# Influence of ferromagnetic thickness on structural and magnetic properties of exchange-biased manganite superlattices

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The structural and magnetic properties of superlattice structures of alternating ferromagnetic $La_{2/3}Ca_{1/3}MnO_3$ (F-LCMO) and antiferromagnetic $La_{1/3}Ca_{2/3}MnO_3$ (AF-LCMO) layers were systematically studied as functions of F-LCMO layer thickness, $t_F$. Samples were grown via a high-oxygen pressure sputtering process. Magnetic hysteresis measurements after field cooling revealed an exchange bias, $H_{ex}$, at low temperatures in such superlattices. We found a correlation of the structural and magnetic properties with $t_F$. In particular, we observed diminished resistance, increased metal-insulator transition temperature, $T_{MI}$, as well as increased Curie temperature with increasing $t_F$. Additionally, we found that the temperature dependence of $H^*_{ex}t_F$ for superlattices with the same antiferromagnetic layer thickness, $t_{AF}$, is a unique function and independent of $t_F$. We also find that the low-temperature saturation magnetization, $M_S$, follows a power-law dependence with temperature, according to $M_0(1-BT^{\alpha})$ with an exponent of $\alpha=2.5\pm0.2$. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2167047]

## I. INTRODUCTION

Superlattices of ferromagnetic (F) and antiferromagnetic (AF) oxide materials have attracted an increased attention given the *exchange-bias* or *exchange anisotropy* phenomenon. Experimentally, this effect manifests itself in a field shift, $H_{ex}$, of the hysteresis loop along the field axis, typically accompanied by an increase of the coercivity. This is due to an interaction between the AF and F materials when cooled in applied magnetic fields (field cooling, FC) below the Néel temperature ($T_N$) of the AF layer.[1] All these issues have taken on technological importance as well, given the use of exchange-bias systems in today's giant magnetoresistance (GMR) sensors for hard disk drive applications.

Research on magnetic oxide superlattices has focused towards the technological goals of increasing magnetoresistance in lower applied magnetic fields and more useful temperatures ranges for various applications. Recent work has been dominated by superlattices of perovskite manganite materials of the form $R_{1-x}A_xMnO_3$.[2] These compounds bear much interest due to the rediscovery of colossal magnetoresistance (CMR),[3] particularly large in thin films. Also, these types of perovskite manganite superlattices support a wide variety of rare-earth and alkaline-earth metals, and numerous systems have been prepared, where F-layer thickness has been varied.[2]

Of particular interest are $La_{2/3}Ca_{1/3}MnO_3$/$La_{1/3}Ca_{2/3}MnO_3$ (F-LCMO/AF-LCMO) superlattices, due to the very high structural compatibility of the AF and F layers in this system, which permits epitaxial coherent growth. In fact, several properties associated with CMR and exchange-bias effects have been recently studied in F-LCMO/AF-LCMO multilayers.[4] Previously, we have reported that saturation magnetization in these superlattices is larger than that in $La_{2/3}Ca_{1/3}MnO_3$ films and even exceeds the bulk value, suggesting that F order extends into the AF regions where the chemical composition would normally produce an AF state.[5]

In the present paper, we report a study of the structural and magnetic properties for a series of [AF-LCMO (7.6 nm)/F-LCMO ($t_F$)]$_N$ multilayers as a function of the ferromagnetically doped layer thickness $t_F$, which was varied from 1.9 to 7.6 nm, while the antiferromagnetic (AF) layer thickness, $t_{AF}$, was kept constant at 20 u.c., (unit cells), corresponding to 7.6 nm. From our study, we derived valuable information on the correlation between the structural and magnetic properties in these layered manganite structures. Furthermore, we present experimental information about of the exchange bias and thermal demagnetization in the low-temperature range. These results have important conse-

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quences in understanding the competitive interaction phenomena in this type of manganite heterostructures.

