Eur. Phys. J. B **45**, 273–281 (2005)
DOI: 10.1140/epjb/e2005-00178-3

THE EUROPEAN PHYSICAL JOURNAL B

Exchange anisotropy in Co/NiO bilayers: time-dependent effects

D. Hrabovský[1], B. Diouf[1], L. Gabillet[2], A. Audouard[3], A.R. Fert[1], and J.F. Bobo[2,a]

[1] LPMC - INSA, 135 Avenue de Rangueil, 31077 Toulouse Cedex 4, France
[2] LNMH-CNRS, ONERA, 2 Avenue E. Belin, 31400 Toulouse, France
[3] LNCMP, 143 Avenue de Rangueil, 31432 Toulouse Cedex 4, France

Received 15 September 2004 / Received in final form 1st February 2005
Published online 28 June 2005 –

Abstract. The time dependence of the exchange anisotropy was studied in Co/NiO bilayers. In order to only observe the relaxation phenomena inside the antiferromagnetic (AF) layer and to eliminate the dynamic behaviour inside the ferromagnetic (F) layer, we have developed an experimental method where a small a.c. magnetic field is applied perpendicular to the main anisotropy axis. All data are obtained by magneto-optical (m.o.) experiments. We observe a logarithmic time dependence of H_{ud}, the exchange unidirectional anisotropy. We prove that the key parameter for the rate of relaxation is the anisotropy of the AF layer which depends strongly of the preparation method. We use the random field model as proposed by Malozemoff and suppose a breakdown of the AF interface into regular domains of size close to the crystallite size (10 nm width). If we further develop a Fulcomer and Charap relaxation model, we can propose from the distribution of relaxation times an analysis in terms of a spread of AF anisotropy energies. High magnetic pulsed field experiments (55 T) complete the experimental study and the results are analysed assuming that the Zeeman energy balances the anisotropy energy of the AF domains and switches them into the opposite direction.

PACS. 75.50.Ee Antiferromagnetics – 75.60.-d Magnetic domain effects, magnetization curves, and hysteresis – 75.70.-i Magnetic properties of thin films, surfaces, and interfaces

1 Introduction

The phenomenon of exchange bias was discovered more than fifty years ago by Meiklejohn and Bean [1]. They observed that fine Co particles, oxidized at their surface, exhibit a shift of the magnetic hysteresis along the field axis, the center of the hysteresis loop being displaced to $H \neq 0$, as though there were an internal exchange field H_{ex}. In recent years, numerous investigations of this effect have been carried out on ferromagnetic (F)-antiferromagnetic (AF) bilayer systems. The hysteresis loop displacement is the most characteristic feature of exchange bias, but also an enhancement of coercivity is often observed. The interfacial exchange coupling gives an efficient method for technical applications like pinning or hardening of some ferromagnetic layer magnetization. Several groups have used FeMn antiferromagnetic films as an exchange biasing layer to study their spin-valve-like response for magnetic sensor applications [2–5]. One can also use NiO as an antiferromagnet, as already reported by several groups [6–10]. Compared to intermetallic antiferromagnets like FeMn, NiO is an insulator with a good resistance to corrosion and a relatively high blocking temperature T_B (the Néel temperature for bulk NiO is 525 K, but clearly lower for thin films). This biasing effect is the consequence of exchange coupling across the AF/F interface. This phenomenon is particularly observed when an external field is applied while cooling the ferromagnetic layer through T_N, the Néel temperature of the AF layer. The role of the external magnetic field is subject of discussion. Despite a great variety of works on this subject, the exact mechanism of exchange bias is still controversial, mainly because the observation of the interfacial magnetic structure is difficult. Meiklejohn and Bean developed a simple model to explain the loop shift when ferromagnetic particles are embedded in an AF. The model considers single domain F particle with uniaxial anisotropy $-K_{ua}\cos^2\theta$ (θ is the angle between the magnetization MF and the uniaxial axis) and subject to an unidirectional field H_{ud} along the same axis which transfers the exchange coupling to a rigid AF. In the case of an ideal F/AF bilayer system, we suppose an uncompensated interface where only one type of AF sublattice spins is present. In a Stoner and Wohlfarth description the switching fields H_{c1} and H_{c2} (in positive and negative direction along the anisotropy axis) do not have the same value because of the additional contribution of the interface coupling and the hysteresis loop is displaced by an amount $H_{bias} = 1/2(H_{c1} + H_{c2}) = -H_{ud}$. The bias field estimated in this simple model where the interface is

[a] e-mail: jfbobo@onecert.fr

uncompensated is, however, two orders of magnitude larger than experimental values. Mauri [11] also assumes a non compensated interface but the AF spins at the interface reverse simultaneously with the F spins when the F magnetization layer is reversed. In fact he supposes that a planar domain wall parallel to the interface is formed inside the AF layer. Malozemoff [12] supposes that roughness at the interface induces a random exchange field between the two layers, much as in the Imry-Ma random field problem. A minimization of energy argument suggests to break up the AF into magnetic domains (lateral size L and depth $h = L/2$) due to random fields. Writing the local unidirectional exchange interface energy (expressed in erg/cm^2) in the form $\sigma_l = \pm z J_i/a^2$ (J_i is the individual exchange constant at the interface and the model assumes a simple cubic structure with lattice parameter a and z the coordination number) in the random field problem, the local interface energy will be random and its average value in a region of area L_2 will go down statistically as $\sigma = \sigma_l/\sqrt{N}$ where $N = L^2/a^2$ the number of sites projected onto the interface plane.

