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COOLING FIELD EFFECT ON EXCHANGE BIAS IN NiO / $NiFe_2O_4$ BILAYERS

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We have studied the exchange anisotropy in $NiFe_2O_4$ / NiO bilayers as a function of the cooling field strength. An unexpected strong variation of the bias upon the cooling field value is observed. Maximum exchange bias fields are observed after cooling in 80 kA/m field. Larger cooling fields induce small, but more stable exchange bias fields. The exchange bias value can be directly correlated with the $NiFe_2O_4$ layer's magnetic state during the field-cool procedure.

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1. Introduction

Exchange anisotropy is observed in coupled antiferromagnetic (AF) - ferromagnetic (F) systems after a field-cool procedure, realized in order to align the AF layer. The most interesting property of this phenomenon is the shift of the hysteresis loop along the field axis, called exchange bias (H_{eb}). This feature can be used as a tool to control the magnetization in such magnetic devices as spin valves [1] or tunnel junctions [2]. Other features that generally occurs in exchange coupled systems are the increase of the coercive field value and the training effect. Training effect refers to the decrease of the coercive field and exchange bias field values upon successive cycling of the applied magnetic field. Although numerous experimental and theoretical studies are focused on this topic, there are still a lot of open questions, such as: the influence of the AF structure or morphology on exchange bias, the interactions type at the F / AF interface or the physical mechanism responsible for the training effect.

Here we present some results concerning the influence of the cooling field procedure on the exchange anisotropy and the training effect in an all-oxide system, the $NiFe_2O_4$ / NiO bilayer. The antiferromagnetic nickel oxide, NiO, is widely used as an exchange bias layer in magnetoresistive devices because it has relatively high anisotropy ($K \sim 0.3 \times 10^6$ J/m^3) and Néel temperature (T_N = 523 K). The ferrimagnetic nickel ferrite, $NiFe_2O_4$, has a saturation magnetization of about $M_S = 3 \times 10^5$ A/m and a Curie temperature T_C = 858 K.

2. Experimental

The $NiFe_2O_4$ / NiO bilayers were prepared by pulsed laser deposition (PLD) on quartz substrates at a partial oxygen pressure of 60 mTorr, using a Nd:YAG laser. The laser was operated at a wavelength of 355 nm with a pulse width of 6 ns and a repetition rate of 10 Hz. Ceramic targets of $NiFe_2O_4$ and NiO were used. The $NiFe_2O_4$ layer was deposited first, at about 900 K. For the NiO layer deposition the temperature was reduced down to about 600 K. No magnetic field was applied during deposition.

X-ray diffraction spectras recorded on selected samples show that the films are essentially polycrystalline. Spectra are consistent with the expected spinel $NiFe_2O_4$ and rocksalt NiO structures.

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Atomic force microscopy images show granular growth of the films, with a typical grain size smaller than 100 nm. Surface roughness is about 1 nm for 20 nm thick single $NiFe_2O_4$ film. This roughness slightly increases for thicker films.

Hysteresis loops were recorded at 10 K in a SQUID magnetometer after cooling from room temperature under various fields, in order to study the influence of the cooling field on the exchange bias value. Training effect was also analyzed at 10 K by successively cycling the external field. The influence of the measuring temperature on the exchange bias field was studied by recording several hysteresis loops at various temperatures in the range 10 - 300 K.

3. Results

At room temperature, the as-deposited bilayers do not show any exchange bias, as expected since no field was applied during samples preparation. In order to align the AF layer in one direction, samples were cooled from 300 K to 10 K with an applied magnetic field. Hysteresis loops, measured after this procedure, are shifted on the field axis.

The variation of the exchange bias field upon the cooling field was analyzed on several samples with a 24 nm thick $NiFe_2O_4$ layer and the thickness of the NiO layer smaller than 40 nm. In this particular system we had shown that 40 nm is a critical thickness for the antiferromagnetic NiO layer [3]. Below 40 nm H_{eb} value increases with the NiO layer thickness and becomes independent of the thickness above 40 nm. This variation is presented in Fig. 1 where the exchange coupling energies, $\Delta\sigma$, calculated at 10 K from two successive hysteresis loops, are represented as a function of the AF layer thickness, t_{AF}. The exchange coupling energy is calculated with the relation $\Delta\sigma = H_{eb}M_St_F$, where M_S represents the saturation magnetization of the system and t_F is the thickness of the F layer. The decrease of the coupling energy after the first cycle shows the training effect in our system.

