

## Exchange bias in a Co/CoO/Co trilayer with two different ferromagnetic-antiferromagnetic interfaces

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(Received 4 November 2011; published 17 January 2012)

The exchange-bias properties of a Co/CoO/Co sandwich structure are examined. The Co/CoO bottom bilayer was obtained by oxidation of a Co layer and the top bilayer was formed by subsequent deposition of Co on top of CoO. The strength of the interfacial coupling is shown to be stronger at the bottom Co/CoO interface than at the top interface. A number of original properties characterize exchange bias of the top bilayer. This includes weak temperature dependence of the bias field, weak coercivity due to exchange bias, and lack of the training effect. These properties suggest that the antiferromagnetic moment configuration is frozen during magnetization reversal of the top Co layer.

DOI: [10.1103/PhysRevB.85.014413](https://doi.org/10.1103/PhysRevB.85.014413)

PACS number(s): 75.30.Et, 75.30.Gw, 75.50.Ee, 75.70.-i

### I. INTRODUCTION

Exchange bias (EB) appears in hybrid magnetic nanostructures due to interfacial exchange coupling between different magnetic phases, most often ferromagnetic (F) and antiferromagnetic (AF) ones.<sup>1</sup> This effect manifests itself as a shift of the magnetic hysteresis loop along the field axis after field cooling the sample down through a certain temperature, which is typically of the order of the Néel temperature  $T_N$  of the AF phase. The bias field  $H_{eb}$  is normally negative (i.e., opposite to the cooling field direction), and it vanishes at a blocking temperature  $T_B$ , such that  $T_B < T_N$ .

In the original Meiklejohn-Bean model, the AF moment configuration is assumed to consist of a stacking of ferromagnetic planes, of which orientation alternates from one plane to the next. The AF moments in interactions with the F ones have all the same orientation, i.e., the interface is fully uncompensated. The associated F-AF coupling is unfrustrated and it may be expected to have some values intermediate between F-F and AF-AF coupling. Assuming that the AF spin structure stays rigid during reversal of the F moments, the calculated  $H_{eb}$  value is typically one to two orders of magnitude larger than the experimental bias field.<sup>2-5</sup> This discrepancy between calculated and experimental EB fields indicates that the interfacial AF moment configuration is more complex than the above naive picture assumes. Actually, the uncompensation of the interfacial AF moments is weak in general and the AF moment configuration is not fully frozen during F reversal. For these reasons, EB is a complex phenomenon and a variety of behaviors are found in different systems, which indicates that different mechanisms may be the source of hysteresis loop biasing.

A number of models have been developed to describe the properties of exchange-bias systems.<sup>6</sup> Among them, one may quote models that assume the formation of a domain wall in the antiferromagnet during F moment reversal<sup>7</sup> or hybrid domain walls at the F-AF interface,<sup>8</sup> the Malozemoff's model derived from the fundamental analogy between F-AF coupling and random anisotropy in disordered systems,<sup>9,10</sup> and the spin-flop models of exchange bias.<sup>11-13</sup> Some of the topics of current interest include the role of interfacial spins in exchange bias,<sup>14</sup> the role of the bulk AF spin structure,<sup>15</sup> asymmetric

reversal due to competing anisotropies in such systems,<sup>16</sup> EB in nanoparticle systems,<sup>17,18</sup> and influence of interface roughness on EB.<sup>19,20</sup>

Despite the variety of behaviors of different systems, a number of phenomena are common to almost all EB systems. This includes (i) EB training effects, which manifest themselves by the fact that, as the hysteresis loop is measured a number of times, the loop progressively evolves and, in particular, a progressive decrease in  $H_{eb}$  occurs.<sup>21-23</sup> Other common features of exchange-bias systems are the following: (ii) the bias field decreases with temperature much faster than the individual magnetization of the ferromagnetic and antiferromagnetic layers involved, and (iii) a large contribution to coercivity is associated with exchange bias.

