



Journal of Applied Sciences

ISSN 1812-5654

science
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Magnetic and Transport Properties of Half-Metallic Ferromagnetic Compounds as the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ Epitaxial Manganite Oxide Thin Films

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Abstract: The aim of this study is to investigate magnetic properties of an example of half-metallic and ferromagnetic compounds, namely, the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) epitaxial manganite oxide thin films using tensile SrTiO_3 (STO) and compressive LaAlO_3 (LAO) substrates. From the magnetic measurements, it can be shown a large magnetic anisotropy in these compounds. The plane of the film is the easy magnetization direction for LSMO deposited onto STO substrate. Also, it seems that the magnetic properties are quite influenced by the strain effects in the films. Furthermore, electrical resistivity and magnetoresistance measurements were performed. The residual resistivity is higher than the massive single crystal sample value due certainly to the crystal defects. The half-metallic character of the LSMO films is not modified when the temperature increases. The magnetoresistance is only strong close to the Curie temperature and substantial in high magnetic field. And these are typical of the colossal magnetoresistance (CMR).

Key words: Spin electronic, half-metallic ferromagnets, epitaxial thin films, manganite oxide

INTRODUCTION

The electric transport in magnetic materials is often strongly modified by the application of a magnetic field. The spin electronics, i.e., the phenomena of electronic spin-dependent transport became in some years a very active research field in magnetism. Conduction electrons behaviour depends on the orientation of their spin in relation to the local magnetization of the material. The physical quantities characterizing the transport become dependent on the majority or minority character of the spin. The fundamental studies in the spin electronics domain are generally focused on two themes. One is concerned with the manganites. The study of the manganites with colossal magnetoresistance (CMR) permitted the electronic correlations comprehension in manganites in general and of the competitions between different energies in the same order of magnitude (magneto-elastic energy, Jahn-Teller distortion, crystalline field, coulombian repulsion). The role of the strain effects and therefore of the magneto-elasticity, has especially been studied for epitaxial manganite films (Ramos *et al.*, 2002; Boujelben *et al.*, 2002; Garcia-Munoz *et al.*, 2002;

Dho *et al.*, 2003; Souza-Neto *et al.*, 2003, 2004a, b; Bergenti *et al.*, 2007). The other is about the highly spin-polarized materials as the polycrystalline half-metallic ferromagnets. The half metals discovered by De Groot *et al.* (1983) are the materials whose band-structure shows a metallic state for up-spin electrons whereas an energy gap occurs at the Fermi level for down-spin electrons. These systems are therefore exemplary materials for the study of the spin electronics (Pierre and Karla, 2000; Borca *et al.*, 2000; Komesu *et al.*, 2000).

In a earlier study, the magnetic properties and the electrical resistivity of a half-metallic ferromagnetic compound as the half-Heusler PtMnSb (Kouacou *et al.*, 2008) were analysed in relation with the second theme that we are described.

In present study, the magnetic properties of another half-metallic compounds examples are to be investigated in relation with the first theme, $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) manganite oxide epitaxial thin films. Electrical resistivity and magnetoresistance experiments have also been performed. These measurements have allowed investigating the strain effects in these compounds.

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MATERIALS AND METHODS

Preparation and crystallographic properties: The $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) films were grown under tensile SrTiO_3 (STO) and compressive LaAlO_3 (LAO) substrates with cubic and pseudo-cubic structures in the [001] direction using Pulsed Laser Deposition (PLD). This technique consists in focusing a pulsed laser beam with a very high power on a target inside a vacuum room. The deposit was made under a controlled oxygen pressure. The laser used for deposition is a pulsed YAD Nd laser tripled in frequency (350 nm). The pulse time is about 10 nsec with 240 mJ per impulsion maximal energy. The laser was adjusted on 10 Hz clock frequency. The LSMO layers were grown at 1023 K (750°C with a heat velocity of $20^\circ\text{C min}^{-1}$) under 40 Pa of oxygen, the laser operating at 1.79 W maximal power. The reflectivity obtained shows the oscillations. After deposit, the samples were cooled under 0.5 bar of oxygen with a constant cooling velocity of $20^\circ\text{C min}^{-1}$. Prior to the deposition, the substrates were heated in oxygen up to the deposition temperature. We have realized squared tablet LSMO films of 7.5 mm side with 60 nm thickness for tensile SrTiO_3 (STO) substrate and 15 nm for compressive LaAlO_3 (LAO) substrate estimated *in situ* by optical reflectometry. The films thickness has been chosen under the critical thickness (of the order of 100 nm for LSMO/STO film and of 30 nm in the LSMO/LAO case) for structural relaxation, in order to keep homogeneous materials.

