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# Step-induced uniaxial magnetic anisotropy of $La_{0:67}Sr_{0:33}MnO_3$ thin films

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The magnetic anisotropy of epitaxial $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) thin films on vicinal, $TiO_2$-terminated $SrTiO_3$ substrates is investigated. Atomic force microscopy shows a regular step-terrace structure on the LSMO surface which is a replication of the surface of the substrate. The films show in-plane uniaxial magnetic anisotropy at room temperature, with the easy axis along the step direction. At low temperature the films show biaxial crystalline anisotropy with easy axes along [110], and hard axes along the [100] direction of LSMO. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2143136]

Since double exchange perovskite manganites, such as $La_{0.67}Sr_{0.33}MnO_3$ (LSMO), show colossal magnetoresistance and have spin polarization close to 100%, this material is a good candidate for miniature spintronic devices.[1] Various properties such as magnetization reversal mechanism, domain structure and coercivity of LSMO are directly or indirectly determined by the magnetic anisotropy. The magnetic anisotropy behavior depends on the structure and morphology of the film, which in turn is related to the substrate used. Thin films of LSMO on diverse substrates show different magnetic anisotropy behavior.[2,3] Room temperature magnetic anisotropy of epitaxial LSMO (001) films on $SrTiO_3$ (STO) substrates were studied and found to be dominated by stress effects over magneto-crystalline anisotropy.[4] Later studies in LSMO (001) films showed that the biaxial anisotropy is magneto-crystalline in nature.[5] Furthermore, also biaxial in-plane crystalline anisotropy of LSMO films at different temperature was investigated.[6]

Within the film surface uniaxial in-plane magnetic anisotropy can be created if the rotational symmetry is broken by regular atomic steps on the surface. Uniaxial anisotropy induced by surface steps on ultra thin films of metallic ferromagnets have been studied previously.[7–9]

Step-induced uniaxial anisotropy in LSMO films on STO substrates with a large vicinal angle of 10° was recently investigated at low temperature (80 K). It was found that the LSMO film showed uniaxial anisotropy with easy axis along the substrate steps.[10] Here, we report on the influence of substrate steps on the in-plane magnetic anisotropy of LSMO thin films of different thickness at various temperatures. We analyzed two representative LSMO films with thicknesses of 7 and 25 nm, which are grown epitaxially on STO substrates with very low vicinal angles of 0.13° and 0.24°, respectively, and with different in-plane orientation of the step direction. We find that at room temperature the films show in-plane uniaxial anisotropy, while at low temperature biaxial anisotropy dominates over uniaxial anisotropy.

The epitaxial LSMO films were grown on STO substrates by pulsed laser deposition at 750 °C from a stoichiometric target at oxygen background pressure of 0.35 mbar. A KrF excimer laser ($\lambda = 248$ nm) is used with laser fluence of 3 J/cm$^2$ and a pulse repetition rate of 1 Hz. The target to substrate distance was fixed at 4 cm. Before deposition, the STO substrates were chemically treated and annealed at 950 °C to obtain single $TiO_2$ termination.[11]

The treated substrate showed straight steps on its surface with terrace width depending on the vicinal angle of the substrate. The vicinal angle and step direction of the substrates were determined using x-ray diffraction (XRD). After LSMO deposition, the films were cooled to room temperature with 10 °C/min ramp rate at 1 bar of oxygen gas pressure. Reciprocal space mapping measurements and XRD 2-theta scans show that the LSMO films grow epitaxially in the [001] direction. The thickness of the films were determined using low angle XRD and surface morphology was analyzed by atomic force microscopy (AFM).

Magnetic measurements, like in-plane magnetic anisotropy behavior, were done using a vibrating sample magnetometer (VSM) at room temperature and 160 K. For each film, hysteresis loops were taken at different in-plane field directions with intervals of 5°. Figure 1 shows two hysteresis loops obtain with the 25 nm thick LSMO film at room temperature, taken along 130° and 40° with respect to one edge of the substrate crystal ([100] direction). These field directions were found to be the in-plane easy and hard directions, respectively. A clear difference in the remanence value can be seen. AFM measurements show that the film surface has steps which are a clear imprint of the treated STO substrate surface. All LSMO films, even with thickness up to 80 nm, show this atomically flat stepped surface, with only unit cell high steps. Note that the step direction of each substrate is

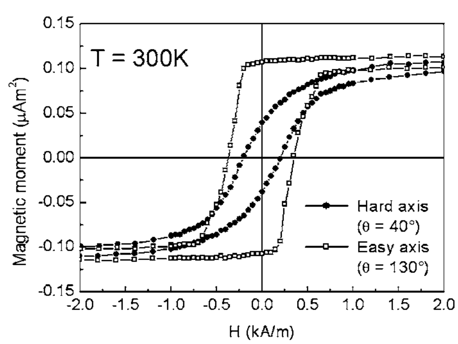


FIG. 1. Magnetic hysteresis loops of a LSMO film of thickness 25 nm, with field applied in-plane along the easy axis (direction of 130° angle with respect to one crystal edge [100] of the substrate) and hard axis (direction of 40°) at room temperature.