This model yields a result very similar to the one of Mauri and explains the order of magnitude of the bias field observed in AF-F interfaces. But in the Mauri model the AF domain wall is parallel to the interface, it is created for one direction of the applied magnetic field and disappears for the opposite direction, exactly as when you exert a rotation strain on a mechanical sample, an elastic stress (a twist) appears to absorb the strain. This dynamic aspect is very interesting. On the contrary, in the Malozemoff model, the AF magnetization breaks down into numerous domains at the interface, most of them being oriented in the same direction as the F magnetization. These domains are a priori static during the reversal of the F layer magnetization, but when the F magnetization remains in reverse position, the AF domains are submitted to a torque that tends to reverse their magnetization. This phenomenon is thermally activated and a relaxation of the exchange anisotropy could be observed, and this will be the presently studied phenomena in Co/NiO bilayers. These last years some experimental studies have demonstrated that the exchange field at the AF/ F interface is time dependent. Several authors have observed a slight decrease or increase in time of the bias depending of the history and final state of the F magnetization. We have observed, in particular when studying magnetic properties of NiO/ Co bilayers, that if we do not pay attention to restore the sample with the same final Co magnetization state the bias is strongly modified a couple of days or a week after. For the two states accessible in zero magnetic field and corresponding to the Co magnetization in the same (forward) or in the opposite (reverse) direction to the bias field, the after-effects are strongly different.

In the present paper, we describe the preparation method of samples Glass/NiO/Co and the experimental procedure based on a small a.c. magnetic field applied perpendicular to the easy anisotropy axis, the measurements being done by magneto optical methods. We develop a simple model based on the relaxation of interfacial AF magnetic domains to analyze the observed after effects.

2 Experimental procedure

We have studied various types of Co/NiO samples corresponding to different preparation methods that we have described in detail in a previous paper [13] where we find out conditions for obtaining strong uniaxial anisotropy in the Co layers grown on nickel oxide. NiO was deposited using an RF facing target magnetron, the standard DC magnetron method being used for the Co layer. For deposition of NiO layer, with a constant 1250 Å thickness, two distinct processes were adopted. For the first one the substrate was in a fixed position (FIX), by this way we obtained a well-defined anisotropy in the AF layer, due to oblique incidence deposition, and a subsequently anisotropy in the Co layer deposited on it. We also prepared samples with desired anisotropy by depositing NiO with a rotation of the sample holder, in order to avoid oblique incidence deposition, but the sample holder was equipped with two permanent magnets (MAG) generating an applied magnetic field of 1 kOe. In this last case the morphology of the interface is smoother. Magnetic tunnel junctions [13] of NiO/Co/Al_2O_3/Co type made with both processes for the bottom electrode show sharp magnetoresistive responses and yield more than 20% of tunneling magneto-resistance (TMR) at room temperature along the easy axis. We will present here the results observed with three samples FIX60, FIX80, and MAG50 (the sample name indicates the type of growth process and the Co layer thickness in Angstrom unit) selected from many others. The whole magnetic study was realized by magneto optical (m.o.) measurements. To measure the Kerr rotation we used a balanced optical bridge composed of a Wollaston prism to divide the reflected light at the sample surface into two orthogonal polarized beams. Their intensities are detected by two matched Si photodiodes. The differential signal is converted using a 16 bit analog-digital converter. A linearly polarized He-Ne laser was used as a probe. The sample is placed in the center of a copper coil and an accurate current supply generates a magnetic field at its center.

First results on the value of the exchange anisotropy were obtained by classical hysteresis loops measurements. The applied magnetic field was along the easy magnetization axis of the Co film, and the observed Kerr rotation was proportional to the Co longitudinal magnetization (component parallel to the field and to the plane of incidence of the light). For these measurements the applied magnetic field was periodic with a 50 Hz frequency and an amplitude up to 1 kOe, but in order to avoid the warming of the coil, the computer limited the number of periods to six and evaluated the hysteresis loop data during the last period of the magnetic field. Moreover it stopped the magnetic field to manage the final direction of the Co magnetization always in the same direction (positive or negative). At the beginning of the experiments the sample was set up and the Co layer magnetized in the

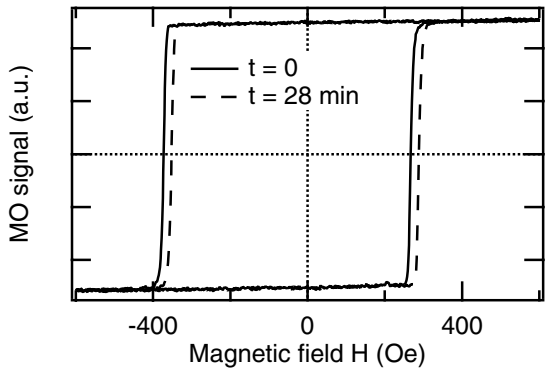

Fig. 1. Hysteresis loops obtained at $t = 0$ and $t = 28$ min after reversal of the Co magnetisation at $t = 0$.

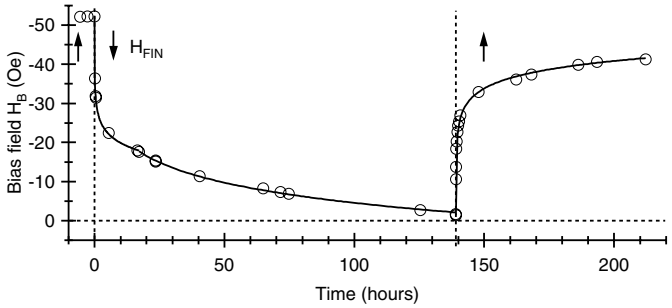

Fig. 2. Time dependence of the bias field HB after reversal of the Co magnetisation (at $t = 0$ and $t = 140$ sec). The arrows indicate the direction of the Co magnetisation.

positive direction, we measured successively the hysteresis loop measurements at regular time intervals and verified that the value of the bias remains constant. After this test and in order to study the relaxation of the bias, we applied, at $t = 0$, the magnetic field in order that the final state of the Co magnetization was reversed (in the negative direction) and we carried out successive hysteresis loop measurements. Figure 1 presents the hysteresis loops for $t = 0$ and $t = 28$ minutes for sample FIX60 deposited without rotation of the substrate. Note that the bias field has relaxed from a value close to 56 Oe to 37 Oe. This kind of experiment is repeated and the time dependence of the bias is shown in Figure 2. We observe a continuous decrease of the bias when the Co magnetization is in a reversed position (the bias is close to zero after 6 days) and an increase of the bias if the Co magnetization is switched back to the first positive direction.