X-Ray Diffraction (XRD) study was carried out to examine the structural properties of the layers. The LSMO films are epitaxially fully constrained and a large epitaxially-induced magneto-elastic anisotropy is present

as we shall see. As in the earlier studies (Ranno *et al.*, 2002; Favre-Nicolin and Ranno, 2004; Singh *et al.*, 2006) the LSMO films are textured with (001) orientations. XRD θ - 2θ diagram in the range $20 < 2\theta < 120^\circ$ is shown in Fig. 1a and shows diffraction peaks corresponding to (002) and (004) directions for LSMO film grown on STO substrate. Another diagram shows only (004) peak (Fig. 1b). The last accessible LSMO/STO film peak is (004) direction that is located to 106.25° . This result differs from the one found at 107° in the earlier study (Favre-Nicolin, 2003). The difference can be explained by the fact that the films have not the same strain effects. In the present case, the film is less strained than the other one. The X ray that gives the same picks as the previous LSMO films (even though their positions differ) shows that our films are homogeneous and do not contain any other picks. The sample characterization by X-ray diffraction is shown in Table 1 where, biaxial strain parameters can be defined as $\epsilon_{\parallel} = \frac{a_{\parallel} - a_{\text{bulk}}}{a_{\text{bulk}}}$ and $\epsilon_{\perp} = \frac{a_{\perp} - a_{\text{bulk}}}{a_{\text{bulk}}}$, a_{\parallel} and a_{\perp} being the in plane and out of plane lattice parameters of films, respectively.

Experimental methods: $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) films magnetic measurements were performed with a Vibrating

Table 1: Samples nomenclature and characteristics obtained by X-ray diffraction

Composition	Strain	Lattice parameter (Å)	Strain parameters	
			ϵ_{\parallel} (%)	ϵ_{\perp} (%)
SrTiO_3 (STO)	Tensile (fully strained)	3.907	+0.85±0.08	-0.95±0.08
LaAlO_3 (LAO)	Compressive (strained)	3.791	-2.14±0.08	+2.22±0.08
$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO)	-	3.874±0.003	-	-

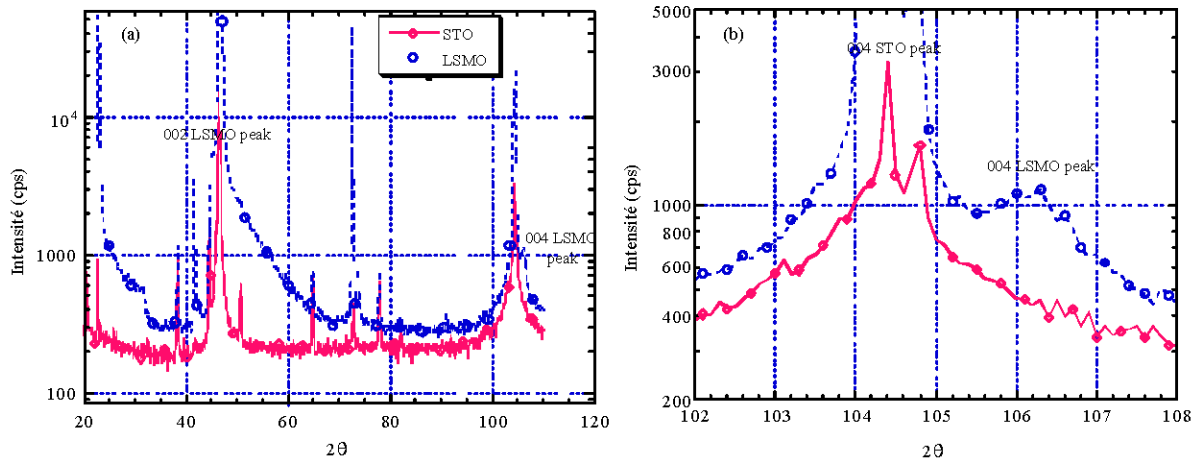


Fig. 1: (a) X ray diffraction diagram of LSMO/STO film and (b) diffraction X spectrum limited to (004) peak

Sample Magnetometer (VSM) below 300 K in magnetic field up to 3 T. For each temperature, a complete magnetization $M(H)$ hysteresis loop was recorded.

