

Research articles

A phenomenological model for the spontaneous exchange bias effect

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ABSTRACT

In this work we propose an alternative model to explain the spontaneous exchange bias (SEB) effect observed in spin glass (SG)-like systems. As in a previously proposed model [1], it is based on the unconventional dynamics of the SG-like moments at the magnetic hysteresis cycle. However, using a reliable estimate of the amount of SG-spins that are relaxing during the cycle, the new model can correctly describe the changes in the SEB observed for measurements performed at different temperatures and different maximum applied fields.

1. Introduction

The exchange bias (EB) effect finds its applicability in magnetic recording read heads and spintronic devices. The phenomena is known since the 1950's, being characterized by a horizontal shift of the magnetic hysteresis loop of heterostructured materials [2]. In general, the exchange unidirectional anisotropy is set at the interface of different magnetic phases after the system is cooled in the presence of an external magnetic field (H) from above its magnetic transition temperature (T). Nonetheless, recently there were discovered some materials manifesting the EB spontaneously, *i.e.* even after being cooled from an unmagnetized state down to low T in zero H [3–5].

Recently we have shown that the presence of a re-entrant spin glass (RSG) state is necessary for the manifestation of the spontaneous EB (SEB) effect, and proposed a model to explain the phenomenon [1]. The model is based on the pinning of the spin glass (SG)-like moments and on their unusual temporal evolution in magnetization as a function of H [$M(H)$] curves, from which it is obtained accurate predictions of the SEB effect observed in two representative SEB materials, $\text{La}_{1.5}\text{Sr}_{0.5}\text{CoMnO}_6$ [6] (LSCMO) and $\text{La}_{1.5}\text{Ca}_{0.5}\text{CoMnO}_6$ [8,7] (LCCMO). More specifically, our model considers an asymmetric magnetic relaxation of the SG-like moments at the regions encompassing the positive and negative coercive fields (H_C) in the $M(H)$ cycle of these materials. It is assumed, as an approximation, that for the region close to the positive H_C , half of SG-like spins are relaxing due to the negative H previously applied in the third quadrant of the $M(H)$ cycle, while the other half is pinned toward the positive H direction. However, in spite of the very good agreement between the theoretical and experimental results observed for LSCMO and LCCMO in $M(H)$ loops carried at $T =$

5 K with a maximum applied field (H_m) of 90 kOe, one cannot rely that this assumption is always correct. By measuring at different T and different H_m , the correlation lengths for the SG-like phase can change, resulting in unique magnetic relaxations and consequently in distinctive EB fields (H_{EB}).

Due to its robust SEB effect [1,6], in this work we chose the LSCMO compound as a representative example of a SEB material to confirm that T and H_m remarkably affect the H_{EB} observed in $M(H)$ loops, complementing the model proposed in Ref. [1]. Here we offer a complementary model, adapted from the previous one, but that is based on a plausible estimate of the SG-like moments pinned toward the positive and negative H directions. This model can capture the evolution of H_{EB} as a function of T and H_m , resulting in theoretical values that are very close to those experimentally observed for LSCMO.

2. Experiment details

The polycrystalline LSCMO sample here investigated was prepared as described elsewhere [1]. The $M(H)$ loops were carried out at several T and H_m using a Quantum Design PPMS-VSM magnetometer. The curves were measured at a H sweep rate of 210 Oe/s, after zero field cooling (ZFC) the system. In order to prevent the presence of trapped current on the magnet and ensure a reliable ZFC process, from one measurement to another the sample was warmed up to the paramagnetic state and the coil was demagnetized in the oscillating mode.

