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Interface defects induced vertical magnetic anisotropy in Sr_2FeMoO_6 thin films

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ABSTRACT

Pulsed laser deposition was used to fabricate high quality Sr_2FeMoO_6 thin films on $SrTiO_3$ and $SrLaAlO_4$ single crystal substrates. The focus of our research has been on the magnetic properties of Sr_2FeMoO_6 thin films and we have observed an outstanding shift from parallel in-plane magnetic anisotropy to perpendicular out-of-plane anisotropy, induced by the substrate. Our results also provide the first experimental evidence of magnetic stripe domain pattern in Sr_2FeMoO_6 thin films. Compared with previous studies for magnetoresistive thin films, we propose a conspicuous mechanism behind the shift in magnetic anisotropy. Rather than being induced directly by the lattice mismatch, we find the possible contribution to perpendicular magnetic anisotropy from over-relaxation at the film substrate interface or from other crystalline defects such as low-angle grain boundaries.

1. Introduction

High Curie temperature (T_C), between 410–450 K, and 100% spin polarization along with magnetoresistive phenomenon have attracted a lot of interest to the double perovskite Sr_2FeMoO_6(SFMO) [1,2]. The magnetic attributes make SFMO an interesting material on its own, but also a valuable candidate as material for spintronics. Therefore, many studies have focused on the optimization of SFMO thin films, which are necessary for multilayer based applications. As a ferrimagnetic material, the magnetic domain structure is an important element for magnetic properties of Sr_2FeMoO_6 thin films. Perhaps the most common embodiment of the effects of domains is the magnetic hysteresis, but domains are also the key component to understand any magnetic behaviour in ferromagnetic (ferrimagnetic) materials. Magnetic properties of SFMO, polycrystalline bulk and thin films, have been studied intensively, both theoretically and experimentally [3–11]. However, there appears to be very little research done to directly image magnetic domains in SFMO films. Only few studies have been carried out with magnetic force microscopy (MFM) [12,13]. Kalanda et al. have reported a correlation between single grains in SFMO thin films and domain structure as well as a correlation between clusters in SFMO thin films and domain structure [12]. MFM has been used in various research projects to study magnetic domain structure in other magnetoresistive thin films, especially in $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) films, but also in other magnetoresistive materials [14–17]. In these materials, the common magnetic stripe domain pattern has been reported and the domain structure has been shown to be affected by the thin film thickness and choice of the substrate material [14,15,17]. Stripe like patterns indicate increased magnetic anisotropy [18]. Directly related to the magnetic domain structure, the magnetic anisotropy will also play a key role in magnetic phenomena in SFMO thin films.

Magnetic anisotropy is usually divided into three separate categories: magnetocrystalline anisotropy, magnetoelastic anisotropy and magnetic dipolar anisotropy, all which contribute in the direction of the magnetic moment in thin films [19]. Magnetocrystalline anisotropy originates from the spin-orbit interaction and results with easy and hard magnetization axes relative to the lattice vectors. Magnetoelastic anisotropy refers to anisotropy where the direction of magnetization is altered by the structural stress in the crystalline lattice. Magnetic dipolar anisotropy, a.k.a. shape anisotropy, usually dominates the anisotropy in thin films and sets the magnetic moment to in-plane of the film surface. Magnetoelastic anisotropy, induced by the lattice mismatch between the substrate and film material, has been used to shift magnetic anisotropy from parallel to perpendicular of the film plane orientation [19–21]. Before the fabrication of SFMO spin valves the

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phenomenon of magnetic anisotropy must be addressed, since anisotropy will have a role in multilayer spinvalves [22,23].

The substrate material and the film thickness have been shown to have a great importance on the structural and the magnetic properties of SFMO thin films [9,24–28] but, according to our knowledge no study has been conducted with MFM to study the effects of substrate material and the film thickness directly to magnetic domains. In this paper, magnetic anisotropy and domain structure of SFMO films on different substrates with different thicknesses has been studied. The results reveal a shift from parallel in-plane easy axis orientation to perpendicular in-plane orientation and also traces of stripe domain pattern. Additional magnetometric measurements provide more information about domain dynamics. The effects of pinning are also analyzed from the magnetic hysteresis measurements.

2. Experimental details

Four SFMO thin films were deposited on $SrTiO_3$ (STO) and $SrLaAlO_4$ (SLAO) single crystal substrates, two films on each substrate with different thicknesses, with pulsed laser deposition (PLD). The lattice parameters for STO are $a = b = c = 3.901$ Åand for SLAO $a = b = 3.756$ Åand $c = 12.636$ Å, which produce the in-plane lattice mismatch values with SFMO of −1.05% and −4.73%, respectively. The in-plane lattice mismatch between the substrate lattice parameter, a_{sub}, and the SFMO lattice parameter, a_{SFMO}, is calculated using the formula $\eta = (\sqrt{2}a_{sub} - a_{SFMO})/a_{SFMO}$ based on the values of SFMO lattice parameters published in [29]. The substrate is heated from room temperature to the deposition temperature at the rate of 25 °C/min in 9 Pa Ar-atmosphere. Deposition temperature was 900 °C, which has shown to result with smooth SFMO thin films [30,31]. We used XeCl excimer laser with laser wavelength of 308 nm and the pulse frequency adjusted to 5 Hz. Laser fluence was 1.41 J/cm^2. Film thickness was controlled by varying the pulse number. After the laser deposition, the annealing treatment was carried out by keeping the temperature constant for 10 min, before the temperature was decreased back to the room temperature with rate of 25 °C/min. Thinner films were deposited with 500 and thicker films with 2000 PLD laser pulses. The thinner SFMO films fabricated on STO and SLAO with 500 pulses are named STO-1 and SLAO-1 and the thicker films fabricated on STO and SLAO with 2000 pulses are named STO-2 and SLAO-2. SFMO target was prepared for pulsed laser deposition using solid state synthesis. Details for target preparation are reported elsewhere [31].