The electrical resistivity (in null applied field), was measured from 5 to 300 K using the conventional four-probe method with AC current. Magnetoresistance (MR) measurements were performed under applied magnetic field parallel to the current direction up to 7 T.

RESULTS AND DISCUSSION

Magnetic measurements: $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) manganite oxide (mixed-valence manganese) with perovskite structure (pseudo cubic) is a half-metallic and ferromagnetic compound that orders at 370 K for massive single crystal sample (Urushibara *et al.*, 1995; Mahendiran *et al.*, 1996; Mahesh *et al.*, 1999).

The magnetization measurements were realized in the plane of the film according to the directions [100] and [110] and in the perpendicular plane. These measurements show a magnetic anisotropy. Figure 2 shows the $M(H)$ hysteresis loop of LSMO/STO film measured at 285 K (a) and 10 K (b), with the magnetic field applied along the substrate [110] axis and along the perpendicular axis. Figure 2 shows that the plane of the film is the easy magnetization direction with [110] as the easy-axis (Favre-Nicolin and Ranno, 2004). In the perpendicular direction, the saturation field $\mu_0 H_{sat}$ found to be 0.7 T at 285 K and 1.4 T at 10 K is not consistent with the earlier studies (Ranno *et al.*, 2002; Favre-Nicolin, 2003). For a comparison, Favre-Nicolin (2003) found 0.5 T at 295 K and 1.56 T at 15 K. The difference between these values could stem from the strain effects of the film. For the LSMO film deposited onto LAO substrate, the saturation field $\mu_0 H_{sat}$ in the perpendicular direction is found to be 0.24 T at 10 K (Fig. 3). The spontaneous magnetization M_{sp} (in null applied magnetic field) determined from the hysteresis loops is reported on Fig. 4. For LSMO/STO film, the saturation magnetization (magnetic moment), $\mu_0 M_{sat} = 0.66$ T (i.e., $M_{sat} = 3.3 \mu_B$ per Mn atom), obtained at 10 K is lower than the 0.74 T (i.e., $M_{sat} = 3.7 \mu_B$ per Mn atom) value observed by (Ranno *et al.*, 2002; Favre-Nicolin, 2003). Such a discrepancy could be explained by the strain effects of the film generated by the difference between the lattice parameters of the deposited layer and the substrate. These strain effects can create fairly important distortions carrying along a change of the magnetic film symmetry which can engender very important modifications of magnetic and transport properties (Kreisel *et al.*, 2002). In the case of LSMO/LAO film, the saturation magnetization $\mu_0 M_{sat}$ found to be 0.58 T (i.e., $M_{sat} = 2.9 \mu_B$ per Mn atom) is yet smaller. These

