Transactions of the Materials Research Society of Japan 28 [4] 1117-1120 (2003)

Growth of LBMO Thin Films by Ion Beam Sputtering - Crystallinities and Ferromagnetic Resonance

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The LBMO thin films were deposited by ion beam sputtering on MgO and LAO at 700 and 750℃ with oxygen molecular and plasma supply under various oxygen partial pressures. The intragrain and intergrain crystallinities, and the surface roughness of deposited films at 700℃ are investigated. The intergrain crystallinity is excellent on LAO and superior for the molecular supply. The surface roughness can be extremely improved by the plasma on LAO. FMR study is carried on the film deposited at 750℃ after consecutive annealings. Ferromagnetic property can be enhanced by the annealing.
Keywords: Ion Beam Sputtering, LBMO Thin Films, Crystallinity, Ferromagnetic Resonance

1. INTRODUCTION

Tunable microwave filters are expected as next generation devices for mobile communication system [1]. High performance microwave filters can be achieved using high temperature superconducting $YBa_2Cu_3O_x$ (YBCO) thin films, and the tunable filters can be fabricated by stacking a ferromagnetic layer on YBCO layer [2]. In this work, we deposited a single layer of ferromagnetic $La(Ba)MnO_3$ (LBMO) thin films as a first step for the multi-layer.

$LaAlO_3$ (LAO) is a superior material as a substrate for crystalline LBMO and YBCO film growths due to lattice matching. However, it is not suitable for high frequency devices owing to its high dielectric constant (15-22) and commercial reasons. While MgO is quite suitable due to its low dielectric constant (9.5) and mass production. Then it is valuable to compare the results of thin film growths on LAO and MgO in terms of fundamental research and applications.

Various researches have done on LBMO thin film crystallinities and surface roughness as well as its magnetic properties [3-5]. We are currently investigating fundamental film growths of LBMO and their crystallinities and surface roughness [6-8]. In this work, LBMO thin films were deposited on LAO and MgO with various deposition conditions, and their crystallinities and surface roughness were investigated in relation to the deposition conditions. Their ferromagnetic properties were studied by means of ferromagnetic resonance. It is known that the ferromagnetic properties are improved by annealing [9,10]. Then we studied the annealing effects on the ferromagnetic resonance.

2. EXPERIMENTAL

LBMO thin films were fabricated by ion beam sputtering (IBS) system shown in Fig.1. A target (T) of $La_{0.94}Ba_{0.39}Mn_1O_x$ was sputtered by 4 keV Ar^+ ions and sputtered particles (SP) were deposited on $LaAlO_3$ (LAO) (100) and MgO (100) substrates (S) simultaneously. Substrate temperatures (T_S) were fixed at 700 and 750℃, and either oxygen molecule (ML) or oxygen plasma (PL) was supplied from a plasma source (PS) on the substrates under various oxygen partial pressures (P_O). The oxygen plasma was produced by discharging oxygen gas in the plasma source. When the oxygen molecules were supplied, oxygen gas was directly emitted from the same plasma source without discharge.

The deposited LBMO thin films were characterized by X-ray diffraction (XRD) on θ-2θ scan using Cu Kα line. The film "intragrain crystallinity" was estimated by full width at half maximum of θ-2θ XRD peaks ($Av.\Delta_\theta$) averaging over (001) and (002) peaks. The film "intergrain crystallinity" was estimated by full width at half maximum of XRD rocking curves ($Av.\Delta_\omega$) averaging over (001) and (002) peaks. The intragrain crystallinity is a measure of distribution of crystallographic plane-distance in the grains which have parallel planes to the substrate plane. The intergrain crystallinity is a measure of distribution of crystallographic

