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**Production and magnetic characterization
of exchange-coupled magnetic
nanostructures**

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Chapter 1

Introduction

The recent discovery of giant magnetoresistance made in 1988 by Grünberg and Fert, allowed scientists to consider the spin of electron as an additional degree of freedom in determining the value of the electronic conductivity of a system containing ferromagnetic (FM) phases. This finding paved the way to a new physics subject, that is the study of the spin-dependent conductivity, the so-called spintronics, and had great impact on technology as well, because it influenced the production of read-heads for hard-disk, of magnetic memories and magnetic field sensors.

From the technological point of view, the interest was initially focused on materials known as “spin-valves” (SVs), formed by two thin FM layers, the electrodes, separated by a thin layer of non-magnetic metal spacer. The two electrodes are prepared so to have different magnetic properties, in particular they are expected to have different switching fields, so that an external magnetic field can change the relative orientation of their magnetization vectors (from parallel to antiparallel and viceversa), determining a variation of the SV electrical resistance. In particular, one electrode is prepared to act as the reference layer, namely the layer whose orientation is not affected by the external field, whilst the other one to act as the sensor layer, viz. the orientation of its magnetization can be more easily modified in the

presence of an external magnetic field. Nonvolatile magnetic random access memories (MRAMs) represent an important example of application of such technological achievements. They are basically composed of an array of SV cells, each one able to store one bit of data.

In magnetic materials, anisotropy is a key aspect to tune the magnetic properties, and it may include different contributions: magnetocrystalline, shape, surface and magnetoelastic. Recently, a new way of tuning and enhancing the magnetic anisotropy of layered magnetic materials has been proposed, which exploits the exchange-coupling at the interface between a FM and an antiferromagnetic (AFM) phase. In these systems, during a hysteresis loop, the interfacial spins of the AFM region may exert a torque on the FM spins, resulting in the appearance of an unidirectional anisotropy, which is revealed by a horizontal shift of the loop, commonly called exchange bias (EB) [1, 2, 3, 4, 5]. In other words, the exchange interaction across the FM/AFM interface gives rise to an extra source of magnetic anisotropy, which affects both the magnetization reversal process of the system and the coercivity. This effect plays a crucial role in several technological applications such as read heads of recording devices [6, 7] and in MRAM cells [8, 9], and a deep understanding of the exchange coupling fundamental mechanism is necessary to make progress in the field of spintronics and related technologies [10, 11].

Moreover, the increasing demand for miniaturization of magnetic devices triggers an increasing interest for the study of the magnetic properties of elements size-confined to the nanometric scale (dots). In fact, going from the continuous film to single-phase dots, strong changes may occur in the magnetic configuration (single-domain, multi-domains or non-uniform magnetization states) and in the reversal mode of the magnetization. Moreover, below a critical size and depending on the anisotropy, the magnetization becomes unstable against thermal fluctuations and

coercivity may vanish.

In this scientific context, the present thesis addresses the study of EB in nanostructures such as thin films and arrays of nanodots. The formers are studied in order to clarify the role of the interface between the FM and AFM phases in the exchange coupling mechanism, the latter to elucidate the influence of size confinement effects on EB, in view of the application of the nanostructures as electrodes in technological devices based on SV cells. This research activity has been made possible thanks to the collaboration with different scientific groups from “Istituto di Struttura della Materia” CNR Rome, “Istituto di Fotonica e Nanotecnologie” CNR Rome and “Università Politecnica delle Marche” Ancona, under the common research framework of the FIRB 2010 NANOREST project “Tailoring the magnetic anisotropy of nanostructures for enhancing the magnetic stability of magnetoresistive devices”.

As described in the next chapters, we exploited $\text{Ni}_{80}\text{Fe}_{20}$ (NiFe) and $\text{Ir}_{25}\text{Mn}_{75}$ (IrMn) alloys as FM and AFM phases respectively, to grow our exchange-biased systems by dc-magnetron sputtering. We performed the magnetic and the structural characterization of the samples and finally we elaborated a model accounting for the magnetothermal properties of our samples. In detail, the work that we present in the next chapters is structured as described below.

In chapter 2, the basic EB phenomenology is presented in terms of the pioneering achievements that its discoverers, Meiklejohn and Bean, obtained in nanoparticles systems. Their basic model is illustrated, in order to clarify how the spin configurations of both the FM and the AFM may provide EB in exchange-coupled systems. Particular attention will be devoted to the description of the effects of the blocking temperature of the nanoparticles on the EB. In fact, the concepts of blocking temperature and blocking temperature distribution will be widely exploited in the formulation of our new model accounting for EB mechanism in our samples.

In chapter 3, the experimental techniques employed for both the production and the magnetic characterization of the samples are briefly reviewed, with particular focus on those exploited in the laboratories of the Department of Physics and Earth Sciences of the University of Ferrara.

In chapter 4, we illustrate the results obtained from the experimental characterization of the magnetic properties of our exchange coupled samples in form of continuous films. In particular, we studied IrMn/NiFe bilayers as a function of IrMn thickness and stacking sequence. In addition, we present an original experimental protocol we devised in order to further study the magnetothermal behavior of the AFM phase, and we show the experimental results obtained from our samples. The experimental observations collected through all these analyses, as well as the results from structural investigations, allowed us to extend the basic model presented in chapter 2 by proposing a precise description of the mechanism to which the AFM interfacial spins take part to provide EB, and that founds its basis on the description of the AFM layer as made of both nanograins and a magnetically disordered interfacial component.

In chapter 5, the temperature dependence of EB is studied in conjunction with spatial confinement effects. To achieve this goal, we proceeded to realize and magnetically characterize arrays of AFM/FM dots with different size, that varies from $1\,\mu\text{m}$ down to $140\,\text{nm}$. The results obtained convinced us to suggest that, to understand the magnetothermal behavior of nanosized exchange-coupled systems in the light of the proposed model, a comparison between the nanostructure size and a characteristic length of correlation of interfacial spins is to be taken into account.

Finally, in the concluding chapter the main results obtained are summarized and discussed.

Chapter 2

The Exchange Bias effect

The theoretical interpretation of the phenomenon of exchange bias is very controversial and many models have been proposed over the years [4, 24]. This chapter mainly describes the original basic model that, according to the classification of Coehoorn [12], belongs to the category of macroscopic models. Macroscopic models are considered those that do not take into account the lateral magnetic structures of the layers, namely the layers are assumed to be homogenous in plane, and consider the coherent rotation of the magnetization as well. As explained in the text, we exploited some main ideas of this model in order to develop our own new model presented in chapter 4. In the same classification due to Coehoorn, other categories are mesoscopic and microscopic. The models belonging to mesoscopic category take account of the possibility that spins are oriented in different ways in plane. One of the main mesoscopic models is the model of Malozemoff, which includes the possibility that the interface FM-AFM is not perfectly flat: the roughness of the interface produces a random field, which leads the AFM phase to break in domains with walls perpendicular to the interface [13]. Finally, in the case of microscopic models, the detailed configuration of the spin of each atom is taken into account inside the volume of the system in order to find the best spin configuration at the interface

between the FM and AFM phases, namely to find the configuration of minimum energy. One of the pioneering microscopic theoretical model was due to Koon, who argued that the configuration with minimum energy is to couple the FM and AFM spins perpendicular to each other [14].

2.1 An Overview of the Phenomenology

The term “exchange bias” (EB) refers to a particular effect that modifies the properties of a ferromagnetic (FM) material due to the additional source of anisotropy introduced by the exchange coupling between the FM material and an antiferromagnetic one (AFM). EB has been universally recognized to be an interfacial effect, and as a consequence, in the last decades, the studies of the EB have been performed mainly on systems consisting of a thin FM layer in contact with an AFM one, exploiting different FM-AFM combinations and several deposition techniques in order to obtain the best control of the interface quality.

The first scientists who observed the effect of this new anisotropy term were Meiklejohn and Bean in 1956 thanks to their studies about Co nanoparticles embedded in antiferromagnetic CoO external shell developed by oxidation [1, 2]. They observed that, decreasing temperature below the characteristic Néel temperature (T_N) of CoO, Co nanoparticles presented hysteresis loops slightly shifted along the field axis and realized that this EB effect must be due to the exchange interaction between the ferromagnetic Co phase and their surrounding antiferromagnetic CoO shell. In the following description of the EB phenomenon, we will briefly look at their experimental results reported in figures 2.1 and 2.5, in order to provide a basic description of the phenomenology.

In figure 2.1 it is shown the first ever observed shift of a hysteresis loop, measured for the case of Co-CoO particles. The system was cooled down from room tempera-

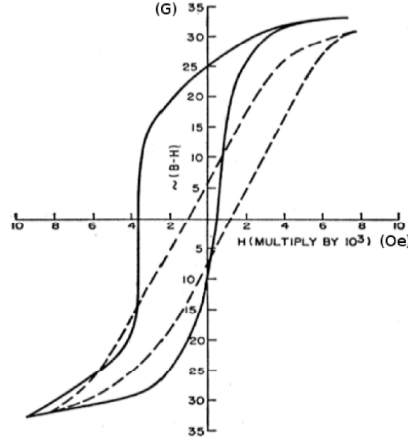


Figure 2.1: Hysteresis loops of Co-CoO particles, both measured at temperature $T = 77\text{ K}$. The dashed line refers to the loop after cooling down it from room temperature in zero external field. The solid line is the hysteresis loop measured after cooling the system in presence of an external field of 10 kOe . Taken from reference [2].

ture to 77 K through the Néel temperature of the CoO ($T_N(\text{CoO}) = 291\text{ K}$) either in presence (field cooled, FC) or in absence (zero field cooled, ZFC) of a large external field applied during the cooling procedure. The magnetization loop reported with a dashed line is measured in ZFC mode and it appears to be centered around the zero value of the field axis.

However, when the sample is cooled in presence of a magnetic field, and we consider the direction of this field to be the positive direction, the hysteresis loop is displaced to negative values (that is, in the opposite direction with respect to the cooling field). The amount of the shift is called the exchange bias field, H_{ex} . So, the spin configuration which gives rise to the EB effect needs an initialization which can be achieved by field cooling the system down to a temperature lower than T_N , when the AFM order is effectively established inside the bulk of the AFM. However, apart from the AFM order, the AFM needs to be stabilized against thermal fluctuations. In this respect, we consider that the AFM may be constituted by different entities, such as independent AFM nanograins, each with a specific temperature associated,

called blocking temperature (T_B). T_B is related to the magnetic anisotropy energy term of the nanograin by the relation $k_B T_B = K_{AFM} V$, where k_B is the Boltzmann constant, V and K_{AFM} are, respectively, the volume and magnetic anisotropy of the nanograin. In this case the AFM must be cooled below each T_B to stabilize the corresponding AFM nanograin against thermal fluctuations, namely to guarantee them stability against thermal fluctuation over a time τ dermined by the Arrhenius law:

$$\tau = \nu_0^{-1} \exp \left(\frac{K_{AFM} V}{k_B T} \right) = \nu_0^{-1} \exp \left(\frac{T_B}{T} \right)$$

where ν_0 is the attempt frequency related to spin-phonon interaction, generally believed to be 10^9 Hz [69].

Blocking temperatures are typically, but not necessarily [21, 22], below the bulk Néel temperature. Accordingly, we prefer to say that temperature needs to be cooled down below some blocking temperature (or blocking temperature distribution, if there are entities with different magnetic properties to consider) characteristic of the AFM phase, in order to have the exchange interaction properly initialized to give a non zero H_{ex} . In this respect, in the model we proposed to account for the EB effect in our samples, we considered the AFM phase to be composed of several entities that can be stabilized against thermal fluctuations at different blocking temperatures, such as nanograins for example. In fact, the volumes of the nanograins are expected to vary according to a volume distribution, and a corresponding distribution of T_B is expected as well.

