Available online at www.sciencedirect.com

ScienceDirect

Physica B 396 (2007) 75–80

PHYSICA B

www.elsevier.com/locate/physb

Annealing effect on the properties of $La_{0.7}Sr_{0.3}MnO_3$ thin film grown on Si substrates by DC sputtering

D.R. Sahu[a,b,*], D.K. Mishra[b], Jow-Lay Huang[a], B.K. Roul[b]

[a]*Department of Materials Science and Engineering, National Cheng Kung University, Tainan 701, Taiwan*
[b]*Institute of Materials Science, Planetarium Building, Bhubaneswar 751013, Orissa, India*

Received 1 December 2006; received in revised form 13 March 2007; accepted 14 March 2007

Abstract

The effect of annealing on the properties of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin film deposited on Si substrates by DC sputtering has been investigated by X-ray diffraction (XRD), electrical and magnetic measurements. As-grown films show a lower metal–insulator transition (T_{MI}) temperature than annealed films. As the annealing temperature increases, significantly higher T_{MI} values are observed up to 270 K. We suggest that the increase of effective hole doping, induced by cationic vacancies due to the excess oxygen, is a possible reason for the observed trend in $T_{MI.}$ Annealing improves the magnetic homogeneity of the grain and grain boundaries. These improvements are favorable to enhance the intrinsic properties of the compound especially the decrease of resistivity. The decrease in resistivity induces the MR ratio to increase. This result is attractive for CMR application studies.

PACS.: 71.30. + h; 75.30. Kz; 75.30.Vn; 75.50.Kj; 75.70.pa

Keywords: Colossal magnetoresistance; LSMO; Sputtering; Magnetic properties and annealing

---

1. Introduction

The interesting interactions among charge, spin and lattice degrees of freedom make that perovskite-based colossal magnetoresistance (CMR) materials are very attractive for theoretical and experimental investigation. These materials have been extensively investigated in the past years, not only because of their rich physical properties but also because of their potential applications in various devices [1–6]. The CMR effects and the corresponding degrees of freedom of the magnetic structure, crystallographic structure and electrical resistivity provide a scope for engineering more sensitive magnetoresistive response.

The basic phenomenon of the CMR effects seems to be the same in bulk and in thin-film samples. However, due to the strain effect of the substrate or oxygen deficiency in a thin film, it is often difficult to reach the same properties as in the bulk [7–9]. Furthermore, it is also well known that the materials should be in the form of thin film in view with the practical application [10]. Although perovskite manganite thin films have been successfully grown on expensive single-crystal substrates such as $LaAlO_3$ and $SrTiO_3$, successful growth and optimization of these films on the mostly used silicon (Si) substrates, the essential materials of the semiconductor industry, are still needed. Though there are some studies on Si substrates [11–14], still several problems occur when Si substrates are used for perovskite manganite thin films: large difference in the thermal expansion coefficient, lattice mismatch and severe chemical reaction between Si and the deposited film layers [15,16].

It is also believed that oxygen content is important for the magnetic and electronic properties in these materials [17–19]. However, there is no quantitative relation between the oxygen content and the magnetic and transport properties in films because of the difficulty in the determination and control of the oxygen content. It was found that preparation conditions such as substrate

---

*Corresponding author. Tel/fax.: +886 6 2754410.
E-mail addresses: sahu@mail.ncku.edu.tw, diptirs@yahoo.com (D.R. Sahu).

0921-4526/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.physb.2007.03.016

temperature, oxygen partial pressure and deposition rate could affect the oxygen content in the resulting film severely [20,21]. Especially in the magnetron-sputtering method, the as-grown $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) films feature a reduction of oxygen content because of the low oxygen partial pressure during deposition [22]. Some previous studies [23–25] on the effect of annealing have been primarily focused on the role of the oxygen content since oxygen annealing on the perovskite manganite can lead to an increase of T_c and the saturation magnetization value.

In this paper, we have studied the effect of annealing on the properties of LSMO films grown on Si substrates by the DC-sputtering technique. The main advantage of the magnetron-sputtering method is its superiority in large-area deposition techniques compatible with microelectronics applications. Targets prepared from nanopowders synthesized by the chemical-solution technique [26] are used for the preparation of the films.