3. Results and discussion

Fig. 1(a) shows the $M(H)$ loop at $T = 2$ K and $H_m = 90$ kOe. The

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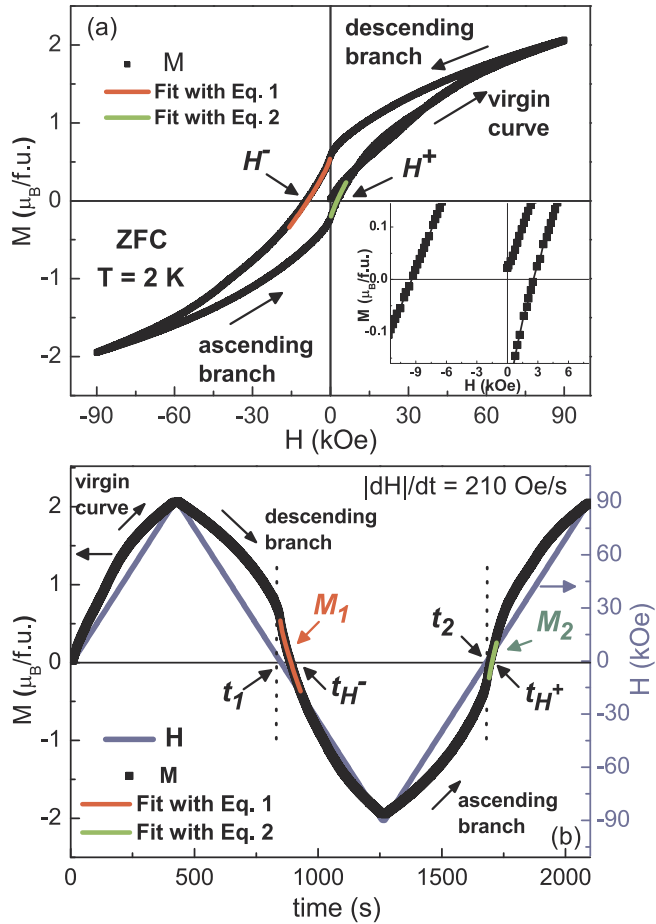


Fig. 1. (a) $M(H)$ loop of LSCMO at $T = 2$ K and $H_m = 90$ kOe. Red and green solid lines are the calculated M_1 (Eq. (1)) and M_2 (Eq. (2)) stretches, respectively. Inset shows zoom in around $M = 0$, evidencing the shift along H -axis. (b) The same hysteresis loop for LSCMO, now displayed in $M(t)$ mode. The blue solid line is the magnetic field as function of time.

shape of the curve is a result of the contribution of three distinct magnetic phases, a ferromagnetic (FM), an antiferromagnetic (AFM) and a SG-like phase [1], producing a closed loop that is asymmetric in respect to the H axis, as evidenced in the inset. In the $M(H)$ cycle the magnetization (M) depends on H , which in turn varies linearly with time (t). Thus, the hysteresis curves can be displayed in the form of M as a function of t [$M(t)$], Fig. 1(b). Since our model is based on the time-evolution of the magnetization of the SG-like phase, this form is suitable for its understanding.

The phenomenological model intends to describe the H_{EB} obtained from $M(H)$ curves, herein defined as $H_{EB} = |H^+ + H^-|/2$, where H^+ and H^- are the positive and negative coercive fields, respectively. For that, the M_1 and M_2 stretches of the curve, encompassing respectively the H^- and H^+ [see Figs. 1(a) and (b)], must be calculated. The material's net magnetization results from the contributions of FM, AFM and SG-like phases, leading to the following equation for M_1

$$M_1(t) = \{M_{sp} + M_0 e^{-(t-t_1)/t_p}\} - \{A(t - t_1) + B(t - t_1)^r\}, \quad (1)$$

which corresponds precisely to the equation proposed in Ref. [1]. The first pair of braces represents the SG-like phase's relaxation from the previously applied positive H_m . In fact, this corresponds to the stretched exponential equation commonly used to verify the isothermal remnant magnetization (IRM) of glassy magnetic systems [9], where M_{sp} represents the spontaneous magnetization of the FM phase, M_0 is the initial magnetization of the SG-like phase at the instant t_1 when $H = 0$ (see Fig. 1), and t_p and n are the time and the time-stretch exponential

Table 1

Main results obtained from the fits of M_1 and M_2 stretches with Eqs. (1) and (2). The H_{EB} results obtained experimentally, the values calculated with Eqs. (1) and (2) and those obtained from the previous model of Ref. [1] are referred respectively as H_{EB}^{exp} , H_{EB}^{new} and H_{EB}^{old} .

