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EXCHANGE COUPLING IN EPITAXIAL FERROMAGNETIC-ANTIFERROMAGNETIC $La_{1-X}Ca_XMnO_3$ HETEROSTRUCTURES

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ABSTRACT

We report exchange coupling in bilayers and trilayers with alternating layer of ferromagnetic (F)-$La_{2/3}Ca_{1/3}MnO_3$ and antiferromagnetic (AF)-$La_{1/3}Ca_{2/3}MnO_3$ compositions, grown on (001) oriented $SrTiO_3$ single crystal by a high-oxygen pressure sputtering process. We observe from isothermal magnetic measurements between 5 K and ~150 K a shift of the hysteretic loops towards negative fields, evidencing an exchange bias mechanism (H_{ex}) in these heterostructures. We applied two different values of the field cooling (FC), 500 Oe and 1500 Oe. H_{ex} exhibits an exponential decrease with temperature below ~100 K. Fitting the experimental data to a exponential function, we found that the fitted parameter H_{ex}^0 is within the error bar the same, whereas the fitted T_o parameter is different indicating that the FC value could affect the magnetic state when the exchange biasing is established in the AF-F interface.

Key words: Exchange coupling mechanism, manganite thin films, ferro-antiferro interfaces.

INTRODUCTION

Ferro-antiferromagnetic (F-AF) exchange coupling effect has been observed in different kind of systems during the last four decades. *Exchange Bias Effect* is one of the phenomena associated with the unidirectional anisotropy created at the interface between a ferromagnetic (F) and antiferromagnetic (AF) material when they are in intimate contact with each other and are cooled in an applied magnetic field through the Néel temperature (T_N) of the AF-material. It is manifested as a displacement of the loop of the F layer away from $H=0$ by an amount known as the *exchange bias field* H_{ex}, and, an increase of coercively field H_C [1,2]. Recently, it attracted much attention and despite the technological interest in these structures there is little basic understanding of the phenomenon [3]. In the case of Ferro-Antiferro interface in thin films, during field cooling, FC, the magnetic moment configuration gives rise to an exchange field (H_{ex}) parallel in the direction of the magnetic moment (M_s) of the ferromagnetic layer that establishes a preferred *direction* of magnetization at the interface. If the thickness of the ferromagnetic (F) layer (t_F) is less than the thickness of a domain wall (δ_{dw}), the response of the F-layer to a field causes an exchange twist of spins in the AF layer near the interface. In this approximation [4] is valid the following relation: $H_{ex} = -2\frac{K_{AF}d_{AF}}{M_s t_F}$

where d_{AF} is the thickness of a domain wall in the antiferromagnetic (AF) layer, K_{AF} is the magnetic anisotropy coefficient.

Oxide materials have attracted increasing attention in recent years. In particular, significant work has been done in efforts to understand and expand upon the observations of colossal magnetoresistance (CMR) in perovskite manganite films of the type $La_{1-x}Ca_xMnO_3$ [5]. To date, a little work has been done on the exchange coupling effect in manganite system of the type $La_{1-x}Ca_xMnO_3$, which shows different ground state as AF or F depending of the *x* values [6]. $La_{2/3}Ca_{1/3}MnO_3$ is ferromagnetic, F-LCMO; while the ground state of $La_{1/3}Ca_{2/3}MnO_3$ is antiferromagnetic, AF-LCMO. Bilayers of F-LCMO and AF-LCMO provide a good candidate to study F-AF coupling, and it may shed light on the nature of exchange coupling due to its special spin structure [7]. In this work, we grew AF-LCMO/-F-LCMO bilayers and [AF-LCMO/F-LCMO/AF-LCMO] trilayer in order to study the exchange coupling effect in this system.

EXPERIMENTAL DETAILS

The films were grown *"in situ"* on (001)-$SrTiO_3$ substrates by a high-oxygen pressure dc-sputtering process. During deposition the substrate temperature was stabilized at 850 °C and the oxygen pressure in the chamber 3.5 mbar, with a potential difference of 300 V, current of 100 mA applied between the electrodes as reported elsewhere [8] . The film thickness was chosen to be around 50 nm. Structural analysis was performed by means of X-ray diffraction (XRD) measurements (θ - 2θ) by using a Rigaku diffractometer. Magnetic measurements were done in a quantum Design^{TM} superconducting quantum interference device (SQUID) magnetometer. The coercive and exchange bias fields were derived from isothermal loops at low temperatures after field cooling in 500 Oe and 1500 Oe for bilayers, and 5 kOe for the trilayers.