The analysis of the hysteresis loop was done using the classical model (Meiklejohn and Bean - *M.B.*) written in the case where a F layer is deposited on an AF layer with an uncompensated interface. Taking into account a ferromagnetic exchange interaction between F and AF spins, the energy of this configuration could be written in the form

$$E = -H_a M_F t_F \cos\theta - K_{ua} t_F \cos^2\theta - J_{int} \cos\theta \qquad (1)$$

where M_F is the F magnetization by unit volume (emu/cm^3), H_a is the applied magnetic field supposed along the easy axis, K_{ua} is the uniaxial anisotropy by unit volume (erg/cm^3), t_F is the thickness of the F layer, J_{int} is the exchange interaction energy at the interface by unit surface (erg/cm^2). In a Stoner-Wohlfarth (S.W.) model description, the switching fields H_{c1} (forward) and H_{c2} (reverse), derived by considering the stability of states $\theta = 0$ and $\theta = \pi$ are

$$H_{c1} = -(2\,K_{ua} t_F + J_{int})/M_F\,t_F$$
$$H_{c2} = (2\,K_{ua} t_F - J_{int})/M_F\,t_F.$$

We could introduce, as was done by M.B., an uniaxial field $H_{ua} = 2K_{ua}/M_F$ and an unidirectional field $H_{ud} = J_{int}/M_F t_F$ which translates the effect of the interface coupling into $H_{c1} = -H_{ua} - H_{ud}$ and $H_{c2} = H_{ua} - H_{ud}$.

Due to the contribution of the interface exchange coupling, the two switching fields are not equal and the hysteresis loop is displaced by an amount $-H_{ud} = -J_{int}/M_F t_F$ which appear inversely proportional to the F layer thickness as expected for an interfacial effect. From hysteresis loop measurements the exchange anisotropy (uniaxial and unidirectional parts) is deduced from the two coercive fields H_{c1} and H_{c2}. Moreover the coercive fields depend strongly on the rate at which the applied field is varied. Many phenomena must be taken into account like nucleation or propagation processes and thermal activation, and a S.W. model is generally not convenient for describing the reversal process. Moreover the hysteresis loops depend on dynamic effects (magnetic viscosity) in the F layer resulting from the rate of change of the applied field. In order to observe only the relaxation phenomena inside the AF layer, the dynamic behavior inside the F layer must be eliminated. For these reasons we have developed an experimental method close to that proposed by Krivotorov [14], probably being more efficient because the magnetic state of the F layer is unchanged during exchange anisotropy relaxation measurements. In the Krivotorov's experiments a small a.c. magnetic field is applied perpendicularly to the main easy anisotropy axis in the plane of the Co/CoO sample and by help of a technique based on the anisotropic magnetoresistance effect (AMR) they obtain the a.c. response of the Co magnetization whose amplitude depends on the exchange anisotropy field $H_{ex} = H_{ua} + H_{ud}$. We have developed an equivalent method based on magneto optical (m.o.) measurements. The anisotropy axis of the sample is oriented perpendicularly to the incidence-reflection plane of the laser beam (see Fig. 3), the a.c. magnetic field (amplitude 10 Oe, frequency 60 Hz) is applied in this plane and in the plane of the sample, by this way the longitudinal Kerr effect (obtained by help of a numerical lock-in amplifier) gives directly the component of the magnetization along the applied a.c. magnetic field (perpendicular to the easy magnetization axis). In our measurements the magnetization of the Co layer is locked in one of the two stable positions corresponding to $\theta = 0$ or π and we observe a small a.c. Co magnetization component resulting from the a.c. applied field. From the amplitude of this component we could deduce the exchange anisotropy field and study its time dependence. With this method, we separate the time

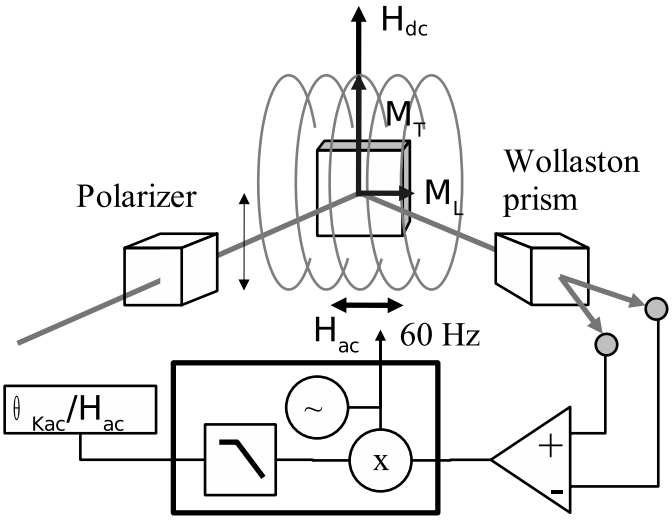

Fig. 3. Experimental configuration for the a.c. perturbation method: position of sample, of the laser beam and of the two applied magnetic field components H_{dc} (static) and $H_{a.c.}$ (alternate).

dependent effects in the AF layer from dynamic effects in the F layer. Using the expression (1), completed by the term $-H_{ac}\ M_F t_F \sin\theta$, we could deduce from the measurements of M_{ac} the value of H_{ex} by the simplified formula (we suppose small angles and a zero dc magnetic field)

$$M_{ac} = M_F H_{ac}/(H_{ua} + H_{ud}) \qquad (2)$$

analogous to expression (2) in Krivorotov's paper. The smaller important is the exchange field, the smallest is the amplitude of transversal magnetization oscillations. For larger amplitudes of oscillation a numerical calculus replaces this simplified formula.

