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Electroresistance of Electrically Nonhomogeneous $La_{0.67}Ca_{0.33}MnO_3$/MgO Thin Films

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Current and electrical field-induced electroresistive effects were investigated for $La_{0.67}Ca_{0.33}MnO_3$/MgO thin films demonstrating nanosized electrical inhomogeneities. Two different models based on enhanced conductivity of intergrain boundaries by injecting spin-polarized carriers from ferromagnetic grains and electrical field-enhanced hopping of carriers in high resistance intergrain media were carried out to explain nonlinear electrical properties of the films.

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1. Introduction

$La_{0.67}Ca_{0.33}MnO_3$ (LCMO) thin films exhibiting paramagnetic to ferromagnetic (PM–FM) transition demonstrate a colossal magnetoresistance (CMR) phenomenon and interesting electrical transport properties. The importance of intrinsic inhomogeneity of the films in a form of co-existing FM and PM phases below the PM–FM transition temperature T_{m} (phase separation phenomenon) for electrical properties of the films has been reported recently [1, 2]. It was found that electrical properties of polycrystalline manganite films containing nanoscale inhomogeneities such as grain boundaries are different from those of high crystalline quality (epitaxial) films. A significant film resistance change with applied voltage ("electroresistance" ER) [3] and applied current (current-induced electroresistive effect) [4] as well as an effect caused by nonuniform Joule heating [5, 6] have been reported recently for electrically inhomogeneous manganite films.

In this work we were focussing on nonlinear electrical properties of structurally and electrically nonhomogeneous textured LCMO films deposited on cleaved (100) faces of MgO crystals. It was our goal to elucidate an influence of various factors as the presence of inhomogeneities, intergrain boundaries, and a possible heating effect on nonlinear electrical properties (electroresistance) of the prepared LCMO films.

2. Sample preparation

MgO substrates demonstrating a significant lattice mismatch (of about 9%) in respect of LCMO lattice were used in this work to introduce possible structural and electrical inhomogeneities in the grown films to reveal the ER effect [3]. The films were fabricated by pulsed laser deposition at $T = 750°\text{C}$ under a fixed oxygen pressure on cleaved (100) faces of MgO crystals. After the deposition, oxygen pressure was increased up to 1 atmosphere and the films were cooled down to room temperature during 3 hours.

$\theta - 2\theta$ X-ray diffraction (XRD) scans measured for the films revealed single--phase material with preferential (100)-texture. Relatively wide XRD reflexes of (100) type certified growth of the material with slightly misoriented grains.

The surface of both the grown films and the substrates was studied by atomic force microscope (AFM). The AFM image of cleaved MgO(100) faces revealed a terrace-like structure with a typical step height ranging from 5 to 25 nm. The AFM image of the grown LCMO film cut (at 200 nm height) in Fig. 1 demonstrates a columnar structure with nanosized grains appearing during deposition. A typical height of the grains was in the range of 250–300 nm. The prominent parts of the grains formed a "rocky" structure in accordance to the observed terrace structure of the substrate surface. Thus, following Fig. 1 we point out significant structural inhomogeneities of the grown films induced by various factors, namely by a significant (9%) lattice mismatch, relatively fast deposition process, and nanostructured step-like substrate surface.

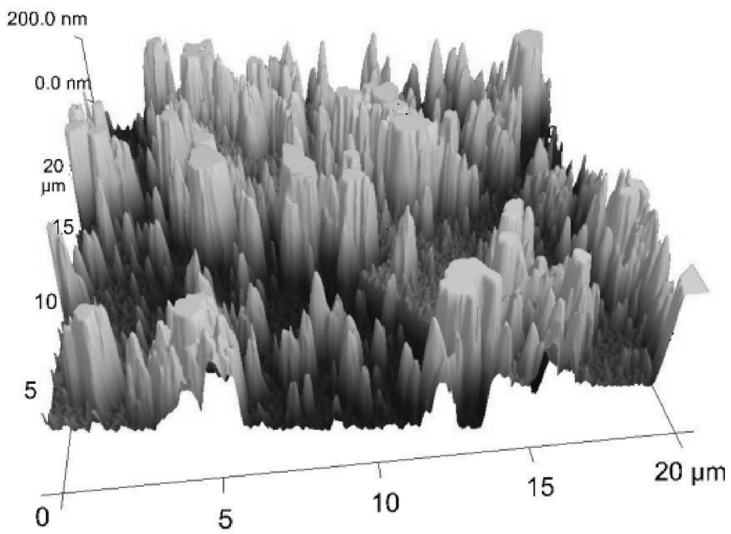

Fig. 1. Sample 1 film surface image of 200 nm height cut obtained by AFM. Area dimensions: $20 \times 20\ \mu\text{m}^2$.

