International Journal of Science and Research (IJSR)
ISSN (Online): 2319-7064
Index Copernicus Value (2015): 78.96 | Impact Factor (2015): 6.391

Interfacial Magnetic Coupling Effect in $LaNiO_3/LaCoO_3$ Bilayer Thin Films Prepared by Pulsed Laser Deposition

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Abstract: ***We report the synthesis of epitaxial individual and bilayer of $LaNiO_3/LaCoO_3$ (LNO/LCO) on (100) $LaAlO_3$ by pulsed laser deposition. The bilayers of LNO/LCO showed increase in magnetic properties with respect to isolated LCO or LNO films. We suggest that the origin of this effect is a strong Ni^{3+}-O-Co^{3+} exchange interaction at the interface. We also shown conducting behavior of bilayers. Our result demonstrates that a bilayer of LNO/LCO is able to produce multifunctional properties which can be useful in spintronic applications.***

Keywords: Interfacial effect, thin films, $LaNiO_3$, $LaCoO_3$, Pulsed laser deposition

1. Introduction

Thin films, multilayers and the phenomena associated to their interfaces have attracted the interest of scientific community due to the possibility of tunning their magnetic/electronic properties by epitaxial strain or by the electronic reconstruction induced at the contact between two different materials. For example, the ferromagnetic (FM) ordering exhibited below 85K in a biaxially strained $LaCoO_3$ thin films [1-2], is surprising given the diamagnetic behavior observed in bulk. A well-known phenomena characteristic of magnetic heterostructures is the exchange bias effect. In general, this interfacial effect involves ferromagnetic and non-ferromagnetic materials, although it has been reported at the interface between two different materials.

Nowadays, the electronic reconstruction at oxides interface is a topic of intense investigation, due to the appearance of unexpected properties in layered structures. R. Pentcheva et..al.[3] showed that besides extrinsic factors as oxygen vacancies and strains, the effect if charge re-accommodation in the atomic layers close to interfaces needs to be taken into account to understand some of the properties of oxide based nanostructures. The observation of exchange bias effect at the ferromagnetic/antiferromagnetic oxide-based super lattices was first reported a few years ago. In spite of the fact that there are still many open questions to solve in this matter. The interest in this effect and the exploration of the leading mechanism has been renewed in the last decades. The possibility of building artificial structure with controlled interfaces has allowed researchers to get a deeper insight in the characteristics and origin of the phenomena [4-5].

In this paper, we report the synthesis of an epitaxial individual and bilayer film of $LaNiO_3/LaCoO_3$ on (100) $LaAlO_3$ (LAO) by pulsed laser deposition.

2. Experimental

The $LaNiO_3$ and $LaCoO_3$ targets were prepared by the conventional solid-state reaction method employing La_2O_3, Ni_2O_3 and Co_2O_3 (Aldrich, [99 %). The reactants were mixed in deionized water medium using planetary ball mill and further calcined in the range 800-1000°C for 12 h. The phase formation was confirmed from the X-ray diffractogram which shows a well-crystalline with no evidence of any secondary phase. The calcined powders were pressed and sintered at the optimized sintering temperature of 1050°C to make a sputtering target of 2 inch in diameter and about 96 % density.

LNO/LCO bilayers were deposited on (100) single crystal LAO substrate by pulsed laser deposition at pure O_2 pressure on the order of 10^{-3}Torr and a substrate temperature of 750°C. An excimer laser KrF operating at 248nm, 5Hz and 20-50 mJ was used. In our studies, LCO thickness was chosen around 40nm and LNO thickness 20nm. X-ray diffraction measurements also showed highly textured layers with preferential growth in thea-axis direction. The magnetic loops and temperature verses magnetic field measurements were carried out in a Quantum Design Physical property measurement system (QD-PPMS-6600) in the range 5 to 300 K. The electrical resistance was measured as a function of temperature using a standard dc four point probe. Before measuring the electrical properties, a Hall bar was defined using ion etching and a stencil mask Ti/Au (5nm/100nm) was deposited to ensure Ohmic contacts.

