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On the temperature dependence of coercivity and exchange biasing field in La–Ca–Mn–O ferromagnetic/antiferromagnetic multilayers

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Abstract

Two series of exchange-coupled $[La_{1-x}Ca_xMnO_3(4\,nm)/La_{1-y}Ca_yMnO_3\ (4\,nm)]_{15}$ multilayers (one with constant $y=0.67$ while $x=0.33$, 0.4, 0.48, and the other with constant $x=0.4$ while $y=0.52$, 0.75) were used to investigate the effect of the field cooling on both, the exchange-biasing field (H_{EB}) and the coercivity (H_c). Large magnetothermal irreversibilities result in spin-glass-like $M(T)$ curves. Exchange bias appears below a blocking temperature $T_B \approx 70\,K$. Between 5 and 70 K, the $M(H)$ loops exhibit an asymmetry between the magnetization reversal from $+M$ to $-M$ and that from $-M$ to $+M$, which seems to depend strongly on the domain structure in the FM layers.

Keywords: Exchange biasing; Magnetization reversal; Multilayers; Perovskite structure

Currently, the technological interest in exchange-coupled ferromagnetic (FM)/antiferromagnetic (AF) junctions shifts to [1] thin-layered structures that contain half-metallic $La_{0.67}A_{0.33}MnO_3$ (A = Ba, Ca, Sr) FM manganites where all the mobile carriers have identical spin states and their conduction band is expected to be 100% spin polarized [2]. Recently, we have reported [3,4] the existence of exchange biasing [5] in compositionally modulated structures consisting of $[La_{2/3}Ca_{1/3}MnO_3(FM)/La_{1/3}Ca_{2/3}MnO_3(AF)]_{15}$ multilayers grown on (0 0 1) $LaAlO_3$ by pulsed-laser deposition. These studies reveal that [3,4] exchange coupling appears below 70 K which is much lower than the magnetic ordering temperatures of the AF (T_N) and the FM (T_c) layers.

In this study, two series of exchange-coupled [3,4] $[La_{1-x}Ca_xMnO_3(4\,nm)/La_{1-y}Ca_yMnO_3(4\,nm)]_{15}$ multilayers (one with constant $y=0.67$ while $x=0.33$, 0.4, 0.48, and the other with constant $x=0.4$ while $y=0.52$, 0.75) were used to investigate the effect of the field cooling on both, the exchange-biasing field (H_{EB}) and the coercivity (H_c). The multilayers were prepared [3,4] by pulsed-laser deposition of bulk stoichiometric targets on (0 0 1) $LaAlO_3$ single-crystal substrates. Both series were deposited on 40 nm thick AF buffer layer to achieve a better lattice matching with the substrate used. For brevity, we named the samples by the Ca concentration ratio x/y used. The epitaxially strained growth of these multilayers, along the pseudocubic (0 0 1) direction of the perovskite unit cell, has been revealed by low- and high-angle X-ray diffraction [4], conventional and high-resolution transmission electron microscopy (TEM) measurements [6].

Magnetic measurements were performed in a Quantum Design MPMSR2 SQUID magnetometer with the field applied in the film plane. The coercive and exchange-biasing fields were derived from isothermal loops at various temperatures after zero-field cooling (ZFC) from 300 K and field cooling (FC) in 50 kOe. Below 70 K the ZFC loops are symmetric around zero whereas the FC loops are shifted towards negative fields, evidencing exchange biasing mechanism in all the samples (Table 1). The H_{EB} is defined as the loop

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Table 1
Typical H_{EB}, H_c^{FC} values from FC loops with $H_{FC} = 50$ kOe at 10 K. T_{bif} is the bifurcation temperature from Fig. 1 and T_C is the Curie temperature of the film

x/y	H_c^{FC} (Oe)	H_{EB} (Oe)	T_{bif} (K)	T_C (K)
0.40/0.52	860	125	215	246
0.33/0.67	960	545	215	215
0.40/0.67	1030	385	185	212
0.48/0.67	955	380	205	224
0.40/0.75	1150	870	155	195

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 hysteresis loop intercept the abscissa, then $H_{EB} = -(H_1 + H_2)/2$ and $H_c = -(H_1 - H_2)/2$. It is worth noting that neither our results nor other studies reveal exchange biasing effects in single FM or AF thin films, whereas exchange-bias appears in AF/FM multilayers without a thick AF buffer layer as well [3].