We illustrate in figure 2.2 some schematic expected hysteresis loops of a FM at different stages during the initialization process of EB. Figure 2.2(a) displays the hysteresis loop of a simple FM-AFM system that would be measured at $T > T_N$. The simple presence of the AFM adjacent to a FM does not affect the FM hysteresis

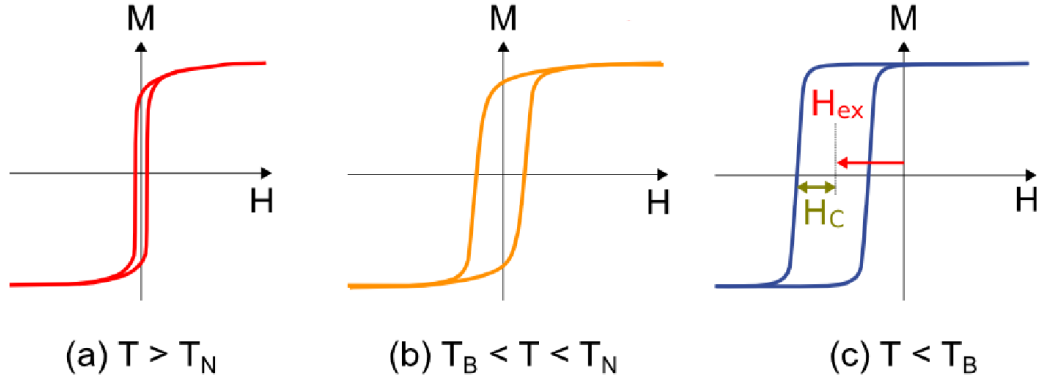


Figure 2.2: Schematic expected hysteresis loops of a ferromagnet coupled to an AFM phase at different regimes of temperature: (a) $T > T_N$, (b) $T < T_N$ (c) $T < T_B$, only the last case shows both a significant exchange bias and a loop enhanced coercivity.

loop in any significant way, due to the lack of both AFM order among the AFM spins and stability over time, above T_N . Hence, the displayed hysteresis loop arises from the intrinsic properties of the FM. Figure 2.2(b) displays the FM hysteresis with increased loop width at $T_B < T < T_N$, due to the drag effect generated by the AFM phase which is now established and interacting with the FM layer, but not rigidly stable in a fixed orientation. Finally, figure 2.2(c) displays a shifted hysteresis loop along the magnetic field axis indicating exchange bias quantified by H_{ex} . The coercivity H_C also increases due to the increment of the drag effect, consequence of an overwhole hardening of the magnetic phases at low temperatures as the K_{AFM} may be expected to increase with the decrease of temperature [23].

In order to give a first intuitive description of the spin configuration that gives rise to the EB [24, 25], we consider the ideal case of a flat interface between pure crystalline FM and AFM thin films, each forming single domain from the magnetical point of view, in a stack sequence that presents the AFM phase grown before the FM one (AFM/FM), and in the simplified hypothesis that T_B is numerically equal to T_N . We can refer to this simplified case as a starting point to evaluate cases of more

complex systems, such as the ones constituted of a larger number of non-interacting single domains, as proposed for our samples.

For this case, the onset of exchange bias is depicted in figure 2.3.

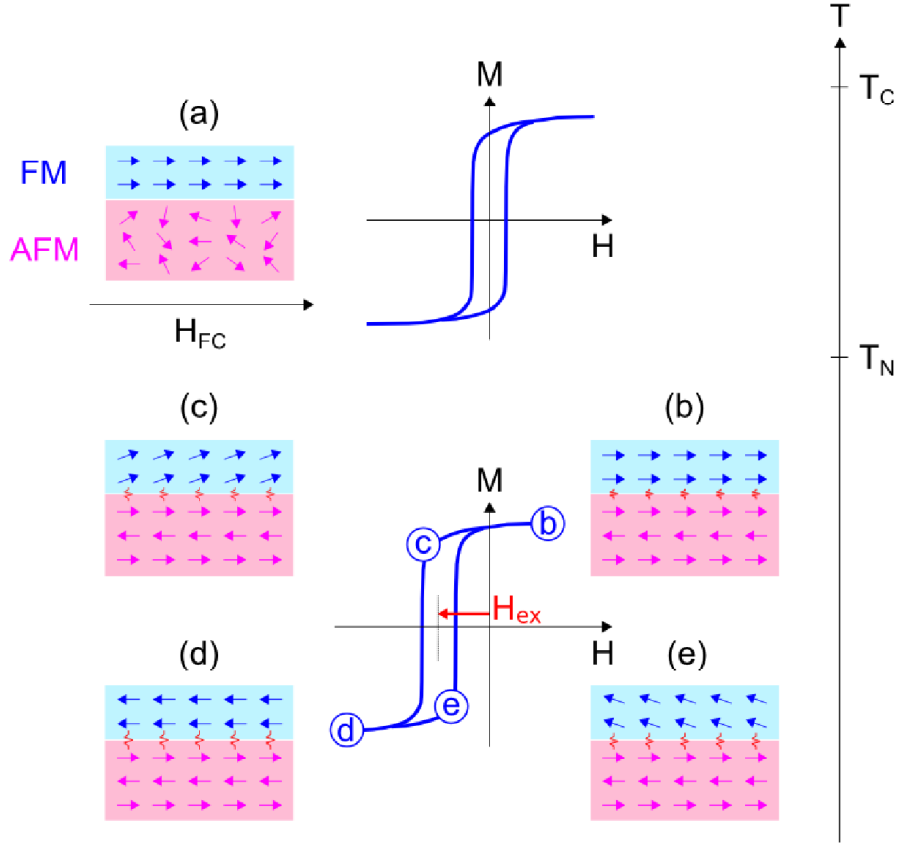


Figure 2.3: For the system AFM/FM, different spin configurations in function of temperature T and external field are shown, in case of a FM layer in close contact to an AFM one.

We consider the case in which the critical temperatures, associated with the two magnetic phases, satisfy the condition: $T_C > T_N$, where T_C is the Curie temperature of the ferromagnetic layer. At a temperature higher than T_N and lower than T_C , the FM magnetization aligns along the direction of an external applied field H_{FC} ; we consider H_{FC} large enough to saturate the FM, whereas the AFM spins remain almost randomly oriented in a paramagnetic state due to the high thermal energy (figure 2.3(a)). The hysteresis curve of the FM layer is centered around zero, not

being affected by the proximity of the AFM layer. Next, without changing the applied field, the temperature is decreased to a finite value smaller than T_N . After field cooling the system, due to the exchange interaction at the interface between the FM and the AFM, the first monolayer of the AFM layer will align ferromagnetically (or antiferromagnetically, it depends on the characteristic exchange coupling of the system) to the FM spins that are located at the interface. The next AFM monolayer will align antiparallel to the previous layer as to complete the AFM order, and so on (see figure 2.3(b)). The AFM interfacial spins so displaced are commonly said to be “uncompensated”, in the sense that they lead to a finite net moment for the AFM layer at the interface (even though EB in polycrystalline systems was also found at perfectly compensated interfaces, namely where the spins at the interface lead to a net averaged moment equal to zero [18]).

It is assumed that both the FM and the AFM are in a single domain state and that they will remain single domains as we reverse the external field in order to measure the hysteresis loop; this implies a coherent rotation of the FM magnetization. So, when reversing the field (figure 2.3(c)), the FM spins will try to rotate to the opposite direction but, being coupled to the AFM which is considered to be rigid and stable here, a stronger external field is needed to overcome this coupling and rotate the FM spins. As a result, the first coercive field (H_{c1}) is higher in modulus than the corresponding one at $T > T_N$, when there was no proper AFM set to interact with. On the way back, from negative saturation to positive one (figure 2.3(d)), the FM spins will need a smaller external force in order to rotate back (figure 2.3(e)) to the original direction. As a result, the hysteresis loop appears shifted to negative values of the applied field thanks to the torque action on the FM spins for all other angles except the stable direction set by the field cooling direction, so implying an unidirectional anisotropy.

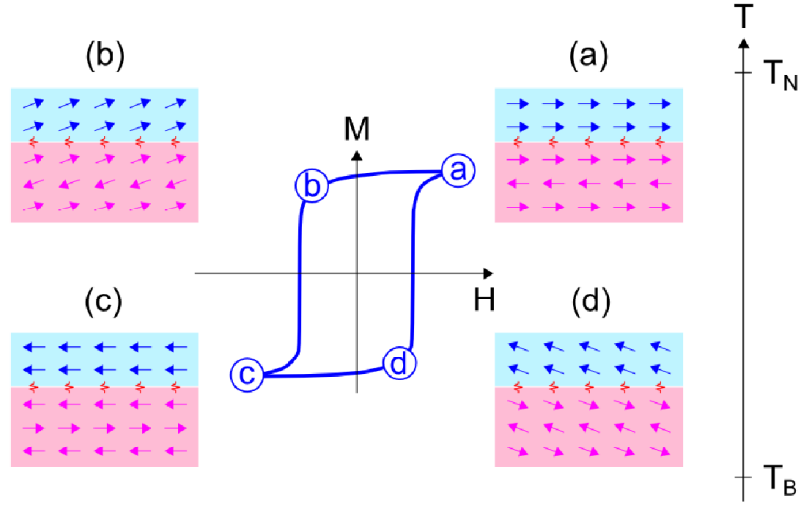


Figure 2.4: For the system AFM/FM, different spin configurations in function of the external field are shown, at temperature $T_B < T < T_N$, illustrating the drag effect that causes an enhancement of coercivity in the hysteresis loop.

The intermediate case in which $T_B < T_N$, and the temperature of the system is $T_B < T < T_N$ is now considered. The related spin configuration is illustrated in figure 2.4.

In figure 2.4(a), we show that the same initial setting of figure 2.3(b) is obtained, through the cooling of the system down to $T < T_N$ in FC mode. But, the temperature condition $T > T_B$ means that the temperature is still high enough to compete against the anisotropy of the AFM, preventing a stable configuration of AFM spins (even though the AFM order is still preserved), and as we reverse the external field we observe that the FM magnetization starts to rotate, dragging the AFM phase (figure 2.4(b)). An analogous behavior is observed after saturating in negative fields (figure 2.4(c)), in fact, on the way back to positive values of the external field, the FM magnetization still drags the AFM phase in the same way it did before. With respect to either a FM uncoupled with an AFM phase, or a AMF/FM system at $T_N < T < T_C$, the work done by the external field during the reversal of the magnetization is larger, and the coercive fields appear greater in modulus for both

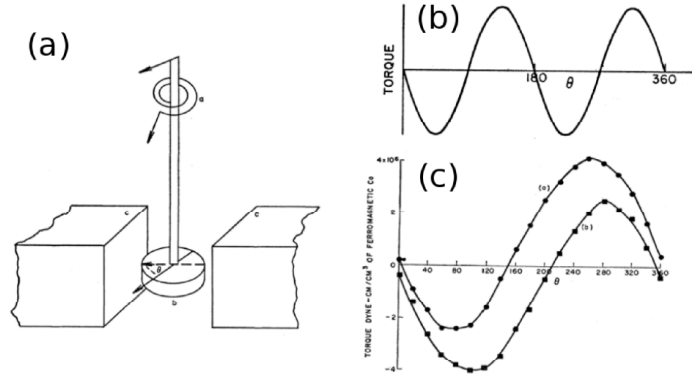


Figure 2.5: Schematic diagram of a torque magnetometer (a). The sample is placed in an magnetic field and a spiraling spring, connected to the sample, measures the torque as a function of the angle θ between some direction integral to the sample and the external field. Torque curve expected for a Co particle at 300 K showing uniaxial anisotropy. (c) Experimental torque curves of Co-CoO particles measured at 77 K showing unidirectional anisotropy. Taken from [2, 26]

the ascending and descending branches of the hysteresis loop, resulting in a general enhancement of H_C , but no shift is observed in this case (it was observed for the previous case, where $T < T_B$).

