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Structural, $1/f$ Noise and MOKE Characterization of Vicinal $La_{0.7}Sr_{0.3}MnO_3$ Thin Films

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This paper presents some structural, magnetic and electrical properties of 40 nm and 75 nm thick $La_{0.7}Sr_{0.3}MnO_3$ thin films deposited on vicinal $SrTiO_3(001)$ substrates. The vicinal angles were 2, 4, 6, 8, and 10° from the [001] direction towards [110]. Standard $SrTiO_3(001)$ substrates were used for comparison and for the growth condition optimization. Structural properties were studied by X-ray diffraction, which indicated that the LSMO films grew with their (001) axis coincident with the (001) axis of the substrate. The surface morphology was carefully studied by atomic force microscopy in tapping mode. Very smooth films and regular step-terrace structures on the $La_{0.7}Sr_{0.3}MnO_3$ surfaces could be observed. The root mean square roughness measured in 2 μm $\times$ 2 μm images was in the 0.130–0.580 nm range for all angles and both thicknesses. Superconducting quantum interference device magnetometer measurements revealed a Curie temperature in the 340–350 K range and magneto-optical Kerr microscopy enabled magnetic domain imaging and hysteresis loop measurements at 300 K. A uniaxial easy magnetization direction was obtained at 300 K for angles above 4°, with the easy axis along the steps. Finally, preliminary $1/f$ noise measurements were performed at 300 K for bias currents along and perpendicular to the step direction, showing a noise level as low as that we typically measured on standard $SrTiO_3(001)$ substrates. All these results are promising for the future realization of room-temperature devices making use of the anisotropy of the magnetization.

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1. Introduction

The rare-earth manganese oxides are attracting considerable interest because of their potential impact on magnetic recording. Among them, $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) shows a Curie temperature above 300 K, thus potentially leading to devices operating at room temperature. Interestingly, magnetic properties, such as magnetic anisotropy, magnetization reversal mode, magnetic domain structures, or coercitive field, can be changed and controlled by surfaces and interfaces when materials are in the form of thin films. All the above-mentioned magnetic properties are linked together. One of the ultimate goal for the design of devices is to be able to engineer them and one simple idea for that could be to artificially modify the surface morphology of the ferromagnetic thin films.

In the case of ferromagnetic LSMO, Tsui et al. [1] have shown that the tensile or compressive strain induced by the film–substrate lattice mismatch can induce in-plane or out-of-plane easy magnetization directions. Furthermore LSMO thin films grown on $SrTiO_3$ (STO) (001) substrates show an in-plane biaxial magnetic anisotropy along the [110] directions, which was determined to have a magneto--crystalline origin [2]. Hyman et al. [3] have shown theoretically that vicinal surfaces can influence strongly the in-plane magnetization reversal of ultrathin ferromagnetic films. Atomic steps on a (001) surface break the fourfold rotational symmetry of the film surface indeed and therefore should induce an in-plane magnetic anisotropy since magnetocrystalline anisotropy obeys the symmetry of the lattice [4]. It has thus been demonstrated experimentally for a variety of magnetic metallic materials that steps can induce a uniaxial anisotropy within the ultrathin films. Step-induced uniaxial magnetic anisotropy has been observed recently at 80 K in ultrathin (12.6 nm thick) LSMO films grown on 10° vicinal substrates [5]. Mathews et al. [6] could obtain an in-plane anisotropy at room temperature in 7 and 25 nm thick LSMO films deposited on STO(001) substrates with very low vicinal angles (0.13° and 0.24°). In that latter case, the substrates were chemically treated and annealed at 950°C to obtain TiO_2 termination.

This paper presents a study of the growth of 40 nm and 75 nm thick LSMO films onto vicinal STO(001) substrates of angles in the 0–10° range. Section 2 describes the deposition conditions and the structural study by X-ray diffraction. The surface morphology was studied by atomic force microscopy (AFM) in tapping mode (Sect. 3). Preliminary magnetic characterization, by magneto-optical Kerr (MOKE) microscopy is presented in Sect. 4. Conclusions and perspectives of this work are finally given.

