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Exchange-coupling effect and magnetotransport properties in epitaxial $La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3$ superlattices

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We have measured structural, magnetic and, magneto transport properties of heterostructures consisting of $La_{1-x}Ca_xMnO_3$ ferromagnetic (FM) layers ($x = 0.33$) and antiferromagnetic (AF) layers ($x = 0.67$). FM/AF superlattices were grown by a high-pressure sputtering technique on (001) oriented $SrTiO_3$ substrates. We have systematically varied the thickness of the ferromagnetic layers, while maintaining the thickness of the antiferromagnetic layers fixed. The total superlattice thickness was held approximately constant. The XRD analysis confirmed the existence of the superlattice structure by the multiple satellite peaks around the 00l manganite Bragg reflections. We have done field cooling (FC) and zero field cooling (ZFC) magnetization, and magneto resistance measurements at temperatures between 10 K and 280 K. The existence of an exchange bias effect at temperatures below the Neel temperature of the AF layer was revealed by magnetization loops after field cooling. The exchange bias field magnitude H_{ex} exhibited exponential temperature dependence below the blocking temperature, as well as an inverse proportionality with the ferromagnetic layer thickness.

1 Introduction

Magnetic films have been the subject of intensive research in the past, due to their technological utilization in magnetic recording media and sensors. Magnetic oxide materials have attracted increased 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 La(Ca)–MnO_3 (LCMO) manganite films [1] and the exchange bias effect (H_{ex}) created at the interface between ferromagnetic (FM) and antiferromagnetic (AF) materials when they are cooled below the Neel temperature (T_N) of the AF layer. Experimentally, this phenomenon is manifested as a displacement of the hysteretic loop of the FM layer along its field axis and typically occurs simultaneously with an increase of coercive field [2]. This loop shift is important because the exchange coupling can be used to "pin" an FM soft layer in low fields, a technique applied in spin-valves and magnetic random access memories. However, despite technological interest in these structures there exists only an insufficient basic understanding of this phenomenon [2]. During the field cooling (FC) procedure, the magnetic moment configuration gives rise to an exchange field (H_{ex}) parallel to the direction of the FM moment (M_s) that establishes a preferred *direction* of magnetization at the interface. The existence of exchange biasing in multilayers with alternating

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layers of FM $La_{2/3}Ca_{1/3}MnO_3$ and AF $La_{1/3}Ca_{2/3}MnO_3$ compositions has been previously reported [3]. These studies reveal that exchange bias appears below a blocking temperature (T_B), which is less than the magnetic ordering temperatures of the AF (T_N) and the T_C of the FM layers. Above the T_B the AF order in the grains is not stable enough to support a net exchange bias effect, which is also referred to as unidirectional anisotropy [4]. Below the T_B the AF order becomes stable or frozen and the unidirectional anisotropy at low T depends on the domain wall energy in or near the interface. Furthermore, it was observed that exchange-coupled layers with $T_B < T_N$ exhibit asymmetric hysteretic loops due to irreversible transitions of the AF order in the AF grains. In this paper, we have studied the exchange bias effect in epitaxial $[La_{1/3}Ca_{2/3}MnO_3(20\ \text{u.c.})/La_{2/3}Ca_{1/3}MnO_3(\text{N u.c.})]_{14}$ superlattices as a function temperature and, the number (N) of unit cells (u.c.) of the FM layers.

2 Experimental details

The FM/AF heterostructures were grown on (001)-$SrTiO_3$ single crystal substrates by a high-pressure sputtering process. Self sintered targets with stoichiometric ($La_{1/3}Ca_{2/3}MnO_3$ (AF), $La_{2/3}Ca_{1/3}MnO_3$ (F)) compounds were used. We had previously reported on the growth of high-quality epitaxial manganite thin films on a variety of substrates [5, 6]. The deposition process took place in a pure O_2 atmosphere at 3.5 mbar and, with a substrate temperature of 850 °C; the deposition rate for both types of oxides was kept constant at approximately 1.5 nm/min. Total thickness was chosen to be around 180 nm for all superlattices. Bilayer thickness modulation, Λ, varied according to FM-layer thickness.

Structural analysis was performed by means of ($\theta - 2\theta$) X-ray diffraction (XRD) measurements by using a Rigaku diffractometer. Magnetotransport measurements have been carried out with the standard four-point probe method, applying the magnetic field parallel to the current flow direction in the film plane. Magnetoresistance isothermal loops at low temperatures were measured after initial cool down in different magnetic fields (from ZFC to 1 T) from 300 to 15 K. Magnetization measurements were done in a Quantum Design™ superconducting quantum interference device (SQUID) magnetometer. The coercive and exchange bias fields were derived from isothermal loops at low temperatures after initial cool-down in a 20 kOe magnetic field applied in the temperature range from 300 to 20 K.

