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Thickness and angular dependent magnetic anisotropy of La$_{0.67}$Sr$_{0.33}$MnO$_3$ thin films by Vectorial Magneto Optical Kerr Magnetometry

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Abstract. We investigate the in-plane magnetic anisotropy in La$_{0.67}$Sr$_{0.33}$MnO$_3$ thin films grown on SrTiO$_3$ (001) substrate using angular dependent room temperature Vectorial Magneto-Optical Kerr Magnetometry. The experimental data reveals that the magnetic anisotropy symmetry landscape significantly changes depending upon the strain and thickness. At low film thickness (12 and 25 nm) the dominant uniaxial anisotropy is due to interface effects, step edges due to mis-cut angle of SrTiO$_3$ substrate. At intermediate thickness, the magnetic anisotropy presents a competition between magnetocrystalline (biaxial) and substrate step induced (uniaxial) anisotropy. Depending upon their relative strengths, a profound biaxial or uniaxial or mixed anisotropy is favoured. Above the critical thickness, magnetocrystalline anisotropy dominates all other effects and shows a biaxial anisotropy.

1. Introduction
La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO) is considered to be a promising candidate for spintronic devices. It exhibits nearly 100% spin polarization and room temperature ferromagnetism ($T_c \approx 370$K). LSMO thin film properties are very sensitive to thickness, strain, temperature etc. Due to the different lattice parameters between LSMO and SrTiO$_3$ (STO) (001), rhombohedral LSMO undergoes an in-plane biaxial tensile strain when grown on STO (001) and imposes a tetragonal distortion in to film. As STO (001) is cubic, the in-plane biaxial tensile strain induced into the film is isotropic and therefore inducing four-fold magnetic anisotropy. For a cubic crystal, the magneto-elastic energy is equal in all the directions. Hence, it does not contribute to anisotropy and the resultant biaxial magnetic anisotropy is due to strong influence from magneto-crystalline effects. Lecoeur, P. et al [1] observed biaxial anisotropy in LSMO film with buckled remanent spin states along $<110>_{pc}$ crystallographic axis. However, other studies also reported that LSMO films can exhibit a uniaxial anisotropy and it is because of the steps transferred into film from the mis-cut...
angle of STO (001) substrate[2]. In addition to above studies, several authors artificially tuned uniaxial anisotropy of LSMO along the step edge direction by growing thin films on vicinal STO (001) substrates[3].

Here, we report thickness and angular dependent magnetic anisotropy studies of strained epitaxial LSMO thin films of various thicknesses (50, 25 and 12 nm) epitaxially grown onto STO (001) substrates by pulsed laser deposition technique.

2. Experimental details

The LSMO thin films were epitaxially grown on single crystal STO (001) substrates by pulsed laser deposition (PLD) technique. The films of three different thicknesses (50, 25 and 12 nm) were grown at 720°C and 0.35 mbar oxygen pressure. After deposition, films were immediately cooled down to room temperature at the rate of 10°C per minute at 7x10^-2 mbar oxygen background pressure. Room temperature magnetic anisotropy measurements in thin films were carried out by using high resolution angular dependent vectorial Magneto-Optical Kerr (v-MOKE)[4] magnetometry.

3. Results and discussion

The structural, electrical and magnetic transport measurements were studied and the results are tabulated in table 1. Thickness and angular dependent in-plane magnetic anisotropy studies of LSMO thin films grown on STO (001) substrates were performed at room temperature by v-MOKE magnetometry in longitudinal geometry. For all the measurements, the samples were mounted on eucentric goniometer that allows keeping constant the reflection plane during the angular measurements. For each film thickness,