2. Deposition conditions and structural study

Two series of LSMO thin films of thicknesses 40 nm and 75 nm, respectively, were deposited by pulsed laser deposition from a stoichiometric target onto commercially available vicinal STO(001) substrates. The vicinal angles were 2, 4, 6, 8, and 10° from the [001] direction towards [110]. Standard (001) STO

substrates were used for comparison and for the growth condition optimization. The laser energy density was 1–2 J/cm^2 (250 mJ), the target-to-substrate distance was 50 mm, the oxygen pressure was 0.35 mbar and the substrate temperature was 720°C. These parameter values were found optimal for producing single-crystalline films as judged by the X-ray diffraction (XRD) study. The latter indicated that the LSMO films grew with their (001) axis coincident with the (001) axis of the substrate. An offset angle of the LSMO film was checked to be equal to the substrate vicinal angle within ±0.05° for all the considered angles. XRD $\theta-2\theta$ patterns are shown in Fig. 1 for the two thicknesses. One can note the satellite peaks around

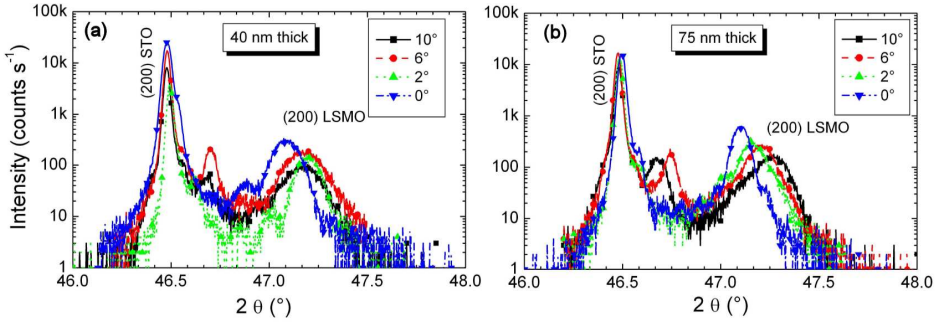

Fig. 1. XRD $\theta-2\theta$ patterns measured using an offset value on θ (a vicinal angle value was added or subtracted, depending on the sample position, to the θ value in order to bring the film planes into their Bragg positions). No peaks could be recorded if no offset was added thus confirming that the LSMO films grew with their (001) axis coincident with the (001) axis of the substrate: (a) 40 nm thick series, (b) 75 nm thick series.

the LSMO(002) peak, which gives an indication of the very low roughness of the films. The average out-of-plane parameter of vicinal LSMO films was 0.3850 nm, which corresponds to a lattice mismatch with STO of 1.396×10^{-2}. It has to be compared to the value of 0.3857 nm (i.e. a lattice mismatch of 1.217×10^{-2}), which is typically measured on our LSMO films on STO(001) substrates. This means that the LSMO cell shows a higher in-plane tensile strain in the case of vicinal films than in the case of LSMO on standard STO(001). The full-width-at--half-maximum of the (002) peak of the vicinal LSMO films was measured in the 0.23–0.31° range, with no clear relation to the increasing angle. These values can be compared to 0.23° which we typically obtained for LSMO films of comparable thickness deposited on STO(001) substrates. Saturated magnetization versus temperature was measured at 0.5 T for the different angles using a superconducting quantum interference device (SQUID) magnetometer. The magnetic field H was applied in the plane of the films and for each angle, the $M(T)$ curve was recorded with H in both directions with respect to the steps, i.e. parallel and perpendicular. The Curie temperature T_C was found in the 340–350 K range, which is compara-

ble to T_C of LSMO thin films in the 40–75 nm thickness range, and no difference could be observed as a function of the field direction with respect to the steps. In addition, the electrical resistivity was in the 1.25–2.25 mΩ cm range at 300 K, thus confirming the overall good quality of our LSMO films.

3. Surface morphology

An AFM study was performed in tapping mode in order to study the surface morphology properties of the films. As shown in Fig. 2, very smooth films and regular step-terrace structures on the vicinal LSMO surfaces corresponding to the replication of the substrate surface were obtained for angles higher than 4°. The