3 Results

Before presenting the direct relaxation study of the exchange anisotropy field H_{ex}, we have realized a series of preliminary measurements in order to test the a.c. method chosen to obtain H_{ua} and H_{ud}. They are to carry carry out, by m.o. measurements, the amplitude of the transversal a.c. magnetization component M_{ac} and using expression (2) we deduce H_{ex}. Figures 4 and 5 present the exchange field versus time for the sample FIX60. The sample was prepared with the Co magnetization saturated in the forward direction, H_{ua} and H_{ud} are in the same direction corresponding to the more stable state of the AF layer and $H_{ex} = H_{ua} + H_{ud} = 941$ Oe. At $t = 65$ seconds (see Fig. 4), we switch the Co magnetization in the reverse direction, the interface coupling changes its sign and the exchange field becomes $H_{ex} = H_{ua} - H_{ud} = 777$ Oe (we have supposed that H_{ud} was not modified during the short time of the Co reversal). By this simple way we can deduce both values of the uniaxial and unidirectional field $H_{ua} = 859$ Oe and $H_{ud} = 82$ Oe before any relaxation

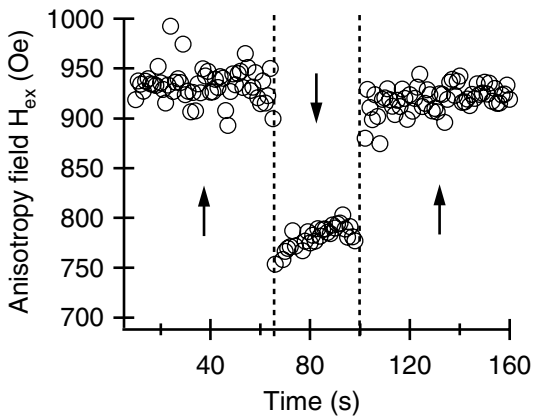

Fig. 4. Exchange field H_{ex} vs. time for sample FIX60. The arrows indicate the direction of the Co magnetisation.

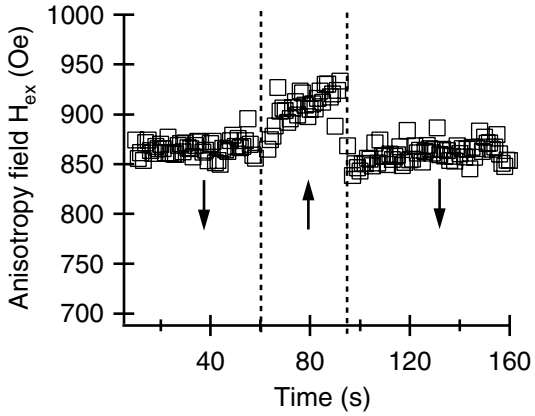

Fig. 5. Same experiment as in the Figure 4, but with opposite initial cobalt magnetization.

phenomenon. These values of H_{ua} and H_{ud} can be compared to those obtained, before any relaxation, by the classical hysteresis cycle method (see Fig. 2). We deduce from the shift of the hysteresis loop a slightly different value of the unidirectional field, $H_{ud} \approx 50$ Oe. As explained above, the hysteresis loop method supposes that the S.W. model is appropriated to describe the switching of the Co magnetization, however, this is not the case.

Moreover, for $65 < t < 100$ seconds, we observe in Figure 4 the onset of the relaxation of H_{ud}. At $t = 100$ seconds, switching back the Co magnetization to the positive direction, we observe $H_{ex} = 930$ Oe. Considering that the uniaxial anisotropy has a constant value, we could deduce the unidirectional field $H_{ud}(100) = 71$ Oe value obtained after the relaxation of the AF domains during 35 seconds. The second preliminary study concerns the same sample, but 3 days after that the Co layer was saturated in the reverse direction. We consider that H_{ua} has not changed and that H_{ud} has relaxed during this time towards a value corresponding to some more complex and stable state of the NiO layer obtained after three days of reverse orientation of Co. Measurements presented Figure 5 show that $H_{ex} \approx 869$ Oe. When reversing at $t = 60$ s the Co magnetization in the forward direction, a very small change

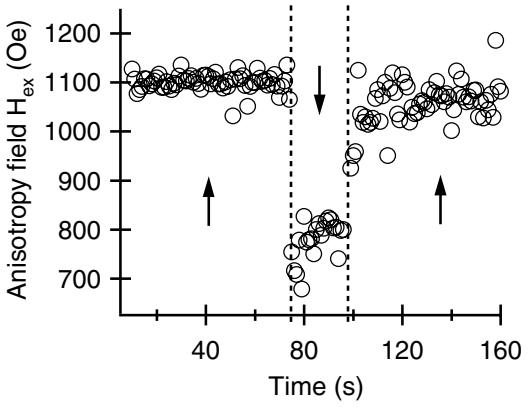

Fig. 6. Exchange field H_{ex} vs. time for sample MAG50. The arrows indicate the direction of the Co magnetisation.

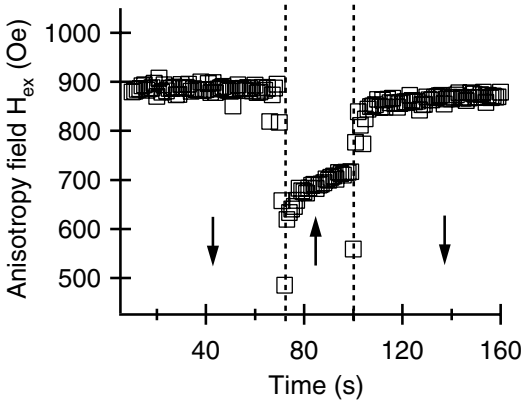

Fig. 7. Same experiment as in Figure 6, but with opposite initial cobalt magnetization. Contrary to sample FIX60 (Fig. 5), it could be observed that the unidirectional field H_{ud} has changed its sign after several days with a reverse Co magnetisation, proving a larger relaxation rate of the bias field.

of the exchange field is observed. As previously, we can deduce both values of the uniaxial and unidirectional field $H_{ua} \approx 875$ Oe and $H_{ud} \approx 6$ Oe. We observed that H_{ua} has kept the same value (the discrepancy between 875 and 859 Oe is not significant) but the unidirectional field has relaxed during the three days to some value close to zero. Moreover, we observed for $60 < t < 95$ s , when the Co is back in a forward position, that H_{ud} begins to increase to recover a positive value.

