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On the magnetic properties of F/AF Ca-doped lanthanum manganite bilayers: Approach to interface effects

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Abstract. Magnetic measurements were performed on Ferromagnetic/Antiferromagnetic (F/AF) Ca-doped manganite bilayers in order to elucidate the interface contribution to the total magnetic moment. A 10-nm-thick La$_{2/3}$Ca$_{1/3}$MnO$_3$ (F) film was used as a ferromagnetic layer and a 60-nm-thick La$_{1/3}$Ca$_{2/3}$MnO$_3$ (AF) film as an Antiferromagnetic. The low thickness ratio (1/6) allowed us to enhance the magnetic moment effects due to the AF layer. The temperature dependence of the saturation magnetic moment ($M_s$) was studied. $M_s$ was extracted from isothermal magnetic hysteresis loops from room temperature down to 10 K. By using the Heisenberg model frame for the saturation magnetization change, a $\tau^{5/2}$ term was found to better describe the experimental data, meaning long-wavelength spin waves dominate the temperature behaviour. By applying a field cooling field (FC), a significant exchange bias field ($H_{ex}$) was found (around 20 Oe at 10 K). The $H_{ex}$ temperature dependence follows an exponential decay similar to that observed in the F/AF superlattice case.

1. Introduction
Modulation doping of La$_{1-x}$Ca$_x$MnO$_3$ is a new approach for combining ferromagnetic/antiferromagnetic systems with sharp interfaces and creating new exchange bias systems. In fact, [La$_{2/3}$Ca$_{1/3}$MnO$_3$/La$_{1/3}$Ca$_{2/3}$MnO$_3$]$_N$ superlattices show exchange bias effect, but we found that saturation magnetization normalized to the ferromagnetic layer thickness exceeds the bulk value of the ferromagnetic component [1]. The effects at the interface between the F and AF layers were found among the main reasons for such a behavior, meaning that at the interfaces – in addition to the type of antiferromagnetic material where research has to focus - the F/AF multilayers show a collective contribution due to the high number of interfaces. No reports on magnetic properties of single bilayers have been shown so far for this system. We studied the single F/AF interface effects on magnetic properties by using La$_{2/3}$Ca$_{1/3}$MnO$_3$ as ferromagnetic layer and La$_{1/3}$Ca$_{2/3}$MnO$_3$ as an antiferromagnetic layer. In the LaMnO$_3$ system, different magnetic behaviors can be found as functions of Ca-doping [2]. We chose this system due to the clean interface and small interface roughness reported [3], down to two unit cells. Here, we report on the magnetic properties of La$_{2/3}$Ca$_{1/3}$MnO$_3$/La$_{1/3}$Ca$_{2/3}$MnO$_3$ bilayers. The ferromagnetic layer thickness was set to 10 nm and the antiferromagnetic layer thickness to 60 nm. We measured magnetic isothermal hysteresis loops at temperatures ranging between 10 K and room temperature. From them, the temperature dependence of the saturation magnetization, $M_s$, and of the exchange bias field was obtained. A Heisenberg model is used to explain the temperature dependence of the $M_s$. 

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Figure 1. Isothermal magnetic hysteresis loops at different temperatures for the F/AF bilayer measured after FC at \( H_{\text{FC}} \) of 10 kOe. In the color scale, red corresponds to the highest temperature (250 K), and blue to the lowest temperature (10 K).

Figure 2. Temperature dependence of the exchange bias field, \( H_{\text{ex}} \). An exponential decay fit is represented by the red dashed line. The arrow depicts the expected antiferro-paramagnetic temperature transition of the AF thin film. Error bar is calculated by standard interpolation methods.

2. Experimental

La\(_{2/3}\)Ca\(_{1/3}\)MnO\(_3\) (F) films with a given thickness, \( t_F \), over La\(_{1/3}\)Ca\(_{2/3}\)MnO\(_3\) (AF) films with a given thickness, \( t_{AF} \), were grown in-situ on (001) oriented SrTiO\(_3\) substrates by reactive dc sputtering technique at high oxygen pressures. The ratio between the thicknesses \( t_F/t_{AF} \) was set to 1/6. Substrate temperature at 850 °C, dc power at 40W and, ultra-high-purity oxygen pressure of 3.5 mbar were maintained during deposition, resulting in a deposition rate of 80 nm/h. No post-deposition annealing was necessary. The above parameters yielded single-phase thin film growth [4] with a \( T_C \) for the ferromagnetic layer around 270 K and \( T_N \) for the antiferromagnetic layer close to 150 K [5].

