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ONSET OF EXCHANGE BIAS IN NiO/$NiFe_2O_4$ BILAYERS

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Exchange coupling in $NiFe_2O_4$ / NiO bilayers is observed after cooling the system through the Néel temperature in the presence of a static magnetic field. To understand the coupling mechanism in more details, we have studied the differential susceptibility curves $\partial M/\partial H$ measured along the hysteresis loops for a set of samples with different NiO layer thicknesses. These curves are well described using lorentzian distribution functions, allowing us to extract both the center and the width of the switching field distribution for both branches of the hysteresis loops. Upon increasing the NiO layer thickness, the onset of exchange bias is associated with a significant increase of coercivity and the broadening of the switching field distribution. While the loops are strongly biased at low temperatures (the exchange bias field may be as strong as 2 kOe), the width of the switching field distribution is roughly the same for both branches of the hysteresis loops. Moreover, this width is weakly dependent upon cycling the magnetic field, whereas the exchange bias field is reduced.

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1. Introduction

Exchange anisotropy is observed in coupled antiferromagnetic (AF) - ferromagnetic (F) systems after cooling through the Néel temperature in the presence of a static magnetic field. The most interesting property of this phenomenon is the shift of the hysteresis loop along the field axis. This feature can be used as a tool to control the magnetization in such magnetic devices as spin valves [1] or tunnel junctions [2]. These applications triggered extensive studies of exchange anisotropy in the last decade, even though it was discovered more than four decades ago [3]. Among several open questions, the onset of exchange biasing when increasing the AF layer thickness is still poorly understood.

2. Experimental

Here we present results on $NiFe_2O_4$ / NiO bilayers. The AF nickel oxide NiO is widely used as an exchange bias layer in magnetoresistive devices because of its relatively high anisotropy ($K \sim 3\ 10^6$ erg/cm^3) [4] and high Néel temperature ($T_N = 523$ K). The nickel ferrite $NiFe_2O_4$ is ferrimagnetic. Its saturation magnetization is about $M_S = 300$ emu/cm^3 at low temperature.

Samples 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. The temperature was then reduced down to about 600 K for the NiO layer deposition, in order to reduce the diffusion through the interface. No magnetic field was applied during deposition.

X-ray diffraction spectra 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. 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 in a SQUID magnetometer. In order to obtain the exchange anisotropy, the samples were heated up to 700 K in the presence of a static magnetic field of 5 kOe, for only a few minutes.

3. Results

Fig. 1a shows hysteresis loops measured at 10 K for a single 12 nm thick nickel ferrite film (full circles), and for a bilayer with the same nickel ferrite thickness and a 18 nm thick NiO layer (open circles). The interface coupling between $NiFe_2O_4$ and NiO layers induces several effects upon the magnetic properties of the bilayer sample. Firstly, the hysteresis loop is shifted by about 1 kOe in the negative field direction. This shift is defined as the exchange bias field H_{eb}. Secondly, a significant increase of coercivity is also associated with the F/AF coupling. While the coercive field of the single layer is about 1 kOe, it increases up to 2.5 kOe for the bilayer. Besides, it appears clearly that the magnetization reversal takes place over a much broader field range compared to the single $NiFe_2O_4$ film. It is worthwhile to notice that the single layer's cycle is also biased by a field of ~0.6 kOe.

In order to study the magnetization reversal in more details, we analyzed the differential susceptibility curves $\partial M/\partial H$ measured along both branches of the hysteresis loops (Fig. 1b). $\partial M/\partial H$ curves are well described by lorentzian distributions functions. Indeed, best fits are obtained using two different lorentzian distributions. This appears clearly on the left part of Fig. 1b, corresponding to the derivative of the upper branch of the hysteresis loop. The solid line shows the best fit using the sum of two lorentzian functions, which are drawn separately using dashed lines. We found that the switching field distribution centered at low fields comes from a parasitic magnetic contribution from the NiO layer. Its origin is still unclear. We did not find any evidence for magnetic impurities within the NiO layer. More precise studies are needed to determine whether a minority phase could be present within the film. Note that the parasitic contribution is also observed on the lower branch of the hysteresis loop. However, it is partly hidden by its convolution with the main $NiFe_2O_4$ contribution.