The same preliminary study was realized with sample MAG50 corresponding to a lower anisotropy of the antiferromagnetic layer. Results are presented in Figures 6 and 7. We observed that, after several days with a reverse magnetization of the Co layer, the unidirectional field H_{ud} has changed its sign which proves that the NiO magnetization was partially reversed.

After these preliminary studies, we have investigated the relaxation of $H_{ud}(t)$ following the experimental protocol:

1. saturation of Co in the positive direction during several days to recover the magnetic state S1 observed after the preparation of the samples,

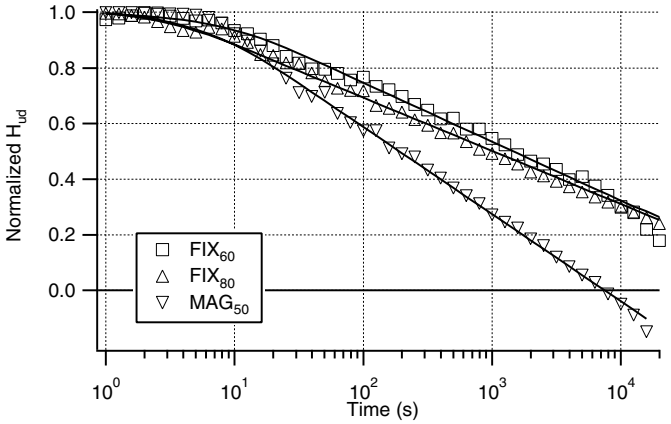

Fig. 8. Comparison of normalised unidirectional field H_{ud} vs. time for samples FIX60 FIX80 and MAG50. Solid lines are fits to equation (3).

2. reversal of the Co magnetization in the negative direction at $t = 0$,
3. measurements of $M_{ac}(t)$ for $t > 0$, $H_{ud}(t)$ being deduced using expression (2).

In Figure 8 we present the results obtained for the three samples FIX60, FIX80 and MAG50. In order to compare the relaxation process for the different samples, $H_{ud}(t)$ was normalized by the value of $H_{ud}(0)$. We observe that H_{ud} is roughly constant during the first instants of measurements $0 < t < t_1$ (t_1 is sample-dependent but is always of the order of magnitude of ten seconds). For $t_1 < t < t_{exp}$ we observe a logarithmic time dependence of H_{ud} that could be written as $H_{ud}(t) - H_{ud}(t_1) = -K \text{ Log } t/t_1$. The time t_{exp} corresponds to the end of the M_{ac} acquisition measurements. Clearly several experiments were done with a greater t_{exp} (for example 12 hours) and seems to prove a logarithmic time dependence for larger time range but the signal to noise ratio of dH/dt becomes smaller and smaller and prevents a precis analysis of the time dependence of H_{ud} for longer times. The various parameters characteristic of the samples FIX60, FIX80 and MAG50 are summarized in Table 1. For each of them, the values of the relaxation speed K, uniaxial and unidirectional fields H_{ua} and H_{ud} are presented for the relaxation from the state S1 which is the quasi stable state obtained after preparation of the samples. Table 1 also shows the main parameters deduced from a next experimental step where the relaxation is studied from an intermediate state, called S2 for simplification, carried out several days after reversal of the Co magnetization. The relaxation is then studied when reversing one more time the Co magnetization in the positive direction, the system coming back to state S1. We observe a value of the slope K larger in the case of NiO deposited with rotation of the substrate, this sample corresponding to lower anisotropy of the NiO layer. The values of H_{ud} are close to zero after 2 hours of measurements, the unidirectional field $H_{ud}(t)$ changes its sign and the reversal of a large part of the AF domain magnetization is achieved. On the contrary, for a sample without rotation of the substrate, the unidirectional field $H_{ud}(t)$ is

Table 1. Table of parameters H_{ua} and H_{ud} resulting of the preliminary studies (short reversal).

sample	state	H_{ua} (Oe)	H_{ud} (Oe)	K (Oe/dec)	K_{norm}
FIX60	S1	865	86	18.7	0.22
	S2	832	4.7	–34.2	
FIX80	S1	515	53	10.4	0.20
	S2	503	–11	–19.8	
MAG50	S1	851	160	51.0	0.32
	S2	881	–116	–98.0	

close to zero only after several days. These results demonstrate that the behavior of $H_{ud}(t)$ depends strongly on the anisotropy of the AF layer. As already discussed, a difficult experimental problem is met when analyzing the behavior of $H_{ud}(t)$ for a very long time (a couple of days or a couple of weeks) after the reversal of the Co magnetization. Moreover most of our tentatives to obtain the final value of H_{ud} seem to indicate that it is obvious to consider that the final AF state is symmetric with respect to the state at $t = 0$ before reversal. Clearly the time dependence of the exchange field results from the relaxation of the AF domains dragged by the reversal of F magnetization and excited by thermal activation. The initial state can be considered as a quasi stable state after the sample preparation process. When switching the Co magnetization a torque appears due to F coupling which tends to reverse progressively the various AF domains. But taking into account numerous crystallites of different size due to the sputtering technique used for preparation but also the roughness of the interface we have to consider that the various intermediate states of AF layer are very complex and numerous, similarly as in frustrated spin systems such as spin glasses, and it is difficult to suppose an exact symmetry of the system even after a very long time.