Magnetic measurements were performed with a physical properties measurement system (PPMS) with a vibrating sample magnetometer technique (VSM) by Quantum Design®. All measurements were taken by using a vibration frequency of 40Hz for the detection coil with amplitude of 2 mm. These parameters allow the system a resolution of \( 10^{-6} \) emu. The following procedure was conducted for the zero field cooling (ZFC) isothermal magnetic hysteresis loops: the sample was introduced into the system at room temperature and pumped out down to 1 Torr and was cooled down at a rate of 1 K/min to 10 K. Once the temperature was stabilized, a magnetic field parallel to the sample surface was swept in four steps: starting at zero field to \( H_{\text{max}} \), back to \( H_{\text{min}} \), set to reach \( H_{\text{max}} \) and finally back to zero field. The field sweep rate was kept at 10 Oe/sec. \( H_{\text{max}} \) and \( H_{\text{min}} \) were set to reach a saturation state, ±10 K.Oe. The sweep field rate was slow enough to properly define the coercive fields. Once the measurement was completed, temperature was increased at 1 K/min to reach the next set temperature. A total of 14 temperatures were set during heating up. For FC, a magnetic field was set at room temperature (the sample was in a paramagnetic state) and then cooled down by using the same procedure presented above.

3. Experimental results

By using the procedure discussed, we performed measurements of the isothermal magnetic hysteresis response of F/AF La\(_{2/3}\)Ca\(_{1/3}\)MnO\(_3\) (10 nm) / La\(_{1/3}\)Ca\(_{2/3}\)MnO\(_3\) (60 nm) (F-LCMO/AF-LCMO) bilayers for temperatures ranging between 10 K and 300 K, at ZFC and FC. All the loops were corrected assuming a diamagnetic contribution from the substrate, holder and tape. No correction from the demagnetization factor was taken into account due to sample geometry considerations. Figure 1 shows...
isothermal normalized magnetic moment vs. magnetic field loops for temperatures ranging between 10 K and 250 K after field cooling at H_{FC}=10 kOe. Magnetization is normalized. It should be noted that the hysteresis area from the FC curves is greater than the area for ZFC at the same temperature. Over 250 K, isothermal curves start to show paramagnetic contribution; probably coming from the AF layer (data not presented). A lower signal/noise ratio appears in some loops at low temperature and it is depending on the FC. The field-cooled loops shift along the negative direction of the field axis, whereas the ZFC ones remain symmetric around the field, indicating the existence of the exchange bias effect in our (F-LCMO/AF-LCMO) bilayer. We plot in Figure 2 the temperature dependence of H_{ex}, defining H_{ex}=(H_{c1}+H_{c2})/2, where H_{c1} and H_{c2} denoted the values of the magnetic field for which the descending and ascending parts of the hysteresis loop intercept the abscissa. The exchange bias field has an exponential decay dependence with temperature as it has also been observed in our F/AF superlattices; nevertheless, the magnitude is much less then that obtained for [F(10 u.c.)/AF(20 u.c.)]_{16} superlattices [6]. We think such tendency is due to the low number of interfaces (one for the bilayer) compared with that for the superlattice. The red dashed line corresponds to the fit of the experimental data to the expression H_{ex}=H_{ex0}exp(−T/T_0). This fitting gives us for the parameter T_0 a value of 79 K. We have obtained a value of 30 K for T_0 in case of epitaxial superlattices field cooled at 2 K0e [7]. It is worth noting that this bilayer exhibits a small exchange bias contribution at temperatures over the assumed Néel temperature value for the AF thin film phase.