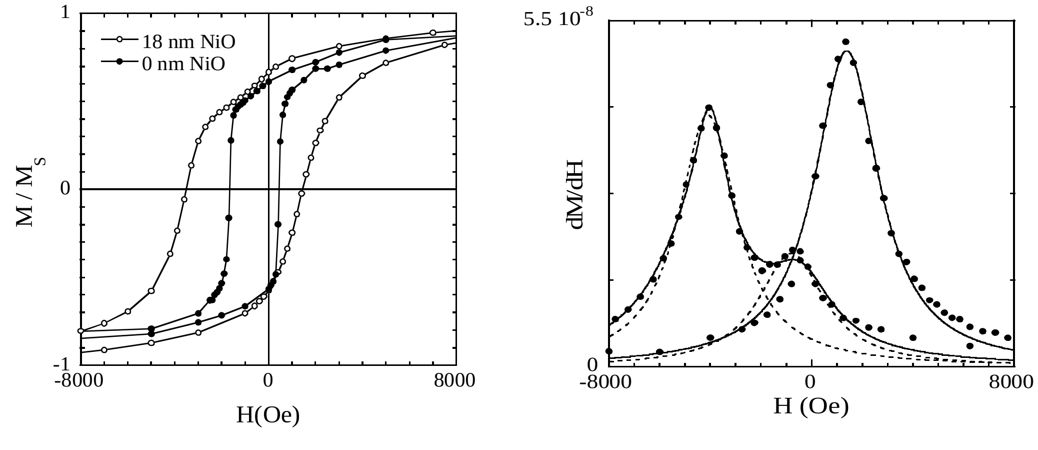

Fig. 1.a – biased hysteresis cycles for a single 12nm $NiFe_2O_4$ layer (filled circles) and a 12 nm $NiFe_2O_4$ / 18 nm NiO bilayer (open circles); b - $\partial M/\partial H$ curves obtained from the 12 nm $NiFe_2O_4$ / 18nm NiO hysteresis cycle

Exchange bias dependence upon NiO layer thickness was studied on a set of bilayers with 12 nm thick $NiFe_2O_4$ layer. As it was already reported [5], H_{eb} increases upon increasing NiO thickness and saturates for films thicker than about 40 nm [6].

Fig. 2 shows ∂M/∂H curves at 10 K for three samples with 0 nm, 9 nm and 18 nm thick NiO layer. Curves are normalized to their maximum for clarity. Upon increasing the NiO layer thickness, the onset of exchange bias is associated with an increase of coercivity and a significant broadening of the switching field distributions for both branches of the hysteresis loops. Along the upper branches of the hysteresis loops, the width (as determined from the lorentzian fits) increases from 75 Oe (single layer) up to 315 Oe (9 nm thick NiO) and 650 Oe (18 nm thick NiO). The same trend is observed along the lower branches of the hysteresis loops. The width increases from 75 Oe up to 425 Oe and 950 Oe upon increasing NiO thickness.

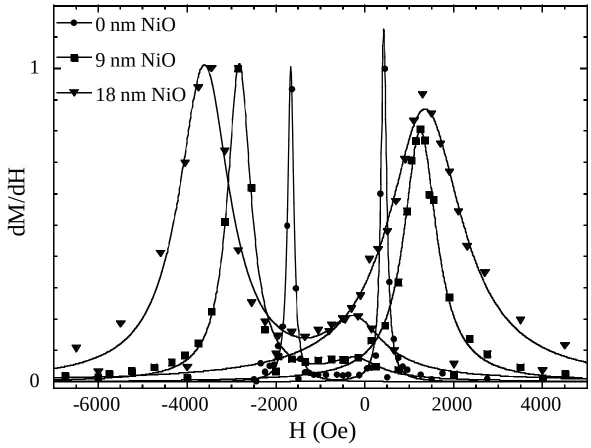

Fig. 2. ∂M/∂H curves for 12 nm NiFe$_2$O$_4$ single layer (circles), 12 nm NiFe$_2$O$_4$ / 9 nm NiO bilayer (squares) and 12 nm NiFe$_2$O$_4$ /18 nm NiO bilayer (triangles)

Exchange bias is strongly reduced upon cycling the external magnetic field. The strongest effect is observed after the first cycle. Further cycling still induces smaller changes before a stable loop is obtained, after about 10 cycles. ∂M/∂H curves are shown in Fig. 3 for the first three cycles, in the case of the 12 nm NiFe$_2$O$_4$ / 18 nm NiO sample. The width of the peaks is only weakly dependent upon cycling the magnetic field, whereas the exchange bias field is strongly reduced.