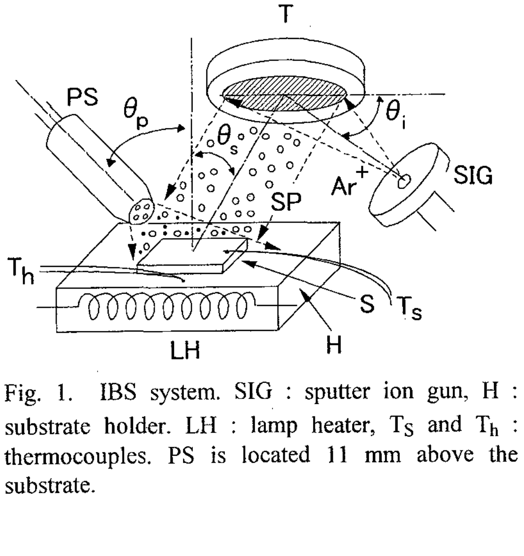

Fig. 1. IBS system. SIG : sputter ion gun, H : substrate holder. LH : lamp heater, T_S and T_h : thermocouples. PS is located 11 mm above the substrate.

plane-direction in the grains deflected from the substrate plane. This is usually called "mosaicity". The film surface roughness was measured by atomic force microscopy (AFM), and it was evaluated by rms values (R_{sf}). These crystallinities were estimated for the deposited films at T_S = 700℃ at P_O = 0.05-4 mTorr.

The ferromagnetic resonance (FMR) was measured using microwave at 9.3 GHz on the deposited films on LAO at T_S = 750℃ with ML at P_O = 1 mTorr. The sample was put in the center of TE_{102} cavity and cooled using liquid nitrogen. A dc magnetic field (H_a) and a modulation field (H_m) were applied to the sample perpendicular ($H_{a\perp}$) or parallel ($H_{a\parallel}$) to the film plane. The dc field was swept from 0 to 8 kOe with H_m = 5 Oe at 100 kHz. A part of the samples were measured at room temperature (r.t.). The effective magnetization ($4\pi M_{eff}$) was calculated using resonance fields (H_r) both for $H_{a\perp}$ and $H_{a\parallel}$. The half width (Γ_{PP}) of resonance peak was defined as a peak-to-peak field interval of the derivative signal. FMR measurement was also done on annealed samples. The deposited film on LAO at T_S = 750℃ with ML at P_O = 1 mTorr was annealed in oxygen gas flow first at 875℃ for 13 h (called 1st-anneal) and consecutively at 800℃ for 16 h (called 2nd-anneal).

3. Results and Discussion

3.1 Crystallinities (T_S = 700℃)

The values of Av.Δ_θ were plotted as a function of P_O on the deposited films at 700℃ with ML or PL on LAO and MgO. These values were roughly 0.15-0.2°, but we found a quite peculiar result that Av.Δ_θ were extremely small in a narrow P_O range around 2 mTorr both for ML and PL only on MgO. We call it a "window effect" hereafter [6,7]. The value is 0.09° for the film with ML. Then the intragrain crystallinity is exceptionally excellent on MgO in this window region of P_O especially when the oxygen molecules are supplied.

The intergrain crystallinity was estimated on the same films. The values of Av.Δ_ω are plotted in Figs. 2 and 3 as a function of P_O for ML and PL, respectively. It must be noticed that the curves of Av.Δ_ω on MgO show peaks at P_O = 2 mTorr both for ML and PL which is just corresponding to the window region. This indicates that the intragrain crystallinity and intergrain crystallinity have inverse correlations on MgO, that is, if the intragrain crystallinity is good in a film, the intergrain crystallinity must be poor in the same film.

There is no remarkable difference between the results for ML and PL, however, there are striking differences between the values of Av.Δ_ω on MgO and LAO both for ML and PL. The values are extremely smaller on LAO such as 0.14° at P_O= 0.5 mTorr for ML (Fig. 2) than on MgO, then the intergrain crystallinity (mosaicity) is much better on LAO.