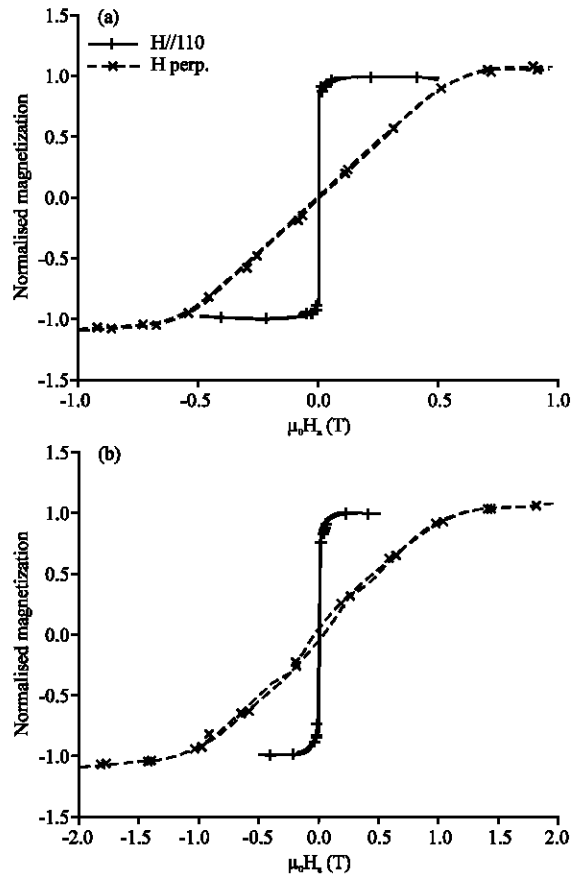


Fig. 2: $M(H)$ hysteresis loop of LSMO/STO film, with the magnetic field applied along the substrate [110] axis and along the perpendicular axis, at (a) 285 K and (b) 10 K

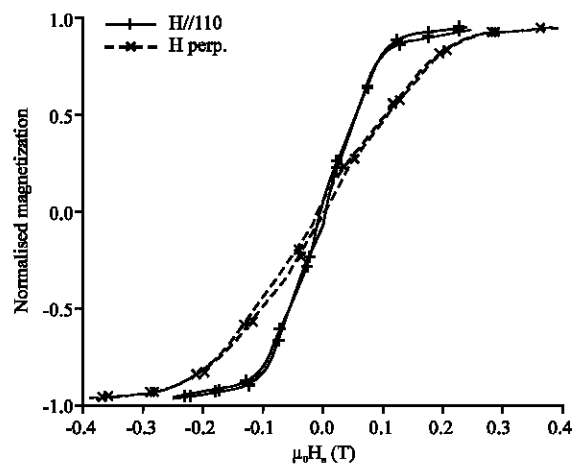


Fig. 3: $M(H)$ hysteresis loop of LSMO/LAO film, with the magnetic field applied along the substrate [110] axis and along the perpendicular axis, at 10 K

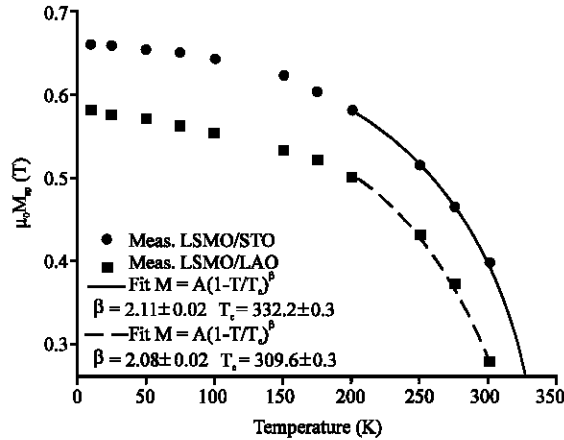


Fig. 4: Temperature variation of LSMO films spontaneous magnetization. The continuous and dotted lines are $M = A(1-T/T_c)^\beta$ fits above 200 K

experimental values remain nevertheless quite lower than most of manganese half-metallic ferromagnets (as for example NiMnSb and PtMnSb) moment which is $4 \mu_B$ per Mn atom (Hames and Crangle, 1971; Otto *et al.*, 1989; Van Engen *et al.*, 1983; De Groot *et al.*, 1983). Moreover, in order to verify the magnetic anisotropy, it is necessary to determine the anisotropy field H_{anis} , when the demagnetizing field H_d is considered in perpendicular direction. This anisotropy field is of magneto-elastic origin and related to the material magnetostriction due to the coupling between the magnetization and the lattice distortions (Hazama *et al.*, 2000; Abramovich *et al.*, 2001; Maurice *et al.*, 2002). In fact, when the saturation field H_{sat} in perpendicular direction is found to be higher than the magnetic moment M_{sat} , the film is epitaxially-strained in stretching (tensile substrate) and in this situation (case of $La_{0.7}Sr_{0.3}MnO_3$ film grown on a $SrTiO_3$ substrate) the total saturation field is given by Favre-Nicolin (2003):

$$H_{sat} = H_{anis} - H_d$$

where, $H_d = -M_{sat}$ (for squared tablet thin film) i.e., $H_{sat} = H_{anis} + M_{sat}$