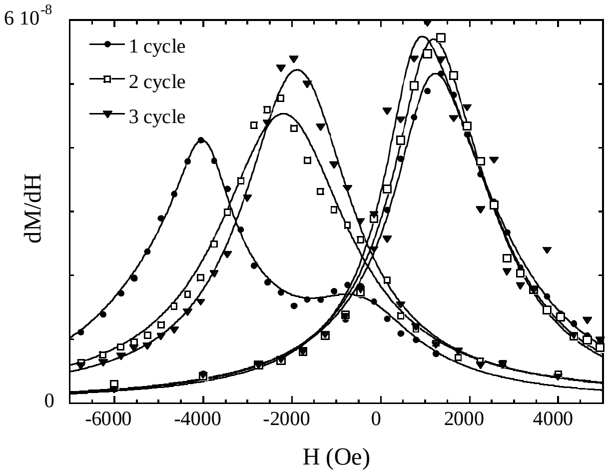

Fig. 3. Training effect in a 12 nm NiFe$_2$O$_4$ / 18 nm NiO bilayer

4. Discussion

Strong exchange biasing is observed in the NiFe$_2$O$_4$ / NiO bilayers at low temperature, indicating that there is significant exchange coupling at the interface between both layers, and that the NiO antiferromagnetic domains are well enough pinned. It is important to notice that the interface exchange coupling should remain roughly unchanged upon increasing the NiO layer thickness.

Indeed, structural changes at the interface are unlikely, since the NiO layer was grown on top of the Ni ferrite layer. Therefore, the main consequence of increasing the NiO layer thickness is the increase of the NiO anisotropy energy, which gets bigger, compared to the interface exchange energy. Indeed, the anisotropy energy per surface unit roughly scales as the NiO layer thickness. For thicker NiO films, NiO domain are better pinned, thus inducing stronger exchange bias. However, we also observe a strong increase of the samples coercivity with increasing NiO thickness, which implies on the contrary that NiO domains are not perfectly pinned. Perfect pinning would only induce biasing without any increase of the coercivity. This partial pinning is also confirmed by the significant training effect shown in Fig. 3.

The strong broadening of the switching field distribution is likely to be related to the distribution of NiO anisotropy axis. During the field-cooled process, AF spins are frozen along the external field direction, which is not generally the local anisotropy direction. Since exchange bias is strongly angular dependent, the local biasing may be distributed throughout the sample. This would result in a broadening of the switching field distribution, whose width should scale roughly with the average exchange bias field, as it is experimentally observed. However, this simple explanation does not account nor for the increase of the coercivity, neither for the negligible dependence of the switching field distribution width associated with the training effect. These results rather suggest that partial reorientation may occur within the NiO layer. As described above, the AF spins are frozen in a metastable configuration during the field cooling process. Upon $NiFe_2O_4$ magnetization reversal, FM spins may drag AF spins, allowing them to reach local energy minima. These reorientations may induce the increase of the effective anisotropy.

It seems to be a paradox to find an increase of both the exchange bias and the coercivity upon varying the AF layer thickness. Most results show on the contrary a decrease of the coercivity when the AF layer gets thick enough to allow the pinning of the AF domain and the onset of exchange bias [5]. Numerical calculations are in progress to help solving this apparent contradiction.

5. Conclusions

$NiFe_2O_4$ / NiO bilayers exhibit significant exchange biasing at low temperature. Exchange biasing strongly depends upon NiO thickness. Differential susceptibility curves $\partial M/\partial H$ measured along the hysteresis loops are well described using lorentzian distribution functions, allowing the determination of the center and the width of the switching field distribution. Upon increasing the NiO layer thickness, the onset of exchange bias is associated with a significant increase of coercivity and a strong broadening of the switching field distribution. The width of the switching field distribution is weakly dependent upon cycling the magnetic field, whereas the exchange bias field is reduced.

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