In order to better characterize the unidirectional property of this new kind of anisotropy, Meiklejohn and Bean studied its behavior by the use of a self-made torque magnetometer (figure 2.5(a)). It essentially consisted of a spring connected to a sample placed in an external magnetic field [2, 26]. Generally, the torque magnetometry is an accurate method for measuring the magnetocrystalline anisotropy of single crystal ferromagnets. The torque on a sample is measured as a function of the angle θ between a certain crystallographic direction and the applied magnetic field. In strong external fields, when the magnetization of the sample is parallel to the applied field (saturation), the torque is equal to $T = -\frac{\partial E(\theta)}{\partial \theta}$ where $E(\theta)$ is the magnetocrystalline energy.

In the case of Co which has a hexagonal structure, the torque with respect to the easy axis (c-axis) follows a $\sin(2\theta)$ function as seen in figure 2.5(b). The torque

and the energy can be written as:

$$T = -A_{mc} \sin(2\theta)$$

$$E = \int A_{mc} \sin(2\theta) d\theta = A_{mc} \sin^2 \theta + B$$

where A_{mc} is a energy term related to the magnetocrystalline anisotropy and B is an integration constant. It is clearly seen from the energy expression that along the c-axis, at $\theta = 0$ and $\theta = 180^\circ$, the system is found in stable equilibrium.

For the CoO-Co particles, at $77^\circ K$, after field cooling, the CoO is in an antiferromagnetic state. Here, the torque curve of the Co-CoO system looks completely different as seen in figure 2.5(c). The torque curve is a function of $\sin(\theta)$, and $E(\theta)$ results in a function of $\cos(\theta)$ accordingly:

$$T = -A_u \sin(\theta)$$

$$E = \int A_u \sin(\theta) d\theta = -A_u \cos(\theta) + B$$

where A_u is a energy term related to the anisotropy introduced by the coupling with the AFM phase. The energy function shows that the particles are in equilibrium for one position only, namely $\theta = 0$. Rotating the sample to any other angle, it tries to return to the original position. This direction is parallel to the field cooling direction and such anisotropy is the unidirectional anisotropy. Now, we can analitically show that the same unidirectional anisotropy observed by torque magnetometry is responsible for the loop shift. In figure 2.6 is shown schematically

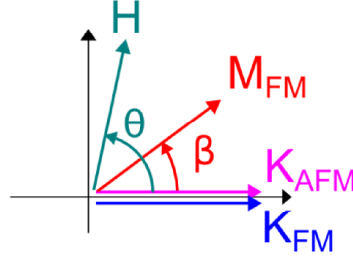


Figure 2.6: Diagram of the angles and vectors used. The AFM layer is assumed to be rigid and no deviation from its initially set orientation is allowed. $\vec{K}_{AFM}$ and $\vec{K}_{FM}$ are the anisotropy axis of the AFM layer and FM layer, respectively, which are assumed to be parallel oriented to the field cooling direction. β is the angle between FM magnetization vector $\vec{M}_{FM}$ and the anisotropy direction of the FM layer, it can vary during the magnetization reversal. $\vec{H}$ is the external magnetic field applied at the angle θ .

the vectors involved in the ideal Meiklejohn-Bean model.

H is the applied external magnetic field, which makes an angle θ with respect to the field cooling direction denoted by $\theta = 0$, K_{FM} and K_{AFM} are the uniaxial anisotropy directions of the FM and the AFM layer, respectively. They are set to be oriented parallel to the field cooling direction. $\vec{M}_{FM}$ is the magnetization of the FM layer during the magnetization reversal. It is assumed that AFM spins do not deviate from their orientation defined during the field cooling procedure (rigid AFM). The applied field is assumed to be parallel to the field cooling direction in the analysis below ($\theta = 0$). In the assumption of coherent rotation of the magnetization, within this model the energy can be written as the sum of a Zeeman energy term, a contribution of the magnetocrystalline anisotropy, and the energy term previously derived from the torque experiment:

$$E = -HM_{FM}V_{FM}\cos(\theta - \beta) + K_{FM}V_{FM}\sin^2(\beta) - A_u\cos(\beta)$$

V_{FM} being the volume of the FM phase. For the case $\theta = 0$, and dividing both members for the interface area, we can rewrite the energy as energy per unit area

expressing the thickness of the FM layer as t_{FM} :

$$\varepsilon = -HM_{FM}t_{FM}\cos(-\beta) + K_{FM}t_{FM}\sin^2(\beta) - J_{FM-AFM}\cos(\beta)$$

where J_{FM-AFM} is called the coupling constant and it is expressed in terms of interfacial exchange energy per unit area (erg/cm^2). From the stability condition $\frac{\partial E}{\partial \beta} = 0$, calculated for the case $\beta = 0, \pi$ the points H_{c1} and H_{c2} where the hysteresis loop intercepts the field axis can be extracted [26]:

$$H_{c1} = -\frac{2K_{FM}t_{FM} + J_{FM-AFM}}{M_{FM}t_{FM}}$$

$$H_{c2} = \frac{2K_{FM}t_{FM} - J_{FM-AFM}}{M_{FM}t_{FM}}$$

And using the expressions above, the coercive field H_C of the loop and the displacement H_{ex} can be calculated according to:

$$H_C = -\frac{H_{c1} - H_{c2}}{2} = \frac{2K_{FM}}{M_{FM}}$$

$$H_{ex} = -\frac{H_{c1} + H_{c2}}{2} = -\frac{J_{FM-AFM}}{M_{FM}t_{FM}}$$

Coercivity is not affected by the exchange coupling, in fact it results in the classical term predicted by the model of coherent rotation for a FM (the Stoner-Wohlfarth model) because the AFM is supposed to be rigid and no drag effect is expected, accordingly. For the case of H_{ex} instead, the last equation derived above is considered to be the master formula of EB effect. Even if it gives the expected

characteristic shift of the hysteresis loop for an ideal case, it is usually exploited as a standard qualitative estimation: it predicts that the sign of EB is related to the characteristic exchange coupling and that the variation of the EB field is direct proportional to the inverse thickness of the FM layer.

Since the discovery of EB, a wide range of samples has been studied and the basic model proposed above to account for EB has been improved. The samples studied range from nanoparticles, where the interface between FM and AFM is not flat [1, 2, 15], to epitaxially grown films where the interface is expected to be the closest approximation of an near ideal flat interface on atomic scale [19, 20], and, as intermediate case, sputtered polycrystalline films where the interface has significant roughness which can lead to both structural and magnetic disorder [16, 17].

Among all three types of samples presented above, the largest exchange bias effects at room temperature are observed in polycrystalline granular films produced by sputtering [17], like the ones studied throughout this thesis work, and this characteristic makes this type of samples the preferred one to be proposed for applications in devices, such as magnetic recording read heads and MRAMs.

Chapter 3

Experimental Techniques

In this chapter we briefly review the main experimental techniques we employed for the production of the nanostructures of our interest in this thesis and for the characterization of their magnetic properties. This chapter is divided into two sections. Section 3.1 briefly describes the dc-magnetron sputtering, namely the technique of deposition we adopted for the production of both continuous films and dot arrays. Finally, section 3.2 describes the experimental setup based on the magnetooptical Kerr effect (MOKE) and the system based on the superconductive quantum interference device (SQUID): we exploited these two techniques to achieve magnetic information from both the continuous films and the dot arrays, in particular about their magnetothermal behavior.

3.1 Production of Exchange-Coupled Nanostructures

We have studied the exchange coupling in AFM-FM bilayers in form of both continuous films and arrays of dots with different shape and size. Both the systems considered have been produced by dc-magnetron sputtering but, for the specific case of the arrays of nanodots, the production process consisted of several steps,

made possible thanks to the collaboration of Annamaria Gerardino from “Istituto di Fotonica e Nanotecnologie” CNR Rome.

The first step consisted in drawing, through a process of electron beam lithography (EBL), the shape of the dots on a thin layer of polymeric material (resist) previously deposited on a substrate. During the lithographic process, information is recorded by means of an electron beam emitted by an electron beam gun: the purpose, as in photolithography, is to create very small structures inside the resist. The spatial resolution with which the drawings can be transferred on the resist depend both on the characteristics of EBL and on the properties of the polymeric material used; in fact an incident electron beam induces chemical reactions in the resist that make it more or less soluble to the action of solvents. The main advantage of EBL resides in the possibility of exceeding the diffraction limit of light in photolithography and, consequently, to draw structures with size down to the nanometric scale. In contrast, the disadvantage is the long time required to expose a large area, such as an entire silicon wafer: a long exposure time makes the process vulnerable to drifts and instability of the electron beam. Anyway, although the process may reveal to be relatively slow, it is a very practical technique for the fabrication of arrays of nano-elements, inside which the dimensions and mutual distances of the elements can be easily controlled.

In our specific case, the substrate, consisting of naturally oxidized silicon Si(100), was spin coated with a poly(methyl methacrylate), PMMA, bilayer: a first PMMA layer of 50k molecular weight, 4% solid content, was spun at 4000 *rpm* and baked at 170°C for 5 minutes. Then a second PMMA layer, 600k molecular weight, 4% solid content, was spun on top of it and baked (by the same process). The exposure dose is 250 $\mu\text{C}/\text{cm}^2$ at 100 *kV*, and the patterns were developed in *MIBK* : *IPA* = 1 : 3 for one minute at 20°C. The PMMA layer with lower molecular weight (50k) is more

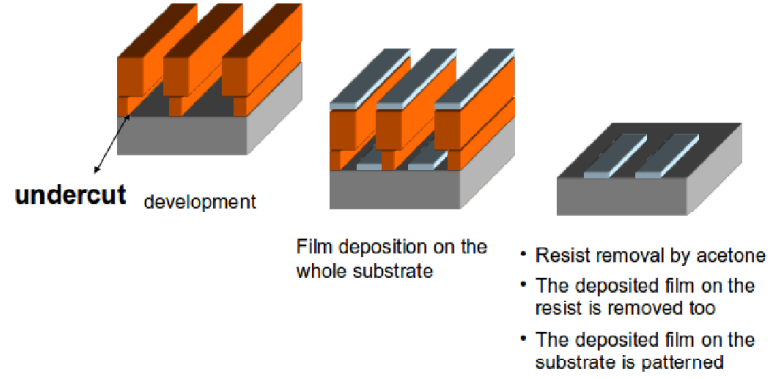


Figure 3.1: Schematic illustration of some steps employed for the fabrication of nanodots: from the resist development to the lif-off procedure. (A courtesy of A. Gerardino.)

sensitive to the electron exposure than the one on the top so, after the development, the resist profile shows an undercut that, along with the presence of a collimator during the growth process, causes the successive lift-off process (see figure 3.1).

Once the patterned areas are developed, the desired sequence of metallic layers were deposited by means of the dc-magnetron sputtering apparatus described in the next sections. Finally, the PMMA bilayer was removed in a lift-off process, leaving the desired arrays of nanodots on the silicon substrate. We fabricated samples with two different dots shape: square and circular. For the square shape we chose different lateral dimensions for the dots, in order to assess how the progressive decrease in size may affect EB properties, and appropriate relative inter-distances, so as to prevent the establishment of mutual magnetic dipolar interactions, as we will describe with more detail in chapter 5.

3.1.1 DC-Magnetron Sputtering

The dc-magnetron sputtering deposition technique is generally exploited for both commercial and research activities, in order to grow thin films of several different metallic materials over a substrate layer. This technique belongs to the class of

physical vapour deposition (PVD¹) techniques because the the metallic vapour of the metal that is to be grown over the substrate is obtained through a mere physical process, based on the ion bombarding of the metallic target material; so, the ion bombarding represents the main feature of the sputtering process [27].