TABLE

Preparation conditions and parameters of the prepared LCMO films.

Sample No.	Deposition temperature [°C]	PM–FM transition temperature [K]	Film average thickness [nm]	Film width, w [mm]	Gap width, d [μm]
1	750	150	120	1	1750
2	750	135	40	0.5	20

Ag coatings were magnetron sputtered onto the tape-like LCMO films to prepare electrical contacts of width w for electrical measurements. Two different gaps between the electrodes ($d_1 \approx 1750$ μm and $d_2 \approx 20$ μm) were prepared to elucidate an influence of current and strong electrical field on electroresistance values of the films. Parameters of the samples used for the investigations are displayed in Table.

3. Experimental

The samples were placed in a broken microstrip line attached to a holder with radio frequency connectors to investigate electrical properties of the films in a wide temperature range. Electrical resistance of the films was measured by using both dc current and nanosecond electrical pulses. The transport characteristics were measured at various temperatures by using combined experimental setup described in [7] and applying nanosecond electrical pulses with 30 ns in duration and repetition rate of 100–150 Hz. The dc circuit was connected to the transmission line through broad band bias tees with blocking capacitors. A regulated voltage source and variable resistor were used for sample current stabilization. Sample resistance was calculated using ampermeter and voltmeter indications.

4. Results

Figure 2 shows resistance versus temperature (R–T) dependence of sample 1 measured at several dc current values. Let us note a significant decrease in film resistance at low temperatures ($T < 150$ K), i.e. below the characteristic PM–FM transition temperature (T_{m}) and a slight shift of the characteristic resistance maximum temperature T_{m} (from 150 K up to about 160 K) with dc current increasing.

The R–T dependences measured at several dc current values for the film 2 are shown in Fig. 3 (curves 1, 2, 3). The curves 4 and 5 displayed in the same figure for comparison were obtained for the same film by applying pulsed electric field of about 1.9 kV/cm and 3.5 kV/cm, respectively. Following this figure we point out the slight shift of T_{m} value with dc current increasing. However, in this case, it is important to note the significant resistance decrease in a wide temperature

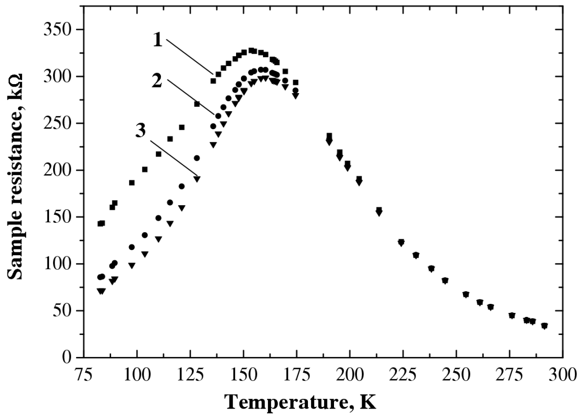

Fig. 2. Resistance versus temperature of sample 1 measured at fixed dc current values: $I = 5$ μA (1), 30 μA (2), and 50 μA (3).

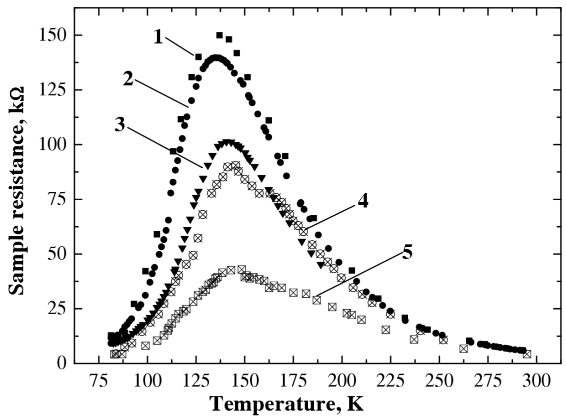

Fig. 3. Resistance versus temperature of sample 2 measured at fixed dc current values: $I = 1$ μA (1), 5 μA (2), 50 μA (3) and by applying pulsed electric field of 1.9 kV/cm (4) and 3.5 kV/cm (5).

range (both below and above the characteristic PM–FM transition temperature) with increasing dc current and especially by applying pulsed electrical field.

