Research Report

Domain Structure in LaFeO$_3$ Thin Films and its Role on Exchange Coupling

Jin Won Seo[1,2], Jean Fompeyrine[2], Heinz Siegwart[2] and Jean-Pierre Locquet[2]

[1]Institut de Physique
Université de Neuchâtel
CH-2000 Neuchâtel
Switzerland

[2]IBM Research
Zurich Research Laboratory
CH-8803 Rüschlikon
Switzerland

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[1]*Institut de Physique, Université de Neuchâtel, CH-2000 Neuchâtel, Switzerland*
[2]*IBM Research, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland*

ABSTRACT

The origin of the exchange bias between a ferro- and an antiferromagnet still is not fully resolved. Many structural parameters such as film thickness, roughness, domain size, domain walls, strain, dislocations, and surface defects play a role. To disentangle their individual contributions, one must correlate each of these structural effects with the exchange bias. Here we report our first such attempts using thin films of the antiferromagnetic orthoferrite LaFeO$_3$ grown by molecular beam epitaxy. We investigate how the epitaxial relationship and strain affect the domain configuration as well as their size and orientation. Next, we explore how these different domains can pin ferromagnetic domains in exchange-coupled systems. We conclude that decreasing the domain size of the antiferromagnet leads to a higher exchange bias. The highest exchange bias was obtained for LaFeO$_3$ films on MgO substrates.

INTRODUCTION

Antiferromagnets (AF) are essential elements in the read heads of current hard-disk drives, where their purpose is to pin the nearest ferromagnetic (FM) layer into one preferred orientation. This coupling between the spins in the AF and FM layers, called exchange bias, largely determines the efficiency and the long-term stability of the giant magnetoresistance ratio. Although discovered a long time ago [1], this phenomenon still is poorly understood. A number of experiments and theoretical models have suggested that the exchange bias is correlated with the presence and morphology of AF domains [2,3].

The AF orthoferrite $LaFeO_3$ (LFO) is an interesting model system to explore this correlation, as the AF axis A is uniquely defined along the **a**-axis of the lattice. The orthorhombicity of this crystal is large enough so that its structural recognition on a local scale leads to a direct determination of A. Recently, we reported that it is possible to observe the local magnetic contrast related to the AF domains in LFO films grown on $SrTiO_3$ (STO) by means of photoemission electron microscopy (PEEM) [4]. These AF domains could be directly correlated with the structural domains, as a detailed comparison between the magnetic sensitive PEEM and the structural measurements using transmission electron microscopy (TEM) has shown [4]. This illustrates the strong anisotropy in this system. Next, after growing a thin polycrystalline Co layer on top of LFO, the magnetic correlation between the AF and FM layer was studied. The results suggest that the FM spins are aligned in-plane, parallel to the projected spin directions of the underlying AF layer [5]. The local exchange coupling is stronger than the anisotropy in Co and takes place on an individual AF domain basis.

In this paper, we investigate how parameters, such as the growth conditions, epitaxial relationship and strain, affect the size, shape and orientations of the LFO domains. We conclude that the domain size plays a major role in determining the value of the exchange bias.

EXPERIMENTAL DETAILS

We used substrates of STO, $LaAlO_3$, MgO, and $MgAl_2O_4$, preannealed in vacuum for about one hour at 800°C. Here we only discuss the results obtained for STO and MgO, as the other two substrates exhibit a similar behavior. The 40-nm-thick LFO films were grown in a molecular beam epitaxy (MBE) system using the block-by-block growth method [6] at 750°C under a beam of atomic oxygen and a partial O_2 pressure of 5×10^{-6} Torr. After cooling to room temperature, a 3-nm-thin Co layer, followed by a 1-nm Pt protection layer, was deposited on top of the LFO film. The structural quality was monitored in-situ by reflection high-energy electron diffraction (RHEED) and ex-situ by D500 x-ray diffraction (XRD). For the TEM study, plan-view samples were prepared by grinding and thinning them with an Ar-ion beam from the substrate side to electron transparency. TEM studies were performed with a 200-kV JEOL2010 microscope. Atomic force microscopy (AFM) was performed using an UHV Omicron VT. The magnetization was measured using a Lakeshore vibrating sample magnetometer before and after annealed the samples in a 5000-Oe applied field for 15 min at 120°C.

