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Magnetic properties of $SrTiO_3/Pr_{2/3}Ca_{1/3}MnO_3$ multilayers

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Abstract. Nanoscale ferromagnetic (FM) clusters embedded within insulating (I) layers of the antiferromagnetic $Pr_{2/3}Ca_{1/3}MnO_3$ (PCMO) are known to improve the magnetoresistance ratio of FM-I magnetoresistive multilayers. Here, we study the magnetic properties of perovskite superlattices comprised of five PCMO layers of variable thickness t separated by 5nm-thick spacers of $SrTiO_3$ (STO). Several multilayers (1.5 nm $\leq t \leq$ 8 nm) were grown on (001) STO substrates by magnetron sputtering. We show that the magnetic moment of PCMO due to the FM inclusions presents a maximum for $t \sim 3$ nm, accompanied by an abrupt increase of coercivity and exchange bias field. The nonmagnetic nature of the neighboring STO layers demonstrates that the observed behavior is not related to any interlayer exchange coupling, but the geometrical matching between layer thickness and FM domain size is the key driver for the enhancement of the FM moment and anisotropy energy. These results open a new door for the optimization of perovskite based spintronic devices.

1. Introduction

Understanding the physical properties of antiferromagnetic (AFM) thin films has become of utmost importance for multiple applications, such as exchange bias and tunneling magnetoresistance (TMR) devices [1-5]. For both these applications the role of the AFM material is not only to establish an interface magnetic coupling or an insulating energy barrier, but the inner magnetic structure of the AFM layer is capable of critically changing the physical behavior of the mentioned devices. In particular, manipulation of the nanoscale magnetic structure of AFM insulating layers has shown to have great impact on the performance of TMR manganite multilayers [3, 4]. In $La_{2/3}Sr_{1/3}MnO_3/Pr_{2/3}Ca_{1/3}MnO_3$ (LSMO/PCMO) superlattices, the presence of nanoscale ferromagnetic (FM) instabilities within the AFM layers of PCMO play a fundamental role in determining the low-field magnetoresistance (MR) ratio [3].

In the LSMO/PCMO multilayers, it has been demonstrated that for PCMO layer thicknesses close to 3 nm a maximum magnetic contribution due to FM nanoscale clusters produces an increase of the MR ratio as well. The origin of this maximum FM moment was suspected of not being related to an interlayer magnetic coupling of the PCMO with the neighboring LSMO ferromagnetic layers, however no direct evidence of this has been provided so far [5].

In this paper we study the magnetic properties of a series of perovskite multilayers, comprised of five repetitions of $SrTiO_3/Pr_{2/3}Ca_{1/3}MnO_3$ (STO/PCMO) bilayers. In this set of superlattices, magnetization data also show a maximum FM moment for PCMO thicknesses ~ 3 nm, in spite of the fact that the neighboring layers are now the non-magnetic strontium

titanate. This enhanced ferromagnetism is also accompanied by an enhancement of anisotropy energy, evidenced by a sudden increase of coercivity and exchange bias field. We discuss these results in terms of the geometrical matching between layer thickness and FM domain size in thin PCMO films, as well as modified magnetic interactions at the PCMO-STO interfaces.

2. Experiment

$SrTiO_3/Pr_{2/3}Ca_{1/3}MnO_3$ superlattices were grown by magnetron sputtering, using a DC source for the PCMO layers and a RF source for the STO layers. Film deposition was performed in an $Ar(90\%)/O_2(10\%)$ mixture at a total pressure of 400 mTorr. The substrate temperature was held at 800 °C during deposition. After deposition the samples were cooled down to 680 °C in 5 min, followed by a slow cooling to room temperature in 2 h in a 100 Torr O_2 atmosphere.

The superlattices are comprised of five STO/PCMO bilayers deposited on top of (001) STO substrates, starting with STO and terminated with an extra STO cap layer. The thickness of all the STO layers was kept constant at 5 nm, while the PCMO thickness (t) was varied between 1.5 and 8 nm (t = 1.5, 2.25, 3, 3.7, 4.5, 6, and 8 nm, respectively). The high quality of the samples was confirmed by the observation of several superlattice reflections in the X-ray diffractograms.

Isothermal magnetization (M) measurements as a function of magnetic field (H) at a temperature of 10 K were performed in a Quantum Design Squid magnetometer, equipped with a Reciprocating Sample Option (RSO) and a 50 kOe magnet. All the samples were cooled from room temperature to 10 K under an applied field of 1 kOe, after which a M-H loop between 10 kOe and -10 kOe was measured with the magnetic field applied parallel to the plane of the films. The magnetization of the PCMO layers was then obtained after subtracting the linear contribution of the STO.