It remains to be determined whether these common phenomena are inherent to EB in general. Since AF moment rearrangement appears to affect the properties of many EB systems, one of the first questions to be answered is how a simpler conceptual exchange bias system would behave, in which the AF moments would remain frozen as the F magnetization is reversed. To approach this question, a model Co/CoO/Co sandwich structure has been prepared and its properties examined in this paper.

### II. SAMPLE PREPARATION

The samples were prepared using dc triode sputtering.<sup>24</sup> For calibration purpose, a Co film was deposited initially for which the thickness was determined by scanning electron microscopy observation of the cross section. The deduced deposition rate was of approximately 1 nm/s. A "bottom" Co layer was deposited then, under the exact same conditions (filament current, target voltage, and Ar partial pressure) as the calibration Co film, on a thermally oxidized Si substrate, which was covered by a 100-nm-thick Ta buffer layer. The deposition duration was determined to obtain a Co thickness of 6 nm. After deposition, the film was exposed to air at 200 °C to form a surface CoO oxide layer. Magnetization measurements revealed that the effective nonoxidized thickness of the bottom Co layer was 2 nm (see below), thus indicating that 4 nm of Co were oxidized to form CoO. From AFM, the roughness of this

CoO layer was determined to be 2.5 nm rms. A 10-nm Co “top” layer was deposited on top of the CoO layer, and covered *in situ* by a 100-nm Ta capping layer. The expectation in preparing this system was that the EB will most likely be different at both interfaces. Assuming that each CoO grain within the AF layer is coupled at the same time to both the bottom and top F layers, it was hoped that the AF moment configuration will remain frozen during reversal of the ferromagnetic Co layer with which the exchange coupling energy will be the weaker. The thicknesses of the bottom and top Co layers were made significantly different so that magnetization reversal of each layer occurs at different field values, and it is possible to identify the magnetization of the layer that reverses from the corresponding magnetization jumps on the hysteresis cycle.

### III. MAGNETIZATION MEASUREMENTS

Magnetization measurements were performed using a Quantum Design MPMS SQUID magnetometer. Figure 1 shows the first (filled circles) and the trained (open circles) hysteresis loops taken at 10 K after cooling down from 300 K under an applied magnetic field of 1 T. Each loop is a superposition of two components corresponding to magnetization reversal of the top and the bottom Co layers, respectively, and each component is characterized by specific magnetization jumps and switching fields (which were derived from the maxima found in the superimposed susceptibility). The smaller magnetization jumps correspond to reversal of the bottom layer and the larger jumps correspond to reversal of the top layer. The top-layer jumps are almost exactly five times larger than the bottom-layer jumps. Knowing that the top-layer thickness is 10 nm, it is derived that the bottom Co layer is 2 nm thick after oxidation. For each component to the hysteresis cycle, the exchange-bias field  $H_{eb}$  and coercive field  $H_c$  were defined as half the sum and half the difference of the switching fields in positive and negative applied fields, respectively. At 10 K, the exchange-bias field of the first hysteresis loop of

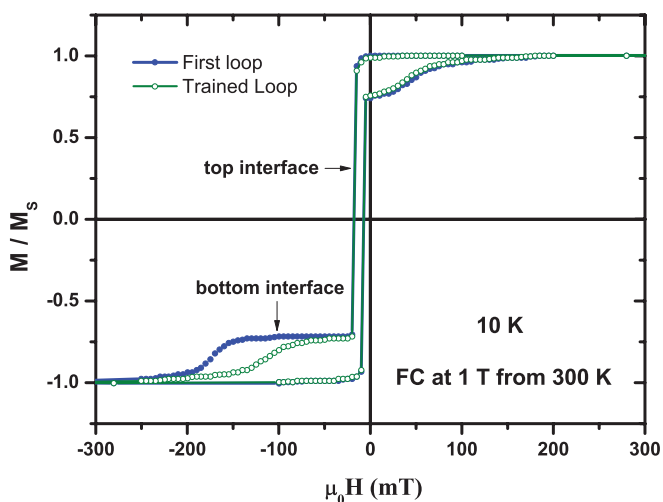


FIG. 1. (Color online) First and trained magnetic hysteresis loops for the sample measured at 10 K after field cooling at 1 T from 300 K. The component with lower  $H_c$  and higher  $M_s$  corresponds to the top Co layer, and the component with larger  $H_c$  and lower  $M_s$  corresponds to the bottom Co layer.