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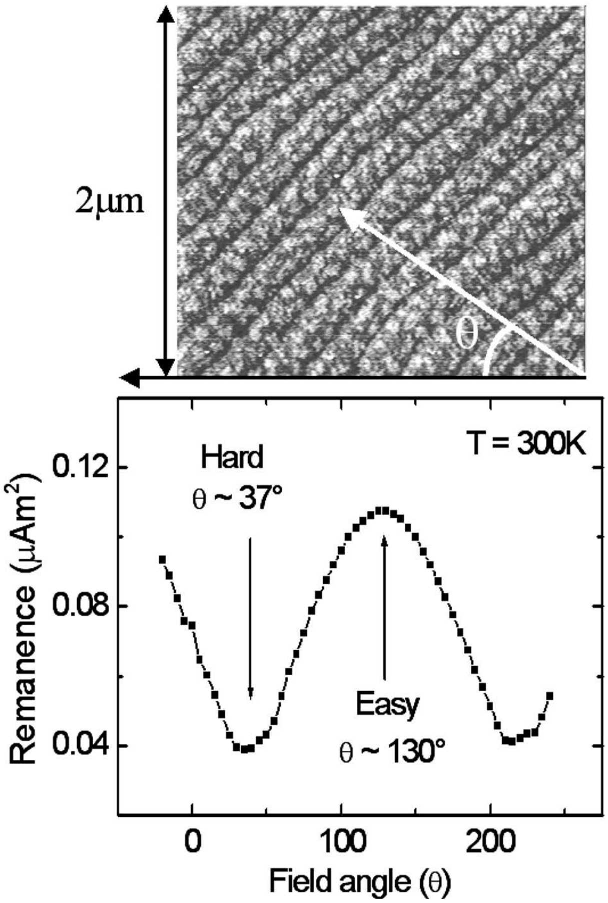


FIG. 2. Remanence vs in-plane field angle of a 25 nm thick LSMO film at room temperature (bottom panel). Arrows denote easy and hard directions. AFM image of the same film (top panel) with the step direction $\theta = 133 \pm 5°$. The gray scale range is from 0 to 2 nm.

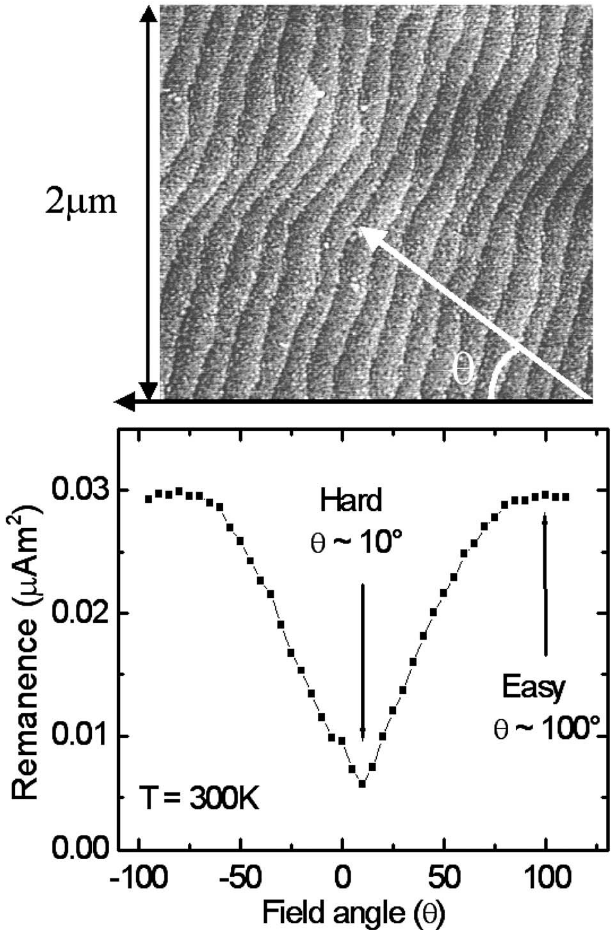


FIG. 3. Remanence vs in-plane field angle of a 7 nm thick LSMO film at room temperature (bottom panel). Arrows denote easy and hard directions. AFM image of the same film (top panel) with the step direction $\theta = 105 \pm 5°$. The gray scale range is from 0 to 2 nm.

different because of the variation in miscut direction. A typical example of an AFM image of the 25 nm thick film is shown in the top panel of Fig. 2. The remanence of this film, taken from hysteresis loops at different field angles, versus in-plane field angle ($\theta$) is also given in Fig. 2 (bottom panel). Here $\theta$ is defined as angle between applied field direction and the [100] STO crystal direction. There is an oscillation with periodicity of 180° with highest remanence at $\theta \sim 130°$ and lowest remanence at $\theta \sim 37°$. From this, we can conclude that this film has a uniaxial anisotropy with the easy direction along 130° and the hard direction along 37°. From the AFM image obtained from the same film, we found that the step direction is along the $133° \pm 5°$ direction, which corresponds with the easy axis, whereas the hard axis is perpendicular to the step direction (top panel of Fig. 2).