5. Discussion

In recent works [3, 4, 8], several models based on current flow through conducting channels (network of filaments) in nonhomogeneous media were carried out to explain magnetic and electrical transport properties of various manganite films. To discuss a role played by various factors as current, electrical field intergrain boundaries in nonlinear properties (electroresistance) of the prepared LCMO films let us estimate the values of mean electrical field (E_d) defined as V/d (here d is the gap between electrodes and V is the corresponding voltage drop). In a case of dc current, the highest E_d values (corresponding to the resistance maxima at T_{m}) for film 1 ($E_d \leq 42$ V/cm) were significantly lower compared to those for the film 2 ($E_d \leq 65$, 325, and 3250 V/cm corresponding to dc current $I = 1$, 5, and 50 μA, respectively).

Taking into account relatively low E_d values for film 1, we assume that resistance variation seen for the film from Fig. 2 should be associated with current-induced effects. At the same time, the significant resistance change for the film 2 (seen from Fig. 3) should be caused mainly by strong electrical field. Further, we assume the presence of both structural and electrical inhomogeneities of the LCMO/MgO films to explain the significant ER values. We believe that in the manganite films containing a great number of misoriented grains, current should flow through highly conductive FM grains separated by high resistance intergrain boundaries.

A slight resistance decrease seen from Fig. 2 for the film 1 with current increasing at $T < T_{\mathrm{m}}$ may be easily understood assuming enhanced conductivity (metallization) of high resistivity media at the intergrain boundaries by injection of spin-polarised carriers from the adjacent low resistance FM grains [4]. Within this model, one can also explain why film resistance does not depend on current at $T > T_{\mathrm{m}}$, i.e. above the PM–FM transition temperature.

It can be seen from Fig. 3 that a similar resistance change and the corresponding R–T curves have been measured for the film 2 in a wide temperature range (both above and below the PM–FM transition temperature) by applying dc current (curve 3) and short electrical pulses (curve 4). Thus, we avoid importance of possible sample heating and concentrate on a key role of electrical field effect in the measured ER values of film 2.

Most of the manganites including LCMO demonstrate semiconductor-like R–T dependence at $T > T_{\mathrm{m}}$ (PM-phase) due to thermally-assisted hopping of carriers between Mn^{3+} and Mn^{4+} states (variable range hopping (VRH) model). A role of strong electrical field in electrical resistivity of the films can be easily understood within the same (generalised) VRH model [9] taking into account electrical field-enhanced tunnelling of carriers between neighbouring Mn^{3+} and Mn^{4+} ions. In a case of nonhomogeneous LCMO films, local electrical field in high resistivity intergrain regions should be significantly higher compared to the average electrical field E_d and thus, for nonhomogeneous films one can expect a stronger ER effect. The highest electroresistance values ($ER = [R(E) - R(0)]/R(0)$, $R(E)$ is sample resistance, E is the mean electrical field value and $R(0)$ is sample resistance at zero E) of about –58% measured for the film 2 at $T = 110$ K ($< T_{\mathrm{m}}$) may, probably, be explained taking into account joint current and electrical field effects on resistivity of the material at the intergrain boundaries.

6. Conclusions

It was found in this work that textured $La_{0.67}Ca_{0.33}MnO_3$ films exhibiting a great number of misoriented grains and intergrain boundaries demonstrate a significant ER effect depending on both applied voltage and current. The current--induced ER effect observed at low electric field (< 50 V/cm) below the PM–FM transition temperature was explained assuming reduced resistance of intergrain boundaries due to injected spin-polarized carriers from low resistance FM grains. At the same time, the origin of ER effect caused by strong electrical field ($E \approx 0.5$ kV/cm) was associated with electrical field-enhanced hopping of carriers in high resistance intergrain media.

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