H_m (kOe)	70	90	90
T (K)	2	2	5
M_{sp} ($\mu_B/f.u.$)	0.220	0.226	0.231
M_0 ($\mu_B/f.u.$)	0.348	0.363	0.370
t_p (s)	4.882×10^{10}	1.983×10^9	1.961×10^{10}
n	0.130	0.122	0.143
A ($\mu_B/f.u.$)	5.2×10^{-4}	6.1×10^{-4}	9.8×10^{-4}
B ($\mu_B/f.u.$)	0.028	0.026	0.023
r	0.78	0.79	0.74
x	0.47	0.43	0.54
H_{EB}^{exp} (Oe)	3517	3716	3114
H_{EB}^{new} (Oe)	3539	3719	3084
H_{EB}^{old} (Oe)	3049	3085	3172

constants, respectively. The second pair of braces account for the contributions of the AFM and FM phases to M_1 , when under the effect of the immediately applied negative H . The A parameter is related to the linear dependence of the AFM phase with H (and consequently with t), while the B and r parameters account for the non-linear contribution of the FM phase to the magnetization.

To get the parameters of the first pair of braces of Eq. (1), the $IRM(t)$ curve must be fitted immediately after H_m is turned on and subsequently turned off in the ZFC sample. Subsequently, by fixing the parameters of the SG-like phase, the M_1 stretch can be fitted with Eq. (1), yielding a very good match, as expected [1]. The main results obtained from the fittings are displayed in Table 1.

For the M_2 stretch, we take into account the unusually slow relaxation of glassy magnetic systems. It is assumed that at M_2 not all spins of the SG-like phase are relaxing due to the negative field previously applied during the $M(H)$ cycle, but it considers that some amount of them are still pinned toward positive direction due to the firstly applied positive H_m . The resulting equation for M_2 becomes

$$M_2(t) = -\{M_{sp} + xM_0 e^{-[(t-t_2)/t_p]^n}\} + \{(1-x)M_0 e^{-[(t-t_1)/t_p]^n}\} + \{A(t - t_2) + B(t - t_2)^r\}, \quad (2)$$

where the first pair of braces represents the decay of the SG-like spins that are relaxing from the negative H previously applied, the second pair corresponds to the relaxation from the positive H applied before, and the third pair represents the contributions of the AFM and FM phases that are under the effect of the just applied positive H . This is similar to the equation proposed in Ref. [1], where the decay of the SG moments pointing toward the positive direction starts at t_1 while the decay of those pointing toward the negative direction starts at t_2 . However, there is a remarkable difference here. While for the previous model it was considered that an equal amount of the moments were relaxing in opposite directions, here the amount of SG-spins pointing toward negative (x) and positive ($1-x$) directions can be estimated directly from the experimental curve. To fit M_2 with Eq. (2), all parameters, with the exception of x , are kept fixed at the values obtained from the fit of M_1 .

Although the previous model succeed in predicting the H_{EB} values of $M(H)$ loops carried at $T = 5$ K and $H_m = 90$ kOe, changes in the T and/or the H_m at which the experiments are carried can lead to deviations from the calculated results in relation to the experimentally observed values. Fig. 2(a) shows a magnified view of loops carried with $H_m = 90$ kOe at different temperatures. It is evident that changing T has a greater impact on H^+ , while H^- keeps nearly unchanged. This can be understood in terms of the gain of thermal energy with increasing T , which

