

# Fabrication of LBMO films and magnetic properties at low temperatures

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## Abstract

Thin films of (La,Ba)MnO<sub>3</sub> are prepared with a supply of oxygen molecules or plasma at various partial pressures. Magnetic properties of the films are investigated by measuring the magnetic susceptibilities and the resistivities, and by ferromagnetic resonance. One of the film shows a Curie transition at 174 K and a metal–insulator transition at 84.2 K. Magnetic homogeneity is studied in relation to the preparation condition.

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## 1. Introduction

Microwave filters are most important superconducting thin film devices for mobile phone system [1]. Next generation devices must be tunable filters which are composed of the superconducting/ferromagnetic stacked thin films [2]. Manganites are most hopeful materials for the ferromagnetic layer. However, we have a difficult demand on the manganite layers, namely, they must be ferromagnetic and insulating phase at device temperatures. Usually, Curie temperature  $T_C$  is close to metal–insulator transition temperature  $T_P$ , then the materials are metallic in the ferromagnetic phase [3]. Fortunately, thin films sometimes show  $T_C$  and  $T_P$  at different temperature regions [4]. It is worthwhile to investigate the magnetic properties of such films in a region of  $T_P < T < T_C$ .

## 2. Experimental

Thin films of La–Ba–Mn–O were prepared by ion beam sputtering (IBS) on LaAlO<sub>3</sub> (1 0 0) substrates with a supply of oxygen molecules (ML) or oxygen plasma (PL) at various oxygen partial pressures ( $P_O$ ). The substrate temperature was fixed at 750°C during the deposition for 6 h [5–7]. Film thickness ranges from 400 Å to 800 Å. A part of the films were annealed at 900°C in O<sub>2</sub> flow for 5 h.

Ferromagnetic resonance (FMR) studies were carried out mainly at 77 K and at various other temperatures ( $T$ ) using EPR spectrometer on the samples prepared with various deposition conditions stated above. The resonance microwave frequency was 9.3 GHz. A modulation field (5 Oe) was superimposed on the DC field ( $H_a$ ) at 100 kHz. Then the derivative of the microwave absorption signals ( $S$ ) with respect to  $H_a$  is taken. Effective magnetization ( $4\pi M_{\text{eff}}$ ) was estimated by the resonance field. We can use the resonance equations of Kittel according to Landau–Lifshitz–Gilbert [8].

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$$\omega_r/\gamma = [H_{r\parallel}(H_{r\parallel} + 4\pi M_{\text{eff}})]^{1/2} \quad (1)$$

for the field  $H_{a\parallel}$  within the film plane and

$$\omega_r/\gamma = H_{r\perp} - 4\pi M_{\text{eff}} \quad (2)$$

for the field  $H_{a\perp}$  applied perpendicular to the plane. In these equations,  $\omega_r$  is a resonance frequency,  $H_{r\parallel}$  is a resonance field for  $H_{a\parallel}$ ,  $H_{r\perp}$  is a resonance field for  $H_{a\perp}$ , and  $\gamma$  is the gyromagnetic ratio. This  $\gamma$  is expressed as

$$\gamma = g\mu_B/\hbar, (\hbar = h/2\pi), \quad (3)$$

where  $\mu_B$  is the Bohr magneton and  $g = 2.00$  is used for  $g$ -factor. We can estimate  $4\pi M_{\text{eff}}$  using values of  $H_{r\parallel}$  or  $H_{r\perp}$  for each magnetic configuration. The magnitudes of  $H_{r\parallel}$  and  $H_{r\perp}$  are defined as a center field between positive and negative peaks on the derivative signal as usual. Half width ( $\Gamma_{\text{pp}}$ ) of the resonance peak was evaluated from peak-to-peak field of the derivative signal. When the signal has a doublet peak,  $\Gamma_{\text{pp}}$  value has some error. Its extent, however, is small and estimated to be 20–30% at most. FMR was measured for  $H_a$  perpendicular ( $H_{a\perp}$ ) and parallel ( $H_{a\parallel}$ ) to the film plane.

