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Dielectric permittivity and conductivity spectra of $La_{0.5}Ca_{0.5}MnO_3$ thin films presenting electric field induced metal–insulator transition

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ABSTRACT

Electric behavior of polycrystalline $La_{0.5}Ca_{0.5}MnO_3$ thin films, pulsed laser deposited on a (100) silicon substrate, is reported and discussed. An electrically induced metal-insulating transition around 150 K is found, which is voltage and thickness dependent. At low temperatures, the film conductivity is non-Ohmic and moderate electric fields lead to resistivity switching towards metaestable low-resistive states. Impedance spectroscopy measurements were also performed in order to determine the film dielectric permittivity and conductivity and to estimate the characteristic metal–semiconductor interface parameters. The obtained results show that the fraction of ferromagnetic metallic regions does not change when a voltage is applied, and that the mechanism responsible for the low temperature metal insulator transition and the conduction behavior is the appearance of connective paths due to the enhanced mobility of carriers activated by the electric field.

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1. Introduction

Since the 1950s, it was established that $R_{1-x}A_xMnO_3$ (R is a rare earth and A an alkali metal) displays great sensibility to different perturbations, such as external magnetic and electric fields, dimensionality, crystalline disorder, etc. [1]. The anomalously high magnetoresistance of LaCaMnO, reported in 1994 by Jin et al. [2], renewed the interest of studying these doped perovskites, mostly motivated by their potential use in field sensors and electronic devices.

In manganites, different electronic and magnetic phases: charge-ordered insulator (COI), paramagnetic, ferromagnetic metallic (FMM), coexist within a single crystal. This phase coexistence, together with the behavior and competition of the COI and FMM phases at different temperatures, determine the transport properties of these materials. Although the thermodynamically stable phase depends on temperature, Levy et al. [3] reported that the relative fraction of these two phases can be "tuned" changing the grain size of the manganite powder controlling, thereby, the electric conductivity.

The $La_{0.5}Ca_{0.5}MnO_3$ is the less studied manganite, probably due its exotic phase diagram. It shows a FMM transition at Tc = 225 K and a charge-ordered antiferromagnetic transition at Tco = 155 K, although coexistence of both phases has been demonstrated by Loudon [4]. This author found micron sized ferromagnetic regions spanning several grains coexisting with similar sized regions with no local magnetization. The competition between these phases leads to extraordinary transport properties since the spatial distribution of the ferromagnetic phase can lead to a percolative or a non-percolative behavior. In the former case the electric behavior will be metallic with electronic transport through a network of conducting paths within an insulating matrix.

Spectacular manifestations of electric field effects in perovskite manganites are the colossal electroresistance effect and the strong current-induced sudden resistivity jumps [5–7]. As a consequence, manganites exhibit the electric pulse-induced resistance switching (EPIR) effect, which belongs to a group of semiconductor oxides with memory properties [8–10]. This effect was interpreted by Rozenberg et al. [11] assuming the existence of three different domains between the electrodes, each one having different electrical properties. The small domains located near the electrode–semiconductor interfaces could undergo a metal–insulator transition (MIT) that has a significant impact on the conductance of the whole system.

In the present work we pay special attention on studying the possibility of MIT in $La_{0.5}Ca_{0.5}MnO_3$ films and on the electrical properties of the electrodes. We report electric impedance measurements providing information on the whole system: bulk material and semiconductor–electrode interfaces. Broad frequency range impedance spectroscopy was used to separate these contributions in order to determine the dielectric permittivity and conductivity of manganite and to estimate the characteristic metal–semiconductor interface parameters.

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2. Experimental details

$La_{0.5}Ca_{0.5}MnO_3$ films of different thickness were deposited on (100) Si substrates using pulsed laser deposition. A Nd:YAG laser beam, operating at 266 nm and 10 Hz, was focused on a rotating target of stoichiometric composition to yield an energy density of $\approx$2.5 J/cm^2. Film deposition was carried out at 700 °C and 10 Pa oxygen pressure. After deposition, the ablation chamber was back-filled with O_2 at atmospheric pressure and the sample was cooled to room temperature at a rate of $\approx$10 °C/min. X-ray diffraction patterns of the obtained films showed a polycrystalline nature with a slight texture in the (100) direction. Elipsometric and spectrophotometry techniques were used to measure the thicknesses of the five films used in this work: 37-nm; 57-nm; 77-nm; and 100-nm for samples deposited on Si substrates and a 200-nm sample deposited on a gold coated Si substrate.

DC electrical measurements were made using a two-probe scheme since the resistance values exceeded $10^9\,\Omega$. Round 1.5 mm diameter Au electrodes were sputtered on top of each film and Cu wires were soldered using Ag epoxy. A fixed voltage was applied across these wires and the current flow was monitored by measuring the voltage drop across a metal film resistor. This scheme allows a sufficiently high stabilization of the DC voltage, which is not affected by dramatic changes of resistivity at low temperatures. Leakage current values were checked to be always below the values of the measurement currents.

In order to separate the bulk semiconductor from the semiconductor–electrode interface contributions to the electrical conductivity, impedance spectroscopy measurements were performed at 20 °C in the 100 Hz to 10 MHz frequency range, using a Hewlett Packard 4192 A Impedance Analyzer under computer control. The 200-nm sample deposited on a 5×5 mm gold coated substrate was used for this experiment. The AC voltage was applied between the bottom gold coating and the top electrode (see Fig. 3a). The sample was connected to the instrument by means of the HP 16047 C Test Fixture.

