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Investigation of Heterostructure Formed from Hole- and Electron-Doped Lanthanum Manganites

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High crystalline quality films of n - $\text{La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3$, p - $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ and related p - n diode structures were grown heteroepitaxially on lattice-matched $\text{SrTiO}_3(100)$ substrates by dc magnetron sputtering and pulsed laser deposition. The $\text{La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3/\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ bilayer was patterned into a strip-like geometry to investigate electrical properties of the interface. Significant magnetoresistance values and nonlinear current-voltage characteristics were indicated for the interface of the p - n diode heterostructure.

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1. Introduction

Hole-doped perovskite manganites referred to by general formula $\text{Re}_{1-x}\text{A}_x\text{MnO}_3$ ($\text{Re} \equiv \text{La}, \text{Nd}$; A is a divalent alkaline-earth element: Ca, Sr, Ba) have been widely investigated in recent years due to their interesting properties such as transition from high resistance paramagnetic (PM) to a metallic ferromagnetic (FM) state, spin-polarized carriers below Curie temperature, T_c , and colossal magnetoresistance (CMR) values measured in the vicinity of T_c . [1]. Recently it has been found that electron-doped manganites exhibiting similar electrical and magnetic properties may be obtained via substituting La^{3+} by tetravalent cations such as Ce and Sn [2]. Thin films of electron-doped single-phase $\text{La}_{1-x}\text{Ce}_x\text{MnO}_3$ (LCeMO) have been grown recently by pulsed laser deposition although a number of investigators pointed out difficulties of introducing Ce ions into LaMnO_3 system in bulk samples.

In this work, we report high quality films of electron-doped $\text{La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3$ and $p\text{-La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3/n\text{-La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3$ diode structures which are promising for various spintronics applications [3].

2. Preparation and characterization of the films and heterostructures

Thin films of n -type $\text{La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3$, hole-doped $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ (LCaMO) with a thickness ranging from 100 to 400 nm, and the LCaMO/LCeMO diode structures were grown heteroepitaxially at 700–800°C on lattice-matched crystalline $\text{SrTiO}_3(\text{STO})(100)$ substrates by applying dc magnetron sputtering (MS) and pulsed laser deposition (PLD). Disk-shaped ceramic LCeMO and LCaMO targets (3 cm in diameter) with La/Ce, and La/Ca ratios (3:1) were used. After deposition, the films and heterostructures were saturated in oxygen followed by slow cooling down to room temperature. The crystalline structure of the prepared films and the LCaMO/LCeMO heterostructure were characterized by X-ray diffraction (XRD) and reflected high-energy electron diffraction (RHEED).

The LCaMO/LCeMO bilayer was patterned in a cross-stripe geometry to investigate electrical properties of the prepared heterostructure. The underlying strip-like LCaMO film (200 μm in width) was patterned by applying a conventional lithography followed by chemical etching. The tape-like LCeMO film (1 mm in width) was subsequently deposited by PLD at 750°C onto the patterned LCaMO layer through a mask. Low resistance electrodes used for electrical measurements were prepared by sputtering Ag onto the films and heterostructures. Electrical resistance, magnetoresistance, and voltage versus current were measured at $T = (78 \div 300)$ K.

3. Results and discussion

X-ray diffraction and reflected high energy electron diffraction investigations revealed epitaxial quality of the LCaMO, LCeMO films and the LCaMO/LCeMO bilayer structures when grown on lattice-matched SrTiO_3 ($a = 0.3905$ nm). The off-plane lattice parameter of a pseudocubic unit cell of about 0.387 nm and 0.394 nm have been estimated from XRD measurements for the LCaMO, LCeMO films, respectively. Results concerning structure and electrical properties obtained in this work for LCaMO are in consistence with our earlier studies [4, 5].

In Fig. 1 we show resistance versus temperature for epitaxial LCeMO films of various thicknesses ($d \cong 200, 300, \text{ and } 400$ nm) at $B = 0$ (points) and $B = 1.2$ T (dotted lines) grown by PLD under the same deposition conditions ($T_s = 750^\circ\text{C}$). Resistivity values ranging from about 3×10^{-3} to 5×10^{-3} Ω cm (at $T = 300$ K) have been estimated for the films. The characteristic resistance peaks seen in the figure and a slight shift of the peak positions to higher temperatures with an applied magnetic field demonstrate clear evidence of paramagnetic to ferromagnetic transition. A slight increase in T_c seen in Fig. 1 with a film thickness decreasing is not clear as yet. Certainly, an increase in T_c with film

