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Epitaxial growth of $La_{0.7}Ba_{0.3}MnO_3$ thin films on $SrTiO_3$ and $LaAlO_3$ substrates by metal-organic deposition process

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1. Introduction

The doped manganese oxides (the manganites) have been the focus of intensive research recently due to their remarkable colossal magnetoresistance (CMR) effect [1]. Compounds exhibiting CMR are half-metallic. As a consequence, they are a potential source of spin polarized carriers that might be used in spin injection studies [2]. The physical properties of the manganite thin film are sensitive to structure, oxygen content, and disorder, therefore, the Curie temperature (T_c) and resistivity of the film are somewhat different from those of the bulk. Consequently, the growth method, the deposition parameters, and also the substrate-induced strain will influence the properties. Understanding the strain is of particular interest since it can be used to advantage in tuning film properties, as has already been demonstrated in cuprates [3]. Alternative stacking of ferromagnetic $La_{0.7}A_{0.3}MnO_3$ (A= Ca, Ba, or Sr) with nonmagnetic insulating $SrTiO_3$ (STO) or $LaALO_3$ (LAO) provides an opportunity to study both magnetic coupling and spin-polarized tunneling. In these multilayers, the interface magnetism has a strong influence on the interlayer coupling (if present) and the spin-polarized tunneling properties.

In previous studies we have successfully prepared epitaxial thin films of $La_{0.7}A_{0.3}MnO_3$ (LCMO) with various thicknesses on different substrates using the metal-organic deposition (MOD) process [4, 5]. So this study is devoted to the epitaxial growth of $La_{0.7}Ba_{0.3}MnO_3$ (LBMO) thin films using the MOD process on $SrTiO_3$ and $LaAlO_3$ substrates. Microstructural and electrical properties of the obtained films were investigated.

2. Experimental

The starting solution was prepared by mixing the constituent metal-naphthenate solution (Nihon Kagaku Sangyo) and diluting with toluene to obtain the required concentration and viscosity. The molar ratios of La, Ba and Mn in the coating solution were 0.7, 0.3 and 1.0, respectively. This solution was spin-coated onto STO and LAO (001) substrates at 4000 rpm for 10 seconds. To eliminate the toluene, the metal-organic (MO) film was then dried in air at 100°C for 30 min. Before the final annealing, a preheating step at 500°C for 30 min is necessary to decompose the organic part. This preheating step is also required to prevent the formation of fissures on the film surface during the final thermal annealing at high temperature. To obtain a satisfactory film thickness for bolometric or spintronic applications, the above procedure (coating, drying, and preheating) was repeated several times (up to 4 times) giving rise to a corresponding number of superimposed layers in the LBMO product film. The final annealing was carried out in a conventional furnace at 1000°C for 60 min in air.

The cross-section transmission electron microscopy (XTEM) observations were performed using a high resolution electron Hitachi H-9000 microscope operated at 300 kV. The XTEM specimens were prepared following the conventional method, i.e., mechanical cutting, face-to-face gluing, mechanical grinding, polishing and dimpling, followed by Argon-ion milling at 4 KV. The resistance–temperature R/R_{300}–T (R_{300}: resistance value at 300 K) curves were measured by the usual DC four-probe method and by cooling the samples from 320 K to liquid nitrogen temperature (77 K).

3. Results and discussion

3.1 Microstructural properties

Figure 1 shows high-resolution transmission electron microscopy (HRTEM) images taken at the interfaces of the LBMO thin films grown on (a) STO and (b) LAO substrates. As can be seen, both LBMO films are epitaxially grown using the MOD process. The LBMO film grown on the STO substrate presents good epitaxial quality. However, the LBMO/STO interface is not so sharp, that's why we indicated it by the white line in the image (Fig. 1(a)). This is due to the very small lattice mismatch (-0.1%) between the LBMO film and the STO substrate. However, in spite of its high epitaxial quality the LBMO film grown on the LAO substrate contains interfacial misfit dislocations (denoted d_1 and d_2) along the interface. Two adjacent interfacial misfit dislocations were found to be separated by a constant distance D~12.2 nm. This distance is expected to be $D=a_1a_2/(a_1- a_2)$, where a_1=3.91 Å and a_2=3.788 Å which are the interplanar distances of the LBMO film and the LAO substrate. The calculated lattice mismatch is ~ -3.2% and requires one interfacial dislocations for every 12.14 nm spacing. The calculated value (12.14 nm) is approximately the same as the observed one (12.2 nm).

Using the MOD process, one deposited layer is 20 nm thick. Thus we prepared LBMO films of different thickness varying from 20 nm to 100 nm on both STO and LAO substrates.