3 Results and discussion

The high angle X-ray diffraction (HAXRD) spectrum of a $[AF_{15\,\text{u.c.}}/FM_{3\,\text{u.c.}}]_{21}$ superlattice is shown in Fig. 1. The substrate and LCMO (001)-, (002)-, and (003)-Bragg peaks can be observed. In addition, clear satellites are visible, which are characteristic of the superlattice modulation. From the superlattice peaks in Fig. 1 we obtained, for this sample, a modulation of 7 nm in good agreement with the thickness of the individual layers determined from the deposition rate. The inset of this figure shows the structure simulation using the SUPREX 9.0 refinement software [7]. Refinements yielded layer thickness fluctuations of about 1.0 u.c. for the manganite layers. We also found no indications of epitaxial mismatch strain as expected from the small lattice mismatch between the FM and the AF layers. X-ray refinement did not show changes in the intra-cell distances along the c direction.

Figure 2 shows isothermal FC magnetoresistance loop for the $[AF_{20\,\text{u.c.}}/FM_{15\,\text{u.c.}}]_{14}$ superlattice taken at 15 K. The loop exhibits asymmetries between the two (descending and ascending) branches: resistance-maximum appears in the positive field range of the ascending (from −10 kOe to 10 kOe) field branch; the position of the resistance maxima are not symmetric around the y-axis at H = 0. These asymmetries probably occur because of interfacial effect between AF and FM layers. This result agrees with other reports on manganite multilayers [3, 8]. The influence of the roughness in the interface resistance, contributes to the total resistance.

The isothermal FC magnetization loops for the $[AF_{20\,\text{u.c.}}/FM_{10\,\text{u.c.}}]_{16}$ superlattice taken at 20 K and 160 K are shown in Fig. 3. For clarity, we only show the loops for these two different temperatures. The 160 K loop is symmetric around zero, whereas the loops for lower temperatures are shifted towards negative fields, evidencing an exchange bias mechanism in these superlattices. This effect disappears at

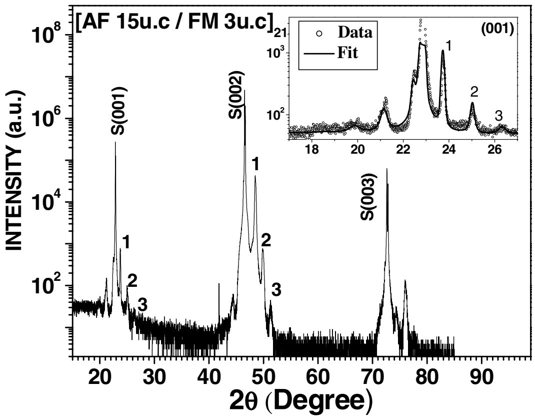

Fig. 1 HAXRD pattern of a $[AF_{15\,u.c.}/FM_{3\,u.c.}]_{21}$ superlattice $SrTiO_3$. Substrate Bragg peaks are labeled as (S). The order of the satellite peaks from the AF/FM superstructure is displayed. The inset shows the SUPREX 9.0 structure refinement on (001)-peak.

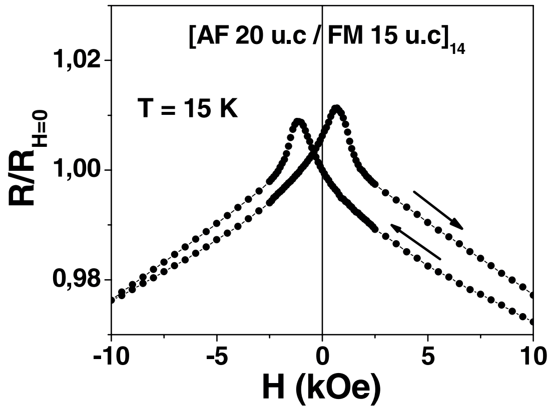

Fig. 2 Normalized magnetoresistance loop measured on the $[AF_{20\,u.c.}/FM_{15\,u.c.}]_{14}$ superlattice at 15 K after cooling from 300 K in a magnetic field of 1 T. Arrows indicate the direction of field change during the loop.

temperatures near 150 K. This finding is consistent with a Neel temperature T_N for the AF layer of approximately 150 K. H_{ex} is defined as the loop shift and the H_c as the half width of the loop. Thus, if H_1 and H_2 are the fields for which the descending and ascending parts of a hysteretic loop intercept the abscissa, then: $H_{ex} = -(H_1 + H_2)/2$. At 20 K, $H_{ex} = 0.229$ kOe.