We can propose that an origin of the striking difference in the intergrain crystallinity depending on the substrates should be lattice mismatching levels. A lattice

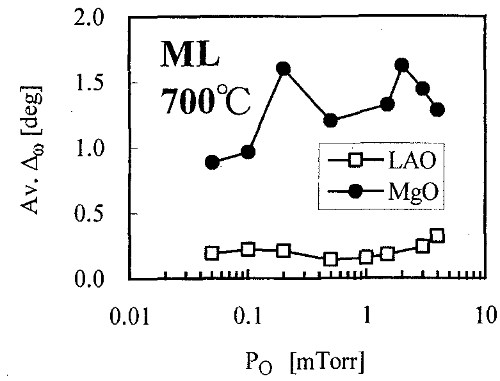

Fig. 2. Intergrain crystallinity for ML

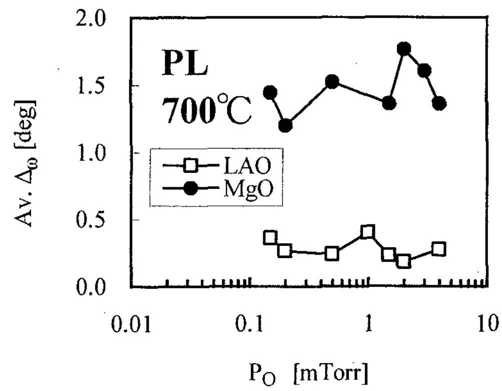

Fig. 3. Intergrain crystallinity for PL.

constant of bulk LBMO is 3.894 Å, and it is 3.79 Å for LAO and 4.21 Å for MgO. Then the lattice matching is better on LAO but quite poor on MgO. Each grain of LBMO grows epitaxially on LAO everywhere on the substrate, then it has the same coherence (crystalline periodicity) with each other. It means that the periodic phase of crystalline structure between the grains is matched when the two grains coalesce. Then the crystallographic planes in the two grains are aligned together along the substrate plane, leading to the smooth mosaicity. We obtained the results that the intragrain crystallinity is almost the same between the films on MgO and LAO. It indicates that the distribution of plane-distance of the film on the large mismatched substrate (MgO) is the same with that on the small mismatched substrate (LAO). This is reasonable because the grains having the parallel plane to the substrate get the excellent intragrain crystallinity but there might be tilted grains between these parallel grains, causing small Av.Δ_θ but large Av.Δ_ω on MgO.

Now we discuss the oxidant effects. Generally speaking, the values of Av.Δ_ω are almost the same for ML and PL as shown Figs. 2 and 3. We have our data which show the crystalline improvement effects by PL at lower T_S. But in this experiments, T_S is higher, then the crystal growth is dominated by thermal energy. Then the plasma energy is not necessary for the crystal growth. On carefully watching the results shown in Figs. 2 and 3, the average values of Av.Δ_ω are rather smaller for ML than for PL both on MgO and LAO. Examining the values on LAO, the smallest value of Av.Δ_ω on LAO for PL is 0.18° at P_O = 2 mTorr which is larger than 0.14° for ML mentioned above. The plasma might supply excess energy for the crystal growth of LBMO thin film at 700℃. Thus it looks as the substrate temperature of 700℃ is high enough to obtain ferromagnetic properties of LBMO. However, the several samples grown at 700℃ did not show FMR at 77 K. Then we fabricated the films at 750℃. The results of FMR are shown in the next section.

We investigated the surface roughness R_{sf} of the 700℃-samples. The values of R_{sf} are plotted in Fig.4 as a function of P_O for PL on MgO and LAO. Completely opposite trend was obtained between MgO and LAO, the curve for MgO shows a peak but it shows a deep dip at P_O = 1 mTorr. Again we got another "window effects". R_{sf} is larger for PL outside the window while it is much smaller for PL inside the window. In this window, the minimum value of R_{sf} is 0.83 nm, it is very small then the film is incredibly smooth even though the growth temperature is very high. AFM image of this sample is shown in Fig. 5. The plasma treatment does not show the excellent effects on the crystallinities but it clearly shows

