Exchange biasing in $La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3$ multilayers

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A series of $[La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3]_{15}$ multilayers, with bilayer thicknesses between 2 and 32 nm, has been prepared by pulsed laser deposition. The study of their magnetic and magnetotransport properties reveals, for the first time in this category of materials, the presence of an exchange biasing mechanism at low temperatures. Zero-field-cooling and field-cooling magnetic measurements reveal a blocking temperature around 70 K that is independent of the bilayer thickness, whereas the average film magnetization becomes zero at 250 K. © *1999 American Institute of Physics.* [S0021-8979(99)27408-7]

I. INTRODUCTION

The existence of unidirectional anisotropy due to exchange coupling between a ferromagnetic (FM) and an antiferromagnetic (AF) phase was first reported in oxide-coated fine particles of Co.[1] Characteristically, exchange anisotropy results in a displaced magnetic hysteresis loop when the sample is field cooled through the Neél temperature of the AF phase. In early studies, this loop displacement has been explained by assuming an ideal AF/FM interface with uncompensated moments in the atomic plane of the AF layer at the AF/FM boundary.[1]

Up to date exchange anisotropy effects have been studied mainly in AF/FM systems consisting of transition metal alloys and metallic oxides e.g., FM═Co, NiFe, Fe_3O_4, and AF═CoO, FeMn,[1–8] where the FM or AF interactions are due to direct-exchange coupling. In this study, our aim is to develop an exchange biasing mechanism in a series of manganese perovskite $La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3$ multilayers, consisting of alternating stacks of FM $La_{2/3}Ca_{1/3}MnO_3$ layers and AF $La_{1/3}Ca_{2/3}MnO_3$ layers[9–11] where the magnetic interactions cannot be described by direct exchange.[9–12] The structural compatibility of the selected AF and FM layers permits coherent growth of the superlattice that satisfy the conditions for magnetic coupling at the interfaces.

II. SAMPLE PREPARATION AND MEASUREMENTS

The films were prepared by pulsed-laser deposition (PLD) of bulk stoichiometric $La_{2/3}Ca_{1/3}MnO_3$ (FM) and $La_{1/3}Ca_{2/3}MnO_3$ (AF) targets on single-crystal $LaAlO_3(100)$ substrates. The targets were prepared by standard solid state reaction from La_2O_3, $CaCO_3$, and MnO_2 powders sintered at 1325 °C for 5 days with two intermediate grindings. The beam of an LPX105 eximer laser (Lambda Physic), operating with KrF gas ($\lambda=248$ nm), was focused on a rotating target. In order to grow a multilayer structure, the AF and FM targets were mounted on a step-motor controlled rotatable carrier that allows different targets to be sequentially exposed in the beam path. The pulse energy was 225 mJ, resulting in a fluence of 1.5 J/cm^2 on the target. The substrate was located at a distance of 6 cm from the target, by the edge of the visible extent of the plume. During deposition the substrate temperature was stabilized at 700 °C and the oxygen pressure in the chamber was 0.3 Torr, resulting in a deposition rate of 0.04 nm per pulse. A series of $La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3$ multilayers with equal AF and FM layer thicknesses, forming bilayers between 2 and 32 nm, were grown along the (001) direction of the simple pseudocubic perovskite unit cell.

X-ray diffraction (XRD) spectra were collected with a Siemens D500 diffractometer using Cu $K\alpha$ radiation. The existence of the superstructure has been confirmed by the presence of low-angle superlattice Bragg peaks and multiple satellite peaks around the (001), (002), and (003) Bragg re-

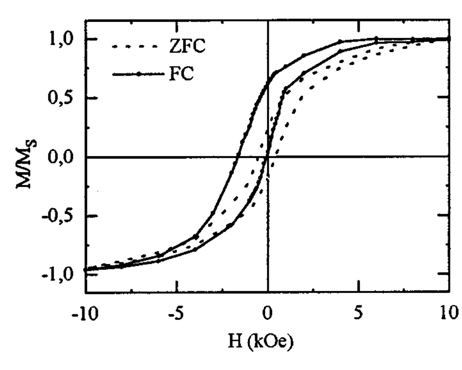

FIG. 1. Hysteresis loops, measured at 10 K after cooling down from 300 K in zero field (ZFC) and in 10 kOe (FC), for a $LaAlO_3$/ $[FM(5\ nm)/AF(5\ nm)]_{15}$ multilayer.