The magnetothermal ZFC and FC curves (Fig. 1) were performed by warming up in 100 Oe after having cooled in zero field and 50 kOe, respectively. The inset of Fig. 1 shows the bifurcation of the FC and the ZFC magnetizations that occurs between 155 and 215 K (Table 1), whereas exchange biasing can be detected only below 70 K. We have observed [6] that in bilayers the bifurcation of the FC and the ZFC magnetizations appears a few degrees below the T_c of the FM layers whereas in multilayers the T_{bif} is much lower than the T_N and the T_c of the AF and the FM layers. This shows that the T_{bif} depends on the number of AF/FM interfaces. Apparently, all the FC curves exhibit a steep increase of M_{FC} below 70 K, that defines [3,4] a blocking temperature T_B. In particular, the magnitude of the M_{FC} at 5 K becomes about three times larger than the M_{FC} at T_B. This large increase below the T_B cannot be solely due to alignment of uncompensated interfacial spins [7], indicating that the interfacial spin order determines the magnetic order within the FM layers.

To investigate the correlation between H_{EB} and H_c six series of FC loops were measured after cooling down to 5 K in six different $H_{FC} = 5$, 10, 20, 30, 40, 50 kOe, and then warming up succesively to 5, 10, 20, 30, 40, 50, 60, 70 K. Each loop was measured at a maximum field equal to H_{FC} used. Fig. 2 shows the temperature dependence of H_1 and H_2 values on the H_{FC} used in the 0.40/0.67 sample. This shows that the different H_{FC} affect the degree of FM alignment in the FM areas below the T_B. Similar results were observed in samples with $(x + y)/2 \geqslant 0.5$. Fig. 3 is a diagram showing the relation between H_{EB} and H_c^{FC} in the 0.40/0.67 sample when: (1) the H_{FC} is fixed while the temperature increases (dashed lines), and (2) the temperature is fixed and the H_{FC} is

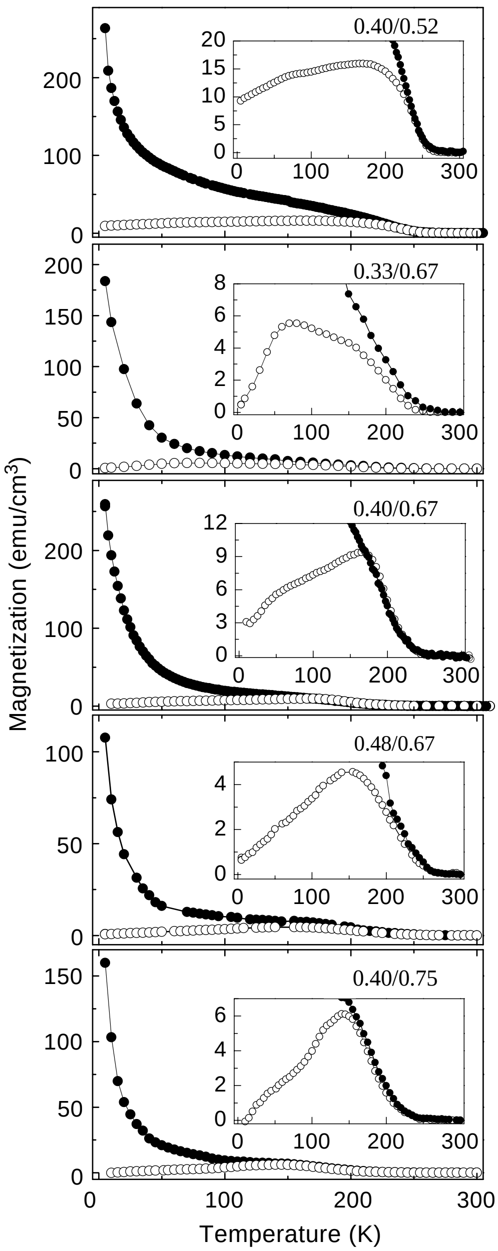

Fig. 1. Magnetothermal measurements, performed by warming up in an applied field of 100 Oe after cooling down from 300 K in zero field (open circles) and 50 kOe (FC, solid circles). The magnetization is normalized to the total FM volume of the film used. The insets show the bifurcation of the ZFC from the FC curves.