## II. EXPERIMENT

Superlattices of F-LCMO and AF-LCMO layers were grown on (001)-oriented $SrTiO_3$ substrates via a high-pressure dc sputtering process, using sintered targets with stoichiometric F-LCMO and AF-LCMO compounds. All $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (t_F)]_N$ multilayers with $1.9\ \mathrm{nm} \leqslant t_F \leqslant 7.6\ \mathrm{nm}$ were prepared under identical deposition conditions, known to give rise to high-quality epitaxial manganite films and superlattices.[6,7] The deposition took place in a pure $O_2$ atmosphere at 3.5 mbars, keeping the substrate temperature constant at 850 °C. The deposition rate for both types of oxides was approximately 1.5 nm/min, and the total thickness was chosen to be around 180 nm for all samples.

Magnetization measurements were carried out in a Quantum Design™ superconducting quantum interference device (SQUID) magnetometer. The coercive and exchange-bias fields were derived from isothermal $M(H)$ loops at low temperatures after initial cool down from room temperature to 5 K in an applied magnetic field of 20 kOe.

## III. RESULTS AND DISCUSSION

Reliable values of the modulation period $\Lambda$ have been experimentally derived from x-ray-diffraction measurements, confirming that for all superlattices actual layer thicknesses were within 10% of the nominal values. The position of the zero-order peak observed in the x-ray-diffraction (XRD) data was used for an estimate of the average multilayer lattice constants $a_{\mathrm{ave}}$ of the superlattice unit cell.[4] We obtained values for $a_{\mathrm{ave}}$ for $t_F$=5, 10, 15, and 20 u.c, ranging from $0.377 \pm 0.001$ nm, for the thinner F layer, to $0.380 \pm 0.001$ nm, for $t_F = t_{\mathrm{AF}}$. The variation of the lattice constant suggests a strain-driven mechanism for the multilayer lattice formation. We can assume a homogeneously strained structure, since the modulation period is <25 nm, the thickness where strain starts to relax by the formation of misfit dislocations.

Magnetotransport measurements were done on these samples to study the variation of the resistance as a function of temperature for a series of $[\mathrm{AF}(7.6\ \mathrm{nm})/\mathrm{F}(t_F)]_N$ multilayers. Resistance data were taken cooling down from room temperature to approximately 50 K either at zero field cooling (ZFC) or during FC at 10 kOe. Figure 1 shows the $T_{\mathrm{MI}}$ transition, given by the maximum of the resistance peak, as a function of F layer thickness $t_F$, extracted from both the ZFC and FC resistance curves. Resistances are normalized to the $R(300\ \mathrm{K})$ value, measured in 10 kOe ($R_H$) and in zero applied field ($R_0$). The $\Delta R/R_H = [R_0 - R_H]/R_H$ ratio (solid line) shows the magnetoresistance. Remarkably, Fig. 1 shows that there is a pronounced F thickness dependence of $T_{\mathrm{MI}}$. The inset shows the resistance (both $R_H$ and $R_0$) curves for the $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (1.9\ \mathrm{nm})]_{18}$ superlattice, i.e., the thinnest F layer. In this case, the resistances increase up to two orders of magnitude, with decreasing temperature, and there are a maxima at around 78 and 90 K for ZFC and FC, respectively. However, for the $[\mathrm{AF}(7.6\ \mathrm{nm})/\mathrm{F}(t_F)]_N$ superlattices,

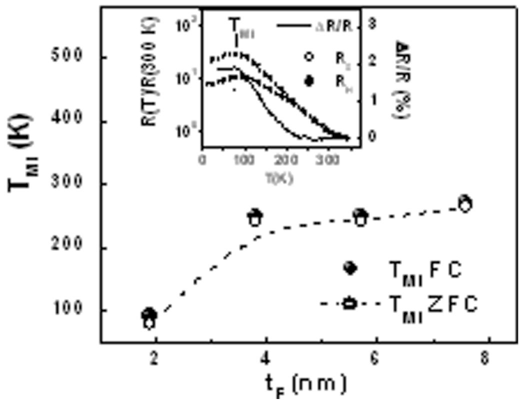


FIG. 1. Metal-insulator transition $T_{\mathrm{MI}}$ for $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (t_F)]_N$ superlattices, as a function of $t_F$, extracted from $R(T)$ data at 10 kOe (solid circle), and at zero field (open circle). The line is a visual guide. The inset shows normalized resistance (symbols, left axis) and $\Delta R/R_H$ (line, right axis) vs $T$ for a $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (1.9\ \mathrm{nm})]_{18}$ superlattice, FC (solid circle), and ZFC (open circle).