Remanence versus field angle of the LSMO film with a thickness of 7 nm and a different step direction is given in Fig. 3. Again, the remanence shows an oscillation with 180° periodicity. The highest remanence value was found at $100° \pm 8°$, whereas the lowest remanence is found in the $10° \pm 8°$ direction. This leads to the conclusion that this film also has a uniaxial anisotropy, with the easy axis along the 100° and the hard axis along the 10° direction. Once more, the easy axis of the uniaxial anisotropy in this film coincides with the step direction of the film as confirmed by the AFM image (step direction is $105° \pm 5°$) of the same film (top panel of Fig. 3). Subsequently, we have examined the anisotropy behavior of the films at low temperature (160 K). In Fig. 4 the remanence versus field angle at 160 K is plotted for the same film that is described in Fig. 2. Again the remanence oscillates with in-plane field angle. However, in contrast to the room temperature data, the remanence at 160 K oscillates with a periodicity of 90° with the high value at 45° and lowest value at 0°. Here, the 45° and 0° are the [110] and [100] crystal directions which are the easy and hard axis, respectively. Hence, at low temperature the film has in-plane biaxial anisotropy instead of uniaxial anisotropy. In this case the easy and hard axes have no relation with the step direction of the film surface. Consequently, the origin of the biaxial anisotropy is not dependent on the direction of the sur-

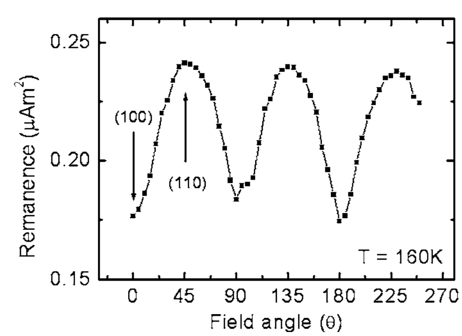


FIG. 4. Remanence vs in-plane field angle at 160 K for the same 25 nm LSMO film as in Fig. 2. Arrows denote easy and hard direction, which corresponds to the [110] and [100] crystal axes, respectively.

face steps of the substrate. This biaxial anisotropy, at low temperature, is crystalline in nature with easy and hard axis along the [110] and [100] directions, respectively. Similar results (not shown) are obtained at low temperature for the film which is described in Fig. 3.

There are several possible interpretations that could explain uniaxial anisotropy on stepped film surfaces. One is the magneto-crystalline anisotropy due to broken bonds and missing atoms at step edges,[7] which creates uniaxial anisotropy with easy axis either along or perpendicular to the step direction, according to Neèl's model. Another would be the magneto-elastic anisotropy due the twofold strain relaxation of the film at the step edges in a direction perpendicular to the steps. If present, this strain relaxation in one direction, only at step edges, would result in a decrease of the LSMO in-plane lattice parameter in the direction perpendicular to the steps, which in turn results in a uniaxial anisotropy. A third explanation could be the magneto-static anisotropy, which is originated from the uniaxial roughened surface of films grown on vicinal substrates.[8,12] Earlier studies on the temperature dependence of the anisotropy constant of LSMO films on different substrates show that the strain induced by the substrates is not affecting the crystalline anisotropy constant.[6] As a consequence, we expect that the uniaxial strain at the step edges of the substrates is not affecting the anisotropy. Magneto-static anisotropy is directly proportional to the magnetization value of the sample. As the temperature increases magnetization of the sample decreases, which means that the magneto-static anisotropy also should decrease. If there is a decrease in uniaxial anisotropy constant with increasing temperature, then it can be concluded that magnetostatic shape anisotropy due to the steps could be the reason for uniaxial anisotropy. In order to analyze this, torque magnetometry at different temperatures is under way. The biaxial anisotropy, which is dominating over uniaxial anisotropy at low temperatures, is determined by the fourfold crystalline anisotropy in the LSMO film on the STO substrate, which increases with decreasing temperature.[6]

In conclusion, at room temperature uniaxial anisotropy is seen in LSMO films grown on STO substrates with low vicinal angle, with easy axis lying along the STO step direction. At low temperature the same film shows biaxial crystalline anisotropy with easy and hard axis along [110] and [100] crystal directions, respectively. This behavior has direct influence on the application of LSMO in magnetic device structures.[1]

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