RESULTS AND DISCUSSION

LFO is orthorhombic with lattice parameters $\mathbf{a}$ = 0.5557 nm, $\mathbf{b}$ = 0.5565 nm, $\mathbf{c}$ = 0.7854 nm. As shown in the schematic drawing (figure 1), the orthorhombicity of the unit cell is induced by the buckling of the Fe-O-Fe bonds to about 155° along the $\mathbf{a}$-axis. The spins of the Fe atoms are aligned parallel to the $\mathbf{a}$-axis, and order antiferromagnetically along the [110] direction. Epitaxial films of LFO can be grown on perovskite substrates owing to their compatible structure and small lattice mismatch (table 1). The perovskite subunit within LFO is indicated by bold lines. The small mismatch with STO ($\mathbf{a}$ = 0.3905 nm) suggests three epitaxial orientations where the $\mathbf{c}$-axis of $LaFeO_3$ is aligned parallel to the three $\langle 001 \rangle$ directions of STO. This results in two orientations with $\mathbf{c}$ in the substrate plane with a mismatch of 0.58% ($\mathbf{c}_{LFO} \simeq 2\,\mathbf{a}_{STO}$), where A points out of plane by 45°, and a third orientation where $\mathbf{c}$ points out of plane (mismatch 0.68%).
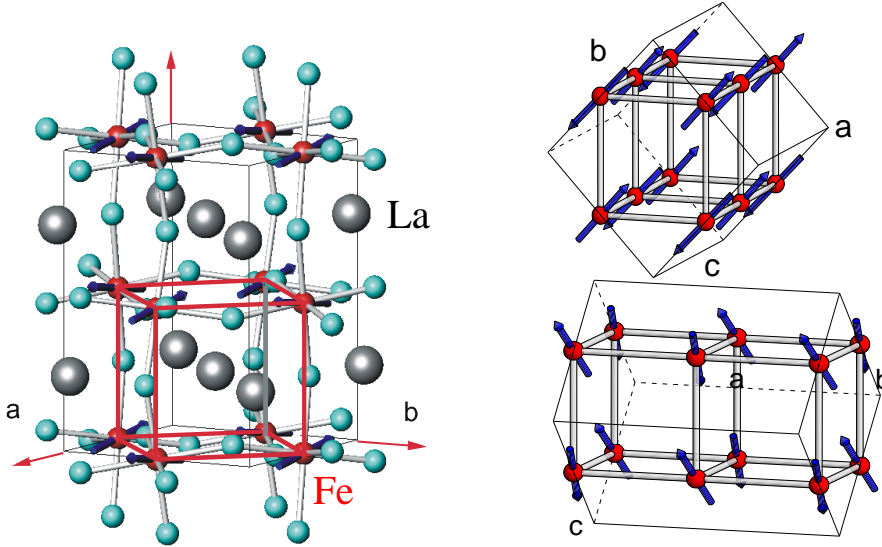

Figure 1. (Left) The LFO unit cell with the perovskite subunit outlined and (right) these subunits with the $\mathbf{c}$-axis in plane, rotated by 90°, and A pointing 45° out of plane.

Table I. Lattice mismatch of LFO compared to STO and MgO. Two orientations of LFO are considered, one having the $\mathbf{c}$-axis in plane and the other having it out of plane.

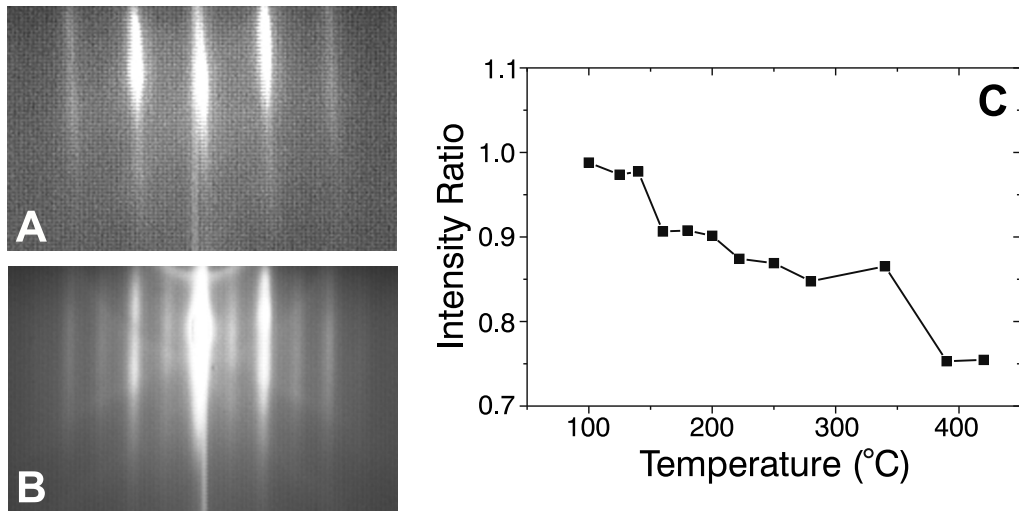|	d(001) SrTiO$_3$	d(001) MgO
d(001) LaFeO$_3$	0.58%	7.00%
d(110) LaFeO$_3$	0.68%	6.88%