3. Results and discussion

Figure 1 presents magnetic moment per unit area of four representative film samples as a function of the applied magnetic field. In these plots a clear FM contribution is observed in all the samples. As it has been previously demonstrated, this FM moment arises from nanoscale FM inclusions that form within the non-FM matrix of the PCMO [5, 6]. These FM nanoclusters are observed either in bulk or thin film samples. However, the behavior evidenced by our samples in Fig. 1 is unique to thin films of PCMO. Usually, one would expect the magnetic moment to increase monotonically with the thickness of the magnetic layer, simply because of the increase

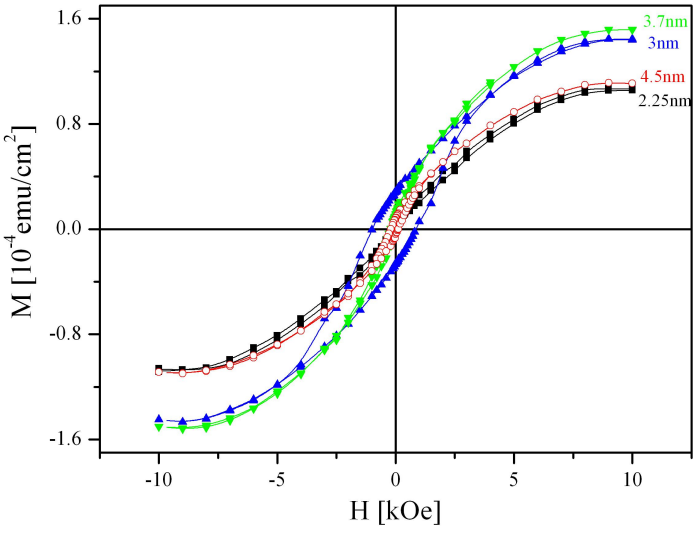

Figure 1. Magnetic moment per unit area vs applied magnetic field at 10 K in four representative samples. Labels indicate the thickness of the $Pr_{2/3}Ca_{1/3}MnO_3$ layers.

of magnetic volume. Nevertheless, our thin films show a maximum magnetic moment for the samples with $t = 3$ and 3.7 nm. It is evident in Fig. 1 that this unexpected enhancement of the FM moment is accompanied by a large coercive field as well.

In order to analyze the thickness dependence of the magnetic moment, in Fig. 2 we show the saturation moment of the films (M_S, the magnetic moment at $H = 10$ kOe) as a function of t. This figure shows that the expected increase of M_S with increasing thickness is only observed for $t > 5nm$. On the other hand, for small thicknesses the maximum magnetic moment is clearly observed for $t \sim 3$ nm. The very same behavior, with a maximum FM moment of PCMO at thicknesses ~ 3 nm was previously observed in another set of perovskite multilayers [5], where the FM $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) manganite was placed between the PCMO layers instead of the STO spacers. Then, these results further confirm the occurrence of an enhancement of the FM moment of the PCMO layers for $t \sim 3$ nm.

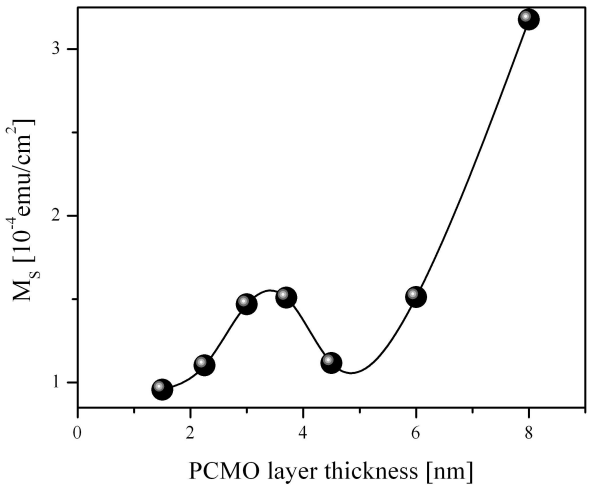

Figure 2. Saturation magnetic moment per unit area at 10 K (M_S) as a function of the thickness of the $Pr_{2/3}Ca_{1/3}MnO_3$ layers in the STO/PCMO superlattices.