Magnetization ( $M$ ) was measured by SQUID as a function of  $T$  down to 4 K on a part of the samples at  $H_a = 1$  kOe for  $H_{a\perp}$  to determine the paramagnetic–ferromagnetic transition temperature ( $T_C$ ). Further, the sample resistance ( $R$ ) was measured as a function of  $T$  down to 4 K on a part of the samples to determine the metal–insulator transition temperature ( $T_P$ ).

### 3. Result and discussion

The temperature dependence of  $M$  is shown in Fig. 1 for the sample prepared with the molecular supply (ML) at  $P_O = 1.5$  mTorr. The result shows  $T_C = 174$  K. (Negative values of  $M$  above 174 K comes from the substrate.) The  $R$ – $T$  curve showed  $T_P = 84.2$  K as indicated in the figure. Then the film is insulating and ferromagnetic in the region  $84.2 < T < 174$  K.

The typical FMR signals for  $H_{a\perp}$  are shown in Fig. 2 for the sample with the plasma supply (PL) at

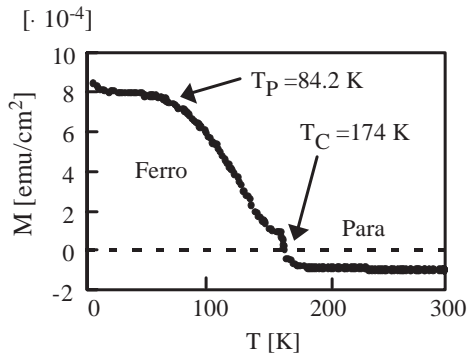


Fig. 1. Magnetization vs  $T$ .

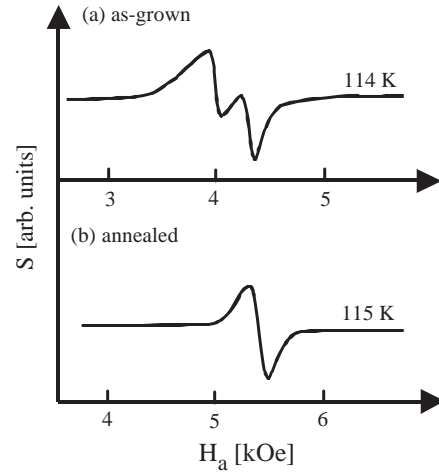


Fig. 2. FMR signals for  $H_{a\perp}$ .

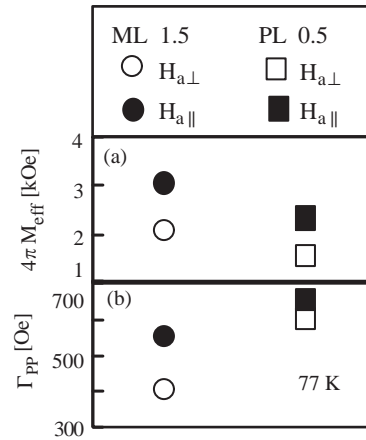


Fig. 3.  $4\pi M_{\text{eff}}$  and  $\Gamma_{\text{pp}}$  for the as-grown samples.

$P_O = 1.5$  mTorr, for (a) the as-grown at  $T = 114$  K, and (b) the annealed sample at  $T = 115$  K. A doublet peak is clearly observed on the as-grown sample, indicating that there are two origins of spins. Probably one comes from interface and the other from “bulk” film. This means that the film has distorted interface phase. After the annealing, this doublet disappears, indicating that the distorted interface phase is rearranged.