Figure 2. RHEED pattern observed along the [001] azimuth (a) before growth on STO at 750°C and (b) during growth of LFO. (c) As the sample is being cooled down after growth, the relative intensity of the additional streaks increases almost linearly with temperature.

Figure 2a shows a RHEED pattern of STO along the [001] azimuth before deposition at 750°C. The RHEED streaks are well defined, indicating a flat substrate surface. As LFO deposition starts, additional streaks of twofold symmetry appear in figure 2b. The position of the superstructure lines indicates a doubling of the perovskite unit and suggests that the long **c**-axis of the LFO film is aligned parallel to the substrate surface. Upon cooling to room temperature, the relative intensity of this superstructure increases almost linearly, as indicated in figure 2c. This could either indicate that the domain size increases or that the structural coherence and orthorhombicity within the domains along the **c**-axis increase. However, high-temperature TEM studies (1000°C) revealed no change in domain size, so that the former hypothesis can be ruled out.

In figure 3 (left), a plan-view TEM diffraction image of the film on STO is shown. The diffraction image reveals superreflections along the [100]* and [010]* directions, which are in agreement with the superstructure seen by RHEED. The fourfold symmetry can be explained by
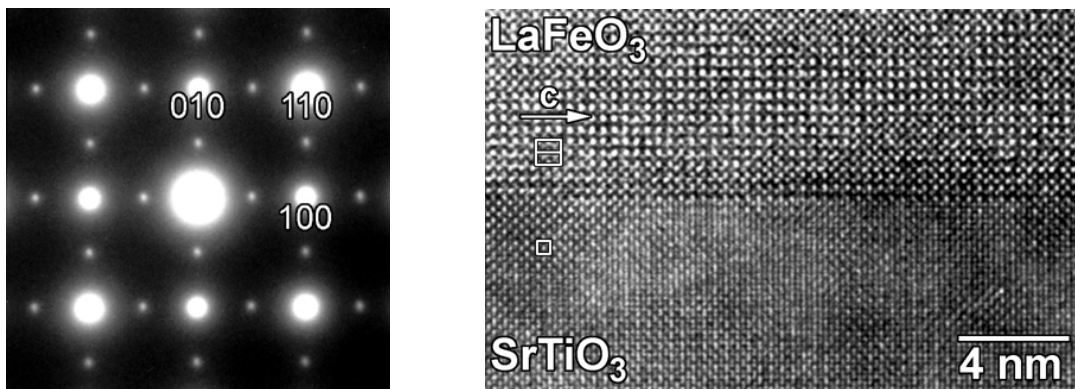

Figure 3. (Left) Plan-view TEM diffraction pattern of LFO on STO, the small spots being the superreflections. (Right) Cross-section TEM image confirming that the **c**-axis is in the plane.

the presence of twins leading to the overlap of two different diffraction patterns. In both twin domains, the **c**-axis lies in-plane, as confirmed by the cross-section image (figure 3, right). The twinning symmetry operation is given by a 90° rotation around the [110] axis of LFO, which is parallel to the substrate surface normal. Using one of the superreflections, the twin structures can be imaged in dark-field mode with the dark and bright contrast (see figure 5) depending on the orientation of the **c**-axis.