4 Analysis

As proposed by Malozemoff, and due to random field created by roughness, we assume the formation of regular AF domains, at the interface. Let us justify, in our case, this Malozemoff approach. Miltényi et al. [21] have proposed that the exchange bias is due to magnetic domains in the volume of the AF layer. This hypothesis is asserted by an experimental study on systems AF/F with diluted AF. In this case, the number of domains depends of the defect density inside the bulk AF layer, each defect being a pinning center for a domain wall (and not on disorder or defects at the interface). Results obtained by A. Mougin et al. [22] on irradiation effect on exchange bias systems are consistent with the Miltényi model. But in our case the structure of magnetic domains is linked to the preparation process of the NiO layer, and we have shown [13] that for non rotating substrate, the inclined deposition induce columnar growth with 10 to 15 nm wide grains (the grains crossing the whole AF layer with a rather constant dimension). We consider that the AF domains size is correlated to the crystallite size and the problem of exchange bias differs, in our case, from the phenomena observed in diluted AF. We suppose, as in the Malozemoff model, that the magnetic configuration in the bulk AF layer (125 nm thick) is fixed when the Co is reversed. The torque exerted by the reversal of the F magnetisation involve only the drag of an interfacial part of the AF domains.

With this hypothesis, each domain presents a weak net magnetic moment resulting from the non total compensation of the spins. As a consequence of the preparation method (cooling with an applied magnetic field) most of these magnetic moments are oriented along the easy anisotropy axis and are ferromagnetically coupled with the adjacent Co layer magnetization. It induces a bias field and the hysteresis cycle is not centered at $H = 0$. When the Co magnetization is reversed (due to negative applied magnetic field) most of the AF domains are submitted to a torque that tends to reverse their magnetization in the same direction as the Co layer. This phenomenon is thermally activated and explains the after-effects observed concerning the bias field. Following the presentation of Fulcomer and Charap [15] we consider that the AF domains are identical and non-interacting and that they are exchange coupled to the magnetization of the Co film. The total energy E_{AF} for one of these identical domains can be written:

$$E_{AF} = K_a^{AF}\ V\ \sin^2\gamma + J \times S\ \cos\gamma$$

where γ is the angle between the AF moment and the common AF and F easy axis, K_a^{AF} is the unit volume anisotropy of the AF layer, V is the AF domain volume, S is the contact area between AF domain and Co film, J is the unit area interface exchange coupling constant (in the paper of Malozemoff J is called σ) and J results from the incomplete compensation of the AF moments at the interface and can be written as inversely proportional to $\sqrt{N}$, where N is the number of interface spins in an AF domain. For $JS < 2\ K_a^{AF}\, V$, there are two equilibrium states for the AF domain magnetization which are $\gamma = 0$ (state 1) and $\gamma = \pi$ (state 2), separated by a maximum of the total energy equal to

$$E_{max} = K_a^{AF}\, V \left[1 + \left(JS/2\ K_a^{AF}\, V\right)^2\right].$$

The switching of the AF domain magnetization above E_{max} is activated by thermal fluctuations. The barrier energies are of distinct amplitudes when going from state 1 to state 2 and inversely

$$\Delta E\pm = K_a^{AF}\, V \left[1 \pm JS/2\ K_a^{AF}\, V\right]^2.$$

The probability to overcome the barrier is thermally activated and consistent with an Arrhenius law $\tau = \tau_0 \exp\left(+\Delta E/kT\right)$ where T is the temperature of the experiment, ΔE the barrier energy and τ_0 being a characteristic attempt time for spin reversal. ΔE being stronger when going from state 2 to state 1, the AF domain magnetization approaches a new equilibrium corresponding to

Table 2. Table of parameters H_{ua} and H_{ud} resulting of the preliminary studies (short reversal).

sample	τ_{min} (s)	τ_{max} (s)	K (Oe/dec)
FIX60	12.1	2.28×10^{10}	18.7
FIX80	4.4	1.55×10^{11}	10.4
MAG50	8.7	2.33×10^{7}	51.0

AF moments in the same direction as the Co magnetization. The two process 1→2 and 2→1 are characterized by two relaxation times τ_1 and τ_2 and finally the total time constant τ will be given by the expression

$$1/\tau = 1/\tau_1 + 1/\tau_2 = 1/\tau_0[\exp(E_1 - E_{max})/kT + \exp(E_2 - E_{max})/kT].$$

The unidirectional field (or bias field) evolves in time according to

$$H_{ud}(t) - H_{ud}(\infty) = [H_{ud}(0) - H_{ud}(\infty)] \exp(-t/\tau)$$

where $H_{ud}(0)$ and $H_{ud}(\infty)$ are the initial and final fields. As often in similar systems an appropriate size, volume or energy distribution leads to a logarithmic time dependence. The unidirectional magnetic field can be then expressed in the form:

$$\begin{aligned} H_{ud}(t) - H_{ud}(t_1) &= [H_{ud}(0) - H_{ud}(t_1)] \\ &\quad \times \int_{\tau_{min}}^{\tau_{max}} \rho(\tau) \exp -t/\tau d\tau \\ &= S \log t/t_1 \end{aligned} \quad (3)$$

where S is the viscosity characteristic of the relaxation rate. The time dependence of H_{ud} observed experimentally in various cases (see Fig. 8) is well described by a logarithmic function of time. These results are in agreement with the existence of a distribution of domain sizes or energies. The parameters τ_{max} and τ_{min}, determining the range of time constant distribution, can be obtained by fitting the experimental $H_{ud}(t)$ to the above equation under the assumption $\rho(\tau) = 1/\tau$ and are presented in Table 2 [16].