The term sputtering refers to the ejection of atoms that we obtain through energetic particles (charged ions) accelerated to collide against the surface atoms of the target. Through these direct collisions of ions with surface atoms, or recoil secondary collisions among surface atoms only, the ejection of atoms from the target material can take place. The name “dc-magnetron” is to emphasize that the deposition process occurs in a continuous current of ions, obtained by applying a constant potential difference between the substrate and the target, and in the presence of magnets at the back of the cathode to increase the efficiency of the sputtering process. The silicon substrate on which the deposition occurs is located above the target, with reference to the schematic representation of the experimental apparatus illustrated in figure 3.2(a).

If the target is negatively charged with respect to the substrate, it will be possible to ionize a gas of argon previously introduced inside the main vacuum chamber. If the argon ions Ar^+ are accelerated with energies sufficiently high, they will be able to sputter the atoms of the target material from its surface. Therefore, if the atoms of the target material are released in the atmosphere of the main chamber, a part of them will be deposited on the substrate surface.

In the main chamber, during the period in which the sputtering system is at rest and in the absence of argon, a pressure of $10^{-7} \div 10^{-8} Torr$ is maintained through a system of vacuum pumps, in order to remove other gases that might otherwise be present during the deposition, such the ones due to the degassing of the chamber

¹other important deposition techniques belong to PVD class, such as those based on thermal evaporation or on pulsed laser.

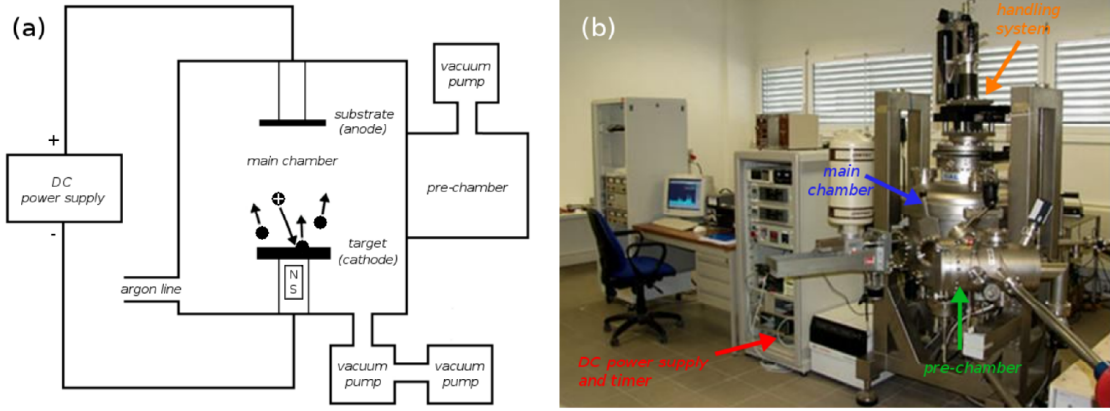


Figure 3.2: (a) Basic scheme of the sputtering deposition apparatus; the physical process that takes place inside the main chamber is also sketched. (b) A picture of the dc-magnetron sputtering apparatus exploited at the laboratories of Ferrara University

walls, which usually tend to release water vapor.

Therefore, to maintain a good quality of vacuum inside the main chamber, the introduction of the substrate from the outside is made through the porthole of a pre-chamber. In the pre-chamber, the substrate holder is screwed on a manipulator which introduces the former into the main chamber. In the main chamber the substrate holder is then hooked to a support connected to a handling system. Thanks to the handling system present in our apparatus it is possible to move the substrate holder so that it is in front of the target source of the material to be grown, so as to have a flux of atoms that is perpendicular to the substrate.

Moreover, the apparatus allows the rotation of the substrate during deposition, guaranteeing a uniform thickness of the sample, and it is equipped with a collimator with a 2:1 (height:width) aspect ratio to reduce the angular spread of the incident sputtered atoms [28] (in this way, it is about 10°). Dealing with the fabrication of nanodots, we have verified that the use of such a collimator greatly favors the lift-off process and offsets the shadow effect, which may alter the dot thickness at the border.

External controls are constituted basically of power supplies (one for each target material), each connected to a timer that interrupts the Ar^+ discharge, and the sputtering process accordingly, once the deposition time passed by. The deposition time was previously set to achieve the desired layer thickness, provided that the deposition rate of the material was previously determined through calibration measurements.

3.2 Magnetic Measurements

3.2.1 Magneto-Optical Kerr Effect

The study of the magnetic properties of a material can be conducted through optics. In this case, we exploited the phenomenon of magnetooptical Kerr effect (MOKE) in order to reveal the state of magnetization of a material during an hysteresis loop. MOKE is determined by the change of the polarization state and/or intensity of a laser beam reflected from the surface of a sample, with respect to the incident beam. In combination with an optical system of lenses, and a microscope with a CCD camera, our apparatus can focus the laser beam down to region $\sim 50 \mu m$ wide, compatible with the size of the arrays of nanodots, and this makes possible to reveal their corresponding magnetic properties.

Another similar effect that exploits the transmission of light is the Faraday effect, in this case both the polarization and the intensity of the beam transmitted through the FM material are expected to vary. In our case, working with metals, the transmission measurements are inhibited because of the high values of the absorption coefficients - this is a typical characteristic of metals - as well as for the presence of a silicon substrate over which the metallic layers are deposited; in other words we will be able to extract information only from the beam reflected by the

surface of the samples studied. Therefore, the collected information relates to the amount of material with which the beam is able to interact. For wavelengths that belong to the visible range, the depth of penetration inside a ferromagnetic material is typically variable between $10 \div 15 \text{ nm}$, therefore, if the thickness of the layer to be analyzed is compatible with this value, the information obtained will concern the whole layer. Conversely, if the thickness of the deposited film is larger, the information collected is limited to the surface layer. On the other hand, if the surface interacts in some way with what lies beneath, we can still extrapolate information relating greater depths, based on the interaction involved between the surface layer and layers located at different depth.

In order to qualitatively understand this phenomenon, it is important to remember that the optical properties of a material are related to its permittivity, and for isotropic materials we can write the equation

$$\epsilon = n^2$$

where n is the refractive index. For anisotropic materials a more general form of ϵ needs to be used, which is given by

$$\hat{\epsilon} = \epsilon_0 \hat{\epsilon}_r$$

where $\hat{\epsilon}$ and $\hat{\epsilon}_r$ are the permittivity and the relative static permittivity tensors, and ϵ_0 is the vacuum permittivity. In a Cartesian coordinate system $\hat{\epsilon}_r$ can be written as

$$\hat{\epsilon}_r = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{xz} & \epsilon_{yz} & \epsilon_{zz} \end{pmatrix}$$

where the off-diagonal terms are zero for an isotropic material. If we take into account the magnetization, this brings non zero terms off the diagonal, and therefore an optical anisotropy that causes the Kerr effect.

We can consider the linearly polarized incident light as a superposition of two circularly polarized components, and as a consequence the Kerr effect can be thought as the result of two processes. First, the two circularly polarized modes propagate with different velocities inside the medium, gaining two different phase shifts that, when recombined at their exit, results in a rotation of the polarization plane of the reflected beam. Second, since the material is dichroic, it absorbs differently the two modes, hence the ellipticity phenomenon is expected. So the tilt angle θ of the polarization axis and the gained ellipticity ε are related to the Fresnel reflection coefficients of the sample. These coefficients depend on the refractive index which contains information about the magnetization since it is linked to the permittivity tensor [30].

The Kerr effect can be observed in three different experimental configurations of the magnetization with respect to both the plane of incidence and the sample reflecting surface: the polar, the longitudinal and the transverse configuration. As shown in figure 3.3(a), in the polar configuration a beam of light with linear polarization falls usually under normal incidence on a surface which is magnetized in a direction perpendicular to the surface. After reflection, the polarization has turned by an angle θ , typically less than a degree, and some ellipticity has appeared. In

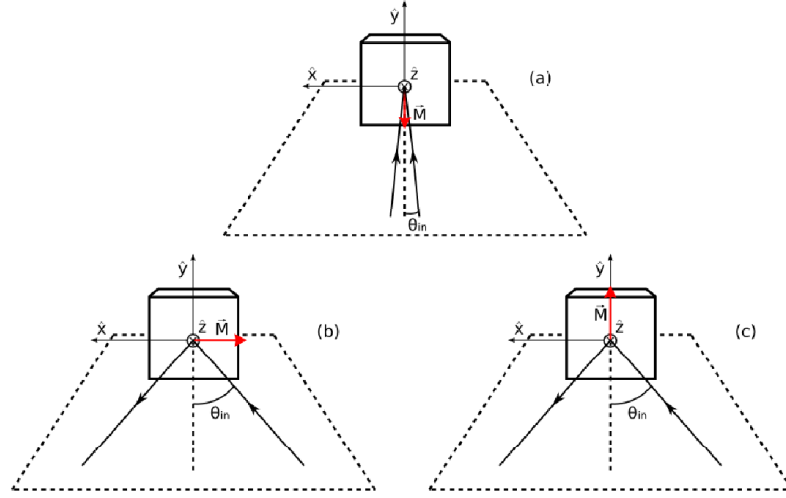


Figure 3.3: Geometries in terms of which the Kerr effect is generally described: (a) polar, (b) longitudinal and (c) transversal configuration. The plane of incidence is enclosed by the dotted line

the longitudinal Kerr effect (figure 3.3(b)), a beam strikes the surface under oblique incidence, this configuration is sensitive to the magnetization belonging to both the plane of incidence and the sample plane. If the polarization of the incident beam is perpendicular (*s* polarization) or parallel (*p* polarization) to the plane of incidence of the light, then the polarization of the reflected beam is slightly elliptical, with a major axis rotated with respect to the incident polarization by an angle generally less than a degree. The third configuration is the transverse one, where the geometry is the same as in the longitudinal case, except that the magnetization, although still in the plane of the specimen surface, is perpendicular to the plane of incidence (figure 3.3(c)) and the reflected beam is observed to change its intensity (but not rotation or ellipticity).

The MOKE measurements have been taken through the system sketched in figure 3.4. The incident light beam originates from a He-Ne laser ($\lambda = 632.8 \text{ nm}$), passes through a rotatable Glan-Thompson polarizer (P1), impacts on the sample surface (S) and then it is reflected. The polarizer P1 is set so that the beam retains a *p*

polarization (parallel to the plane of incidence). The reflected light beam passes through a photoelastic modulator (PEM) vibrating at an angular frequency $\omega = 50\text{ kHz}$, then through a second rotatable Glan-Thompson polarizer (P2) and finally it is detected by a photodiode (D). P2 is set almost at extinction with respect to P1, namely it is positioned at an angle $\alpha \sim 5^\circ$ with respect to the extinction axis, and the PEM is used for modulating the polarization of the light beam at the set frequency. The PEM function is based on the photoelastic effect, in which a mechanically stressed sample exhibits birefringence proportional to the resulting strain. Given the PEM optical axis, a linear polarization of the light parallel to that axis has a slightly different speed from that of a linear polarization perpendicular to the same axis. Defining Δn the difference between the two indices of refraction, $\Delta n = A \cos \omega t$ where A is proportional to the amplitude of the voltage applied to the piezoelectric transducer causing the vibration. As a consequence, the phase angle introduced between two waves with polarization parallel and perpendicular to the optic axis and traveling through the PEM is $\varphi = \frac{2\pi l \Delta n}{\lambda} = \varphi_0 \cos \omega t$, where l is the thickness of the optical element, and $\varphi_0 = \frac{2\pi l A}{\lambda}$. The components of the detected signal modulated at ω and 2ω are measured with two lock-in amplifiers, as they contain information about ε and θ respectively. The sample is positioned between the two poles of an electromagnet whose voltage can be controlled via computer. The electromagnet can be rotated in order to produce a field H that lies in the sample surface plane and that can be either parallel or perpendicular to the plane of incidence.