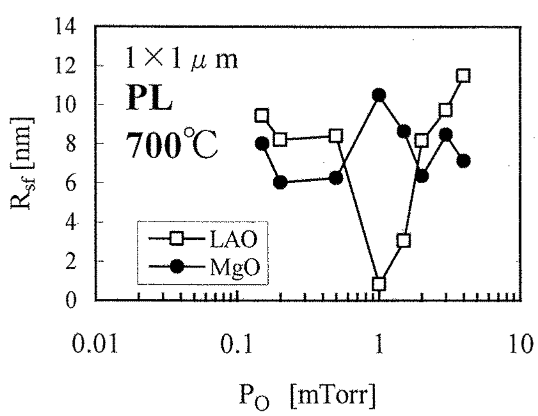

Fig. 4. Surface roughness of the PL-films.

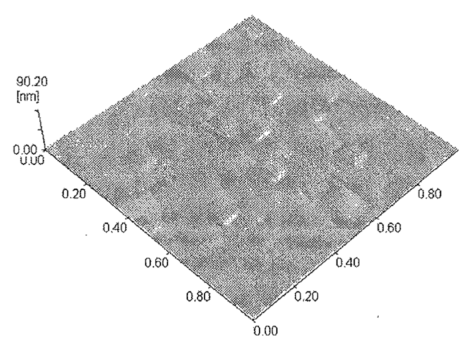

Fig. 5. AFM image of the film deposited on LAO at 700℃ with PL at 1 mTorr.

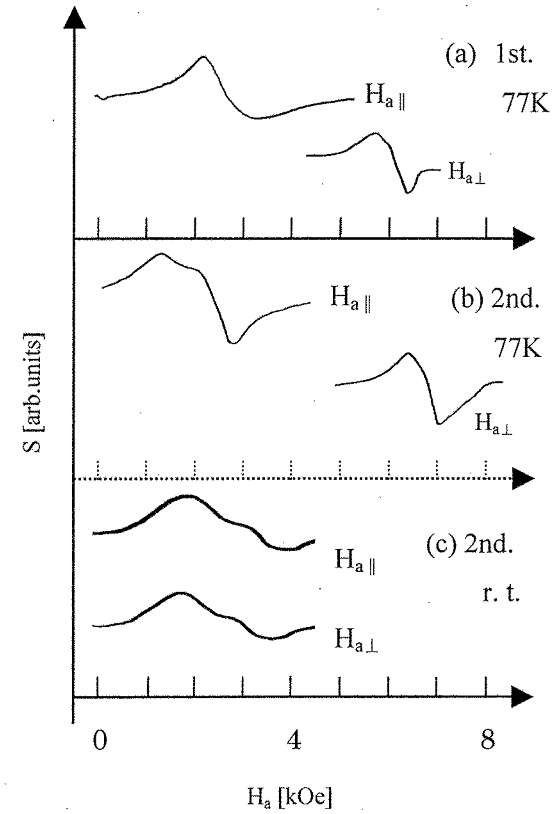

Fig. 6. FMR signals (S). (a) 1st-anneal, 77 K, (b) 2nd.-anneal, 77 K and (c) 2nd-anneal, room temperature.