different (solid lines). In agreement with the thermomagnetic curves in Fig. 1, the H_{EB} approaches zero at about 70 K($= T_B$) whereas the H_c^{FC} converges at similar values above T_B, indicating that the applied H_{FC} affects in a different way the micromagnetic state only when the exchange coupling interaction sets in the AF/FM

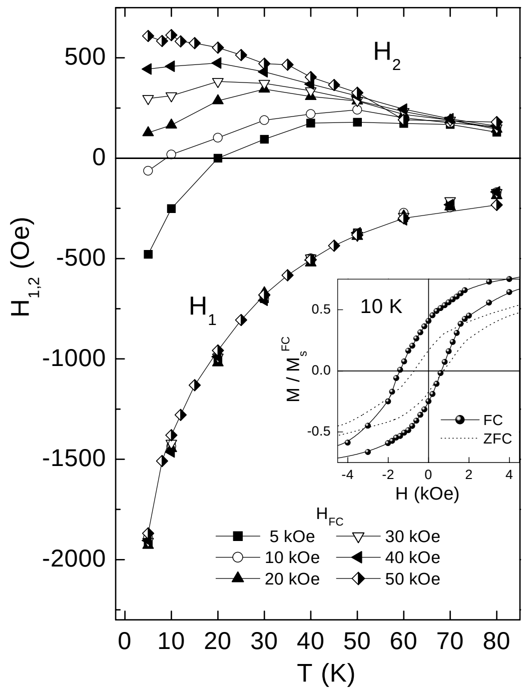

Fig. 2. Sample 0.40/0.67: Temperature dependence of H_1 and H_2 at six different cooling fields. The maximum field in each hysteresis loop is the same as the cooling field used. Lines are guides to the eye. The inset shows the loop-shift observed at 10 K.

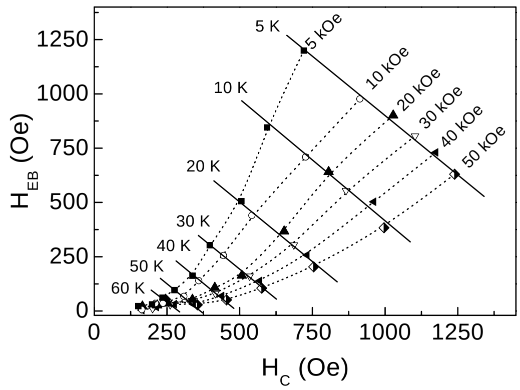

Fig. 3. The relation between the H_{EB} and the H_c^{FC} is shown for the 0.40/0.67 sample. Dashed lines correspond to fixed H_{FC} in a sequence of temperatures whereas the solid lines are the emanating isothermal curves for different H_{FC} values. The solid lines are linear least-squares fits resulting in a slope of about −1 for each temperature.

interfaces. Thus, it is shown that at low temperatures the H_{EB} values are enhanced as the applied H_{FC} decreases and vice versa for the corresponding H_c^{FC} values. Initially, it seems that the dependence of H_{EB} on H_{FC} can be attributed to minor-loop effects. However, in Fig. 2 the H_2 is strongly enhanced below ~70 K by increasing the applied H_{FC} from 5 up to 50 kOe whereas the H_1 is the same, resulting in a constant sum $H_{EB} + H_c^{FC} = H_1$ as a function of H_{FC} at a given temperature.

In summary, it was shown that at the heart of exchange coupling in (La,Ca)MnO_3 AF/FM multilayers is the steep decrease of M_{FC}, observed between 5 and 70 K. Figs. 2 and 3 show that the FC-$M(H)$ loops exhibit an asymmetry between the magnetization reversal from $+M$ to $-M$ and that from $-M$ to $+M$. This loop asymmetry is a characteristic property associated with the elusive mechanism of exchange biasing that concerns the shape and the location of the domain wall in the FM or the AF layer. A comparative study of exchange-biasing properties in bilayers and multilayers [6] indicates that magnetization reversal depends strongly on the domain structure in the FM layers.

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