Conversely, when the magnetic moment M_{sat} is found to be higher than the saturation field H_{sat} in perpendicular direction, the film is epitaxially-strained in compression (case of $La_{0.7}Sr_{0.3}MnO_3$ film grown on a $LaAlO_3$ substrate), the anisotropy field H_{anis} value is then negative and the total saturation field is given by:

$$H_{sat} = |H_{anis}| - M_{sat}$$

Table 2: Magnetic properties of the $La_{0.7}Sr_{0.3}MnO_3$ films

Composition	$\mu_0 M_{sat}$ (T)	$\mu_0 H_{sat}$ (T)	$\mu_0 H_{anis}$	T_c (K)
$La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$	0.66	1.40	0.74	332
$La_{0.7}Sr_{0.3}MnO_3/LaAlO_3$	0.58	0.24	-0.82	309

Thus, the anisotropy field $\mu_0 H_{anis}$ found to be 0.74 T for LSMO/STO and -0.82 T for LSMO/LAO at 10 K shows the presence of a magnetic anisotropy (of magneto-elastic origin). When the anisotropy field is modified only by the lattice distortions, the magneto-elastic theoretical model predicts that this anisotropy field is proportional to the biaxial strain parameter (in plane) $\epsilon_{||}$ (Favre-Nicolin, 2003). We can thus deduce the following relation:

$$H_{anis(LSMO/LAO)} = H_{anis(LSMO/STO)} \times \frac{\epsilon_{||(LAO)}}{\epsilon_{||(STO)}}$$

which gives 2.5 factor between the $H_{anis(LSMO/STO)}$ (for LSMO on STO film) and $H_{anis(LSMO/LAO)}$ (for LSMO on LAO film) module, whereas the experimental result is 1.11 factor (Table 2). This quantitative disagreement between the theory and the experiment could be resulted from the parameters stemming from the different samples for the numeric applications. Another possible explanation of this disagreement could stem from the shape strain of the LSMO on LAO film which is not known as well as in the LSMO on STO film case. In order to know better the shape strain of LSMO on LAO films, it is necessary to analyze a series of films with thicknesses lower than 15 nm.

On the other hand, as our measurements are limited to 300 K, considering the following formula (spontaneous magnetization variation fit above 200 K):

$$M = A.(1 - \frac{T}{T_c})^\beta$$

we are able to obtain 332 K Curie temperature in the case of LSMO/STO film and 309 K for LSMO/LAO film. Let us notice the modification of the films Curie temperature when the substrate changes confirming the role of the strain effects as above-mentioned. Moreover, the magnetization measurements results shown in Table 2 show that the magnetic properties are quite influenced by the strain effects of the LSMO films.

Electrical transport measurements: The LSMO films electrical resistivity $\rho(T)$ is represented in Fig. 5. The residual resistivity with $112.5 \mu\Omega$ cm value in the case of LSMO/STO film and $238 \mu\Omega$ cm value for LSMO/LAO film is higher than the massive single crystal sample one with

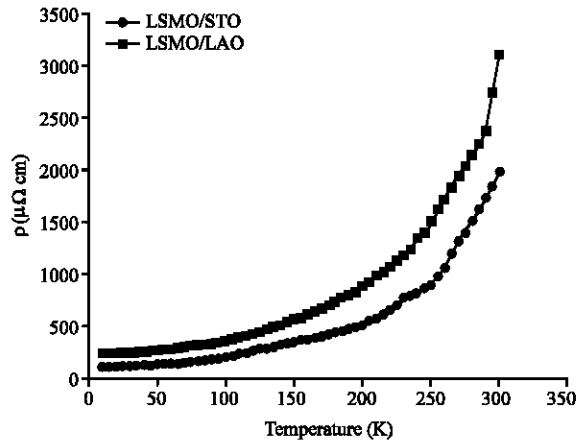


Fig. 5: Temperature dependence of the LSMO films electrical resistivity

40 $\mu\Omega$ cm value (Tokura and Tomioka, 1999). Generally, the electrical resistivity of a ferromagnetic material is characterized at $T = 0$ K by the residual resistivity due to the impurities, crystallographic disorder or crystal defects and above 0 K the magnetic contribution due to spin-disorder scattering (with the spin fluctuations and/or with the spin waves) and electron-phonon scattering appears. These higher values obtained in the present case are not due to the impurities but could stem from the crystal defects due to the strain effects of the films. In all the temperature range, no distinct anomaly appears in the resistivity curves, showing that the half-metallic character of the LSMO films is not modified when the temperature increases contrarily to the half Heusler and half-metals NiMnSb (Hordequin *et al.*, 1996, 2000; Ristoiu *et al.*, 2000) and PtMnSb (Kouacou *et al.*, 2008) cases where a transition from a half-metallic to metallic state occurs at 100 K due to the spin fluctuations.