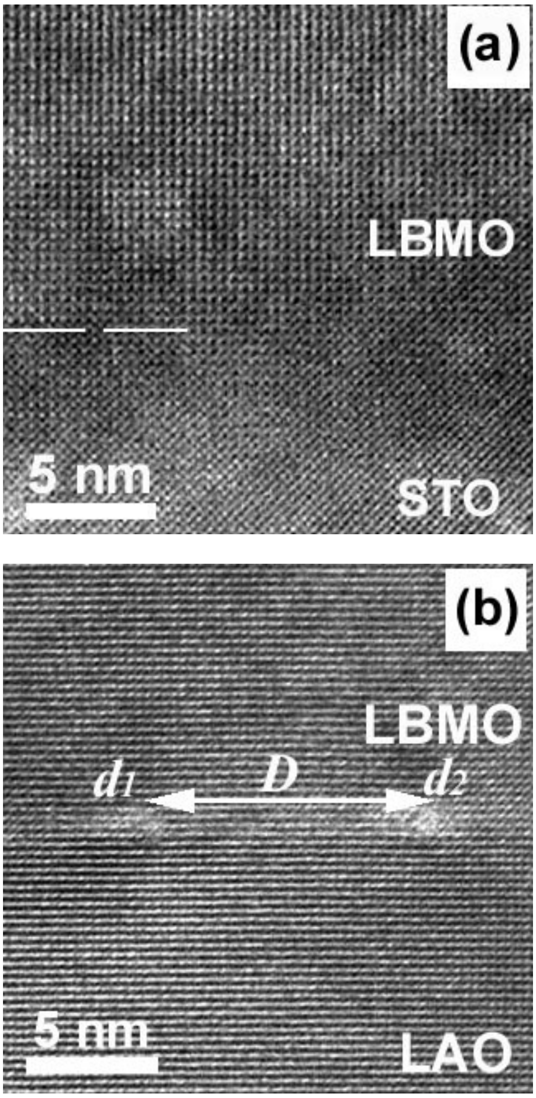

Figure 1: HRTEM images showing details of the LBMO films grown on (a) STO and (b) LAO substrates.

3.2 Electrical properties

Figure 2 shows the temperature dependence of the resistance (*R/R_{300}*) of the LBMO (2 times coating) films grown on the STO and LAO substrates and annealed at 1000°C for 60 min. Both LBMO films showed the typical characteristics curves of the CMR materials with the transition from the paramagnetic-insulator to the ferromagnetic-metallic state at the T_p temperature (close to the Curie temperature T_c). The T_p values are 320 K and 275 K for the LBMO/STO and LBMO/LAO films, respectively. As can be seen, the LBMO film grown on the STO substrate present the highest value of T_p which is in the same range of those reported elsewhere [6]. On the LAO substrate, the temperature peak (T_p) of the LBMO film is relatively low compared to that of the bulk material. This is probably due to the high lattice mismatch between LBMO and LAO. The misfit dislocations along the interface probably damaged the electrical properties (low T_p values).

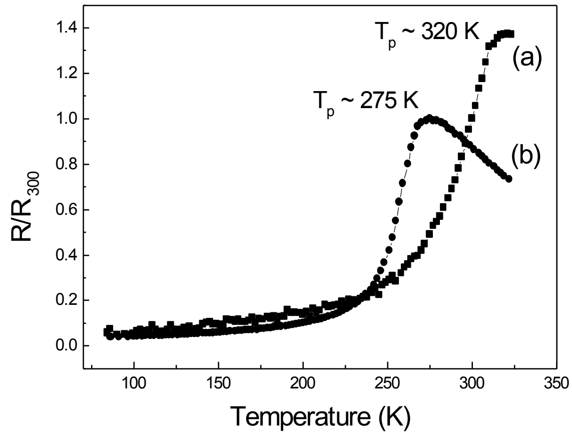

Figure 2: Temperature dependence of the resistance (*R/R_{300}*) of the LBMO (2 times coating) films grown on (a) STO and (b) LAO substrates.

4. Conclusion

We have successfully prepared epitaxial thin films of LBMO on STO and LAO substrates using the metal organic deposition process. The MOD is simple and low cost process and very useful for the epitaxial of metal oxides thin films. The microstructural and electrical properties of the obtained LBMO films were found dependent on the substrate material. On the high lattice mismatched substrate (LAO), the LBMO film presents interfacial misfit disloca-dislocations along the film/substrate interface. Heterostructures of the type LBMO/STO and LBMO/LAO prepared by the MOD process are very promising for spintronic applications.

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