Temperature dependence of the saturation magnetization, M_s, is shown in Figure 3 for an (F-LCMO/AF-LCMO) bilayer. Vertical axis is in emu units; due to the difficulty of assuming that all moments measured come from the 10 nm F layer alone. We found a level off in saturation magnetization at high temperatures, above the T_C for the F layer, around 250 K. This background could be related to the paramagnetic signal coming from the AF layers, such contribution was proven correct by using the hysteresis loops around these temperatures. This paramagnetic signal starts to develop at 150 K (T_N for the AF) and becomes stronger as the F layer loses its magnetic moment.

To understand the M_s behavior, we fit our results to different models in order to identify the source of the main contribution to the saturation magnetization. In the low-temperature limit, the magnetization of a typical ferromagnet is expected to be explained in a good approximation by the spin-wave theory [8]. According to the Heisenberg model, the change in the saturation magnetization due to the excitation of long-wavelength, non-interacting spin waves can be written as M_s = M_{os}(1-\beta^{3/2}), where M_{os} is the magnetization (or moment) at zero temperature; B gives the magnitude of the thermal demagnetization (how fast the system loses its moment per Kelvin) and \tau, represents the reduced temperature T/T_C. In many systems, the low-temperature limit, which corresponds to 0.3T_C, is characterized by a \beta-value of 3/2 (\tau^{3/2} term), while the high-temperature region is better described by a \beta value of 5/2. We fitted our experimental data (symbols) taking into account the temperature limits described above, as noted in Figure 3 (a). We assumed a purely \tau^{5/2} term (green dotted line), a \tau^{3/2} term (red dashed line). We also let the power exponent \beta as a free parameter. Fitting gives us a value of \beta=3.0 ± 0.1. To understand the fits, we separated the contribution to the magnetization of the layers individually. Below the T_N (150 K) of the AF layer, magnetic interactions contribute additionally with the F layer to the total saturation magnetization. We expected a difference from the bulk material behavior in this region, due to the presence of the AF interactions. Above T_N, the paramagnetic behaviour of the AF layer adds a background signal to the magnetization, reducing the demagnetization rate. In order to be factual, we fit our data below T_N. As can be seen from Figure 3, the best fit is obtained at a fixed \beta value of 5/2 (green dotted line). The arrow indicates the T_N limit. We also tried to fit the data assuming two terms, \tau^{5/2} + \tau^{3/2}, but it did not fit better. We found no significant difference between the ZFC and 10 kOe FC data.

In order to picture the results presented above, we grew a 100 nm thick ferromagnetic La_{2/3}Ca_{1/3}MnO_3 layer. M_s versus-temperature curve is displayed in Figure 3(b). The change on the magnetic behavior is clearly shown by the zero value at 300 K. No paramagnetic background is present in the measurement. The magnetic moment decays slower than in the bilayer system. To compare with the fit performed on the bilayer, we fit the data by first using the low-temperature
regime (below 150 K), but no significant difference in the parameters was found. We found no good match using values \( \frac{3}{2} \) (red dashed line) and \( \frac{5}{2} \) (green dash-dot line) for \( \beta \). The best fit gives a \( \beta \) value close to \( \frac{9}{2} \), around 4.3. This significant difference between the bilayer and the F layer is related to the presence of high interactions at the interface and can not be explained by using thickness-dependence arguments for the F layer. The values found from the fit in the bilayer system agree with those found for the epitaxial superlattice system [9], which means no additional mechanism is present when the number of interfaces is increased.

In summary, we grew in-situ \( \text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3(10 \text{ nm})/\text{La}_{1/3}\text{Ca}_{2/3}\text{MnO}_3(60 \text{ nm}) \) epitaxial bilayers via high-pure-oxygen pressure sputtering technique. We found that the F(10 nm)/AF(60 nm) bilayer exhibits a weak exchange bias effect with a magnitude around 20 Oe. The temperature dependence of the saturation magnetization fitted to a power law dependence yielded values for the \( \beta \) exponent of \( \beta > \frac{5}{2} \). Thus, F-LCMO(t\text{F})/AF-LCMO(t\text{AF}) bilayer with a thicknesses ratio \( t\text{F}/t\text{AF}=1/6 \), shows a \( (T/T_0)^{5/2} \) characteristic behavior in the temperature dependence of the saturation magnetization, indicating probably an influence of AF layer on the thermal demagnetization mechanism.

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