Each step of this system can now be analyzed considering that a possible representation of the light beam physical state is the one given in a bidimensional Hilbert space with a basis formed by the s and p polarizations. Using the Jones formalism one can associate a matrix to every optical element, and study the light beam at

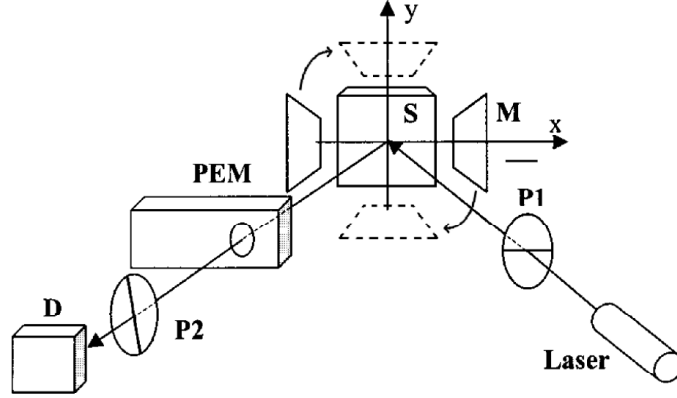


Figure 3.4: A sketch of the MOKE measurements set-up, taken from [31]

the detector stage assuming the initial state was the one of p polarization. The detailed derivation can be found in [32], what we are interested in is the main result, according to which the measured intensity I in a first order approximation on the Fresnel reflective indices is

$$I = \alpha^2 |r_{pp}|^2 + 2\alpha |r_{pp}|^2 (\theta \cos \varphi - \varepsilon \sin \varphi)$$

where it has been explicitly written the Fresnel reflective index r_{pp} relative to a p polarized incident beam and p polarized reflected beam. It can be noticed that I is given by the sum of two main terms, and the second one includes both θ and ε . Thus, we can write both $\cos \varphi$ and $\sin \varphi$ in Fourier series using the Bessel functions:

$$\cos \varphi = J_0(\varphi_0) + 2 \sum_{m=1}^{\infty} J_{2m}(\varphi_0) \cos(2m\omega t)$$

$$\sin \varphi = 2 \sum_{m=0}^{\infty} J_{2m+1}(\varphi_0) \sin[(2m+1)\omega t]$$

where $J_{\xi}(\varphi_0)$ are the Bessel functions of ξ order with φ_0 as argument. Inter-

rupting the expansion in terms of Bessel functions at their second-order, namely neglecting other terms not because subsequent orders are small, but because they will not be detected by selective lock-in amplifiers, we obtain:

$$I = I_0 + 2\alpha\theta |r_{pp}|^2 J_2(\varphi_0) \cos(2\omega t) - 4\alpha\varepsilon |r_{pp}|^2 J_1(\varphi_0) \sin(\omega t)$$

namely a signal constituted of both constant (I_0) and two variable components. The two variable components are found to be proportional to the rotation θ and ellipticity ε and modulated at the two different frequencies 2ω and ω , respectively. With the help of two lock-in amplifiers set at the same frequency values, it is then possible to sense the two contributions, which are expected to be proportional to the longitudinal magnetization.

3.2.2 Superconductive Quantum Interference Device

The Superconducting Quantum Interference Device (SQUID) is the main component of the quantum design Magnetic Property Measurement System (MPMS) that is recognized to be one of the most sensitive instruments for measuring magnetic properties, with applied external field that can reach 5 Tesla of intensity, and a broad range of working temperatures (from 2 K to 400 K in our system). One of the differences between MOKE and SQUID magnetometry is that the latter allows to directly determine the overall magnetic moment of a sample in absolute units, with sensitivity down to about 10^{-7} emu [33]. SQUID combines the physical phenomena of flux quantization and Josephson tunneling, it does not detect directly the magnetic field from the sample. Instead, the sample moves through a system of superconducting detection coils which are connected to the SQUID with superconducting wires, allowing the current from the detection coils to inductively couple

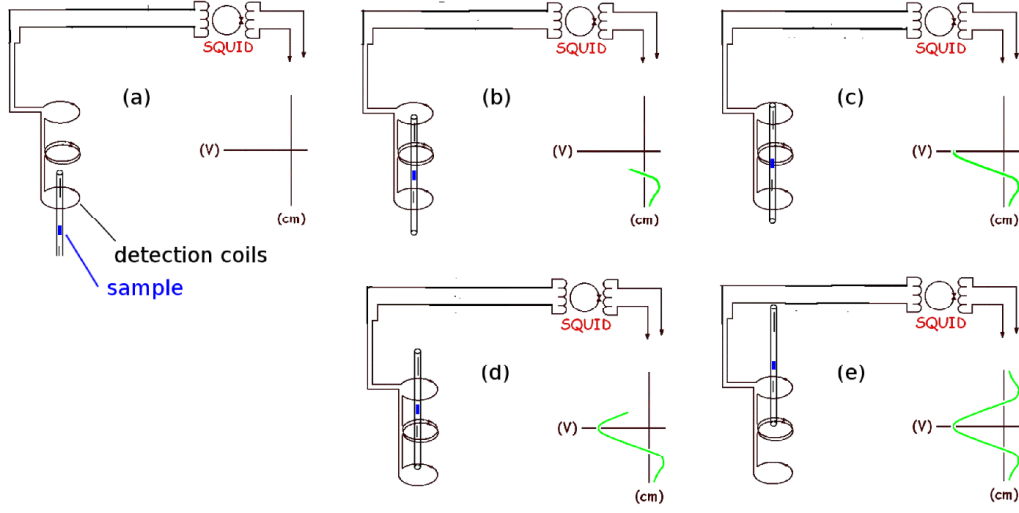


Figure 3.5: A sketch of the MPMS measurement process, from (a) to (e), that brings to the development of the output voltage (in green). Taken from [34]

to the SQUID sensor. When properly configured, the SQUID electronics produces an output voltage which is strictly proportional to the current flowing in the SQUID input coil. Hence, the thin film SQUID device, which is located inside a superconducting shield, essentially works as an extremely sensitive current-to-voltage converter [34].

A measurement (figure 3.5) is performed in the MPMS by moving a sample through the superconducting detection coils, which are located outside the sample chamber and at the center of the magnet. As the sample moves through the coils, the magnetic moment of the sample induces an electric current in the detection coils. Because the detection coils, the connecting wires, and the SQUID input coil form a closed superconducting loop, any change of magnetic flux in the detection coils produces a change in the persistent current in the detection circuit which is proportional to the change in magnetic flux. Since the SQUID functions as a highly linear current-to-voltage converter, the variations in the current in the detection coils produce corresponding variations in the SQUID output voltage which are proportional

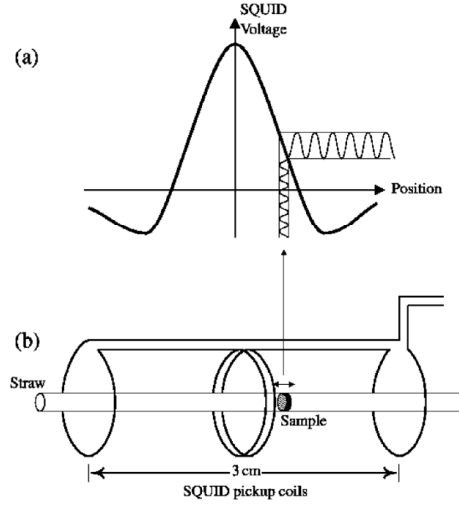


Figure 3.6: Illustration of a measurement with the RSO option. (a) shows the ideal SQUID response for a dipole and (b) shows the movement of the sample within the SQUID pickup coils. Taken from [34]

to the magnetic moment of the sample. In a fully calibrated system, measurements of the voltage variations from the SQUID detector as a sample is moved through the detection coils provide a highly accurate measurement of the sample's magnetic moment. The system can be accurately calibrated using a small piece of material having a known mass and magnetic susceptibility.

The commercial MPMS we exploited at the laboratories of the University of Ferrara is further equipped with the reciprocating sample oscillation (RSO) option. Unlike typical measurements where the sample is moved through the coils in discrete steps, the RSO option allows the sample to oscillate inside the detection coils (figure 3.6). By locking the frequency of the readout to the frequency of the movement, the magnetometer system can achieve the highest sensitivity described above for ultra small magnetic signals.

Chapter 4

Continuous Film Systems

In the introduction of EB presented in chapter 2, we pointed out that the magnetic properties of exchange coupled (EC) systems are strictly related to the magnetic behavior of the AFM phase. So, a deep knowledge and understanding of that is mandatory in the study of EB. However, it is usually difficult to directly access the magnetic behavior of an AFM phase due to the inherent weakness of the magnetic response of AFM materials. However, the soft NiFe that is exchange coupled to the AFM phase can be considered as a tool to gain an insight into the magnetic properties of the IrMn layer. In the following sections we present a preliminary characterization of exchange coupled systems that motivated us to develop an original measurement protocol to understand the role of the AFM layer in our samples. All the experimental results obtained, and presented in the next sections of this chapter, guided us in the formulation of a phenomenological model, illustrated in detail in section 4.2.4, that accounts for the magnetothermal behavior of the AFM phase.

4.1 Exchange Coupling and Exchange Bias

As previously mentioned in this thesis, we exploited a $\text{Ni}_{20}\text{Fe}_{80}$ (NiFe) alloy as FM phase, and a $\text{Ir}_{25}\text{Mn}_{75}$ alloy as AFM phase, to produce exchange coupled bilayers via dc-magnetron sputtering. The samples were grown in a 0.5 Pa Ar atmosphere and in the presence of a static magnetic field, $H_{\text{dep}} = 400 \text{ Oe}$, necessary to properly set the exchange coupling anisotropy along a preferential direction. The different bilayers considered are grown on a silicon substrate, with a 5 nm -thick Cu layer as both underlayer and overlayer, to favor the polycrystalline growth of the AFM phase and prevent the bilayer from oxidation.

The samples we studied are divided in two series of samples, A and B, with constant thickness of NiFe layer of 5 nm , but different values of thickness of the IrMn layer, generally referred as t_{AFM} . The capital letter A (B) is a convention to address the case in which the AFM grows “A”fter (“B”efore) the FM in the layer stack sequence. We considered these two opposite stacking sequences, B and A, in view of exploiting such bilayers as electrodes in spin valve systems, the reference and sensor layer respectively (see figure 4.1).

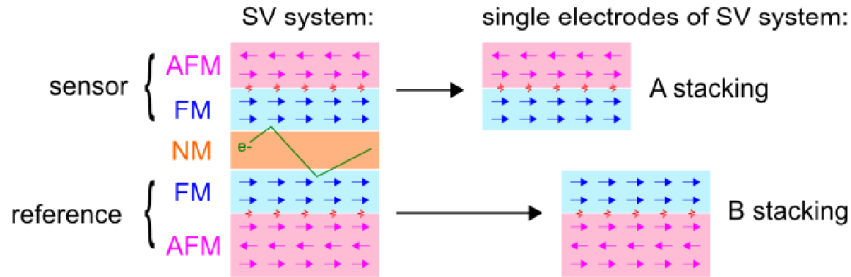


Figure 4.1: Spin valve system (SV) and the two (A and B) sample stacking sequences considered in our study.

With reference to t_{AFM} , the samples have been labeled as A3, A6, A10 and B3, B6, B10. The comparison between the A and B stacking is intended to highlight

which is the effect of the NiFe phase, grown either below or above the AFM phase, on the magnetic configuration of the AFM layer and on the strength of the exchange coupling. We changed the AFM thickness t_{AFM} to reveal the best condition for developing the EB effect.