In the following, a detailed comparison of the roughness, domain size, and exchange bias of LFO films deposited under identical conditions on STO and MgO is made. AFM investigations of substrate and film surface have been carried out. The results reveal that the STO surface has an average peak-to-valley roughness of about 0.4 nm (one unit cell), whereas the MgO surface has a roughness of about 1.2 nm. The roughness of the respective LFO films remains 0.4 nm on STO, but increases to 3 nm on MgO as shown in figure 4. The increased roughness of the film grown on MgO can be explained by (i) the initial substrate roughness and (ii) the roughness induced during growth and domain formation. Whereas the two in-plane LFO domains on STO have the same out-of-plane structure, the third type on MgO has the **c**-axis out of plane. The LFO lattice parameter difference along [001] and [110] is 0.13%, increasing the roughness by 0.5 nm. If growth takes place on a unit cell-by-unit cell basis, then the longer **c**-axis itself can automatically lead to an enhanced roughness.

This increased surface roughness does not necessarily mean that the LFO/Co interface is also much more disordered. In fact, the well-defined streaks in the RHEED data during growth on MgO even suggest that the top surface of each domain is well ordered. The surface roughness is mainly due to the height difference between domains, and will not lead to increased disorder at the AF/FM interface. This should not significantly affect the value of the exchange bias.
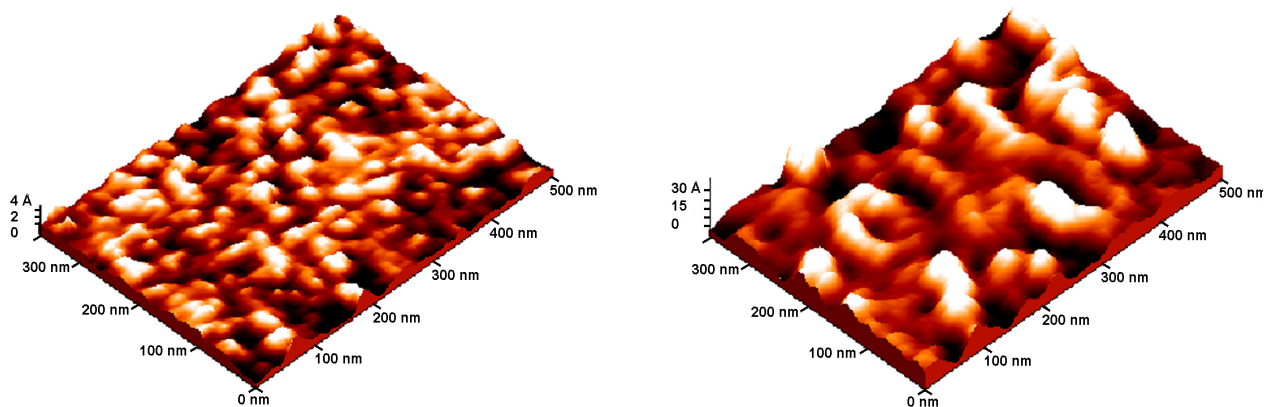

Figure 4. AFM images of a 40-nm LFO film on STO (left) and on MgO (right).
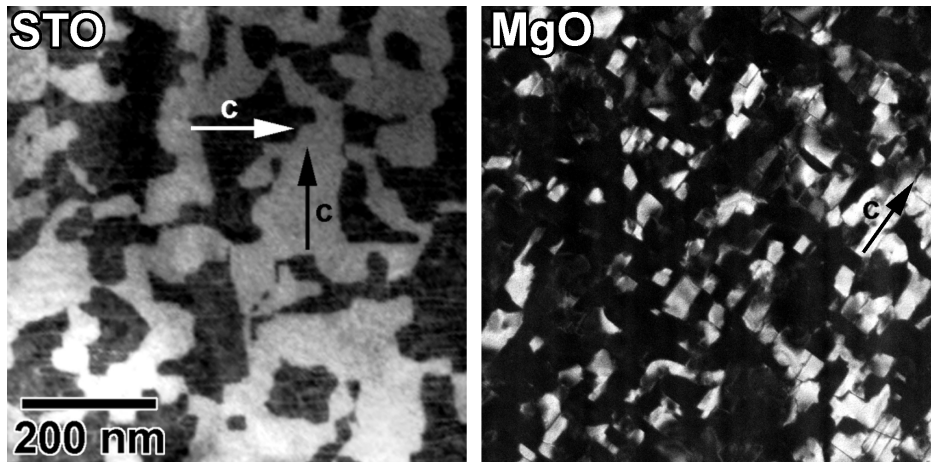
Figure 5. TEM dark-field images of LFO grown on (a) STO and (b) MgO.