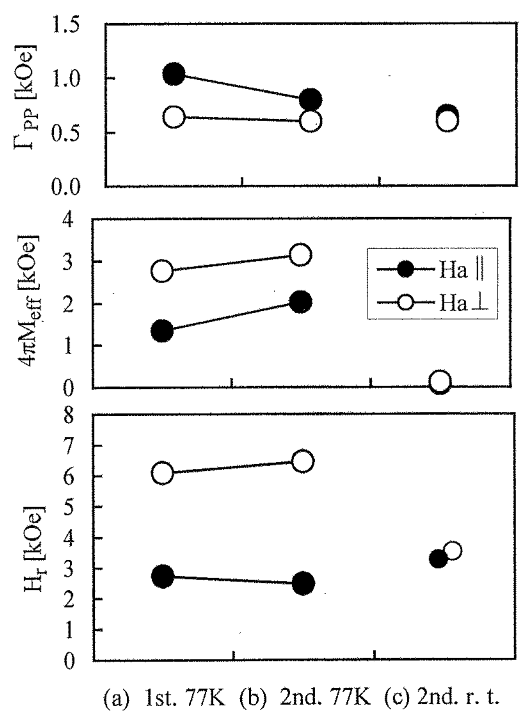

Fig. 7. H_r, $4\pi M_{eff}$ and Γ_{PP} for (a) 1st-anneal (77 K), (b) 2nd-anneal (77 K) and (c) 2nd-anneal (r. t.).

the superior effect on the smoothening of film surface. This gives an important result relating to the stacked tunable microwave filters, because the smoothest surface is an inevitable factor for the microwave devices.

3.2 Ferromagnetic resonance ($T_S = 750$℃)

LBMO films were deposited at 750℃. Some of the as-grown samples showed FMR signals (S) but some other as-grown films did not show FMR [11]. In this work, we investigated the annealing effects on the samples which did not show FMR in as-grown state. FMR signals at 77 K are shown in Fig. 6 on the (a) 1st-anneal and (b) 2nd-anneal samples for $H_{a\parallel}$ and $H_{a\perp}$, and the signals at room temperature (r.t.) are shown only for the 2nd-anneal sample in (c). In Fig. 6 (a) and (b), the resonance field (H_r) is higher for $H_{a\perp}$ than for $H_{a\parallel}$. The higher shift of H_r is caused by demagnetizing effect, and the lower shift of H_r is caused by field concentration effect owing to the ferromagnetic nature of the film.

The values of H_r, $4\pi M_{eff}$ and Γ_{pp} are plotted in Fig. 7 for the 1st-anneal and 2nd-anneal samples. H_r for $H_{a\perp}$ is shifted to higher fields and H_r for $H_{a\parallel}$ is shifted to lower field from the 1st-anneal to 2nd-anneal, indicating that the ferromagnetic property is enhanced by the consecutive annealing. This phenomenon is clearly observed in the increases in the effective magnetizations ($4\pi M_{eff}$) both for $H_{a\perp}$ and $H_{a\parallel}$ by the consecutive annealing. This enhancement is stronger for $H_{a\parallel}$ than for $H_{a\perp}$. The effective magnetizations are larger for $H_{a\perp}$ than for $H_{a\parallel}$. The spin homogeneity is better for $H_{a\perp}$ than for $H_{a\parallel}$ in the 1st-anneal sample because the value of Γ_{pp} is smaller for $H_{a\perp}$. After the consecutive annealing, Γ_{pp} decreases faster for $H_{a\parallel}$ than for $H_{a\perp}$, then the parallel spin homogeneity is improved more than the perpendicular spin homogeneity.

FMR signal can be detected at room temperature in the 2nd-anneal sample, but $4\pi M_{eff}$ is extremely small. So this sample is actually ferromagnetic at room temperature but it has very weak ferromagnetic property. All the samples do not have absolutely small Γ_{pp}, then further examinations of the film depositions and annealing conditions are necessary to obtain the excellent LBMO thin films having spin homogeneous magnetic property at room temperature.

4. CONCLUSION

The LBMO thin films were deposited by IBS on MgO and LAO at 700 and 750℃ with oxygen molecular or plasma supply with various P_O. The intragrain and intergrain crystallinities, and the surface roughness of deposited films at 700℃ were investigated. The intergrain crystallinity is excellent on LAO and superior for ML. The results are interpreted in terms of the lattice matching. The surface roughness can be extremely improved by PL on LAO. FMR study was carried on the film deposited at 750℃ after consecutive annealings. Ferromagnetic property can be enhanced by the annealing.

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(Received February 5, 2003; Accepted June 30, 2003)