With respect to the classical hysteresis loop measured on a thin film of 5 nm thick NiFe alloy that shows low coercivity at room temperature, $H_C \sim 5\text{ Oe}$, and no bias effect $H_{ex} = 0$, the coupling with an AFM layer, such as a 10 nm thick IrMn alloy, results in a shift of about $H_{ex} \sim 240\text{ Oe}$, coercivity $H_C \sim 90\text{ Oe}$ and a clear evidence of the established uniaxial anisotropy. From the master formula $H_{ex} = -\frac{J_{FM-AFM}}{M_{FM}t_{FM}}$ it is possible to estimate the intensity of the exchange coupling to be $J_{NiFe-IrMn} \sim 0.1\text{ erg/cm}^2$, for the considered sample with 5 nm -thick NiFe layer. The calculated value is a reasonable value for AFM/FM system based on IrMn, in fact it collocates in the observed range $0.01 \div 0.19\text{ erg/cm}^2$ reported in literature [3].

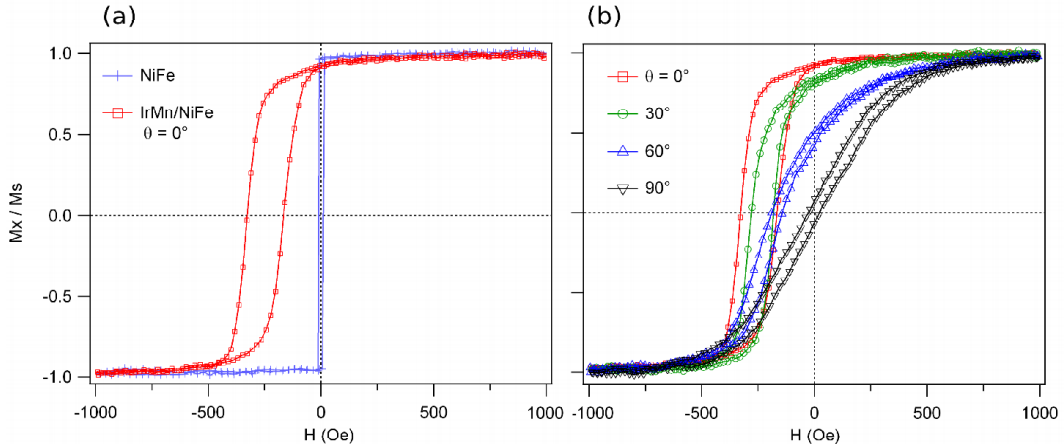


Figure 4.2: Hysteresis loops measured for the sample IrMn[10 nm]/NiFe[5 nm] along the unidirectional anisotropy axis, $\theta = 0^\circ$ compared to the classical hysteresis loop measured for 5 nm -thick NiFe (a), and at other directions, $\theta = 30, 60, 90^\circ$ (b).

In figure 4.2(a), we show the hysteresis loop measured along the direction of application of the external field exploited to set the unidirectional anisotropy during the deposition, namely the external field and the unidirectional exchange anisotropy

form an angle $\theta = 0^\circ$, we measured it with MOKE magnetometry in longitudinal configuration (see section 3.2 for a brief description of this technique). In figure 4.2(b) the same hysteresis loop is compared to other hysteresis loops measured at different orientations, namely varying the angle θ between the external measuring field and the direction of the unidirectional anisotropy. As we increase θ from 0° to 90° we observe a decrease of the H_{ex} to zero value, and the hysteresis loop gradually transforms in the hysteresis loop associated to hard axis in classical Stoner-Wohlfarth model.

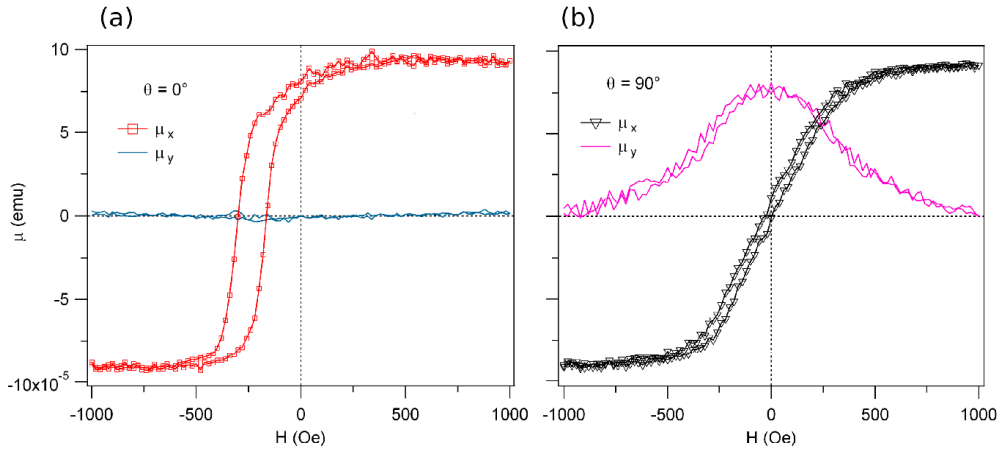


Figure 4.3: Hysteresis loops measured via VSM for the bilayer IrMn[10 nm]/NiFe[5 nm] along the unidirectional anisotropy axis, $\theta = 0^\circ$ (a), and at orientation $\theta = 90^\circ$ (b).

On the same sample, a more detailed analysis of the in-plane components of the magnetic moment μ , parallel (μ_x) and orthogonal (μ_y) to the direction of the applied external field, has been performed by Gaspare Varvaro et al., from “Istituto di Struttura della Materia” CNR Rome, exploiting a vibrating sample magnetometer (VSM). For the case $\theta = 0^\circ$ they observed that the magnetic moment μ of the sample does not show any component along the y direction (see figure 4.3(a)). As we do not expect any component along the out-of-plane direction as well, due to shape anisotropy of thin film structures, we concluded that the magnetization, to achieve

its reversal, breaks into magnetic domains at the coercive fields. They also provided measurements, in quantitative terms, for the μ_y component at other several different values of θ . If the external field is applied at $\theta = 90^\circ$, they observed two identical branches for μ_y in accordance with the coherent rotation of the magnetization inside the half-plane delimited by the direction of the applied external field that contains the unidirectional easy axis (see figure 4.3(b)).

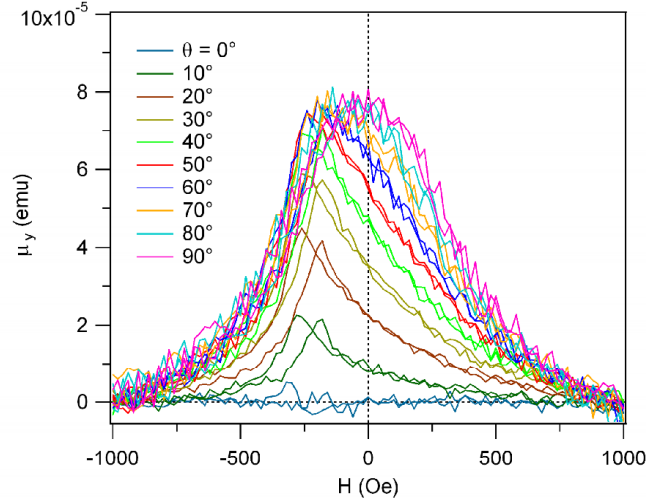


Figure 4.4: Transversal component of magnetic moment μ_y , measured via VSM, for the bilayer IrMn[10 nm]/NiFe[5 nm] at several orientation θ of the applied external field with respect to the unidirectional anisotropy axis.

As we progressively deviate from the case $\theta = 0^\circ$, in correspondence to other intermediate values of θ such as $\theta = 10^\circ, 20^\circ, \dots$ (see figure 4.4), during the magnetization reversal the magnetic moment reveals its tendency to rotate in such an half-plane, as it gradually shows increasing values of its y -component suggesting the gradual transition of the reversal process from the case in which it is obtained by domains formation to the one in which the magnetization rotates coherently.

4.2 Magnetothermal Behaviour of EC bilayers

As a preliminary characterization of the magnetothermal behaviour of exchange coupled bilayers we studied the reversal of the magnetization at different values of temperature for the two series A and B. From the hysteresis loops, we have derived the thermal dependence of the exchange field H_{ex} and of the coercivity H_C .

Both H_{ex} and H_C as functions of T for A3, A6, and A10 samples are shown in figure 4.5. At $T = 300\text{ K}$, no EB effect is observed in A3 (it appears below $T = 250\text{ K}$), whereas $H_{ex} \sim 100$ and 155 Oe in A6 and A10, respectively; H_C varies between 15 and 60 Oe (the largest value is measured in A6). With decreasing T from room temperature, both H_{ex} and H_C increase, especially for $T < 100\text{ K}$, suggesting a change of magnetic regime (in sample A3, the effect is clearly visible with reference to H_C and less evident for H_{ex}). At $T = 5\text{ K}$, $H_{ex} \sim 1000\text{ Oe}$ in A3 and $H_{ex} \sim 640\text{ Oe}$ in A6 and A10; H_C is in the $530 \div 640$ range, A3 showing the largest value.

When the B samples are considered (figure 4.6), for $T > 100\text{ K}$ both H_{ex} and H_C display values similar to those observed for the A samples. The main differences are observed for $T < 100\text{ K}$, where H_{ex} and H_C values obtained for A samples are higher than those measured on B of samples. This possibly suggests that the quality of the crystalline structure of the AFM layer of the B samples may be slightly different than that of the A samples.

The dependence on T of both H_{ex} and H_C for the samples having $t_{AFM} = 6\text{ nm}$ and $t_{AFM} = 10\text{ nm}$ appeared to be similar, irrespectively of the stacking, suggesting that no great differences should be expected for larger values of t_{AFM} .

Anyway, qualitative behaviours are similar in both series, and we decided to focus our attention on A samples because the differences between low and high temperatures appear more pronounced.

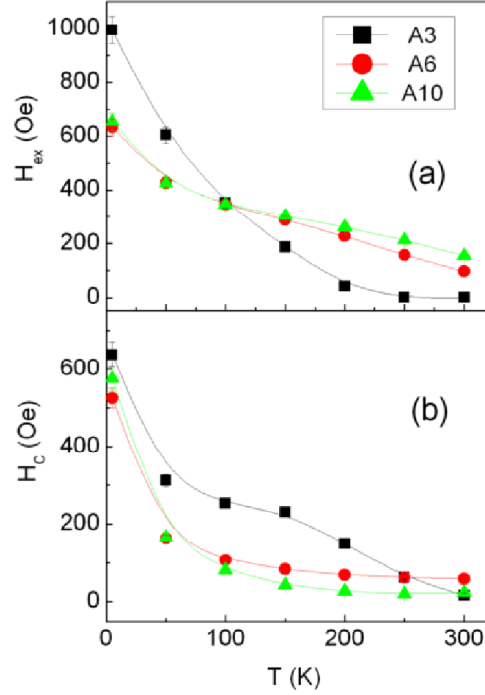


Figure 4.5: H_{ex} (a) and H_C (b) as functions of T for samples A3, A6 and A10. In most cases, the error bar is smaller or comparable to the size of the dots. Solid lines are guides to the eye.

4.2.1 How to disclose Energy Barriers Distribution

As we observed two high and low temperature regimes, we developed a new experimental procedure to deeply investigate the magnetothermal behavior of the AFM phase, in particular we decided to access the distribution of anisotropy energy barriers associated to the antiferromagnetic phase [36]. The procedure is based on the study of the magnetization of the NiFe/IrMn bilayers as a function of temperature, T , at different values of an external magnetic field, H_{inv} , antiparallel to the unidirectional easy axis imposed by the exchange anisotropy. This analysis allowed us to probe the effective distribution of anisotropy energy barriers of the antiferromagnetic phase as it is sensed by the ferromagnetic layer, and its results gave us important guidance to the formulation of a phenomenological model.

