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Effect of strain in $La_{0.7}Sr_{0.3}MnO_3$ epitaxial films with different crystallographic orientation

U. Scotti di Uccio [a,b,*], B. Davidson [c], R. Di Capua [b], F. Miletto Granozio [b], G. Pepe [b,d], P. Perna [a], A. Ruotolo [b,d], M. Salluzzo [b]

[a] *DiMSAT, Università di Cassino, Dip. di Meccanica Strutture Ambiente e Territorio, via Di Biasio 43-03043 Cassino, Italy*
[b] *Coherentia-INFM, Napoli, Italy*
[c] *TASC-INFM, Trieste, Italy*
[d] *Dip. Scienze Fisiche, Università di Napoli Federico II, Italy*

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Abstract

We investigated the role of strain on the magnetic properties of $La_{0.7}Sr_{0.3}MnO_3$ epitaxial films grown on substrates with different crystallographic orientation. The magnetic characterization confirms the large magnetostriction due to the tensile stress applied by the $SrTiO_3$ substrates. The data demonstrate that the easy direction of magnetization is aligned to the elongated, in-plane axis both in (1 1 0)- and (0 0 1)-oriented films. The in-plane anisotropy of (1 1 0) films is interpreted in the frame of a simple structural model. We provide evidence of a spatial separation between a conducting ferromagnetic and an insulating paramagnetic phase at the Curie temperature. The possible effect of such phase separation on the coercitive field and on the magnetization versus temperature is discussed.

Keywords: Magnetic films and multilayers; X-ray diffraction; Magnetic measurements

1. Introduction

$La_{0.7}Sr_{0.3}MnO_3$ (LSMO) films have attracted considerable interest due to the potential application in various spintronics devices, that can take advantage of the colossal magnetoresistance and/or the high degree of spin polarization of charge carriers [1]. The success of the new ideas is mainly based on the control of the physical properties of thin films. Usually, the (0 0 1)-oriented perovskites such as $SrTiO_3$ (STO) are employed as substrates in studies regarding $La_{0.7}Sr_{0.3}MnO_3$, and much less work has been devoted to different crystallographic orientations, such as the (1 1 0). Yet, there are several interesting properties of LSMO films grown on (1 1 0) perovskites. For this reason, we considered in our research both (0 0 1) and (1 1 0) substrates. We report in a separate paper [2] on the detailed structural characterization and on the effect of strain on the transport properties of LSMO grown on STO. Here we mainly consider the effect of strain on the magnetic properties. It is well known that the magnetic properties of LSMO epitaxial films are quite different from those of bulk or single crystals. Early works [3] proved that single crystals show an anisotropic magnetic behavior due to the crystal field contribution, and that this effect is overcome by the magnetostriction in samples subject to stress. The biaxial stress due to the substrate is very effective in LSMO deposited on perovskites. Detailed discussions have been reported in several reviews [1]. Briefly, it was found that LSMO films under compressive strain (i.e. on $LaAlO_3$) have an easy axis perpendicular to the substrate plane, while the opposite is found in the case of tensile stress (i.e. on STO). We present here data regarding films grown on STO with different crystallographic orientations, with the aim to critically discuss the possible different mechanisms leading to the observed behaviors.

2. Experimental results and discussion

The films are deposited by rf magnetron sputtering from a single target. The substrates are heated up to 840 °C and kept in vacuum for 30 min before film deposition. The sputtering atmosphere is a mixture of argon and oxygen, with

* Corresponding author. Fax: +39 07762993711.
E-mail address: scottidu@unicas.it (U.S. di Uccio).

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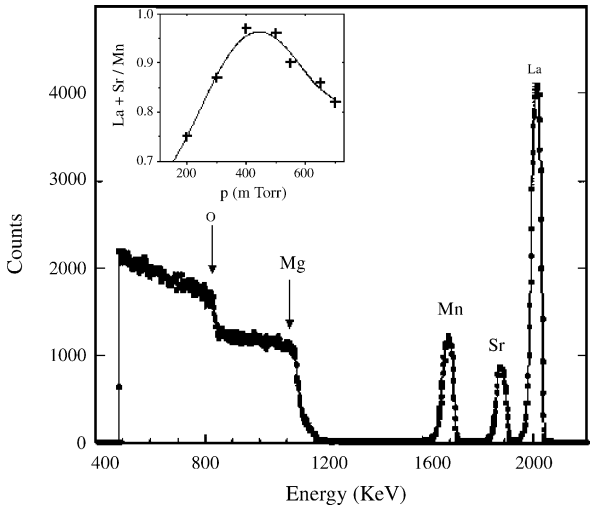

Fig. 1. Typical RBS spectrum. Inset: stoichiometry vs. pressure during deposition.

