the film on LAO substrate (Fig. 3(a)). On the other hand, ML supply gives better crystallinity at 0.5 mTorr on MgO substrate (Fig. 3(a)). There is not much change in Av.Δ_ω of the films with the variation of Po on both LAO and MgO substrates as shown in Fig. 3(b). The mosaicity of the film on LAO is much better than on MgO substrate again. The changes in R_{sf} with P_0 show complicated behavior (Fig. 3(c)). However, it is clear that there is a significant improvement in R_{sf} of the film on LAO substrate for PL supply at 1 mTorr as indicated by an arrow in Fig. 3(c).

Figures 4(a) and (b) show the AFM images of the films grown on LAO and MgO substrates, respectively. It is worthy to mention that the film on LAO substrate shows step and terrace type growth whereas on MgO substrate it shows usual grain type growth. The step and terrace in the AFM image clearly indicate the 2D growth characteristic of LSMO film on LAO substrate. The initial growth study of Sr-substituted film by Ba (LBMO) on both the substrates revealed that at the beginning of the deposition nucleation takes place on LAO substrate, whereas on MgO substrate it remains as amorphous layer (image not shown here). This initial growth experiments clearly indicate that the nucleation is easier on LAO than on MgO due to lattice matching. This must be the origin of the 2D type step and terrace growth of LSMO on LAO substrate.

In Fig. 5(a) we show the AFM image of the same film shown in Fig. 4(a), and show the corresponding cross-sectional profiles in Fig. 5(b). We have measured the terrace length and step height. The terrace length is around 2.45μm and the step height is around 0.72 nm at 550°C. We have also observed the step and terrace type growth on LAO at a higher temperature of 750°C as well. In Fig. 6, we plot the terrace length and step height as a function of T_S. The step height defined as a vertical height in Fig.5(b), corresponds to two unit cell length at 550°C, however it corresponds to 1.5 unit cell length at 750°C. The reason is not know yet but probably they are thermodynamically stable under the deposition conditions. The longer terrace length at 550°C signifies large surface migration of LSMO on LAO substrate. At higher temperature both terrace length and step height decrease.

4. DISCUSSION

We have shown that at higher temperature the crystallinity is degraded, but using PL supply one can improve it (Fig. 2 (a)). Figure 2(b) shows that using PL supply at 700 °C mosaicity is improved on MgO substrate. Again in Fig. 3 we have shown that Av.Δ_θ, Av.Δ_ω and R_{sf} show complicated behaviors with the variation of P_0. However, by PL supply at 1 mTorr of P_0, Av.Δ_θ and R_{sf} on LAO substrate are improved considerably. This behavior is related to the optimized condition among all the parameters, which played a major role during the film growth. In our IBS system the crucial parameters are (1) thermal energy, (2) sputtered particle kinetic energy, (3) supplied oxygen density, (4) collision between the sputtered particles and the supplied oxygen molecules, (5) collision

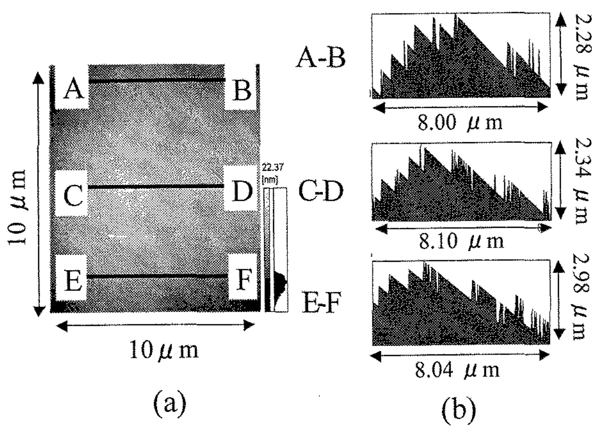

Fig.5. (a) AFM images, and (b) corresponding cross-sectional profiles of the LSMO film on LAO substrate grown at 550℃. The tilted terrace does not affect our estimation of the step height.

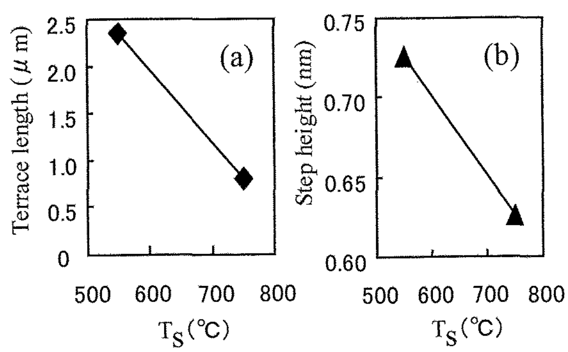

Fig.6. Plot of terrace length and step height against T_s.

excited oxygen species, (6) collision-reduced kinetic energy of the sputtered particles, and (7) oxygen plasma excited energy. Basically all of these are energy parameters. When the total energy is balanced it gives good quality film.

The better lattice matching of the film on LAO substrate gives nucleation at the initial growth stage and it shows 2D type growth. This 2D growth is reflected by the observed good mosaicity, and step and terrace morphology of the LSMO film on LAO substrate. Whereas due to larger lattice mismatch on MgO substrate nucleation does not take place at the initial stage, rather amorphous layer is formed. On continuing deposition nucleation takes place on the amorphous layer. This amorphous layer is remained at the interface between the substrate and film and called dead layer. This leads to the grain type growth with poor mosaicity of the films on MgO substrate. The variation of terrace length and step height with temperature is not clear at this point.

5. CONCLUSIONS

In summery we have fabricated the LSMO thin films on both LAO and MgO substrates at different substrate temperatures and oxygen partial pressures. The plasma supply has definite roles on crystallinity at higher temperatures. The smaller lattice mismatch of LSMO on LAO gives 2D step and terrace type growth, whereas on MgO substrate due to larger lattice mismatch grain type growth is observed. The step height and terrace length decrease with increasing T_S.

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