The measurement procedure, whose steps are basically illustrated in figure 4.7,

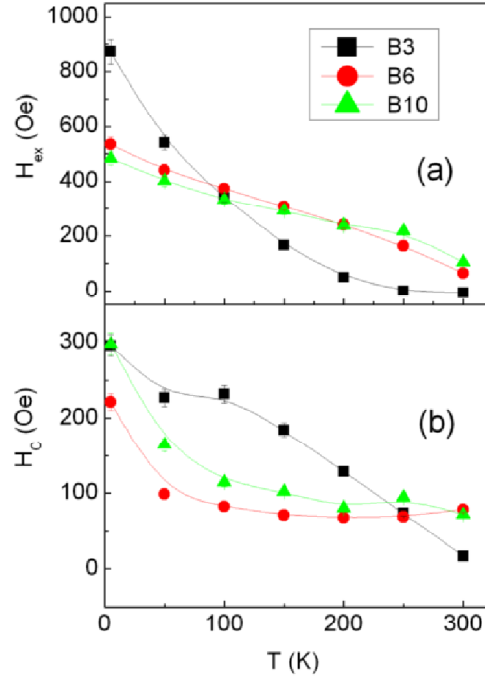


Figure 4.6: H_{ex} (a) and H_C (b) as functions of T for samples B3, B6 and B10. In most cases, the error bar is smaller or comparable to the size of the dots. Solid lines are guides to the eye.

is the following. The sample is inserted in the SQUID magnetometer at room temperature, brought to $T = 400\text{ K}$ (the maximum temperature) and cooled down to $T = 5\text{ K}$ in a saturating magnetic field applied along the direction of H_{dep} ; then, at $T = 5\text{ K}$, a negative magnetic field H_{inv} is applied and the magnetization M is measured as a function of T (heating rate = 3 K min^{-1}). The value of H_{inv} is chosen so as to fulfil two main requirements. First, it must be large enough to saturate the reference soft FM sample in the all temperature range; through such a requirement, we provide the system enough energy to overcome the energy barrier related to the magnetocrystalline anisotropy contribution of the FM phase, that is, to disentangle the inherent anisotropy due to the FM from the exchange anisotropy introduced by the coupling of the FM with the AFM phase. The second requirement is that, at $T = 5\text{ K}$, the value of the magnetization at H_{inv} is reduced by less than 10% with

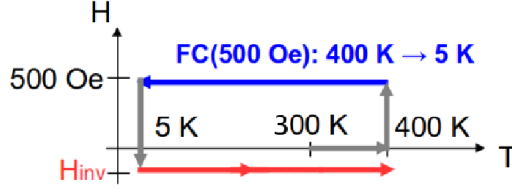


Figure 4.7: Steps employed to analyze the magnetothermal behavior of the sample, with cooling field $H = 500 \text{ Oe}$. As T increases from 5 K (red branch), we record the magnetization $M_{H_{inv}}$.

respect to M_S , in order to consider H_{inv} such as a perturbative term that doesn't interfere too much with the energy barriers distribution (in figure 4.10 it can be observed that a change to the energy barriers distribution is indeed determined by H_{inv}). Both conditions will be discussed afterwards in practical terms.

As T increases from 5 K we keep trace of the magnetization change in presence of H_{inv} . In fact, as temperature increases, the magnetization is expected to gradually decrease when the system, due to H_{inv} and the increase of thermal energy, gradually overcomes its energy barriers. Eventually the reversal of the magnetization is expected, if the anisotropy, at a certain temperature T^* , becomes low enough so as to fulfil the condition $H_{inv} > H_{ex}(T^*) + H_C(T^*)$.

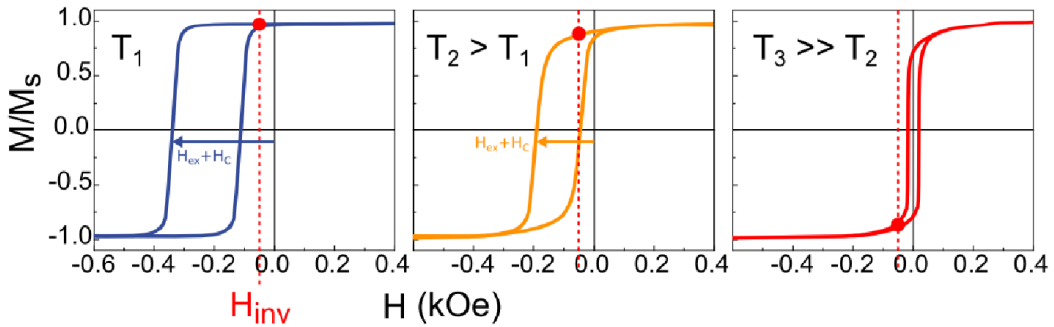


Figure 4.8: As T increases, $H_{ex} + H_C$ is expected to reduce its value, and the magnetization at H_{inv} , represented by the red dot, may eventually reverse, as it is schematically illustrated to happen at temperature T_3 .

The reversal of the magnetization in presence of H_{inv} may be understood in

terms of the hysteresis loops. Figure 4.8 schematically shows how we should expect the hysteresis loop to be in correspondence with different temperature values, T_3 being large enough to provide for the magnetization reversal in presence of H_{inv} .

We have studied the magnetization reversal through the before-mentioned procedure for samples A3, A6, A10. A first magnetic characterization of NiFe, the soft FM, revealed that at $T = 5\text{ K}$ it features $H_C \sim 20\text{ Oe}$, the maximum value observed, and squareness $M_r/M_S \sim 0.95$, so $H_{inv} > 20\text{ Oe}$ is needed to fulfil the first procedural condition. Typical loops at $T = 5$ and 300 K , relative to sample A6, are shown in the inset of figure 4.9 and, in an enlarged view, in the main frame.

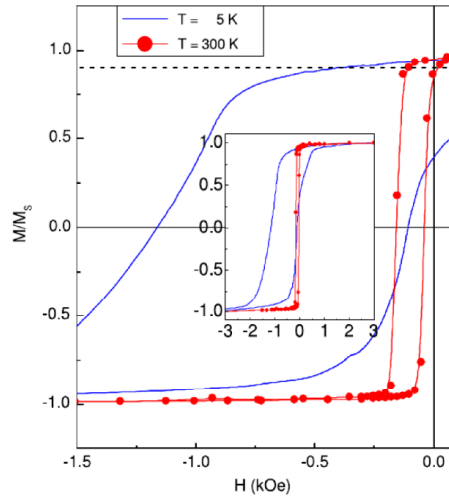


Figure 4.9: Enlarged view of magnetic hysteresis loops measured on sample A6 at $T = 5$ and 300 K (the complete loops are displayed in the inset). The curves are normalized to the saturation magnetization value M_S . A dotted horizontal line is drawn at $M/M_S = 0.9$ so as to identify the second requirement of the procedure.

In figure 4.9 a dotted horizontal line has been drawn at $M/M_S = 0.9$ to identify the upper limiting value of H_{inv} , being $\sim 300\text{ Oe}$ in modulus, to fulfil the second requirement of the procedure.

The result of the analysis performed on sample A6 is shown in figure 4.10(a) for $H_{inv} = -50, -100$ and -200 Oe , respectively, as those values fulfill the requirement $-300\text{ Oe} < H_{inv} < -20\text{ Oe}$. The curves are normalized to the magnetization value

at $T = 5\text{ K}$, M_0 (and in the following, we will indicate M/M_0 as $M_{H_{inv}}$). In the same figure, the normalized saturation magnetization of the reference NiFe sample, $M_{S_{Ref}}$, is also displayed as a function of T (open symbols; the measurement has been carried out in the saturating field $H = 50\text{ Oe}$), representing the natural decay of the FM magnetization with increasing temperature.

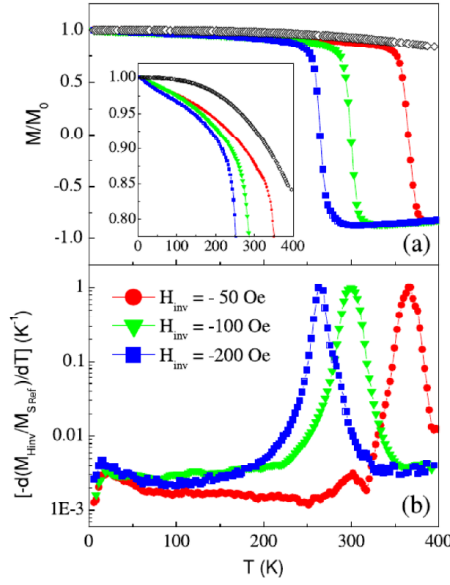


Figure 4.10: (a) M versus T measured on sample A6 at different values of H_{inv} ; the open-symbol curve refers to the reference NiFe sample, measured in a saturating magnetic field $H = +50\text{ Oe}$. All the curves are normalized to the magnetization value at $T = 5\text{ K}$, M_0 . (b) Temperature derivative curves $[-d(M_{H_{inv}}/M_{S_{Ref}})/dT]$ relative to sample A6, normalized to their peak value. $M_{H_{inv}}$ indicates the parameter M/M_0 shown in (a), relative to A6; $M_{S_{Ref}}$ is the parameter M/M_0 shown in (a), relative to NiFe.

The magnetization of sample A6 decreases with rising T and, at the temperature at which $H_{inv} > H_{ex} + H_C$, the magnetization reversal is experienced (hence, in figure 4.10(a), M/M_0 becomes negative in sign). The magnetization decrease is to be ascribed to the progressive reduction of the effective magnetic anisotropy acting on the NiFe layer, arising from the exchange coupling with the IrMn layer. In fact, as the intensity of the effective anisotropy - which keeps the NiFe magnetization along the positive field direction - reduces with rising T , an increasing fraction of

NiFe spins align accordingly to H_{inv} .

Recent studies on polycrystalline AFM films assume that the AFM layer consists of essentially non-interacting nanograins, that may be described by a distribution of magnetic anisotropy energy barriers [41, 42, 43, 44]. In line with this approach, we consider that $M_{H_{inv}}$ is connected to the distribution f of anisotropy energy barriers ΔE acting on the FM component by the relation we empirically derived:

$$\left(\frac{M_{H_{inv}}}{M_{SRef}} + 1 \right) \propto \int_{\Delta E_{crit}(T, H_{inv})}^{\infty} f(\Delta E) d\Delta E \quad (4.1)$$

where $\Delta E_{crit}(T, H_{inv})$ is the critical energy barrier, namely the energy barrier above which, at temperature T , the reversal of the magnetization can be induced by H_{inv} . In equation 4.1, $M_{H_{inv}}$ is divided by M_{SRef} so as to counterbalance the usual thermal decay of the magnetization of the FM phase, and the +1 occurs so as to make the left side of the equation positive. The thermal energy supplied by increasing T shifts the value of ΔE_{crit} across the barrier distribution. Therefore, for a fixed value of H_{inv} , the energy barrier distribution can be obtained by differentiating the $M_{H_{inv}}/M_{SRef}$ versus T curve, with respect to T .

Except for the presence of H_{inv} , this formalism is very similar to that concerning the thermal evolution of the remanent magnetization in an assembly of FM nanoparticles [45]. In that case, after the FM nanoparticles have been field cooled, the dependence of their remanence M_r versus T is measured. The derivative of $M_r(T)$ with respect to T provides the anisotropy energy barrier distribution of the system, which is strictly associated with the nanoparticle volume distribution, if they are magnetically non interacting, and may be perturbed by interparticle magnetic interactions [46]. With respect to that method, we actually probe the energy barrier distribution that can be associated to the AFM nanograins that constitute the AFM phase by studying how, due to the exchange coupling, it determines the insurgence

of an effective energy barrier distribution also in the FM component, ruling its magnetization reversal process. Hence, we obtain information on the anisotropy energy barrier distribution of the AFM as it is felt and mirrored by the FM component.