the Co bottom layer is equal to  $\mu_0 H_{eb} = -63.5$  mT and the coercive field to  $\mu_0 H_c = -111.5$  mT. The corresponding values for the Co top layer are  $\mu_0 H_{eb} = -12.5$  mT and  $\mu_0 H_c = 5.5$  mT. The relative widths of the switching field ( $H_{sw}$ ) distributions have similar values for both layers ( $\delta H_{sw}/H_{sw} \approx 0.1$ ).

It is usual to express the interfacial exchange-coupling energy per unit surface area  $\gamma$  of EB systems as

$$\gamma = \mu_0 M_s H_{eb} t_F. \quad (1)$$

Here,  $\mu_0 M_s$  is the spontaneous magnetization, equal to 1.82 T at 10 K for a Co ferromagnetic film, and  $t_F$  is the ferromagnetic thickness. From  $\mu_0 H_{eb} = -63.5$  mT and  $t_F = 2$  nm, one derives  $\gamma_b = 0.184$  mJ/m<sup>2</sup> for the bottom-layer interfacial energy. Similarly, for the top layer, the interfacial coupling energy according to expression (1) is derived to be  $\gamma_t = 0.181$  mJ/m<sup>2</sup>. These two values are very close to one another. We will show in the following that the coupling energy, normalized to a given grain, is actually larger at the bottom layer. The coupling energy averaged over the entire sample is identical at both interfaces because a fraction of AF moments at the bottom interface reverses with the F layer and do not contribute to EB.

As temperature is increased,  $H_{eb}$  for the bottom bilayer decreases by a factor of 30 from 63.5 mT at 10 K to 2 mT at 160 K, as shown in Fig. 2. In the same temperature range,  $H_{eb}$  for the top bilayer decreases by a factor of 5 from 12.5 mT at 10 K to 2 mT at 160 K. The following other features emerge from magnetization measurements. (i) Large training effects are associated with magnetization reversal of the bottom layer as usually observed in Co/CoO exchange-biased systems, whereas they are almost negligible for magnetization reversal of the top layer. (ii) The coercive field  $H_c$  (see Fig. 2) for the bottom bilayer decreases from 111.5 mT at 10 K to 7.5 mT at 250 K (15-fold increase), whereas for the top bilayer it decreases from 5.5 mT at 10 K to 2.1 mT at 250 K (threefold decrease). Note that the exchange-bias blocking temperature ( $T_B$ ) has very similar values for the two bilayers (around 220 K). It has been argued that  $T_B$  in polycrystalline exchange-bias layers is essentially determined by the size of the grains in the film.<sup>25</sup> This is consistent with the present results since the grain sizes for the two Co layers are expected to have very similar values due to the identical deposition conditions used.

Let us consider now training effects. Most generally, these are due to moment rearrangement in the AF layer: via their coupling with the F moments, some of the AF moments may overcome the local anisotropy energy barrier and follow F moment reversal. The absence of training during reversal of the top layer implies that the AF moment configuration is frozen at the corresponding interface. For three different systems, which unfortunately did not include Co/CoO/Co, Ohldag *et al.*<sup>14</sup> showed that a small fraction of pinned AF spins were responsible for EB. Obviously, it would be very meaningful to check whether any AF moment rotation is found during reversal of the top layer in the present Co/CoO/Co trilayer. With top layer reversal occurring in lower absolute fields than bottom layer reversal, one may conclude from the fact that the AF grain magnetization remains frozen during reversal of the top layer, that the F(Co)-AF(CoO) interfacial coupling