RESULTS AND DISCUSSION

A typical x-ray diffraction spectrum is shown in Fig. 1 for the [AF(23nm)/F(3nm)/AF(23nm)] trilayer. It displays (00*l*) peaks only, indicating a textured heterostructure with the *c*-axis perpendicular to the surface of the substrate.

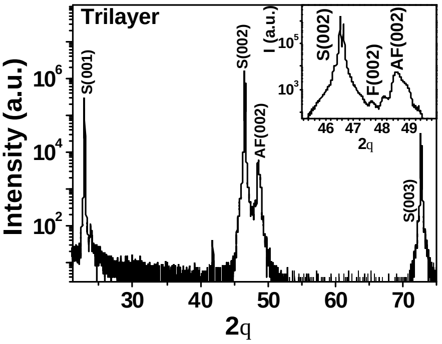

Fig. 1. X-ray diffraction for a [AF(23nm)/F(3nm)/AF(23nm)] trilayer grown on $SrTiO_3$ substrate. The inset shows the(002) substrate, F and AF peaks

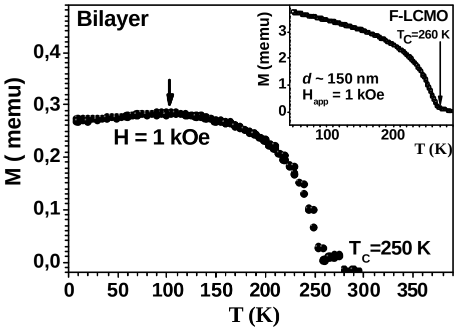

Fig. 2. ZFC temperature dependence of the magnetization for a [AF(36 nm)/ F(15 nm)] bilayer, in a field of 1 kOe. The inset shows the ferromagnetic behavior of 150nm thick a F-LCMO film.

The lattice parameters for the substrate ($SrTiO_3$), F-LCMO ($La_{2/3}Ca_{1/3}MnO_3$) and AF-LCMO ($La_{1/3}Ca_{2/3}MnO_3$) extracted from the observed (00*l*) Bragg-peak positions, were 0,389 nm, 0,385 nm, and 0,372 nm respectively. In Fig. 2 we show the magnetization measurement as a function of temperature for a [AF(36 nm)/F(15 nm)] bilayer. The curve was taken by warming up in 1 kOe in zero-field-cooled, ZFC. It shows a Curie point around 250 K, close to the expected value of 260 K, extracted from the magnetization curve for a 150 nm thick $La_{2/3}Ca_{1/3}MnO_3$ film, shown in the inset [8]. The magnetization shows apparently no dependence with temperature ranged between 10 K and 120 K. However, detailed analysis of it shows us that, there is broad hump with a maximum value around 100 ± 5 K. It is important to compare with the temperature dependence of the magnetization for a F-LCMO film, displayed in the inset, that shows a typical ferromagnetic behavior in this range of temperature. This hump in the bilayer could be due to the presence of the AF-layer and the AF/F interface [9].

Field cooling (FC) magnetization loops were carried out in range between 75 K and 10 K for the [AF(23nm)/F(3nm)/AF(23nm)]-trilayer, and between 230 K and 5 K for the [AF(36 nm)/F(15 nm)]-bilayer. The Fig. 3 shows two isothermal FC (at 500 Oe) magnetization loops for the bilayer, at 150 K and 5 K. The 150 K loop is symmetric around zero, whereas the loops for lower temperature are shifted towards negative fields, evidencing an exchange bias mechanism due to the interface AF/F of this bilayer. The magnetization curve for the $SrTiO_3$ substrate indicates diamagnetic behavior at 294 K. That means $SrTiO_3$ is no sensible even at high applied magnetic fields.