where $t_F$ increases from 3.8 to 7.6 nm, both the ZFC and FC $R(T)$ curves resemble the typical curves observed for the individual F films. Interestingly, $T_{\mathrm{MI}}$ increases up to ~270 K for thicker ferromagnetic layer, $t_F$=7.6 nm. This value corresponds to approximately $T_{\mathrm{MI}} \cong 270$ K, expected for a single F film, and also coincides with the Curie temperature $T_C \sim 265$ K, typical for such a film.[6] Apparently, for thicker F layers, the AF layers did not influence transport properties. However, the stability of the ferromagnetic order decreases for smaller $t_F$ as the AF phase becomes dominant.[4] We also extracted $T_C$ from magnetization measurements in both ZFC and FC for the superlattices, obtaining a $T_C \cong 260$ K for thicker $t_{\mathrm{AF}}$, whereas a diminished $T_C$ was observed for the lowest F layer thickness.

For our samples, exchange-bias fields were derived from isothermal hysteresis loops after field cooling in a 2 kOe field. Typical loops are shown in the inset of Fig. 2 for an $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (3.8\ \mathrm{nm})]_{16}$ sample measured at 20 and 160 K. There is a clear exchange bias at low temperature, reflected in the asymmetric shift towards negative fields of $H_{\mathrm{ex}}$=230 Oe at 20 K. This shift vanishes for all measured superlattices at around 150 K and the hysteresis loop becomes symmetric as evident in the 160 K curve in the inset of Fig. 2.

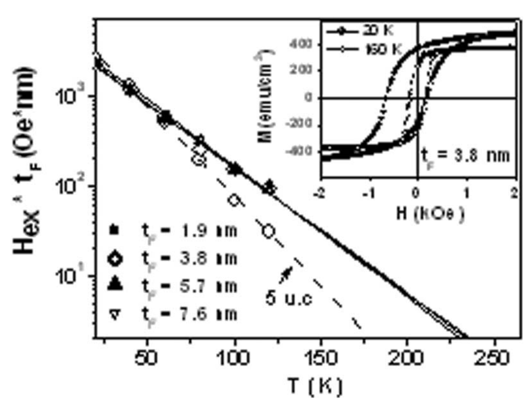


FIG. 2. Log $(H_{\mathrm{ex}}^{*} t_F)$ vs $T$ for a series of $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (t_F)]_N$ superlattices. The inset shows magnetic hysteresis loops at 20 (solid symbols) and 160 K (open symbols) for a $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (3.8\ \mathrm{nm})]_{16}$ superlattice measured after field cooling in 2 kOe.

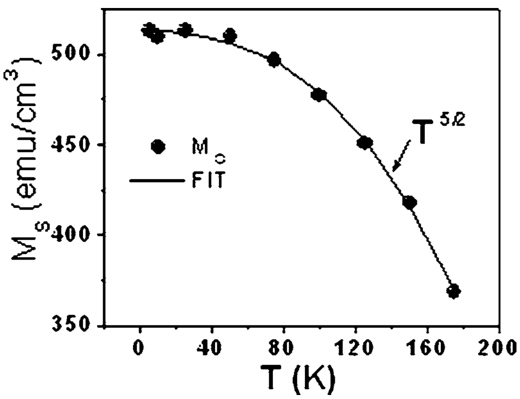


FIG. 3. Saturation magnetization as a function of temperature for a $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (3.8\ \mathrm{nm})]_{16}$ superlattice (solid symbols), and a power-law fit (line), of the form $M_S = M_0(1 - BT^{\alpha})$ with $\alpha = 5/2$.