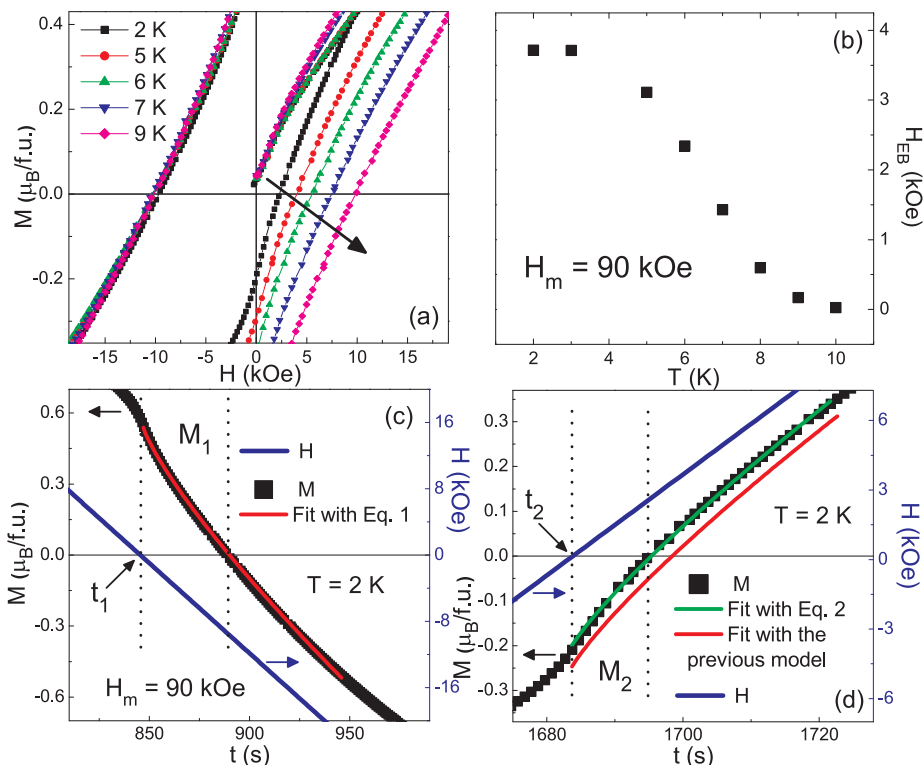


Fig. 2. (a) Magnified view of the $M(H)$ loops of LSCMO carried at $H_m = 90$ kOe at different temperatures. (b) H_{EB} as a function of T . (c) and (d) show magnified views of the experimental and calculated M_1 and M_2 stretches of the loop carried at $T = 2$ K. The blue solid line represents the H time dependence.

favours the flipping of spins toward the negative field direction at the third quadrant of the cycle. As T increases the amount x of SG-spins flipped enhances, leading to the increase of H^+ and consequently to the decrease of H_{EB} , as can be seen in Fig. 2(b). Since the previous model assumes a fixed number of SG-spins in each direction, it cannot capture such changes. Fig. 2(d) shows the fittings of the M_2 stretch of the 2 K $M(H)$ curve for both the previous and the alternative model here proposed, and Table 1 displays the H_{EB} obtained from each model. The difference is remarkable.

The alternative model can also capture changes in $M(H)$ curves measured at different H_m . Fig. 3(a) shows that H^- is significantly influenced by changes in H_m , while H^+ keeps nearly unchanged. In this case, as larger is H_m greater will be the amount of spins pinned toward positive direction. This is translated in the equations by the decrease of x and by the increase of both M_{sp} and M_0 , as can be seen in Table 1. This leads to the increase of H^- , resulting in the enhancement of H_{EB} . Again, the previous model cannot account for such changes because it considers a fixed number of spins pointed toward opposite directions.