2. Experimental

Thin films of LSMO were grown on Si (1 0 0) substrates by DC-sputtering technique using nanosized powder of compacted target. LSMO target was prepared from the chemically processed nanopowders. Details of powder and target preparation were described elsewhere [26,27]. Highly dense and homogeneous CMR target of LSMO were taken for DC sputtering. The sputtering chamber was evacuated and purged with high-purity Ar and then O_2 separately. Low-power (100 W-max) DC sputtering was carried out using a gas mixture of 70% argon with 30% oxygen as the plasmagen gas with chamber pressure of 1.6×10^{-6} Torr. Mirror-polished and ultrasonically cleaned Si (1 0 0) substrates were taken for deposition. Temperature of the substrates was monitored at different temperatures inside the sputtering chamber. During film deposition, the substrate temperature was kept at 650 °C. In order to improve the oxygen content, the samples were annealed ex situ at 900 and 1000 °C temperatures in oxygen flow for 1 h. The heating and the cooling were done at 5 °C/min. The crystal structure of the LSMO films was studied by X-ray diffraction (XRD). Electrical transport properties were measured by a standard four-probe method and their magnetization properties were obtained using a vibrating sample magnetometer (VSM).

3. Results and discussion

XRD patterns of as-grown and annealed LSMO films on Si (1 0 0) substrate are presented in Fig. 1. It is noted from Fig. 1 that both the films are single phases without any impurity peaks. With the increase of annealing temperature, out-of-plane lattice parameter contraction is detected in the films. The c-axis out-of-plane lattice parameter for the as-grown film is 3.88 Å and the film annealed at 1000 °C is 3.83 Å. Similar types of XRD results were observed for the LCMO films grown on Si substrates [27]. The single-phase deposited LSMO film onto Si (1 0 0) has not been reported so far. Earlier report revealed that the deposited LSMO films have multiphase [28,29] even though films were formed epitaxially. Fig. 2 shows the cross-section TEM photograph of LSMO film annealed at 1000 °C. The TEM image indicated that there is a small inter-diffusion of Si and LSMO in the interface due to interfacial reaction during the annealing process.

The temperature-dependent resistances of as-grown and post-annealed LSMO films in zero fields are shown in Fig. 3. Film showed systematic increase in metal–insulator transition temperature (T_{MI}) with an increase in the post-annealing temperature. With increase of the annealing time, the film resistance decreases, while T_{MI} increases up to 270 K for 1000 °C annealed sample. Despite of small structural disorder, the oxygen incorporation improves the

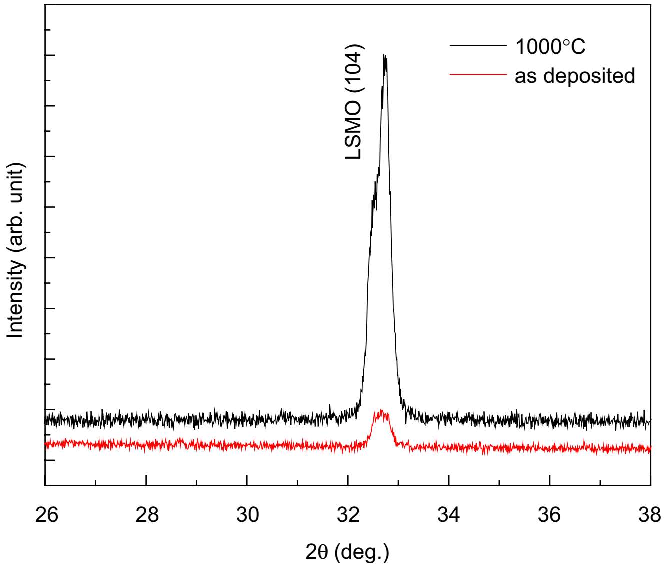

Fig. 1. The XRD pattern of as-grown LSMO film on Si substrates and film annealed at 1000 °C.

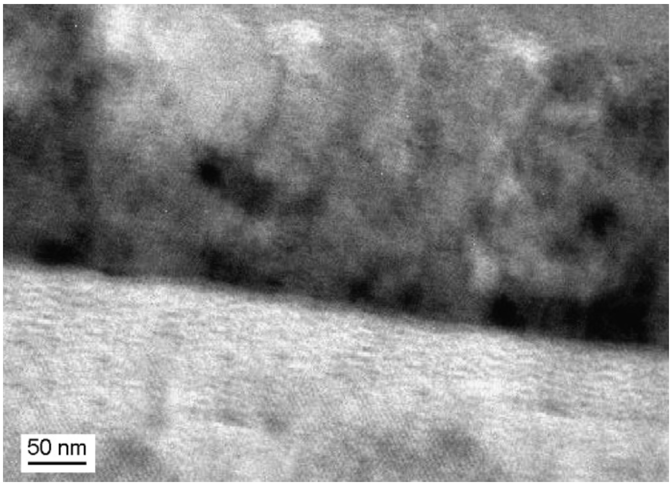

Fig. 2. Cross-section TEM photograph of LSMO film annealed at 1000 °C.