5 Discussion

The Fulcomer and Charap model [15] can be developed or applied to our system when assuming some hypothesis. The main one concerns the size of the AF domains and this point will be discussed at first. In a second part, taking into account the distribution of τ (relaxation characteristic time) which results from the experimental study, we try to deduce most of the characteristics of the NiO/Co systems. As in most of the models, the ferromagnetic layer is regarded as a single domain. This assertion is not obvious because the F domain size, during magnetization reversal, has been observed to be much smaller in an exchange biased bilayer than in a single F layer. Nevertheless we have observed for systems Glass/ NiO/ Co [17] F domains of sufficient large size (in the range 1 μm – 1 mm) compared to the size of the AF domains to consider the F layer as single domain. Moreover we consider that the domain size of NiO may be strongly correlated to the crystallite size. By HREM we have noticed regular columnar grains of 10 to 15 nm wide. The NiO films are in our experiments 125 nm thick, but taking into account the work of Malozemoff the reversal does not concern the whole AF film, only a thickness of order of 5 nm (the half width of the domains) is involved. Consequently the mean AF domain volume could be estimated to $V = 0.5 \times 10^{-18}$ cm^3. This small size of the AF domains is confirmed by the work of Wang et al. [18]. When they pattern in micronic dots a NiO/NiFe bilayer, if the NiO film is epitaxially deposited the exchange field increases when reducing the pattern size proving that, in this case, the AF domains are larger than one micron. On the contrary, the exchange field of patterned polycrystalline NiO on Si does not depend of the pattern size, demonstrating a considerably smaller AF domain size, close to the grain size. The order of magnitude of the AF domain size can be also approached considering that the domains tend to contract, as explained by Malozemoff, until the size equals the wall width, whose classical expression is $\delta = \pi\sqrt{A_{AF}/K_a^{AF}}$. Taking into account the absolute values of the exchange constants $A_1 = 1.6 \times 10^{-7}$ erg/cm between first neighbours, $A_2 = 6.7 \times 10^{-7}$ erg/cm between second next neighbours and a mean anisotropy constant $K_a^{AF} = 3.3 \times 10^6$ erg/cm^3, we obtain the value $\delta = 14$ nm. This value is consistent with the above mentioned order of magnitude obtained when considering that the domain is similar to the grain size (10 to 15 nm). Nevertheless the breakdown of the AF layer, close to the interface, into small conventional magnetic domains is clearly a strong simplification of the physical reality. Due to interface roughness there is a strong magnetic frustration in the AF layer leading to magnetic disorder at the interface as in any system with random anisotropy. The after effects reported above, when we reverse the Co magnetization, is characteristic of frustrated spin system. Complex transitions between metastable states are thermally activated and the simple model of regular AF domains at the interface is a rough approximation. If one further allows that the interface exchange coupling constant J between the Co layer and a perfectly uncompensated AF plane is close to the value $J_0 = 10$ erg/cm^2 [19], one can expect, as in the Imry-Ma random field problem, that the effective coupling energy per unit area would go down statistically as $J = J_0/\sqrt{N}$ where N is the number of Ni magnetic sites at the interface between the AF domain and the Co layer. Taking into account the mean surface of each AF domain at the NiO/Co interface we can deduce an averaged coupling energy associated with an AF domain surface: $J_S = 0.2 \times 10^{-12}$ erg.

We now discuss the experimental results in terms of relaxation time range. As shown in Figure 8, the typical relaxation time range characteristic of the decrease of $H_{ud}(t)$ is roughly equal to $1 < \tau < 1000$ s. Taking into account $\tau_0 = 10^{-9}$ s, and $JS = 0.2 \times 10^{-12}$ erg, assuming that

all the AF domains are equal in size and a distribution of anisotropy energy, we can deduce the range of anisotropy energies $0.72 \times 10^6 < K_a^{AF} < 0.84 \times 10^6$ erg/cm^3. These values are completely in the order of magnitude that we would expect taking into account that K_a^{AF} (associated to magnetocrystalline anisotropy) of NiO is at maximum equal to 3.3×10^6 erg/cm^3. Note that we have not observed a possible texture in the NiO film and that consequently the easy (111) planes are randomly distributed. We recall that the anisotropy is in our case, and for non rotating substrate, the result of oblique incidence deposition. In case of a rotating substrate the anisotropy results from magnetoelastic effects due to deposition under 1 kOe applied magnetic field. When comparing these results with results obtained by Dubourg [17] in the case of a purely rotating substrate we observe that he obtained a value roughly ten times smaller $K_a^{AF} = 0.1 \times 10^6$ erg/cm^3.

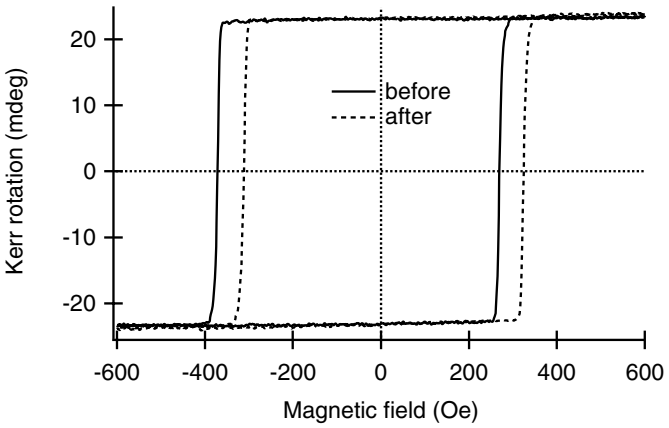

Fig. 9. Comparison of hysteresis loops for sample FIX60, before and after a reverse pulsed field.

6 High magnetic field experiments results and analyze

These experiments were done subsequently to our recent results [20] for various exchange-biased systems exposed to pulsed magnetic fields. A 55 T magnetic field pulse was applied on the same series of samples in two directions, namely along the exchange unidirectional field and opposite to it. In the first case, and for sample FIX60, no change in exchange bias was detected after the pulse, its value remaining 56 Oe (as observed by hysteresis cycle measurements). In the second case, where the 55 T pulsed field reverses the Co magnetization, we observed, as shown in Figure 9, an immediate bias field decrease to zero, the decay time being probably reduced to the field pulse duration (less than 500 ms).

This evidences that, during the high magnetic field pulse, the Zeeman energy on non-compensated moments of the AF domain balances or overcomes the anisotropy energy barrier and switches some of the AF domain magnetization into the opposite direction.