Figure 4 presents the data for H_{ex} as a function of temperature for the $[AF_{20\,u.c.}/FM_{10\,u.c.}]_{16}$ superlattice. The data basically shows an exponential decrease of the exchange bias H_{ex} with the increase of temperature. The existence of frustration due to competing interactions is known to lead to an exponential decay of H_{ex}, and has been experimentally observed [9] and modeled, for example, by an incomplete ferromagnetic domain-wall [10] in metal–metal oxides interfaces. Our experimental results show very good agreement to the following exponential function:

$$H_{ex} = H_0 \exp\left(-T/T_0\right) + C\,. \tag{1}$$

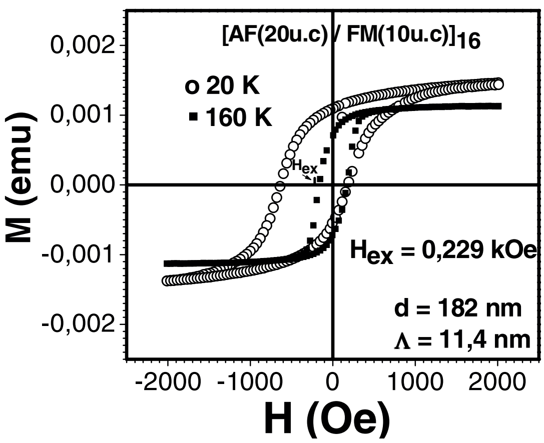

Fig. 3 Magnetic hysteretic loops of $[AF_{20\,u.c.}/FM_{10\,u.c.}]_{16}$, measured at 20 K (circle symbols) and 160 K (square symbols) after cooling from 300 K in 20 kOe.

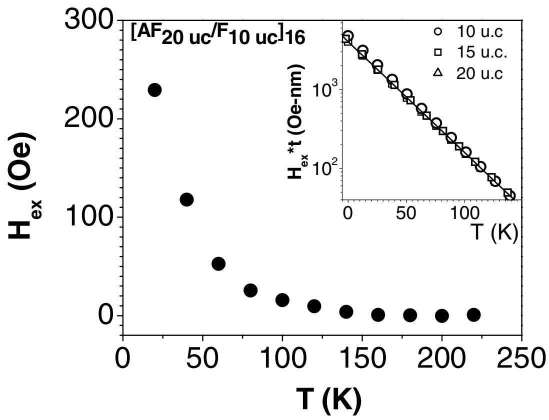

Fig. 4 Temperature dependence of H_{ex} after field cooling in 20 kOe. The inset shows the $H_{ex} \times t$ product as a function of temperature for all FM layer thicknesses.

The inset in Fig. 4 shows the temperature dependence of the product of the measured exchange bias field H_{ex} with the ferromagnetic layer thickness t_F for samples with different FM-thicknesses (10 u.c., 15 u.c and, 20 u.c.). All data fit very well on one line indicating a constant internal exchange bias mechanism that is independent from the FM film thickness. If the thickness of the FM layer (t_F) is less than the thickness of a domain wall of the AF material (δ_{AF}), the response of the FM layer to a field causes an exchange twist of spins in the AF layer near the interface [11]. In this approximation, $H_{ex} \times t_F = -2K_{AF}\delta_{AF}/M$, where K_{AF} is the magnetic anisotropy coefficient of the AF layer and, M_s is the saturation magnetization. Which term of the second member of this equation has exponentially dependence with temperature for this kind of superlattices is not clear yet. We have reported [5] a T_C-distribution in $La_{2/3}Ca_{1/3}MnO_3$ films attributes predominately to the degree of sample inhomogeneity. Studies on T_N distribution in $La_{1/3}Ca_{2/3}MnO_3$ and exponentially temperature dependence of $H_{ex} \times t_F$ for superlattices varying the thickness of the AF-layer are currently carrying on.

4 Conclusions

In summary, we have grown epitaxial [AF (20 u.c.)/F(N u.c.)] superlattices with a unit cell interfacial roughness via a high oxygen pressure sputtering technique. The superlattices show an exchange bias effect at low temperatures, below the Neel temperature of the AF-layer. We observe an exponential behaviour for the temperature dependence of the H_{ex}, similar to previous experimentally results [3, 8, 9] and theoretical models [10] in other F-AF systems. In the exchange bias regime, all experimental data for the product of $H_{ex} \times t_F$ fall onto one line for FM layer thickness ranging from 10 to 20 u.c. FC-magnetoresistance measurements show also asymmetries in the isothermal loops at low temperatures confirming these findings which are attributed to the effect of exchange biasing in F-AF superlattices.

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