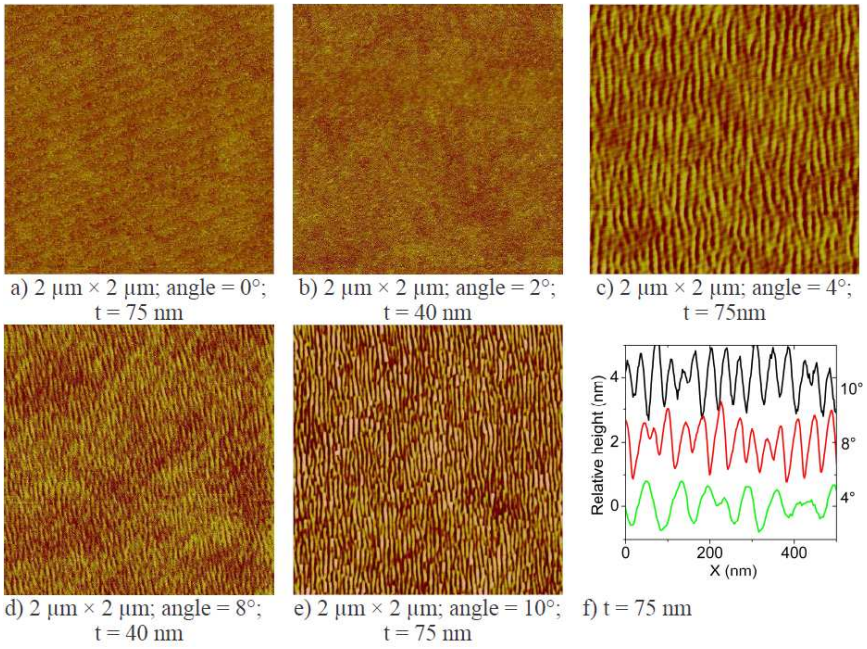

Fig. 2. Selection of 2 μm × 2 μm AFM images in tapping mode (z scale = 3 nm) of 40 nm and 75 nm thick LSMO films for various vicinal angles. Thickness is denoted by t, the vicinal angles — angle (a)–(e). Figure 2f shows an example of section profiles measured in AFM images on 75 nm thick films for different vicinal angles.

root-mean-square (RMS) roughness was measured in 2 μm×2 μm AFM images and was in the 0.130–0.580 nm range for all films. These values have to be compared to the typical value of 0.130 nm that we obtained for non-vicinal LSMO films. One can note that for angle below 2° the thickness has no clear effect on roughness, but above 4° the 75 nm thick film series is systematically rougher than the 40 nm series for each angle. For example, at 10°, the RMS roughness was 0.264 nm for the 40 nm thick film and 0.579 nm for the 75 nm thick film.

In the case of LSMO deposited on standard STO(001) substrates, we systematically observed large steps (about 80 nm wide) corresponding to a step-flow growth mechanism and reproducing the very small miscut (of the order of 0.1°) of the substrates (Fig. 2a). No step could be observed by AFM for LSMO thin films deposited on 2° vicinal substrates (Fig. 2b). As expected, above 4°, it was observed that the step width decreased with increasing vicinal angle, i.e. 60 nm, 50 nm, 36 nm, 32 nm for 4°, 6°, 8°, and 10°, respectively, and for the two considered thickness values, the step width did not depend significantly on the thickness (Fig. 2c–e). Figure 2f shows 500 nm long profiles extracted from the AFM images and showing the regular steps of varying width depending on the vicinal angles. These AFM images confirm the XRD study that demonstrated the vicinal growth of the LSMO films, even for relatively high thicknesses (up to 75 nm), which is important for future devices, since the magnetization value will be directly related to the ferromagnetic volume involved. Published data on vicinal LSMO films so far had concerned ultrathin films only (12–25 nm) [5].

4. Preliminary magnetic characterization

Longitudinal MOKE microscopy was used for investigation of the magnetic domain arrangement of vicinal LSMO films that were patterned by UV photolithography and argon ion etching to form 50 μm wide lines and varying lengths (L = 100, 150, 200 or 300 μm) depending on the voltage probes used.

In our MOKE setup, the sample is positioned on a plane inclined at 45° with respect to the incident polarized light. Two polarizers are used, one for polarizing the incident light, the other for analyzing the reflected one. Due to the dependence of the Fresnel coefficients on the local magnetization, the light polarization state changes after reflection from the sample surface. In proper conditions and after some image processing the setup enables us to measure the in-plane longitudinal magnetization. The images were acquired by a Hamamatsu 4880-80 CCD camera operating in a 14-bit mode with 324000 pixels. The pixel size was 4 μm and numerous images were accumulated in order to increase the signal-to-noise ratio. Magnetic hysteresis loops can be calculated by averaging the measured MOKE signals over the specified image area. Figure 3 shows an example of such cycles, measured on the 40 nm thick 10° vicinal LSMO film, when H was applied parallel or perpendicular to the step directions. The loop of a 40 nm thick LSMO film deposited on standard STO(001) substrate was added for comparison. As expected from simulations made by Zhao et al. [7], an easy direction was found when the field was applied along the steps and a hard direction when it was applied across the steps, i.e. the coercive field is the largest when the external field is along the step direction and vanishes when external field is perpendicular to the step direction. In the easy direction, the coercitive field was 11.6 Oe, which, is much higher than the one obtained on STO(001), which is 2.7 Oe in the [110] or [1–10] directions.