As the measuring temperature is increased, the exchange bias value decreases. At room temperature exchange bias vanishes in the samples having the NiO layer thickness smaller than 30 nm. The temperature at which exchange bias value becomes zero is called the blocking temperature. The decrease of the blocking temperature when the AF layer thickness decreases was already observed in other exchange biased systems [4], [5].

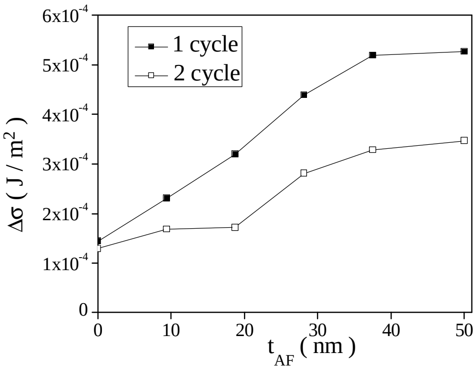
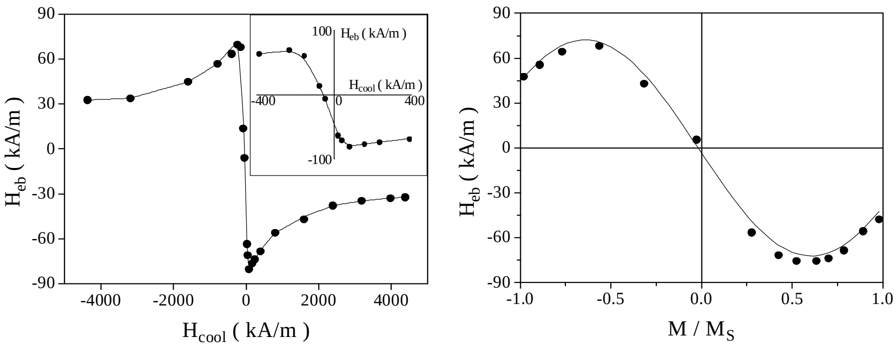


Fig. 1. Exchange coupling energy ($\Delta\sigma$) variation upon the NiO layer thickness measured at 10 K for the first and the second hysteresis cycles.

We have analyzed the influence of the cooling conditions upon the exchange bias in samples having the blocking temperature smaller than 300 K. The samples were annealed by cooling from 300 K to 10 K in different magnetic fields. At 10 K only one hysteresis loop was measured. The variation of H_{eb} upon the cooling field value is presented in Fig. 2a for a sample with a 20 nm thick AF layer. By difference with the generally observed independence of the exchange bias upon the cooling field value [6], in these samples H_{eb} strongly decreases when the cooling field value increases up to 4400 kA/m. Maximum H_{eb} value, ~80 kA/m, is obtained for the 80-160 kA/m cooling fields, while for the 4400 kA/m cooling field the hysteresis loop is shifted with only 30 kA/m.

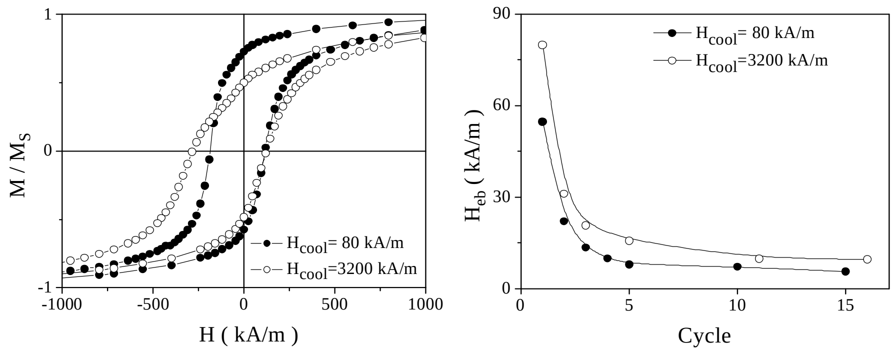
The inversion of the cooling field direction changes the hysteresis loop shift direction. The inset in fig 2a clearly shows that the change of the exchange bias sign is delayed compared with the cooling field inversion. At 0 A/m cooling field a ~50 kA/m exchange bias field is still observed. The change of the bias sign is obtained after applying -80 kA/m during the cooling procedure. This delay can be explained considering that the AF film does not interact directly with the magnetic field, but through the F layer. In fact, the external field orients the F moments which, by exchange interactions at the interface, aligns the AF moments in the direction of the field. The AF magnetic configuration is frozen during the cooling procedure, inducing the exchange anisotropy in the system. The exchange bias field is then directly correlated with the F magnetization value during the thermo-magnetic process, as already observed in other systems [7], [8].