Surface structure and domain structure of the films were studied with an Innova atomic force microscope provided by Bruker. The atomic force microscopy (AFM) and the magnetic force microscopy (MFM) scans were performed at room temperature. The AFM imaging was done with 20 μm × 20 μm, 10 μm × 10 μm, 5 μm × 5 μm, 2 μm × 2 μm scan sizes from four different areas on the SFMO thin films using the tapping mode. The surface roughness was determined as the root mean square (RMS) roughness from 20 μm × 20 μm, 10 μm × 10 μm, 5 μm × 5 μm scans. The magnetic images were obtained from 20 μm × 20 μm, 10 μm × 10 μm and 5 μm × 5 μm scans with 25 nm, 50 nm, 100 nm and 200 nm lift-heights.

X-ray diffraction (XRD) measurements were done using a Philips X'Pert Pro MPD diffractometer with a Schulz goniometer. To check for the possible impurity phases and to determine the *c*-axis parameter, $\theta - 2\theta$ measurements were done between 20°–114°. A detailed $\theta - 2\theta$ scan was done for the (336) to determine the *a*-axis parameter and a texture $\phi - \psi$ scan for (204) peak at $2\theta = 57.106°$ to check the *c*-axis orientation. A detailed $2\theta - \phi$ scan was performed for the (204) peak and used for lattice defect analysis to obtain the scale of in-plane lattice distortion. The lattice parameter values are used to calculate the strain in the films. Film thicknesses were determined with X-ray reflectivity measurements (XRR), conducted on STO-1 and SLAO-1 both fabricated with 500 pulses. The results indicate 30 nm thickness for the 500 pulse films on both STO and SLAO. In this thickness range, we assume linear growth rate, which means that 2000 pulses correspond to thickness of 120 nm, which is slightly less than in our earlier films grown with different growth parameters [10,28].

Besides MFM, we used a MPMS XL SQUID magnetometer provided by Quantum Design to study the magnetic properties. The zero field cooled (ZFC) and the field cooled (FC) magnetizations were measured as a function of temperature in 100 and 500 mT magnetic fields between 10–400 K temperature for all films. The Curie temperature was determined from the minimum of the first order derivative of 100 mT FC curve. Magnetic hysteresis loops were measured between ±1 T at 10 K. Saturation magnetization, M_{sat}, and coercive field, B_c, were obtained from the hysteresis loops. The SQUID measurements were conducted twice, magnetic field aligned differently respect to the SFMO lattice vectors, for all the films with magnetic field parallel first to [110], the diagonal of *a* and *b* lattice parameters, and second to [001], *c* lattice parameter.

3. Results

3.1. Lattice strain and crystalline defects

The $\theta - 2\theta$ scans between 20° – 114° are presented for all our SFMO thin films in Fig. 1(a). The results show clear (00*l*) peaks arising from SFMO thin films and the substrates. No impurity phase peaks are observed since the additional the small peaks observed around 44° and 52° arise from the sample holder. The pole figures of the texture analysis showed clear (204) and (132) peaks as is expected for fully textured *c*-axis oriented SFMO films [8,30]. The XRD results show films to be phase pure, highly textured and *c*-axis oriented.

$2\theta - \phi$ scans for (204) are presented in Fig. 1(b) and (c) for STO-2 and SLAO-2, respectively. $2\theta - \phi$ scans were also performed for thinner films but due to slight overlapping between the peaks of STO substrate and the SFMO film, accompanied by the smaller intensity from the thinner SFMO films, the analysis of the (204) peak in STO-1 becomes inconvenient. The broadening, in ϕ angle, results from low angle grain boundaries in the in-plane orientation of the crystal lattice. The broadening was determined as ϕ–FWHM (full width at half maximum) by fitting a Gaussian distribution function with the section of $2\theta - \phi$ scan corresponding to the 2θ value of the highest intensity of the SFMO (204) peak. The Gaussian fit resulted with ϕ – FWHM = 0.692° and ϕ – FWHM = 0.914° for STO-2 SLAO-2, respectively. SLAO-1 showed ϕ – FWHM = 1.013°. These results are consistent with our previous work also showing larger ϕ – FWHM values and therefore indicating larger spread of in plane crystalline orientations, i.e. more low angle grain boundaries, in the SFMO thin films fabricated on SLAO substrate [10].

Lattice parameters *a* and *c* were obtained by fitting a Gaussian distribution function to (336)- and (008)-peaks in the $\theta - 2\theta$ scans, respectively. The results are presented in Fig. 1(d). The dotted lines represent the values of $a_{SFMO} = 5.575$ Åand $c_{SFMO} = 7.893$ Å [29]. The results show that thinner STO-1 film lattice parameter deviates from bulk values and film is strained due to compressive in-plane lattice mismatch, −1.05%, between the substrate and SFMO, showing strain value of −0.86%. The in-plane and out-of-plane strain, ϵ_a and ϵ_c, were calculated as $\epsilon_l = (l - l_{SFMO})/l_{SFMO}$, where *l* is the SFMO lattice parameter, *a* or *c*, obtained from our XRD results and l_{SFMO} represents the unstrained SFMO lattice parameter according to [29]. Once the thickness increases, the SFMO film on STO becomes more relaxed, with ϵ_a value of −0.46%, and the lattice