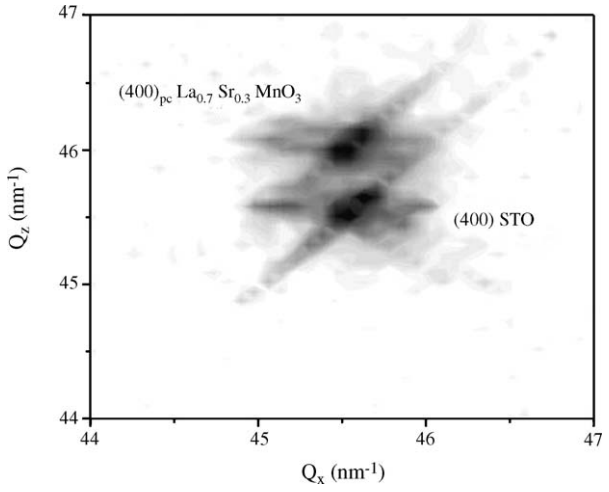

Fig. 3. Reciprocal space map around the $(4\,0\,0)_{pc}$ reflection. The coincidence of the Q_x position for film and substrate indicates the in-plane matching of the structures. Aliasing of the map is due to the Cu Kα_2 radiation.

equal partial pressures. The deposition rate is 0.03 nm/s at 100 W rf incident power. After deposition, the chamber is slowly vented in O_2 up to 400 mbar. The cooling is completed in about 2 h.

Careful Rutherford Backscattering (RBS) analyses were performed on thin samples deposited on MgO substrates at different pressure, in order to find the best deposition conditions. Thin films have been employed in order to get well separated contributes from the different cations in the spectra (Fig. 1). At $P_{tot} = 500$ mTorr, the target stoichiometry is reproduced in the films within the experimental error.

The samples show a high quality crystal structure. Here we report the data for a typical film grown on (1 1 0) STO (Figs. 2 and 3). There are two main results, that are quite general in our experiments: the films are perfectly matched to the substrate lattice and their cell volume is somewhat expanded (about 1.5%) with respect to bulk LSMO. We attribute this last feature to a small amount of oxygen vacancies (a stoichiometry of about 0.05 in formula units) that are left after the deposition and cooling process.

The Curie temperature T_C of LSMO is determined by the magnetization versus temperature plots. The $\rho(T)$ and $M(T)$ for two 30 nm thick samples, grown on (1 1 0) STO and (0 0 1) STO, respectively, are shown in Figs. 4 and 5. The ZFC magnetization is measured by applying a magnetic field of 1 kOe. The coincidence of the temperature T_S of maximum dR/dT with the temperature of maximum dM/dT is remarkable in both cases. The difference of T_C between the samples is mainly attributed to the different growth conditions, even though we always observe that the T_C of (0 0 1) films is higher.

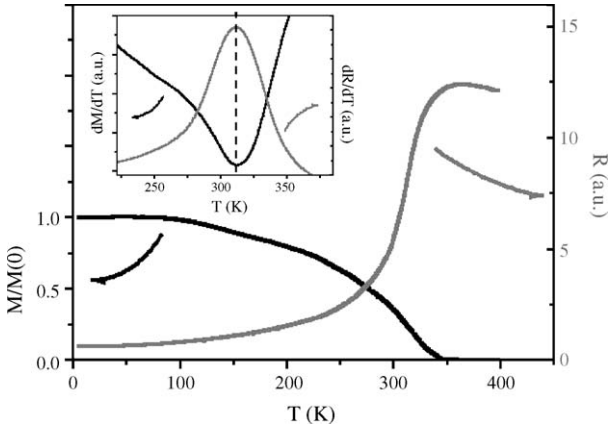

Fig. 4. $\rho(T)$ and $M(T)$ at $H = 1$ kOe for a sample grown on (1 1 0) STO.