$H_{ex}$ as a function of temperature shows an exponential decrease of exchange bias $H_{ex}$ with increasing temperature, which was fitted with

$$H_{ex} = H_{ex}^{0}\exp(-T/T_0) + C, \quad (1)$$

where $H_{ex}^{0}$ is the $H_{ex}$ value at $T=0$, $C$ is a constant, and $T_0$ is a parameter related to the temperature decay. This decay has been interpreted for a variety of $t_F$ and $t_{AF}$ thicknesses, in terms of thermal fluctuations stemming from several potential sources including a spin-glass-like disorder or a distribution of superparamagneticlike domains.[8]

Even though, for all superlattices, $H_{ex}$ becomes zero at approximately 150 K, we observed a dependence of the shift value with the F thickness $t_F$, to low temperature. Basically, the exchange-bias effect increases for the thinnest F layer, becoming almost four times larger at 5 K, than that multilayer with thicker F layer, measured at the same temperature. An interesting result about the dependence of $H_{ex}$ with F layer thickness is shown in Fig. 2, where we plot the product $H_{ex}t_F$ as a function of temperature. Clearly, all data fit very well on a unique line, revealing an intrinsic mechanism of the exchange bias independent of $t_F$. But for very small $t_F$ (1.9 nm) and high temperatures, there is a significant deviation from the exponential decay law.

Temperature dependence of the saturation magnetization $M_S$ is shown in Fig. 3 for a $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (3.8\ \mathrm{nm})]_{16}$ superlattice. $M_S$ for all samples is obtained by applying a magnetic field high enough to reach the saturation (up to 10 kOe). In the low-temperature limit, the magnetization of a typical crystalline ferromagnet is expected to be explained in a good approximation by spin-wave theory.[9] According to the Heisenberg model, the change in the spontaneous magnetization due to the excitation of long-wavelength, noninteracting spin waves can be written as $M(T) = M(0)[1 - BT^{\alpha}]$, where $M(0)$ is magnetization at 0 K, and the coefficient $B$ gives the magnitude of the thermal demagnetization. Usually, the low-temperature range ($T < 0.3T_C$) is dominated by a $T^{3/2}$ term, whereas the observation of $T^{5/2}$ behavior requires relatively high temperatures.

We fitted the saturation magnetization data, obtained at low temperatures (from 5 to 160 K), by using the least-squares fit. The best fit obtained corresponded to $\alpha = 2.5 \pm 0.1$ and $B \cong 5 \times 10^{-7}\ \mathrm{K}^{-2.5}$, with correlation coefficients ($R^2$) of $\sim 0.999$. Figure 3 shows the good fit for a $[\mathrm{AF}\ (7.6\ \mathrm{nm})/\mathrm{F}\ (3.8\ \mathrm{nm})]_{16}$ superlattice. The quality of the fits for all other samples is comparable to the one shown in Fig. 3. However, $\alpha$ obtained for the superlattice with thinner F layer was slightly larger, $\alpha = 2.8 \pm 0.1$.

It is worth noting that we fitted our experimental data considering the $T^{3/2}$ term in the power-law function. Nevertheless, the negative (unphysical) values obtained for the coefficients and the correlation ($R^2$) of $\sim 0.950$ led us to reject for the current discussion the results corresponding to that fit.

The $T^{5/2}$ term is associated with long-wavelength spin waves, and corresponds to the anharmonic second-order expansion of the magnetization.[9] We obtained the best fit, without the $T^{3/2}$ term, expected to dominate the demagnetization behavior at $T < 0.3T_C$. Probably, the AF layer introduces an anharmonicity in the magnon dispersion relation, and thus the contribution of the $T^{5/2}$ term is similar in all superlattices. The variation of parameters for the thinner F layer, $t_F = 1.9$ nm, can be related to the effect of interface roughness.[9]

In summary, we have grown a series of $[\mathrm{La}_{1/3}\mathrm{Ca}_{2/3}\mathrm{MnO}_3]/\mathrm{La}_{2/3}\mathrm{Ca}_{1/3}\mathrm{MnO}_3]_N$ epitaxial superlattices via a high-oxygen pressure sputtering technique; we have studied the variation of structural and magnetotransport properties as a function of ferromagnetic layer thickness. We observed that the $c$-axis lattice parameter, the Curie temperature, and metal-insulator transition are affected when the ferromagnetically doped layer thickness is decreased. However, we observed that exchange bias fits very well to an exponential decay for temperatures below approximately 150 K in all superlattices, indicating an intrinsic mechanism independent of $t_F$, except for a very thin F layer. We also present experimental information on thermal demagnetization in the low-temperature range obtained from the temperature dependence of the saturation magnetization. This demagnetization due to spin waves shows the same power-law dependence, independent of the ferromagnetic layer thickness.

## ACKNOWLEDGMENTS

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