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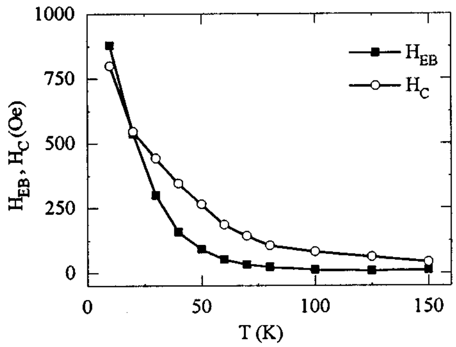

FIG. 2. Temperature dependence of exchange biasing field (H_{EB}) and coercive field (H_C) for the $LaAlO_3$/[FM(5 nm)/AF(5 nm)]$_{15}$ multilayer.

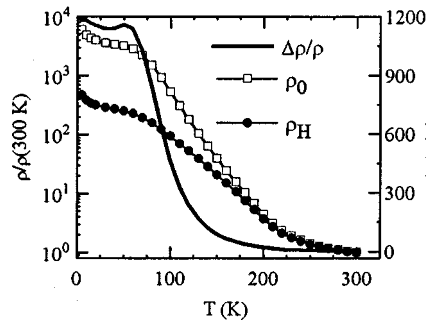

FIG. 4. Resistivity, normalized to the 300 K value, as a function of temperature, measured in 50 kOe (ρ_H) and in zero applied field (ρ_0) for a $LaAlO_3$/[FM(5 nm)/AF(5 nm)]$_{15}$ multilayer. The CMR ratio $\Delta\rho/\rho_H=[\rho_0-\rho_H]/\rho_H$ is plotted as a solid line.

flections of the constituents. Magnetic measurements were performed with a Quantum Design MPMSR2 superconducting quantum interference device (SQUID) magnetometer. The magnetotransport measurements have been carried out with the standard four-probe method, applying the magnetic field parallel to current flow direction.

III. RESULTS AND DISCUSSION

Magnetic hysteresis loops, measured at 10 K after cooling down from 300 K in zero field (ZFC) and in 10 kOe (FC), for a $LaAlO_3$/[FM(5 nm)/AF(5 nm)]$_{15}$ sample are shown in Fig. 1. It is evident that the ZFC loop is symmetric around the zero field, while the FC loop is shifted towards negative fields. This effect can be attributed to exchange biasing at the AF/FM interface, since single-layered FM films do not exhibit any loop displacement after the FC process. If H_1 is the lower and H_2 is the higher field value where the average film magnetization becomes zero, then the exchange biasing field is defined as the loop shift $H_{EB}=-(H_1+H_2)/2$ and the coercivity as the half width of the loop $H_C=(H_1-H_2)/2$. Thus, we calculate for the FC loop an $H_{EB}=880$ Oe and a $H_C=800$ Oe which is almost double compared to the H_C value obtained from the ZFC loop. Additional magnetic measurements were performed in order to investigate the origin of this effect. The temperature dependence of H_{EB} and H_C values is shown in Fig. 2. These values were estimated from isothermal loops measured in constant temperature intervals, after FC the sample from 300 K down to 10 K in 10 kOe and then warming up. It is evident that H_{EB} decreases and disappears around the so-called blocking temperature T_B of 70 K. The H_C values exhibit a similar trend, indicating a connection between the mechanisms that give rise to coercivity and loop displacement.

In Fig. 3 are shown the ZFC and FC measurements of the average film magnetization as a function of temperature. Both measurements were performed by warming up in 1 kOe after having cooled in zero field and 10 kOe, respectively. The ZFC and FC curves coincide at temperatures higher than 100 K and become zero at about 250 K, where the Curie point T_c of the FM layers is expected. The ZFC curve exhibits a broad peak around the $T_B\sim 70$ K, whereas the FC curve exhibits a steep increase just below T_B. It is reasonable to assume that the increase of magnetization in the FC measurement results from the alignment of interfacial magnetic moments, giving rise to unidirectional anisotropy below T_B.[5] Hence, the observed hump below T_B in the ZFC curve can be attributed to thermaly activated magnetic rotation over energy barriers caused by random exchange coupling at the AF/FM interfaces.