In Fig. 2b is presented the variation of the exchange bias field upon the system's magnetization at 300 K, measured before cooling. H_{eb} variation is symmetric to zero value of the magnetization. Unexpectedly, the maximum of the bias is not obtained when thermo-magnetic treatment is applied in the saturated state of the system, but for a magnetization of only 0.6 M_S.



a) b)

Fig. 2. a) Exchange bias field variation upon the cooling field value for the sample having 20 nm thick NiO layer, measured at 10K. The inset of the picture shows a detail of the curve; b) Exchange bias field variation upon the sample's magnetization measured at 300 K, before starting the cooling field procedure. The solid lines are just a guide for the eyes.



a) b)

Fig 3. a) First hysteresis loops measured at 10 K after cooling in 80 kA/m and 3200 kA/m fields, from room temperature for a sample with 20 nm NiO layer thickness; b) H_{eb} variation upon the number of cycles for the same sample.

Comparing the hysteresis loops measured after cooling in a 80 kA/m field, that induces a magnetization of 0.5 M_s, and in a 3200 kA/m saturating field (Fig. 3a), for the sample with the NiO layer thickness of 20 nm, it can be observed that the increase of the cooling field value determines the decrease of the coercive field. In the same time, the loop's squareness increases when the cooling field value increases, indicating a better alignment of the NiO film. In order to better understand the exchange properties of the system, the stability of the bias on successive cycling of the applied field was verified. The training effect, measured after 80 kA/m and 3200 kA/m field-cool procedures, is presented in Fig. 3b. The bias established after alignment in 80 kA/m is very unstable, its value decreasing rapidly during the first 5 cycles, to stabilize after 10 hysteresis cycles at about 1/10 from the initial value. When the system is annealed in a stronger field, training effect is still observed, but bias seems to stabilize after only 5 cycles. Final exchange bias values are 9.5 kA/m and 7 kA/m, respectively. We can consider that, in spite of the high exchange bias field value observed after the thermo-magnetic treatment in 80 kA/m field, the exchange anisotropy is not well established.

4. Discussions

In Fig. 1 it can be observed that exchange bias appears even in samples with zero NiO layer thickness. This means that in the studied samples, besides the bias induced by the interfacial coupling between the NiO and the $NiFe_2O_4$ layer, there is a second contribution due to the $NiFe_2O_4$ material. The origin of exchange bias in the single $NiFe_2O_4$ films probably resides in the compositional non-uniformities. The unexpected strong variation of H_{eb} when the cooling field value increases seems to be related to these imperfections of our system.

Fig. 1 shows that for the samples having the NiO layer thickness smaller than 30 nm, the exchange bias value decreases to that of the single $NiFe_2O_4$ layer, after measuring one hysteresis cycle. This fast relaxation of the anisotropy generated by the coupling with the NiO layer indicates that the training effect is more probably related to the interfacial exchange coupling than to the intrinsic bias of the $NiFe_2O_4$ film.

In our experiment an important training effect is observed after cooling the system in small fields. Consequently, it can be supposed that in this case the high initial bias is essentially due to the coupling between the NiO and the $NiFe_2O_4$ layers. This anisotropy relaxes quickly with the reversal of the magnetization, so that the final exchange bias value is probably generated only by the $NiFe_2O_4$ layer. When bias is set after cooling the system in high fields, the coupling contribution decreases compared to that of the $NiFe_2O_4$ layer, leading to a higher stability of the bias. As for the previous discussed case, after several hysteresis cycles exchange bias induced by the coupling is strongly reduced, leading to a dominant contribution of the intrinsic $NiFe_2O_4$ bias. In Fig. 3.b. it can be observed that final H_{eb} values are not essentially different.

It is difficult to distinguish between the two contributions, but we suppose that the bias given by the ferrite layer increases with the cooling field value similarly with the magnetization, while the coupling related bias decreases in high cooling fields. This second variation is probably generated by the fact that in higher cooling fields NiO spins are obliged to align in the direction of the field. Because this direction differs from the local anisotropy directions, the system is not in an energetically favorable situation, and induces a smaller value of the bias.

5. Conclusions

$NiFe_2O_4$ / NiO bilayers exhibit a strong dependence of exchange bias upon the cooling field value. Maximum of exchange bias is obtained when samples are cooled in 80 kA/m field, but very important training effect is observed in this case. The increase of the cooling field value up to 4400 kA/m induces the decrease of the exchange bias field, but bias becomes more stable, indicating a better alignment of the NiO film.

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