It has to be noted that used m.o. method does not allow to reveal constant contribution to the magnetization (magnetic moment from non-compensated AF domains), and hysteresis loops in Figure 9 are manually centered along vertical axis.

By these high magnetic field experiments we can expect a new possible estimate of the energy barrier and have a confirmation of the previous model. At first we can evaluate the mean magnetic moment associated with the non compensation of the AF domain volume whose size was discussed earlier. We know the average number N of spins in a volume corresponding to the AF magnetic domains $N = 2.5 \times 10^4$. The statistically non-compensated magnetization of each AF domain could be written in the form $M = 2\ \mu_B \sqrt{N} = 29 \times 10^{-22}$ J T^{-1}.

When a 55 T high magnetic field pulse is applied along the uniaxial axis but opposite to the unidirectional field, we have observed an immediate bias field decrease to zero. An estimate of the Zeeman energy of the AF domain (volume V and magnetization M) gives the value

$$E_{Zeeman} = 2\ \mu_B \sqrt{N}\, H = 1.5 \times 10^{-12} \text{ erg}.$$

This energy is larger than the barrier energy due to the anisotropy. It could explain the immediate reversal of a lot of AF domains in the direction of the Co magnetization inducing the observed decrease of the bias field. The method consisting to apply a series of pulsed magnetic field of increasing amplitude could be an efficient experimental process to probe the distribution of barrier energies.

This conclusion could be debated, if we consider that, in high magnetic field, the spin flop phase could be induced in a large part of the antiferromagnetic domains. This effect, probably favor the simultaneous reversal of AF domains with the Co magnetisation in the direction of the applied magnetic field.

7 Conclusion

We have developed a magneto-optical method to study the relaxation of the exchange anisotropy. The results obtained with Co/NiO systems show a systematic logarithmic time dependence of the unidirectional field H_{ud}. The relaxation rate of H_{ud} is directly correlated to the uniaxial anisotropy of the AF layer which results of the deposition method. For example, when NiO is obliquely deposited, the characteristic time of relaxation is very long, the bias tends to a value close to zero after about several days when the Co magnetization is reversed. Assuming the analysis developed by Malozemoff based on random field model and the model of Fulcomer and Charap, we propose a possible first description of this phenomenon. Clearly real systems are more complex and the AF/F interface presents more the characteristics of a frustrated spin system.

References

1. W.H. Meiklejohn, C.P. Bean, Phys. Rev. **105**, 904 (1956)
2. K.Y. Kim, S.H. Jang, K.H. Shin, H.J. Kim, T. Kang, J. Appl. Phys. **89**, 7612 (2001)
3. R. Jungblut, R. Coehoorn, M.T. Johnson, J. van de Stegge, A. Rinders, J. Appl. Phys. **75**, 6659 (1994)
4. D.H. Hempstead, S. Krongelb, D.A. Thompson, IEEE Trans. Magn. **14**, 521 (1978)
5. W.J. Antel Jr., F. Perjeru, G.R. Harp, Phys. Rev. Lett. **83**, 1439 (1999)
6. C.H. Shang, G.P. Berera, J.S. Moodera, Appl. Phys. Lett. **72**, 605 (1998)
7. M. Cartier, S. Auffret, P. Bayle-Guillemaud, F. Ernult, F. Fettar, B. Dieny, J. Appl. Phys. **91**, 1436 (2002)
8. H.D. Chopra, D.X. Yang, P.J. Chen, D.C. Parks, W.F. Egelhoff Jr., Phys. Rev. B **61**, 9642 (2000)
9. H.D. Chopra, D.X. Yang, P.J. Chen, H.J. Brown, L.J. Swartzendruber, W.F. Egelhoff Jr., Phys. Rev. B **61**, 15312 (2000)
10. T. Kimura, Y. Itagaki, F. Wakaya, K. Gamo, Appl. Phys. Lett. **78**, 4007 (2001)
11. D. Mauri, H.C. Siegemann, P.S. Bagus, E. Kay, J. Appl. Phys. **62**, 3047 (1987)
12. A.P. Malozemoff, Phys. Rev. B **35**, 3679 (1987); A.P. Malozemoff, Phys. Rev. B **37**, 7673 (1988)
13. B. Diouf, L. Gabillet, A.R. Fert, D. Hrabovsky, V. Prochazka, E. Snoeck, J.F. Bobo, J. Magn. Magn. Mat. **265**, 204 (2003)
14. I.N. Krivotorov, T. Gredig, K.R. Nikolaev, A.M. Goldman, E. Dan Dahlberg, Phys. Rev. B **65**, 180406/14 (2002)
15. E. Fulcomer, S.H. Charap, J.A.P. **43**, 4184 (1972)
16. D. Hrabovský, Thesis Manuscript, INSA Toulouse, France, 2003
17. S. Dubourg, Thesis Manuscript, INSA Toulouse, France, 2001
18. Y.J. Wang, C.H. Lai, J. Appl. Phys. **89**, 7537 (2001)
19. M. Fraune, U. Rüdiger, G. Güntherodt, S. Cardoso, P. Freitas, Appl. Phys. Lett. **77**, 3815 (2000)
20. J. Nogués, J. Sort, S. Suriñach, J.S. Muñoz, M.D. Baró, J.F. Bobo, U. Lüders, E. Haanappel, M.R. Fitzsimmons, A. Hoffmann, J.W. Cai, Appl. Phys. Lett. **82**, 3044 (2003)
21. P. Miltényi, M. Gierlings, J. Keller, B. Beschoten, G. Güntherodt, U. Nowak, K.D. Usadel. Phys. Rev. Lett. **84**, 4224 (200)
22. A. Mougin, T. Mewes, M. Jung, D. Engel, A. Ehresmann, H. Schmoranzer, J. Bassbender, B. Hillebrands, Phys. Rev. B **63**, 060409(R) (2001)