The TEM analysis of both samples (figure 5) also shows the different domain sizes and orientations. The image reveals domains with a very irregular shape and an average size of about 200 nm. The domain walls are perpendicular to the substrate and sharply defined with the main facets along both **c**-axes. They extend from the film surface down to the substrate. Taking the spin orientations into account, these structural twins lead directly to two different AF domains whose *A* vectors are rotated by 90° relative to each other. The AF domains can be directly observed with PEEM microscopy [4], and we conclude that the AF domains are indeed seeded by the structural domains. For LFO on STO, the average domain size is 200 nm, compared to 25 nm on MgO. Moreover, on MgO the third domain type with the **c**-axis out of plane occupies about a third of the total area. What can explain the difference in domain size and orientation? The lattice mismatch suggests preferential alignment of the **c**-axis in plane compared to out of plane for growth on STO and MgO, respectively. As domain walls lead to a strain relaxation, one expects more domains to be nucleated on MgO. If this is the main relaxation mechanism, then the domain size should be inversely proportional to the mismatch. Hence, the ten times larger mismatch on MgO can qualitatively explain the much smaller domain size on MgO.

The appearance of these domains is quite unexpected, as both lattice mismatch and orthorhombicity are very small ($< 1\%$). In many perovskite-related compounds, the epitaxial strain in thin films grown on a cubic substrate is usually sufficient to induce a tetragonal lattice. The dislocation densities along and perpendicular to the **c**-axis differ by a factor of 3 to 5, suggesting an orthorhombic lattice, even for ultrathin films. Because the domain size does not change with temperature, they must appear during the initial nucleation phase.

Figure 6 shows the magnetization versus applied magnetic field of these two LFO/Co bilayers grown on STO and MgO. Both samples were set for 15 min at 120°C in a 5-kOe field. As the data show, the LFO films on STO and on MgO have an exchange bias of 12 and 37 Oe, respectively. In general, for samples grown under identical conditions, the exchange bias on MgO is about a factor three to five larger than that for those on STO, the highest value obtained so far (3-nm Co) being about 50 Oe. If the pinning takes place at uncompensated moments sitting on or at the domain walls, then the exchange bias should scale with the total domain wall length. For square domains, when the domains size (edge) is reduced by a factor L, the total domain wall length will increases by a factor of $(2L + 2)/4$. When the domain size decreases by a factor of ten, then the total wall length increases only by a factor of 5.5. Although our domains
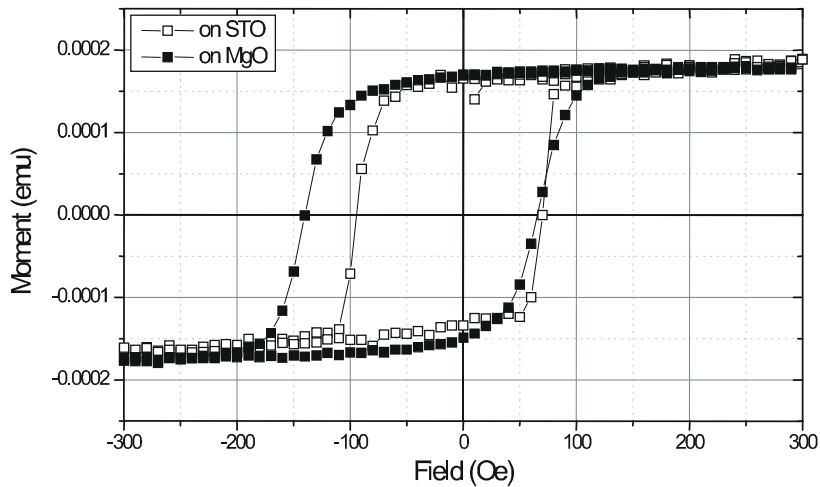
Figure 6. *M*(*H*) measurement of the LFO/Co bilayers grown on STO and MgO.

are not square, this simple estimate is in qualitative agreement with the observed results, and suggests that the domain size could be the main controlling factor for the exchange bias in our experiments.

CONCLUSION

We have shown that decreasing the structural domain size of LFO films by growing them on substrates having a larger lattice mismatch leads to increased exchange coupling with a FM film.

ACKNOWLEDGMENTS

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