3. Metal–insulator transition induced by electric field

A series of measurements of the temperature and thickness dependence of the electric resistance under different voltages applied across the top electrodes were performed. Fig. 1a, b, c and d shows the dependence of the normalized resistance with temperature for the different film thicknesses. In all cases, the cooling and heating rate values were 2 K/min while the applied voltage values were: 1, 2.7, 30, and 50 V, which correspond to electric fields of around: 4, 11, 120 and 200 V/cm, respectively. The thicker films (77 and 100 nm) exhibited an insulator behavior under low applied voltages: 2.7 and 1 V (not shown in Fig. 1). However, a metal–insulator transition was observed for 30 and 50 V. Below the transition temperature, the resistance as a function of temperature is almost constant over a wide temperature range. The thinner films presented huge resistances at low temperatures and did not display a M–I transition for the voltage values used in this work. However, a significant change in the slope of the *R*–*T* curves was observed for the highest 30 and 50 V applied voltages.

Zero field cooling (ZFC) and field cooling (FC) magnetization measurements performed on the thicker samples (see inset in Fig. 2 for the 200 nm sample) show that the paramagnetic to ferromagnetic transition occurs in the 233 to 160 K temperature range. In this work, the MIT transitions occur near 160 K for the thicker samples and the larger resistance instabilities occurs in the 50 to 160 K temperature range, which is coherent with a phase separated system. Furthermore, hysteretic behavior in this range of temperatures for similar samples was already reported by our group [13].

It was established [14] that at low temperatures a competition between double exchange and Jahn–Teller effects generally leads to

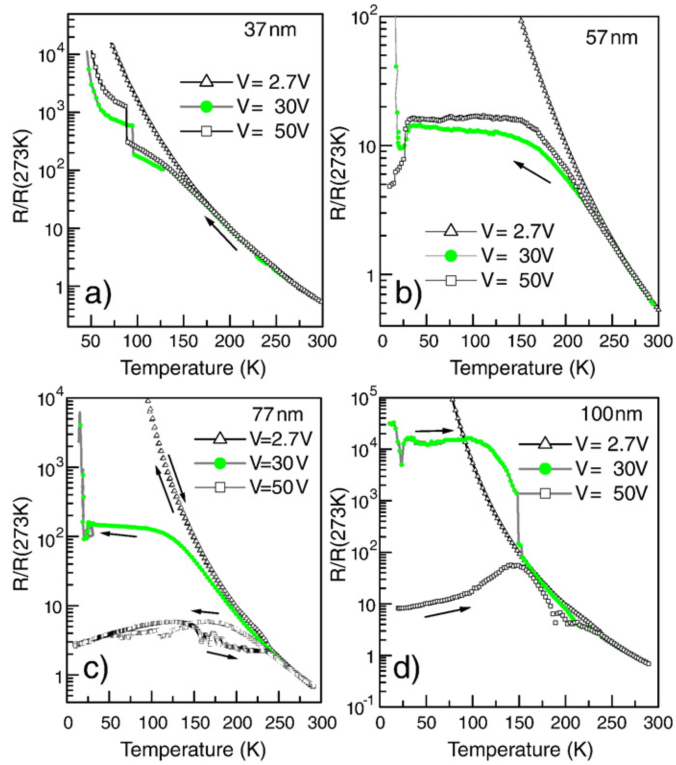

Fig. 1. Dependence of the resistance of a: 37-nm; b: 57-nm; c: 77-nm, and d: 100-nm film samples on temperature for three values of the applied voltage: 2.7, 30, and 50 V.

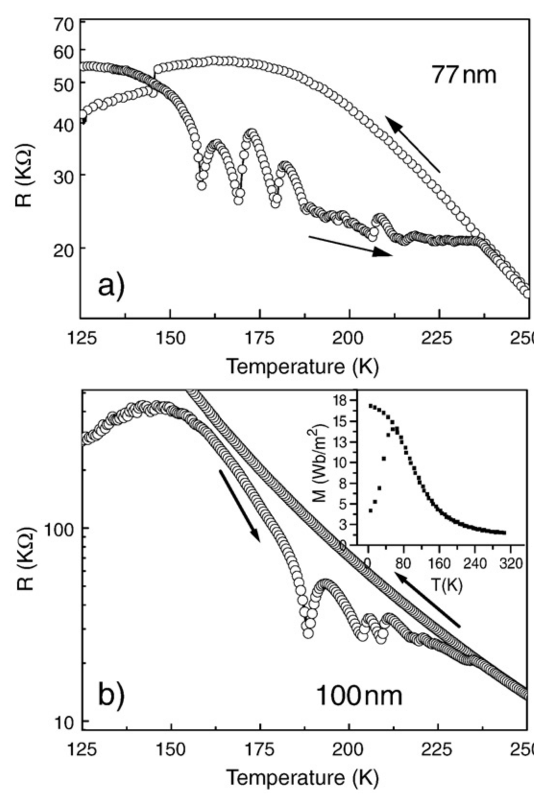

Fig. 2. Temperature dependence of the resistance of: a) 77 nm and b) 100 nm films showing oscillations and instabilities. Inset: zero field cooling (ZFC) and field cooling (FC) magnetization measurements performed on the 100-nm film.

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