Using FMR signals, we obtained  $4\pi M_{\text{eff}}$  and  $\Gamma_{\text{pp}}$  at 77 K on various samples to get relationship between the magnetic property and the deposition condition. They are plotted in Fig. 3 for the as-grown samples, and in Fig. 4 for the annealed samples. In the case of as-grown sample which shows the doublet (Fig. 3),  $4\pi M_{\text{eff}}$  and  $\Gamma_{\text{pp}}$  values are shown for the higher-field peak which corresponds to the “bulk” film part. Then their values can be compared with those for the annealed samples. For the as-grown samples,  $4\pi M_{\text{eff}}$  are larger for ML

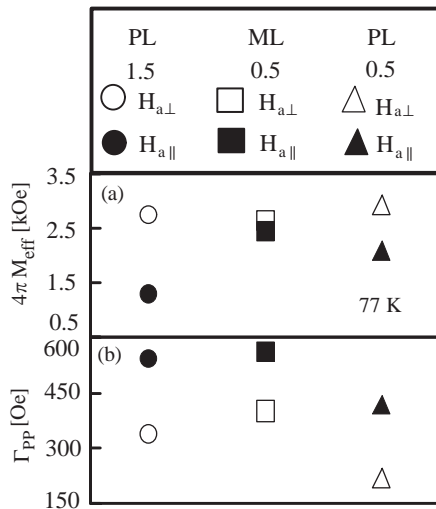


Fig. 4.  $4\pi M_{\text{eff}}$  and  $\Gamma_{\text{PP}}$  for the annealed samples.

(1.5 mTorr) than for PL (0.5 mTorr), while  $\Gamma_{\text{PP}}$  are smaller for ML than for PL as shown in Figs. 3(a) and (b). Then we can obtain the film with stronger ferromagnetic and more magnetically homogeneous nature by the conditions of ML (1.5 mTorr). The values of  $4\pi M_{\text{eff}}$  for  $H_{a\parallel}$  and  $H_{a\perp}$ , are larger for  $H_{a\parallel}$  than for  $H_{a\perp}$  for the both as-grown samples. The magnetization may be larger for the perpendicular field as observed in Fig. 4, then there are some origins which suppress the spin alignment to the perpendicular direction in the as-grown films. One of the origins might be the interface phase. The value of  $\Gamma_{\text{PP}} = 400$  Oe for  $H_{a\perp}$  for the sample with ML (1.5 mTorr) is considerably small, then the film is considerably homogeneous.

After the annealing, the magnitudes of  $4\pi M_{\text{eff}}$  are increased for  $H_{a\perp}$  as a whole (Fig. 4(a)), compared with those for as-grown samples, while their values for  $H_{a\parallel}$  are not increased or rather smaller than those before the annealing. Then the annealing has the particular effect on the anisotropic spin nature, that is, it rearranges the magnetic structure so that the spins become easier to be aligned to the perpendicular direction than to the parallel direction at low  $T$  of 77 K. The preparation condition which gives the largest  $4\pi M_{\text{eff}}$  for  $H_{a\perp}$  is PL (1.5 mTorr). However,  $4\pi M_{\text{eff}}$  for  $H_{a\parallel}$  of this annealed sample is as small as 1300 Oe. Then this preparation condition leads to the extreme magnetic anisotropy.

The magnitudes of  $\Gamma_{\text{PP}}$  are smaller for  $H_{a\perp}$  than for  $H_{a\parallel}$  for all the samples (Fig. 4(b)), which have the larger  $4\pi M_{\text{eff}}$  for  $H_{a\perp}$  than for  $H_{a\parallel}$  (Fig. 4(a)). Then the spins are easier to be aligned and more homogeneous for  $H_{a\perp}$  than for  $H_{a\parallel}$  after the annealing. The film prepared by PL (0.5 mTorr) has very small  $\Gamma_{\text{PP}} \sim 220$  Oe for  $H_{a\perp}$ . Then this film has the extremely homogeneous spins in the perpendicular field.

The data shown in this paper are preliminary results. We will report full data elsewhere.

#### 4. Summary

La–Ba–Mn–O thin films were grown by IBS with the oxygen molecular (ML) or plasma (PL) supply. The as-grown film shows  $T_C = 174$  K and  $T_P = 84.2$  K, then it is ferromagnetic but insulating between  $T_C$  and  $T_P$ . FMR properties are studied on the various samples before and after the annealing, in the fields parallel and perpendicular to the sample plane in relation to the preparation conditions. More detailed magnetic properties are now under investigation.

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