3. Results and Discussion

The XRD patterns of LNO, LCO and LNO/LCO bilayer films grown on LAO (100) substrate is shown in figure 1 (a-c) respectively. Only (l00) reflections are observed, demonstrating the orientated growth of the films; no secondary reflections of the impurity or missorientations are observed. These symmetric reflections are fitted in a pseudocubic indexation to obtain the out of plane pseudocubic a-axis parameter of the films. All the films are identified to be pseudocubic crystal structures whose

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strong (100), (200) and (300) peaks can be observed. The crystallite size is estimated by the Scherrer's equation [6].

$$D=\frac{k\lambda}{\beta\cos\theta} \quad (1)$$

Where D is the particle diameter, λ is the X-ray wavelength, β is the full width half maximum of the X-ray diffraction peak θ is the diffraction angle and k is the Scherrer constant and set as 0.89 here. The crystallite size of the LNO, LCO and LNO/LCO thin films are 22, 17 and 20 nm respectively.

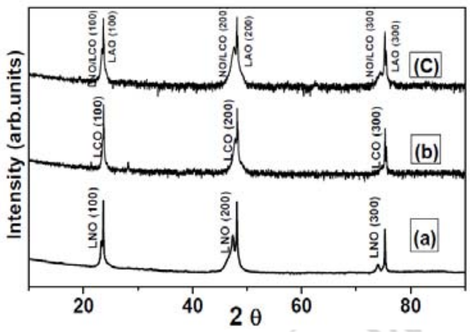
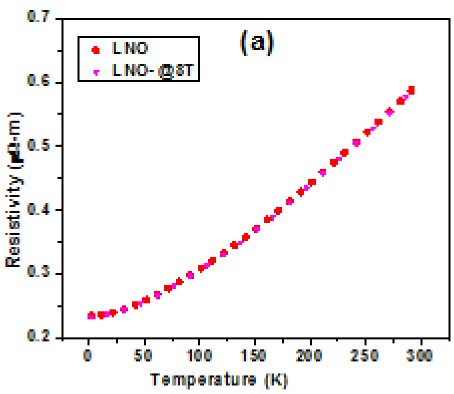
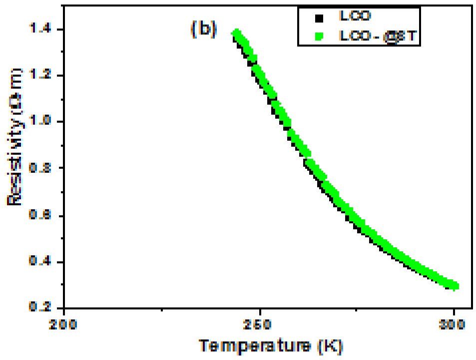


Figure 1(a-c): XRD patterns of the (a) LNO (b) LCO (c) LAO/LNO/LCO thin films

The temperature dependence of the electrical resistivity of LNO, LCO and bilayer thin films were shown in fig 2 (a-c). LNO thin films exhibited the metallic nature as temperature increases. The value of the electrical resistivity of LNO film is 0.5 μΩ-m at 300K. Whereas, the resistivity of LCO films decreased as the temperature increased this is nature of semiconductor materials. Interestingly, bilayer films observed as metallic behavior which is increasing the resistivity as the temperature increased. The values of resistivity are almost equal to pure LNO films (0.45 μΩ-m). There is no metal-insulator transition observed in this range which indicates La-deficient is not there in deposited films [7]. This ensures that LNO and LCO films must be very close to its stoichiometric compositions. The more resistive behavior of LCO layer is consistent with the observation of less resistivity of bilayer, with respect to LNO film. Therefore, although the top layer of LCO shows insulating behavior, bilayer films exhibited the good enough transport properties.