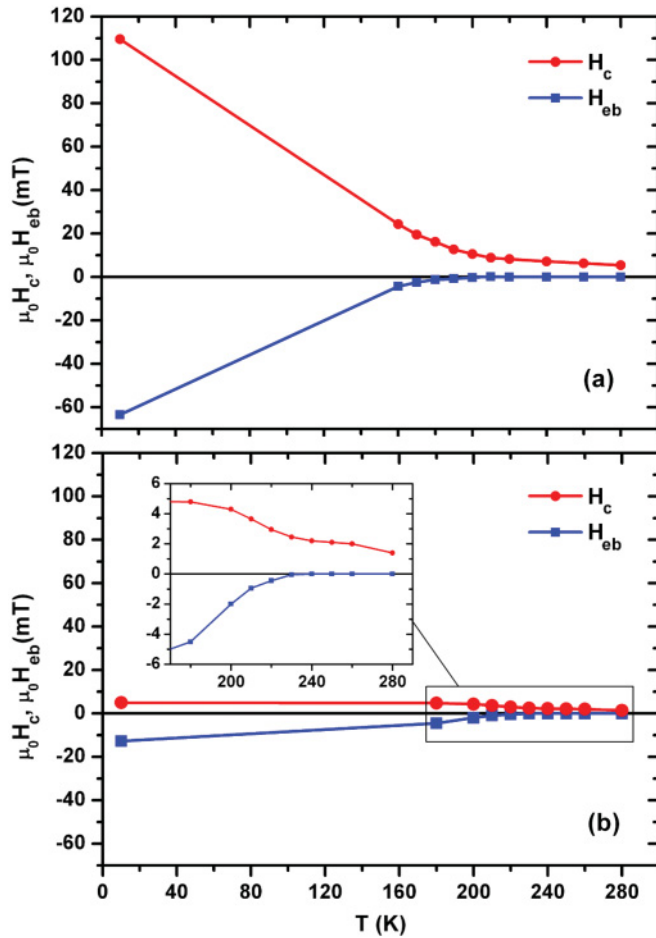


FIG. 2. (Color online) (a), (b) Temperature dependence of the coercive field (circles) and exchange-bias field (squares) for the first and trained loops for the bottom (a) and top (b) layers. The measurements at each temperature were performed after field cooling from 300 K under 1 T applied field.

is stronger at the bottom interface than at the top interface. Applying relation (1), it was concluded above that the coupling energy has almost the same values at both interfaces. However, the interfacial coupling energy in relation (1) is implicitly normalized to the surface area of the F layer. This is valid for the top layer in the present case. All AF moments contribute to EB since the AF moment configuration is frozen at this interface. For the bottom layer, the AF grains, which are most strongly coupled to the F layer and reverse with it, do not contribute to the bias field. Renormalized to the surface area of the frozen AF grains, the bottom-layer interfacial coupling energy is necessarily larger than the associated top-layer coupling energy.

Note that the larger interfacial coupling found here for the bottom F layer does not imply necessarily larger exchange-bias field. The bias field is directly linked to the thickness of the F layer [see relation (1)]. In case the bottom F layer would be much thicker than the top layer, reversal of the bottom layer would occur before reversal of the top layer.

We will show now that the qualitative differences observed in the exchange-bias properties for the top and the bottom layers, respectively, are consequences of the fact that the AF moment configuration is frozen at the top interface and not

at the bottom interface. Let us consider first the temperature dependence of  $H_{eb}$ . In usual EB systems, it is much larger than the expected temperature dependence of the AF staggered magnetization  $M_{AF}$ . This phenomenon may be related to the modification of the AF moment configuration accompanying F moment reversal. The decrease of the magnetocrystalline anisotropy with increasing temperature is normally much stronger than that of the F-AF exchange coupling. Thus, with increasing temperature, a larger fraction of the AF moments follow F moment reversal and the bias field is correspondingly reduced. To illustrate this difference between the temperature dependence of the magnetocrystalline anisotropy and that of the interfacial exchange coupling, consider the bottom bilayer in the present system. The CoO anisotropy, assumed to be dominated by second-order terms, is expected to decrease approximately as  $M_{AF}^3$ ,<sup>26</sup> whereas the F-AF coupling varies as  $M_{AF}$  (the Curie temperature of Co, 1392 K, being much higher than room temperature, the temperature dependence of the F magnetization, may be neglected).