Fig. 6 shows the hysteresis loops at 100 K for a sample grown on (1 1 0) STO, with the field aligned along the two in-plane principal axes. The easy axis is parallel to the [0 0 1] direction, while the [1 $\bar{1}$ 0] is a hard axis, characterized by a smaller hysteresis and a higher saturation field. Quite remarkably, the anisotropy field H_K in the loop with $\vec{H}$//[1 1 0] (not shown) is equal to

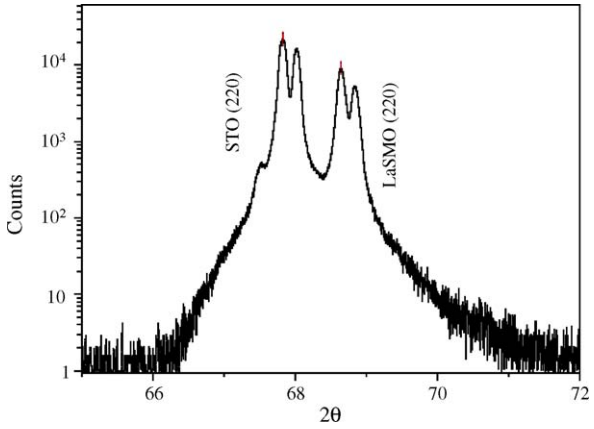

Fig. 2. θ–2θ plot of the $(2\,2\,0)_{pc}$ peak of a sample deposited on (1 1 0) STO.

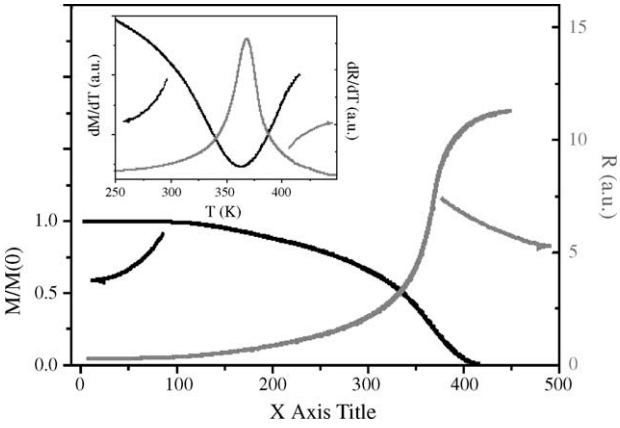

Fig. 5. $\rho(T)$ and $M(T)$ at $H = 1$ kOe for a sample grown on (0 0 1) STO.

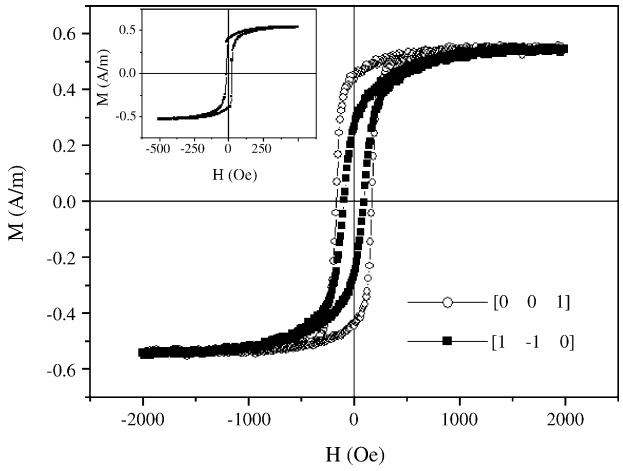

Fig. 6. $M(H)$ at 100 K for a film grown on (1 1 0) STO. $\vec{H}$ is aligned to the in-plane axes [0 0 1] and [1 $\bar{1}$ 0]. Inset: $M(H)$ for a different film grown on (0 0 1) STO (H//[1 0 0], T = 100 K).

the demagnetizing field of $4\pi M_S$ within the experimental error. The agreement indicates that there is no perpendicular uniaxial anisotropy in (1 1 0) LSMO grown on STO. This is very different from the case of films grown on (0 0 1) STO (see inset of Fig. 6). Such samples, as already reported [1], have an easy in-plane axis (i.e. the [0 0 1]) and a hard vertical axis. On the other hand, the saturation magnetization of the two samples is the same (about $3.4\mu_B$ per Mn site).

The magnetic properties of our samples closely resemble those reported in [3] and confirm that LSMO has strong magnetostrictive effects. The results can be interpreted in the framework of the fundamental mechanisms of ferromagnetic ordering in manganites, i.e. by the double exchange [5]. The ferromagnetic coupling is determined by the nature of the Mn—O—Mn bonds, that are affected by the strain. The details of cell deformation are however different for samples grown on (1 1 0) and (0 0 1) STO. In the former case, the tensile biaxial stress acts by applying a shear stress to the LSMO cell. Shear is instead absent in the latter case. Therefore, the Mn—O bond length and on the Mn—O—Mn angles are modified in different ways. In particular, the distance between nearest neighbors Mn cations along the [1 −1 0] direction is scarcely modified in the strained LSMO grown on (1 1 0) STO, because the stress does not change the length of the (1 0 0) and (0 1 0) axes, but instead their angle (Fig. 7). Thus, we can explain the in-plane anisotropy of (1 1 0) samples, assuming that the magnetostrictive effect depends on the axes length, and only to a minor extent on cell angles. Therefore, the in-plane easier axis is expected in the [0 0 1] direction, that is an elongated axis, as it is in fact observed, while the [1 $\bar{1}$ 0] is comparatively harder.