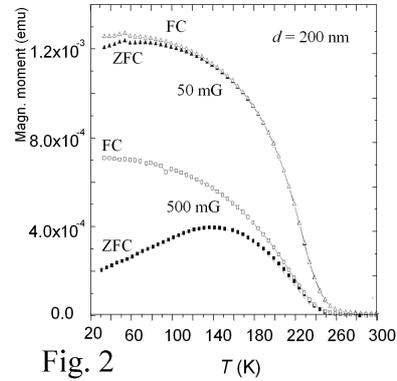
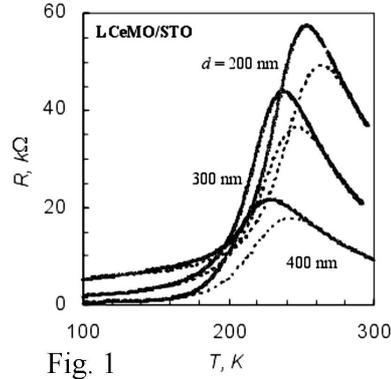


Fig. 1. Resistance versus temperature of epitaxial $\text{La}_{0.67}\text{Ce}_{0.33}\text{MnO}_3/\text{SrTiO}_3$ films of various thicknesses ($d \approx 200, 300,$ and 400 nm) measured at $B = 0$ (points) and 1.2 T (dotted lines).

Fig. 2. Low-field magnetization ($B = 50$ and 500 mG) of $\text{La}_{0.67}\text{Ce}_{0.33}\text{MnO}_3/\text{SrTiO}_3$ film with a thickness $d \approx 200$ nm measured in a field cooling (FC) and zero field cooling (ZFC) regimes.

thickness is usually observed either for Sr or Ca doped manganite films [4]. Figure 2 shows a typical magnetization versus temperature for the LCeMO/STO film ($d \approx 200$ nm). The measurements were carried out during film heating with an applied magnetic field ($B = 50$ and 500 mG) and without magnetic field using a SQUID magnetometer.

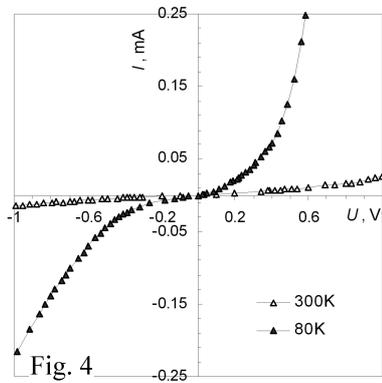
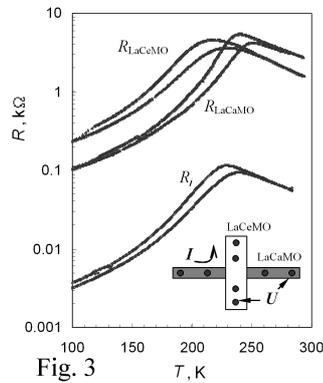


Fig. 3. Resistance versus temperature of the $\text{La}_{0.66}\text{Ca}_{0.34}\text{MnO}_3$ and $\text{La}_{0.66}\text{Ce}_{0.34}\text{MnO}_3$ films of the patterned bilayer structure and that of the $\text{La}_{0.66}\text{Ca}_{0.34}\text{MnO}_3/\text{La}_{0.66}\text{Ce}_{0.34}\text{MnO}_3$ interface. The sample used for vertical electrical transport (interface) investigation is shown in the inset.

Fig. 4. Current versus voltage of the $\text{La}_{0.66}\text{Ca}_{0.34}\text{MnO}_3/\text{La}_{0.66}\text{Ce}_{0.34}\text{MnO}_3$ interface measured at 300 K and 80 K for the LCaMO/LCeMO bilayer film structure patterned in cross-stripe geometry.

Results of electrical measurements for the LCaMO/LCeMO bilayer structure patterned in cross-stripe geometry are presented in Fig. 3. This geometry allows one to study vertical electrical transport between the adjacent n - and p -type manganite layers in order to investigate resistance of the interface, R_I . It can be seen from this figure that both the LCaMO and LCeMO films and their interface exhibit a similar resistive transition at 235 K, 210 K, and 215 K, respectively. Both the intermediate position of the PM–FM transition temperature corresponding to the interface and the relatively low R_I values show the absence of high resistance interlayer and a possible oxygen outdiffusion from the interface. Furthermore, an increase in R_I with a magnetic field ($B = 1.2$ T) in the vicinity of T_c demonstrates a significant magnetoresistance of the p – n manganite interface. Figure 4 demonstrates nonlinear I – U characteristics of the LCaMO/LCeMO interface at $T = 78$ and 300 K as well as asymmetry of the I – U characteristics in respect to current direction which is typical of p – n junctions.

Summarizing results of our investigations we conclude that electrical properties of the $\text{La}_{2/3}\text{Ce}_{1/3}\text{MnO}_3/\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ bilayer structure, namely, magnetoresistance and nonlinear electrical properties of the interface as well as the observed asymmetry of the I – U characteristics may be promising for various applications.

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