By contrast at the top interface, the AF moment configuration being frozen, the bias field is expected to be approximately proportional to  $M_{AF}$ , in agreement with the observed much slower decrease of  $H_{eb}$  with increasing temperature. Such temperature dependence of  $H_{eb}$  is expected to agree with usual models of exchange bias, which neglect AF moment rearrangement. In a recent study,<sup>27</sup> the Néel temperature of a 4-nm-thick CoO layer corresponding approximately to the thickness of the CoO layer in the present system was found around 280 K, to be compared to the  $T_B$  value of 230 K found here. The difference between this  $T_N$  value and  $T_B$  may be ascribed to the fact that the Néel temperature is actually determined by the CoO grain size rather than by the film thickness, and that it may depend on the preparation conditions. Further,  $T_N$  at the interface may differ from  $T_N$  in the middle of the film,<sup>28</sup> and the interfacial F-AF exchange field may induce a reduction in  $T_N$ .

F-AF interfacial exchange coupling is usually a source of an additional coercivity for reversal of the ferromagnetic layer of EB systems. This represents the contribution of AF moments, which overcome their local anisotropy barrier to follow F moments reversal. Accordingly, the large increase in  $H_c$  found for the bottom layer below  $T_B$  [Fig. 2(a)] may be related to CoO AF grains, which are driven over their magnetocrystalline anisotropy energy barrier<sup>29</sup> via coupling with the Co layer. For the top layer, however, the increase in  $H_c$  below  $T_B$  is marginal [Fig. 2(b)]. By extrapolation to low temperature, the EB contribution to the coercive field is derived to be around 25 Oe. This is 40 times smaller than the coercive field for reversal of the bottom layer, to be compared to the ratio of 5 between the corresponding bias fields. This very small  $H_c$  contribution accompanying EB is another illustration of the fact that the AF moment configuration is frozen at the top interface.

The concept of exchange-bias inducing temperature  $T_{ind}$  has been introduced recently as a new probe for EB systems.<sup>28</sup>  $T_{ind}$  is determined according to the following procedure: the sample is cooled from 300 K down to a certain temperature  $T_{switch}$  under an applied negative field. At  $T_{switch}$ , the applied field is inverted and the sample is further cooled to the temperature  $T_m$  at which the hysteresis loop is measured. Differences between two measurements characterized by different  $T_{switch}$  values