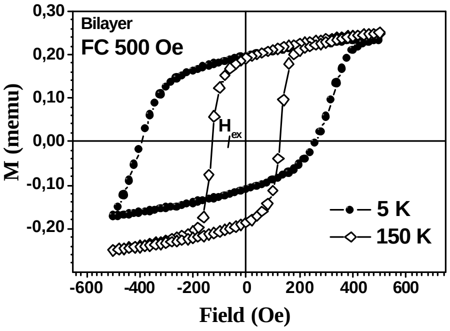

Fig. 3. Magnetic hysteresis loops measured at 5 K and 150 K after FC in 500 Oe, for [AF 36 nm/F 15 nm] bilayer. The inset shows M(H) measurement on the $SrTiO_3$ substrate taken at room temperature.

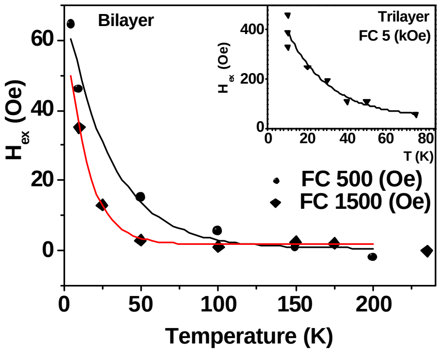

Fig. 4 H_{ex} vs. T for a [AF 36 nm/F 15 nm] bilayer; and for [AF 23nm/ F 3nm/ AF 23nm] trilayer, inset. We show two different FC applied on the bilayer. The parameters found from the fit are summarized in table1.

We observed in the Fig. 3 that at 5 K, the loop is shifted $H_{ex} \approx 66$ Oe, in contrast with the loop at 150 K where H_{ex} is almost zero. The temperature where H_{ex}=0 can indicate the approximated value of Néel temperature of the antiferromagnetic layer, in this case, around 150 K. H_{ex} is defined as the loop shifted and the H_c as the halfwidth of the loop. Thus, if H_1 and H_2 are the fields for which the descending and ascending parts of a hysteresis loop intercept the abscissa, then $H_{ex} = -(H_1+H_2)/2$.

Fig. 4 plots the temperature dependence of the H_{ex} for the bilayer film for two different FC values, 500 and 1500 Oe. The data show basically an exponential decrease of the exchange bias H_{ex} with temperature. The existence of frustration due to competing interactions is known to lead to an exponential decay of H_{ex} with temperatures, as we can fit the data to an exponential decay with temperatures; previously observed in amorphous/ crystalline $NiFe_2O_4$ ferrite, according to

$$H_{ex} = H_{ex}^{0} * \exp\left({}^{T}\!/_{T_0} \right) + C$$

Where C is a small constant field, which, probably comes from the trapped flux in the super-conducting coils. The fitted data H_{ex}^{0}, the magnitude of H_{ex} at T=0; and T_0, the inverse of the factor in the exponential argument, are summarized in the table 1. We observe the following: for the bilayer the term H_{ex}^{0} is the same for the two FC values (500 and 1500 Oe), but T_0 has a dependence with the field cooling field. Similar procedure was done to the trilayer film. The temperature dependence of the H_{ex}^{0} is shown in the inset of Fig 4, and, the corresponding fitted data in table 1. The FC in this case was 5 kOe, and this magnitude could explain the high values of the fitted H_{ex}^{0} for the trilayer.

Heterostructure	$\mathbf{H_{ex}^{0}}$*(Oe)*	***T_0 (K)***
Bilayer - FC 500 Oe	*71*	*30*
Bilayer - FC 1500 Oe	*69*	*14*
Trilayer - FC 5 kOe	*562*	*21*

Table 1. Parameters found from the temperature dependence of H_{ex} fit (Eq. (2)).

CONCLUSIONS

In summary, we have grown "*in situ*" epitaxial [AF/F] bilayer and trilayer heterostructures of La–Ca–Mn-O system by a high oxygen pressure sputtering technique. The heterostructures showed exchange bias effect due to the existence of the interface AF/F at temperatures below ~100 K. This is an indicative of approximate value of the antiferromagnetic transition temperature also. We have observed an exponential behavior for the temperature dependence of the H_{ex} and found from exchange bias fit, that the magnitude of the applied field cooling affects the magnetic state of the exchange coupling in the AF/F established interface.

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