| Substrate | LSMO film Thickness | Out-of-plane lattice parameter (nm) and strain (%) | FWHM (°) | RMS Roughness (nm) | Tp (K) | Tc (K) | Magnetic Anisotropy |
|-----------|---------------------|------------------------------------------|---------|------------------|-------|-------|-------------------|
| STO 50 nm | 0.38511 (-0.642)    | 0.119°                                   | 0.159   | >425             | 348   |       | Biaxial + off-axis Uniaxial |
| STO 25 nm | 0.38512 (-0.645)    | 0.117°                                   | 0.127   | 369              | 345   |       | Uniaxial           |
| STO 12 nm | 0.38492 (-0.6914)   | 0.159°                                   | 0.17    | 349              | 340   |       | Uniaxial           |

hysteresis loops were measured at different magnetic field directions within the film plane with an interval of 4.5° each for about 360°. Figure 1 shows the angular dependence of normalized remanence magnetization of LSMO/STO (001) films of varying thickness with easy axis (e.a) and hard axis (h.a) indicated by arrows respectively. Figure 1(a) shows the 2D plot of angular dependent remanence fields of LSMO film of 50 nm thick has biaxial anisotropy with easy axes aligned along <110> axis. Figure 1(b) shows the polar plot of remanence magnetization that has butterfly structure with four lobes showing a biaxial anisotropy. One should observe that the strength of easy axes is not same in all the directions and also the periodicity between easy axis and immediate hard axis is not exactly 45°, suggesting that there is an additional anisotropy present in the film (unless very weak) that is induced by the surface symmetry breaking due to the step formation. Therefore, there exists a competition between a biaxial (strong) due to magnetocrystalline anisotropy and uniaxial (weak) due to steps formation with a small offset angle. Moreover, as the thickness of the thin film decreases from 50 to 12 nm, there is a change in anisotropy from biaxial to uniaxial. The results are clearly visualized in the angular dependent 2D plots of the remanence magnetization (figure 1(d)) of the LSMO film of 25 nm thick. There are periodic oscillations observed with 180° periodicity and the easy axis is aligned along [100], whereas the hard axis is aligned along [010] axis. Figure 1(g) shows the remanence field at different angles for LSMO film of 12 nm thickness also shows uniaxial anisotropy. Also, there is a change in easy axis direction as compared to 25 nm film and this can due to the different mis-cut angle and direction of STO (001) substrate. Figure 1(e
and h) shows the remanence polar plots of 25 and 12 nm respectively, with symmetrical lobes clearly indicating a well-defined uniaxial anisotropy.

Figure 1: (a, d and g) v-MOKE angular dependent remanence magnetization of LSMO films of varying thickness grown on STO (001) substrate; (b, e, h and c, f, i ) are the corresponding polar plots and AFM topography respectively.

As STO (001) is cubic, the LSMO film undergoes an in-plane biaxial tensile strain. The magneto-elastic energy (due to strain) is constant in the film plane. Therefore, the cause for anisotropy is not due to magneto-elastic effects but it is owing to defects in crystal structure. Due to step formation, there are broken bonds along the step edges and direction (as observed in AFM topography in figure 1(c, f and i). As the thickness of LSMO film decreases, magnetic anisotropy exhibits a strong uniaxial anisotropy. This suggests that the film at lower thickness has strong influence of steps and mis-cut angle from STO (001) substrates in determining the magnetic anisotropy. In order to obtain clear insight about the anisotropy, low temperature v-MOKE and in-plane XRD studies are underway.

Conclusion
In summary, we have presented the angular dependent in-plane magnetic anisotropy properties of LSMO thin films of different thicknesses grown on STO (001). The magnetic anisotropy behavior of LSMO thin is thickness dependent. At lower thickness, uniaxial anisotropy is more predominant in LSMO films and is due to the step edges and mis-cut angle of STO (001) substrate. In the intermediate thickness range, there exists a competition between uniaxial and biaxial anisotropy. Depending upon their relative strength, either a uniaxial or biaxial or mixture of both is present. At higher thicknesses, the magneto-crystalline anisotropy dominates all other effects and leads to a bi-axial anisotropy.

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