In Fig. 8, the $M(H)$ loops at 100 and at 300 K for a sample grown on (1 1 0) STO are compared. The field is aligned to the [1 $\bar{1}$ 0] direction in this case. It is found that H_C decreases from 170 Oe at 100 K to 60 Oe at 300 K. This noticeable reduction of the loop width with temperature is indicative of an enhanced mobility of the domain walls, that is most probably due to a thermal activation mechanism. This effect may be partially contrasted by the phase separation that takes place in a temperature region around the Curie temperature, and that we demonstrated by scanning tunneling measurements

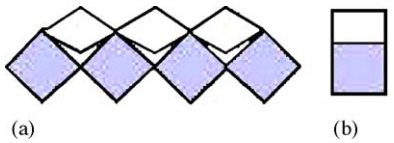

Fig. 7. Sketch of the strained structure of LSMO on: (a) (1 1 0) STO and (b) (0 0 1) STO.

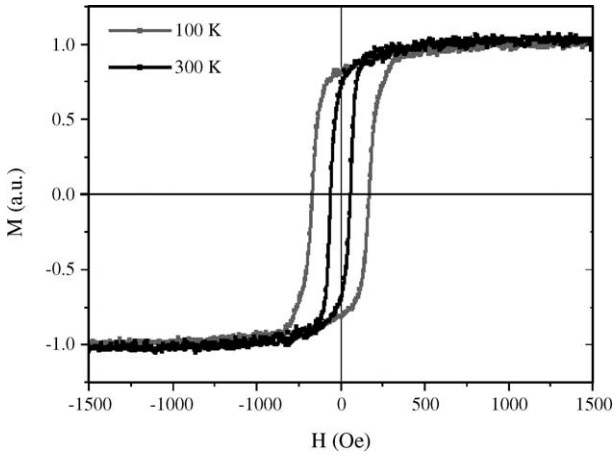

Fig. 8. $M(H)$ for a LSMO film on (1 1 0) STO at 100 and 300 K.

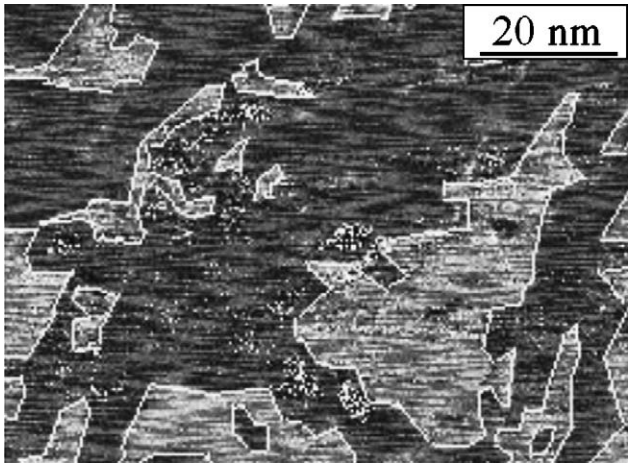

Fig. 9. STM conductance map of a LSMO film on (1 1 0) STO at 300 K.

[6]. The map of tunneling conductance in Fig. 9, taken at 300 K, reproduces in light color the highly conductive regions, that according to the present understanding are ferromagnetic, while dark regions are insulating and hence paramagnetic. A naïve approach within the Stoner–Wohlfarth model would then suggest that the separated, ferromagnetic regions, behave as ferromagnetic nanoparticles, and therefore possess an enhanced coercive field until, reducing the temperature, the percolation of ferromagnetic regions is complete. As a final consideration, let us turn to the data in Fig. 4. It is known that the phase separation can influence the $\rho(T)$ dependence [5]. In a crude model, both the conductibility $\sigma = 1/\rho$, and M, are proportional to the volume of the ferromagnetic phase. The coincidence of the temperatures of maximum slope of $\rho(T)$ and $M(T)$ appears therefore consistent with a phase separation model for LSMO.

3. Conclusions

The properties of LSMO films are sensitive to stoichiometry and to stress. We investigated the magnetic properties of well characterized samples, confirming the magnetostriction due to the tensile stress applied by the substrate. We proposed a possible interpretation of the data based on the knowledge of the details of strain and of the phenomenon of phase separation at the Curie temperature.

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