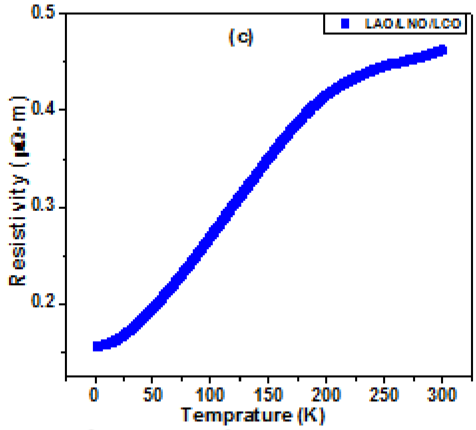


Figure 2 (a-c): Electrical resistivity as function of temperature of the (a) LNO (b) LCO (c) LNO/LCO bilayer films

The magnetization response as a function of temperature of the LNO, LCO and bilayer films are shown in fig. 3 (a-c). LNO films exhibited the paramagnetic behavior throughout the temperature. There is clear magnetic transition observed at 83 K in LCO films which is close to ferromagnetic transition temperature reported for epitaxial grown thin films [8]. In bilayer also we observed the same transition which is corresponds to ferromagnetic transition of LCO films. The magnetic hysteresis loops at 40K for both LCO and bilayer is displayed in figure 4. The pure LCO films exhibited the low saturation magnetization and high coercive field (1.32 emu/cm^3, 3368 H). In case of bilayer, the high saturation magnetization and less coercive filed (3.76 emu/cm^3, 2215 H). This could be related to an interfacial Ferromagnetic coupling of the LNO and LCO layer. However, the coercive remains high above the Curie temperature of LCO films, suggesting that the effect comes from a direct atomic coupling between Ni-O-Co at the interface. It is known that Co^{2+} in an octahedral environment still shows a pronounced orbital magnetic moment and a large magnetic anisotropy due to spin-orbit coupling. At the interface, Ni^{3+} ions will be in contact with Co^{3+}. We suggest that there would be a few unit cells across the interface in which strong ferromagnetic interaction and possible redox reactions of the form Ni^{3+}-O-Co^{3+} which leads to improve its original compound properties.

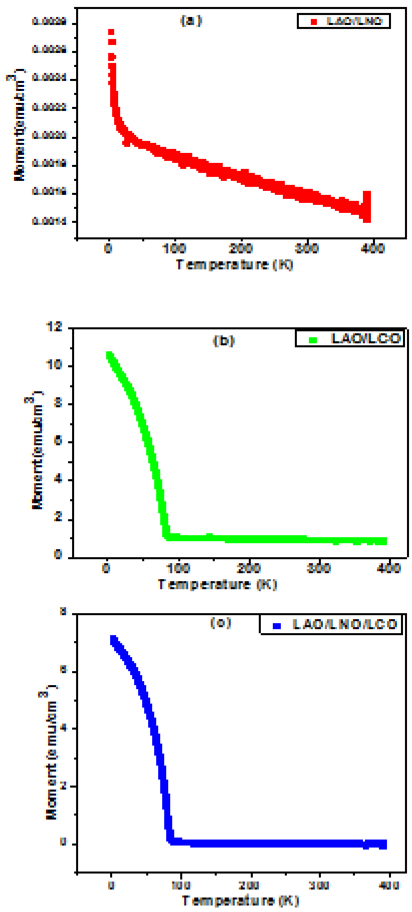


Figure 3 (a-b): Magnetization curves as a function of temperature (a) LNO (b) LCO (c) LNO/LCO bilayer thin films

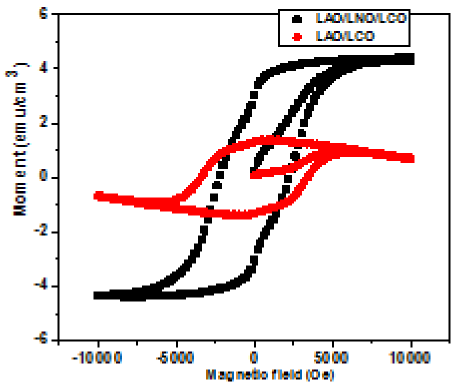


Figure 4: M-H loops of LCO and bilayer thin films

4. Conclusions

In summary, the epitaxial bilayers of different oxides were grown on LAO (100) by pulsed laser deposition. XRD confirms the crystallinity of the films. The bilayer films exhibited the multifunctional properties. We suggest that this could be due to redox reaction and subsequent exchange interaction occurring at the interface. This kind of multifunctional property materials will be useful for many electronic and magnetic applications.

Acknowledgement

The authors gratefully acknowledge the financial support from Jawaharlal Nehru Centre for Advanced Scientific Research of DST.

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