As one can observe it on Fig. 6 and 7 the magnetoresistance (MR) is negative like in all the manganese oxides. The magnetoresistance $[R(T,0) - R(T,H)]/R(T,0)$ at 6 Tesla is shown in Fig. 6 as a function of temperature. It can be seen that the MR is strong above 280 K (30% or 40% at 300K) but decreases rapidly at low temperature. This is in accordance with the study of the manganites which indicates a maximum close to the Curie temperature T_C . The resistance variation as a function of applied magnetic field (Fig. 7) shows that the MR is observed in high magnetic field. Hence, the fact that the MR is only strong close to T_C and substantial in high field indicates that the resistance variation is typical of the CMR (Tokura and Tomioka, 1999).

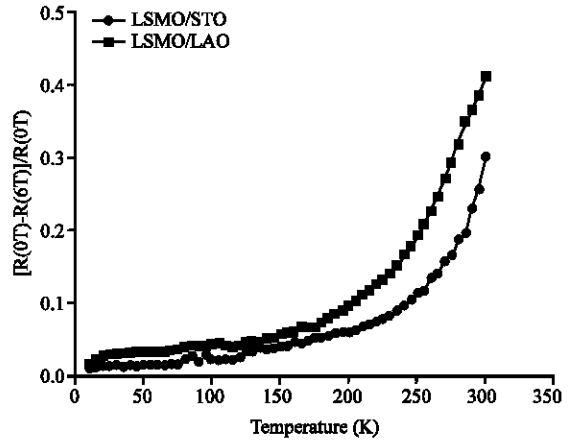


Fig. 6: Magnetoresistance at 6 Tesla as a function of temperature for LSMO films

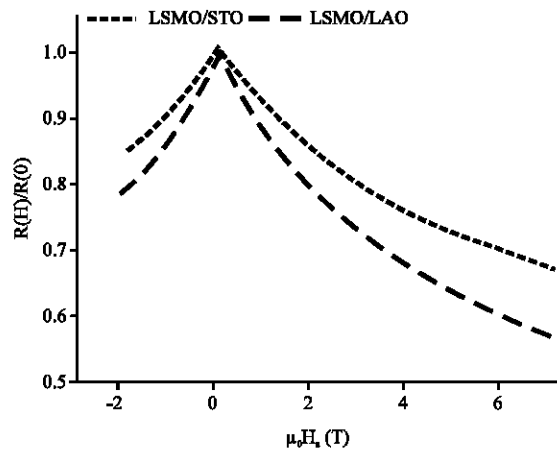


Fig. 7: Resistance variation as a function of applied field at 300 K for LSMO films

CONCLUSION

The LSMO films magnetization measurements, carried out in this present work, noticeably confirm the previous investigation results; the film plane is the easy magnetization direction (in the case of tensile STO substrate) and a large epitaxially-induced magneto-elastic anisotropy is present. However, the magnetic properties and the residual resistivity are influenced by the strain effects of the films. The resistivity measurements do not show any transition with the temperature variation showing that the half-metallic character of the LSMO films is not modified when the temperature increases. The magnetoresistance is only strong close to the Curie temperature and substantial in high magnetic field like in

the most manganite oxides. These features are typical of the CMR. Additional experiments such as anisotropic magnetoresistance (AMR), Hall resistivity and thermoelectrical power are required to investigate the transport properties in more detail.

ACKNOWLEDGMENTS

We wish to thank L. Ranno to have permitted the measurements in laboratory of magnetism Louis Néel, CNRS Grenoble, France. Thanks also to the director, the staff of this laboratory, the first author thesis supervisor J. Pierre for their help and E.K. Yankey from INP-HB for his invaluable contribution.

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