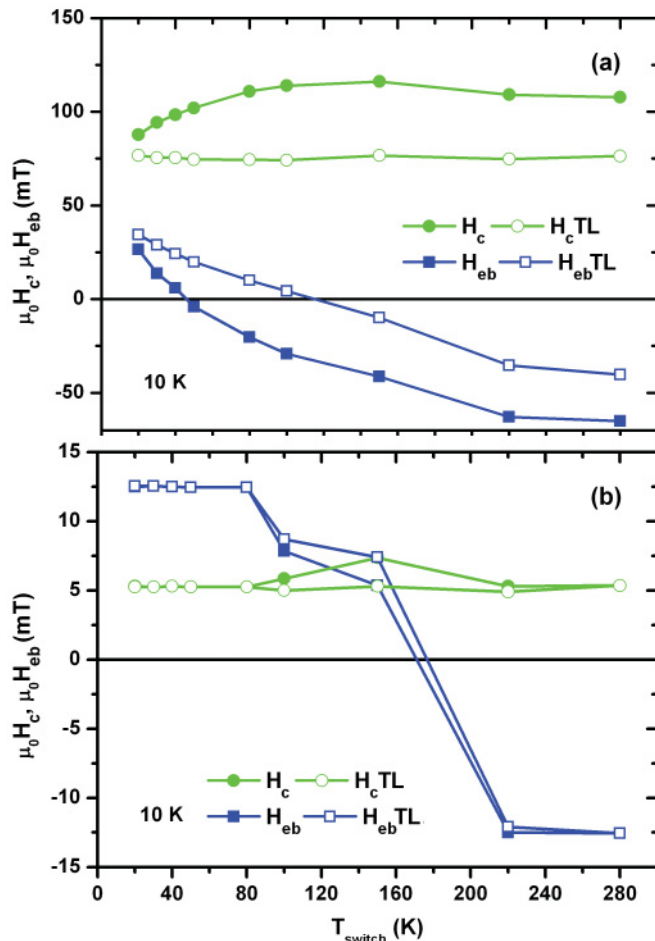


FIG. 3. (Color online) Inducing temperature measurements for the bottom (a) and top (b) layers. The main and trained hysteresis loops were measured at 10 K after switching the sign of the cooling field from  $-1$  T, under which the sample was cooled from 300 K to  $+1$  T at  $T_{switch}$ .

are direct manifestations of different configurations of the AF moments. In an ideal system, the sign of the cooling field at  $T_{switch} > T_{ind}$  leads to the change of the sign of EB (measured at  $T_m$ ), whereas inverting the sign of the cooling field at  $T_{switch} < T_{ind}$  has no effect on EB. In a real system, the bias field (again measured at  $T_m$ ) varies progressively with  $T_{switch}$ , and  $T_{ind}$  is defined as the temperature at which half the AF magnetization is reversed so that the bias field measured at  $T_m$  is zero. In the present case, the Co/CoO/Co trilayer sample was cooled under  $-1$  T down to  $T_{switch}$ , where the field was inverted to  $+1$  T. The low- $T$  hysteresis loops were measured at  $T_m = 10$  K. The  $T_{switch}$  dependence of  $H_{eb}$  for the bottom and the top bilayers are shown in Fig. 3. For the bottom bilayer,  $T_{ind} \approx 40$  K. Further,  $H_{eb}$  varies progressively as  $T_{switch}$  is reduced and it

is affected down to the lowest temperature of measurement (10 K). This implies that a non-negligible fraction of the AF moments never become frozen and follow F moment reversal. For the top bilayer,  $T_{ind}$  is much larger, of the order of 170 K. For  $T_{switch} < 80$  K, the AF moment configuration being frozen, inversion of the applied field has no effect on the bias field. For  $80 \text{ K} < T_{switch} < 220$  K, the exchange-biased loops measured at  $T_m$  show weak training effects. In this  $T_{switch}$  range, the EB field at the bottom interface being close to zero, the AF grain magnetization is not efficiently blocked.

In a previous study of a trilayer Ni/FeF<sub>2</sub>/Py EB system,<sup>15</sup> the role of the bulk spin structure on exchange bias was used to explain the influence of coupling at one F-AF interface on EB at the other F-AF interface. The analysis developed in this paper arrives at a similar conclusion. A given AF grain (or a given AF domain) is frozen due to coupling at the F-AF bottom interface and this affects the EB properties at the other interface. In Ref. 15, however, the coercive field was found not to depend on the coupling between the F layers. The difference with the present Co/CoO/Co system might rely on the much smaller thickness of the CoO AF layer in the latter case ( $\approx 5$  nm), whereas it was 200 nm in the former case (FeF<sub>2</sub>). One may suggest that the coercive field is determined by the smaller AF grains for which the energy barrier is reduced. In the case of FeF<sub>2</sub>, these small grains would not be coupled to the other F layer, unlike the bigger ones.

#### IV. CONCLUSIONS

In conclusion, we have examined the EB behavior of two Co/CoO bilayers, which involve the same CoO layer but for which the strength of the interfacial coupling differ. Properties usual to those of many EB systems were found for the bottom bilayer characterized by the stronger interfacial exchange coupling. By contrast, the EB properties of the top bilayer characterized by the weaker interfacial exchange are specific. In particular, the temperature dependence of the bias field is strongly reduced with respect to the one observed in usual EB systems, the coercivity increase due to EB is marginal, and training effects are almost negligible. These properties can be understood when considering the fact that the AF moment configuration is frozen at this interface. We suggest that the thorough study of the EB properties in systems incorporating such frozen AF interfacial configuration may contribute to a better understanding of exchange bias.

#### ACKNOWLEDGMENT

Financial support from the Foundation Nanosciences is acknowledged.

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