Despite the very good adequacy of the alternative model proposed here, one must stress that it results from approximations and simplifications, most of them concerning the dynamics of the SG-like phase. For instance, the varying H may alter the balance between the distinct magnetic phases present in the system, in a way that in principle several parameters of the equations should be functions of t . To minimize this effect, we fitted M_1 and M_2 for short time-intervals. Another clear approximation comes from the fact that the second brace of Eq. (1) and the third brace of Eq. (2) take into account the influence of the immediately applied H only on the FM and AFM phases, but not on the SG-like one. In principle, a term should be added in the equations to regard it. However, since the two terms in these braces were enough to yield precise fits of M_1 and M_2 , we have disregarded a third term in order to make the model as simple as possible. Despite these simplifications, Eqs. (1) and (2) can successfully reproduce the H_{EB} observed in LSCMO. Since the RSG state is a common feature of all known SEB materials, in principle the model could be applied to any similar compound. Having

established that the glassy magnetism is imperative for the manifestation of SEB, the model here proposed can guide the search for new materials presenting robust SEB effect at higher T .

4. Conclusions

In summary, we proposed an alternative phenomenological model to explain the SEB effect observed in RSG systems. It is based on the pinning and on the dynamics of the SG-like moments relaxing during a magnetic hysteresis measurement. Differently of the model previously proposed [1], here we considered a reliable estimate of the amount of SG-like moments pinned toward the positive and negative H directions during the $M(H)$ cycle. We used LSCMO as a representative example of SEB to check the model and show that it allows an accurate calculation of the H_{EB} and a correct description of the changes observed in the SEB effect for $M(H)$ curves measured at different T and H_m .

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

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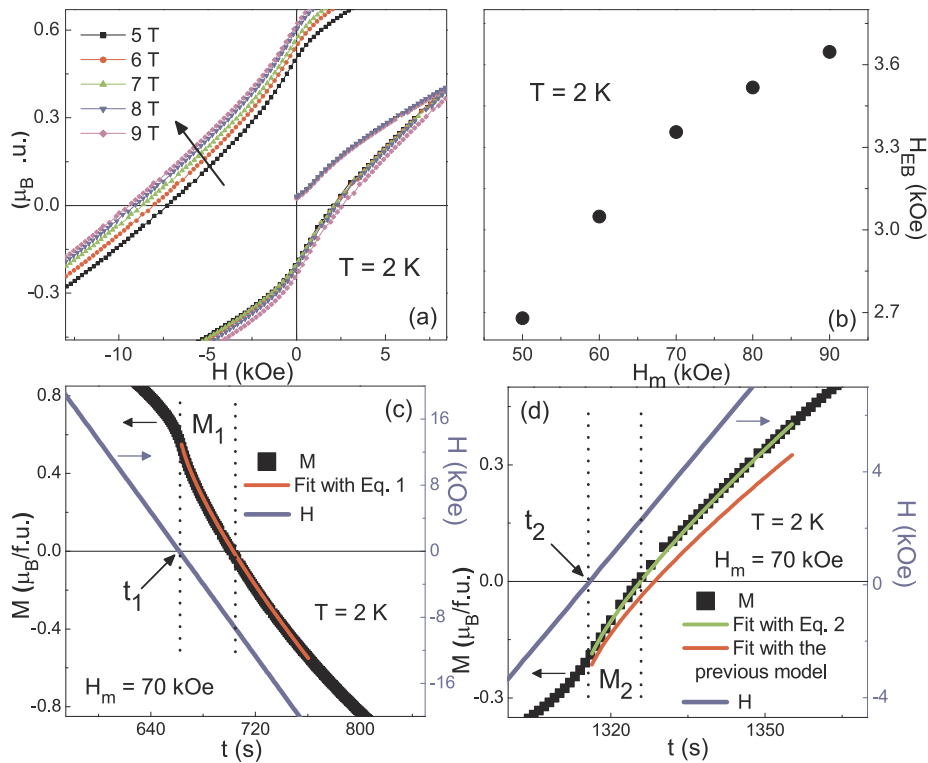


Fig. 3. (a) Magnified view of the $M(H)$ loops of LSCMO carried at $T = 2$ K with different H_m . (b) H_{EB} as a function of H_m . (c) and (d) show magnified views of the experimental and calculated M_1 and M_2 stretches of the loop carried at $H_m = 70$ kOe. The blue solid line represents the H time dependence.

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