Figure 4 shows the variation of the normalized resistivity as a function of temperature, measured in 50 kOe (ρ_H) and in zero applied field (ρ_0). The resistivity increases drastically as we cool down from 300 K, spanning almost four orders of magnitude. The $\Delta\rho/\rho_H=[\rho_0-\rho_H]/\rho_H$ ratio gives an estimate of the collosal-magnetoresistance (CMR) effect.

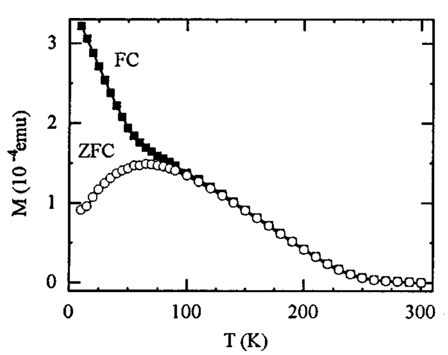

FIG. 3. Magnetization as a function of temperature for a $LaAlO_3$/[FM(5 nm)/AF(5 nm)]$_{15}$ multilayer. The measurements were performed by warming up in 1 kOe after having cooled down to 10 K, in zero field (ZFC) and 10 kOe (FC), respectively.

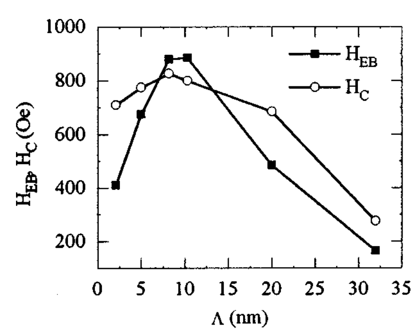

FIG. 5. Exchange biasing field (H_{EB}) and coercive field (H_C) as a function of the bilayer thickness Λ for a series of $LaAlO_3$/[FM(Λ/2)/AF(Λ/2)]$_{15}$ multilayers.

This ratio becomes maximum in the temperature range below $T_B(=70\,\mathrm{K})$. Remarkably, the characteristic peak expected at the ferromagnetic T_C of $La_{2/3}Ca_{1/3}MnO_3$ films[9] is not observed in the ρ_0 versus temperature curve. This behavior is in agreement with the magnetothermal measurements (Fig. 3) where it is evident that the most drastic change of the average film magnetization does not occur near the T_c of the individual FM layers but at T_B.

Since exchange biasing is an interface related phenomenon a strong dependence on the individual FM and AF layer thicknesses is expected. However, our magnetothermal measurements indicate that the T_B does not change in the examined range of bilayer thicknesses and occurs at 70 K for all samples. The observed H_{EB} and H_C values at 10 K, are plotted in Fig. 5 as a function of the bilayer thickness Λ for a series of multilayers with equal AF and FM layer thicknesses $LaAlO_3/[FM(\Lambda/2)/AF(\Lambda/2)]_{15}$. The maximum H_{EB} is observed for the sample with $\Lambda=10\,\mathrm{nm}$ while for thicker and thinner bilayers decreases. Again, the H_{EB} follows the variation of H_c with Λ, indicating that there is a significant contribution in H_c from the exchange anisotropy at the AF/FM interfaces.

In summary, we have studied the variation of exchange biasing and coercive field as a function of Λ and temperature in $La_{2/3}Ca_{1/3}MnO_3/La_{1/3}Ca_{2/3}MnO_3$ multilayers grown by PLD. The maximum $H_{\mathrm{EB}}=880\,\mathrm{Oe}$ was observed for the sample with $\Lambda=10\,\mathrm{nm}$. The exchange biasing mechanism sets in below a blocking temperature of 70 K and induces: (i) an enhancement of H_C in the FC hysteresis loops, (ii) an increase of the CMR ratio.

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