The fact that the maximum FM moment of PCMO at $t \sim 3$ nm is observed either in the STO/PCMO or the LSMO/PCMO multilayers indicates that its occurrence is independent of the material that separates the PCMO layers and of the sample deposition method (note that the LSMO/PCMO superlattices were prepared by Pulsed Layer Deposition) [5]. Furthermore, this indicates that the observed maximum is a property inherent to the $Pr_{2/3}Ca_{1/3}MnO_3$ manganite. Previously, it was speculated about two possible mechanisms for the appearance of this maximum [5]. The first proposed mechanism in the LSMO/PCMO multilayers was an interlayer magnetic coupling of the PCMO with the strongly ferromagnetic system LSMO, i.e. a sort of proximity effect. Although this mechanism was mentioned to be unlikely, no direct evidence was provided. In the present study, however, it becomes clear that such a proximity effect is not possible, since the neighboring STO layers are not ferromagnetic. This points to the second proposed mechanism, i.e. the accommodation of the FM clusters within the PCMO would be favored by the geometrical matching between the size of the clusters and the thickness of the PCMO layers. This is based on previous neutron scattering observations that show a typical size of the FM nanoclusters on the order of 3 nm [6].

Another feature that accompanies the enhancement of the FM moment of PCMO for $t \sim 3$ nm is a steep increase of the anisotropy energy. This is not only shown by the large coercive field of the $t = 3$ nm sample (see Fig. 1) but also by the thickness dependence of the exchange bias (EB) field, shown in Fig. 3. The EB phenomenon describes a shift of the $M(H)$ curves towards the negative field axis when the sample is cooled with an applied magnetic field [1], such as the 1 kOe cooling field that we used in our measurements. As an example, the inset of Fig. 3 presents the low-field portion of the $M(H)$ data for $t = 3.7$ nm, where the EB response is easily observed.

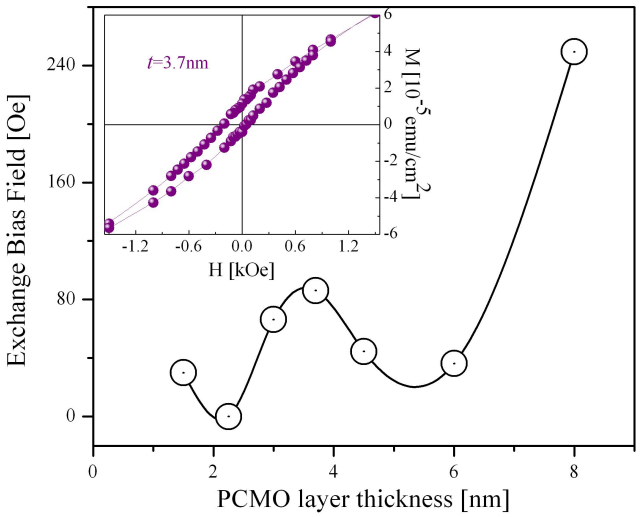

Figure 3. Dependence of the Exchange Bias Field of the STO/PCMO superlattices with the thickness of the PCMO layer. Inset: M vs H at low fields for the $t = 3.7$ nm sample, showing the shift of the curve towards the negative field axis.

EB occurs because of the interface exchange coupling between a FM phase and a non-FM phase. This non-FM phase is usually antiferromagnetic, but EB is also commonly observed with *glassy* phases, where the spins at the interface exhibit a large degree of disorder and multiple ground-state configurations [1, 2]. In our case the FM phase corresponds to the nanoscale clusters inside the PCMO. Although the EB phenomenon would be expected to occur because of the coupling of these clusters with the non-FM matrix of the PCMO, the appearance of a maximum effect at thicknesses ~ 3 nm evidences again the geometrical matching between layer thickness and FM cluster size. It is natural to expect that the spins at the PCMO-STO interfaces posses a large degree of disorder due to their location at the structural interface. Since precisely for $t \sim 3$ nm the surfaces of the clusters most likely coincide with these structural interfaces, the exchange coupling of the FM clusters with the interfacial disordered spins are very likely to be responsible for the enlarged exchange bias field [2].

4. Conclusions

In summary, in our $SrTiO_3/Pr_{2/3}Ca_{1/3}MnO_3$ multilayers we observe an enhancement of the FM magnetic moment for a PCMO layer thickness $t \sim 3$ nm. This is accompanied by an increase of the anisotropy energy, as evidenced by the coercive and exchange bias fields. Since the STO layers are not ferromagnetic, both these phenomena can be interpreted on the basis of the matching condition between the PCMO layer thickness and the size of the FM clusters that occur in PCMO. Our observations imply the possibility of controlling the performance of magnetic multilayer devices simply by imposing appropriate geometrical constraints to the adequate magnetic materials.

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