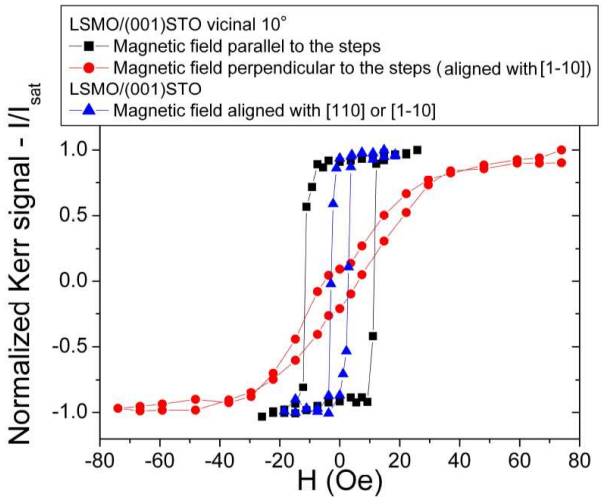

Fig. 3. MOKE hysteresis loops measured at 300 K on a patterned 40 nm thick 10° vicinal LSMO film, with H applied parallel or perpendicular to the step directions (H always perpendicular to the patterned line, see Fig. 4). The loop of a 40 nm thick LSMO film deposited on standard STO(001) substrate is added for comparison.

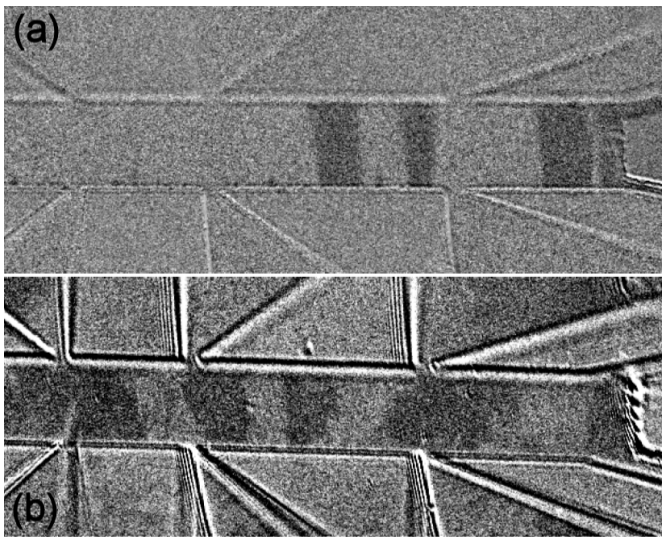

Fig. 4. MOKE images of magnetic domains in 40 nm thick films recorded with an applied magnetic field in the plane of the films and perpendicular to the patterned lines (i.e. vertical in these images). (a) 10° vicinal LSMO film; (b) LSMO on standard STO(001). Black and white regions represent magnetic domains with magnetization of opposite direction.

At last Fig. 4 shows two examples of MOKE images. Black and white regions represent magnetic domains with magnetization of opposite direction. The domain arrangement was visualized by subtracting two images. An image was first acquired in an applied field higher than 80 Oe, high enough for reaching the saturated state (i.e. with all domains pointing in the same direction), and then subtracted from a second image taken in an applied field lower than the coercive field in the transition region (for which some distribution of the domains is expected). The magnetic field was applied in the plane of the film and perpendicular to the line. The first observation to be noted in Fig. 4 is the shape of the observed magnetic domains. It is a straight line perpendicular to the bridge length in the case of the 10° vicinal films, whereas the domain boundary is either perpendicular or at 45° in the case of the LSMO on standard STO(001). These qualitative results confirm the different magnetic domain arrangement in vicinal and non-vicinal LSMO films as can be expected from the hysteresis loops.

5. Conclusions and perspectives

We showed that even with relatively high thicknesses (40–75 nm), very smooth LSMO films and regular step-terrace structures corresponding to the replication of the substrate surface could be obtained for angles higher than 4°. The AFM images confirmed the vicinal growth as indicated first by the XRD study. The step-induced magnetic anisotropy could be demonstrated for 40 nm thick films at 300 K from magnetization hysteresis loops measured by MOKE. At last, preliminary $1/f$ noise measurements were earned out on the 40 nm thick 10° vicinal LSMO. The normalized Hooge parameter is commonly used for comparing noise level of different materials of various size and at various frequency. It is defined as the ratio of the voltage spectral noise density times sample volume over the square of the sample voltage at 1 Hz. Values in the $4 \times 10^{-29} - 4 \times 10^{-28}$ m^3 range were found, which are promising results since they are comparable to the values measured in LSMO of the same thickness deposited on standard STO(001) [8]. Therefore, in addition to the uniaxial magnetic anisotropy observed in the 40 nm thick films deposited on the 10° vicinal STO(001) substrates, no noise in excess was measured neither parallel nor perpendicular to the steps. Both results are of high importance for future device fabrication making use of step-induced magnetic anisotropy, since the magnetization value is directly related to the ferromagnetic volume involved and a low signal-to-noise ratio is expected. Next steps are to measure systematically the magnetic and electrical properties of the films for all angles and thicknesses.

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