The temperature derivative curves $-d(M_{H_{inv}}/M_{S_{Ref}})/dT$ for three different values of H_{inv} are shown in figure 4.10(b). They show two main features: a large peak, centered at $T \sim 365\text{ K}$ for $H_{inv} = -50\text{ Oe}$ and shifting towards lower temperature with increasing $|H_{inv}|$; in fact the presence of H_{inv} implies the existence of an additional energy term favoring the magnetization reversal (i.e., shifting ΔE_{crit}), altering the energy barrier distribution to a certain extent. Depending on the value of H_{inv} the energy barrier distribution may appear more or less shifted, and as a consequence a low value must be preferred in accordance with the second requirement.

Below $T \sim 100\text{ K}$, the curves exhibit a tendency to increase slightly and there is a small peak at $T \sim 20\text{ K}$, whose position does not change significantly with H_{inv} . The shift of the main peak with changing $|H_{inv}|$ supports the picture of energy barriers associated with magnetic entities relaxing independently or weakly interacting; the presence of the small peak, independent of H_{inv} , is consistent with the stabilization of a collective frozen magnetic regime in the AFM phase with glassy features [48, 49, 50].

Both the high and low temperature regimes we have distinguished here, from the analysis of the data collected and reported in figure 4.5, find physical correspondence in the measured energy barrier distribution. The derivatives $[-d(M_{H_{inv}}/M_{S_{Ref}})/dT]$ for the three samples at $H_{inv} = -50\text{ Oe}$ are shown in figure 4.11.

The large peak is present in all the distributions and moves towards higher temperatures with increasing t_{AFM} (in fact, it is not fully visible for A10). It is to be noted that, due to the low value of H_{inv} , the peak temperature approximately corresponds to that where H_{ex} vanishes. This can be verified in the case of A3,

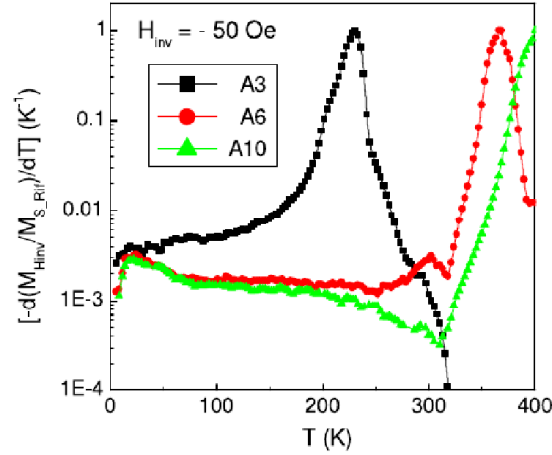


Figure 4.11: Temperature derivative curves for A3, A6 and A10 in $H_{inv} = -50 \text{ Oe}$, normalized to their peak value. The curve for sample A6 is the same also shown in figure 4.10(b).

where H_{ex} goes to zero between 200 and 250 K (figure 4.5(a)). For this sample, above $T \sim 320 \text{ K}$, the derivative curve shows no significant features that could be related to the barrier distribution of the AFM phase, since the FM layer no longer senses its presence. The features observed in the distribution of A6 at $T < 100 \text{ K}$, including the small peak at $\sim 20 \text{ K}$, are perfectly reproduced also in A10; in A3 they are more difficult to distinguish because of the larger population of energy barriers at low temperature.

4.2.2 Magnetic relaxation measurements

The existence of two different dynamical magnetic regimes has been also confirmed by magnetic relaxation measurements in a field H_{inv} . Sample A6 was cooled from 400 K down to 50 K in $H = 500 \text{ Oe}$; at $T = 50 \text{ K}$, $H_{inv} = -200 \text{ Oe}$ was applied and the variation of M with time was recorded. At the end of the measurement, $H = 500 \text{ Oe}$ was applied again, the temperature raised to 70 K, and a second relaxation curve in H_{inv} was measured. The procedure was repeated again at $T = 100, 150, 200$ and 300 K. The results are shown in figure 4.12; the curves are normalized to

the magnetization value at the beginning of the measurement, M_{time0} .

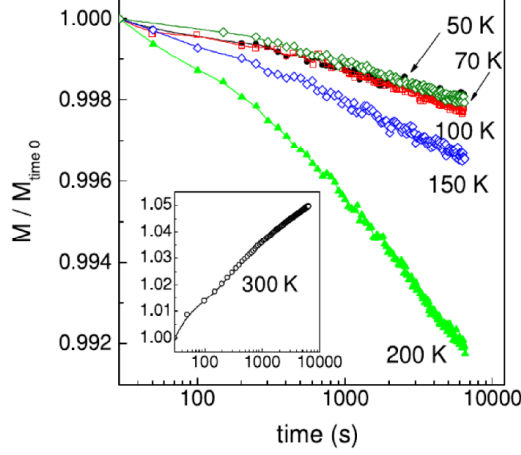


Figure 4.12: Magnetization versus time measured on sample A6 in $H_{inv} = -200 Oe$ at the indicated temperatures. The curves are normalized to the magnetization value at the beginning of the measurement, M_{time0} . The curve at $T = 50 K$ (full circles) is hardly visible because of the superposed curves measured at 70 and 100 K.

A very weak relaxation is experienced for $T \leq 100 K$, indicating that the spin dynamics is governed by long relaxation times with respect to the time scale of the experiment, consistent with the existence of a frozen state with high anisotropy energy barriers [50]. With rising T above 100 K, the dynamics of the system changes, and the extent of the relaxation increases. At $T = 300 K$, most of the NiFe spins are already aligned along H_{inv} at the beginning of the measurement (see figure 4.10(a)). Hence, M_{time0} is negative in sign and the normalized relaxation curve shows a growing trend (inset of figure 4.12), unlike those measured at lower temperature.

4.2.3 Structural investigations through HR-TEM

We performed a structural investigation via High Resolution Transmission Electron Microscopy (HR-TEM) to understand if there is any evidence of possible structural features that may be correlated to the two different dynamical magnetic regimes observed. HR-TEM analysis has been possible thanks to the collaboration of Andrea

Notargiacomo from “Istituto di Fotonica e Nanotecnologie” CNR Rome, and Gianni Barucca from “Dipartimento SIMAU, Università Politecnica delle Marche” Ancona, who provided for the preparation of the TEM lamella and the electron microscopy measurements, respectively. In figure 4.13 we show the main result of the structural investigations performed on a cross section of the B10 sample.

The structural analysis indicates that the “bulk” of the IrMn layer consists of nanograins with mean size $\sim 10\text{ nm}$, whereas the NiFe phase appears amorphous. In particular, the TEM analysis definitely reveals the existence of a structurally disordered IrMn region ($\sim 2\text{ nm}$ thick) at the interface between the FM phase and the bulk of the AFM layer where a lighter contrast is visible with respect to the amorphous NiFe.

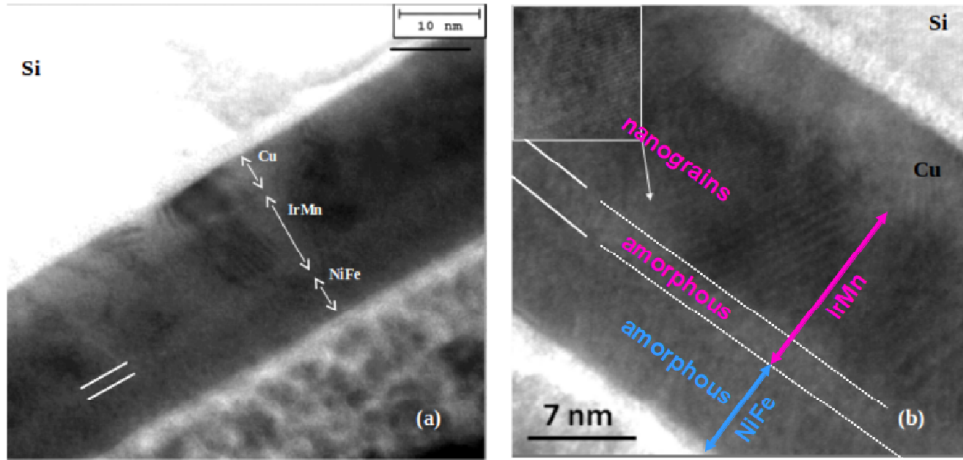


Figure 4.13: (a) TEM image of the IrMn/NiFe continuous film. The stack sequence is Si/Cu[5 nm]/IrMn[10 nm]/NiFe[5 nm]. The short white lines underline the existence of a thin amorphous region (2–3 nm thick) with a light contrast, located between the IrMn crystalline phase and the amorphous NiFe layer. (b) High resolution image of the sample. The inset in the top-left corner is an enlarged view of a IrMn grain. The parallel white lines are used as in (a).

Hence, we are convinced that the AFM entities giving rise to the large peak in the anisotropy energy barrier distribution are the nanograins in the bulk AFM. This conclusion is supported by the rough estimation of the size of these AFM entities

through the relation $V = 25 k_B T_B / K$, linking the critical volume V of spherical nanoparticles undergoing superparamagnetic relaxation, the blocking temperature T_B and the magnetocrystalline anisotropy K (for IrMn, $K_{IrMn} = 1.8 \cdot 10^6 \text{ erg cm}^{-3}$ [51], k_B is the Boltzmann constant). For instance, with reference to the sample A6, where the peak of the distribution is fully visible inside the temperature range, taking $T_B = 365 \text{ K}$, that is the temperature at which the large peak is centered, a size $\sim 9 \text{ nm}$ is calculated for a cubic approximation for the nanograin, in agreement with the value reported for the nanograins of IrMn layers of similar thickness [52] and close to the value observed by TEM analysis for the sample with 10 nm thick IrMn phase. Moreover, the peak shifts towards higher temperature with increasing t_{AFM} (figure 4.11): this behavior can be explained by considering that the thicker the AFM layer, the bigger the volume the nanograins may develop to, and, accordingly, higher values of the associated blocking temperatures may be expected.

As for the disordered AFM “interlayer”, the structural disorder observed may imply magnetic disorder and the insurgence of a spin-glass-like magnetic character, resulting in the onset of a frozen collective regime below a critical temperature [48, 53, 54].

4.2.4 The Phenomenological Model

In the light of the experimental results we have shown up to this point, we propose a phenomenological model to account for the magnetothermal behaviour of the IrMn/NiFe samples. Briefly, in our model, at temperature $T < 100 \text{ K}$, the interfacial IrMn spins are frozen in a glassy state and are collectively involved in the exchange coupling with the NiFe spins. At $T \sim 100 \text{ K}$ the collective state gradually breaks up; thus, above this temperature, only the interfacial IrMn spins which are tightly polarized by the IrMn nanograins, forming the “bulk” of the layer, are effectively

involved in the exchange coupling mechanism, and this is the reason why the FM layer still feels the presence of the bulk AFM nanograins, despite the presence of the disordered AFM “interlayer”. Due to that, for $T > 100\text{ K}$ the exchange coupling is ruled by the anisotropy energy barriers of the bulk IrMn nanograins, namely by the layer thickness.

According to the description given by Mydosh for a canonical spin glass, when the freezing temperature is approached from higher T , clusters of interacting spins develop out from the collection of paramagnetic spins [55]. These clusters may display a characteristic correlation length λ which rigidly couples together spins pointing along random anisotropy directions under the action of competitive magnetic interactions. Thus, the correlation length increases progressively with reducing T . The onset of the glassy state for the interfacial AFM spins, and hence the insurgence of a strong effective anisotropy, explains the observed increase in H_{ex} at low temperature.

Making explicit reference to the results reported for the samples A3, A6, A10, we will now discuss how they are understood according to this model. At low temperature the spins of this disordered AFM component are collectively frozen, and this is consistent with the very slow magnetic relaxation observed (figure 4.12) and with the presence of the small peak in the barrier distribution (figures 4.10(b) and 4.11). Their equilibrium configuration minimizes the energy associated with the mix of competing magnetic interactions they are subjected to, including the exchange interaction with the NiFe spins, on one hand, and with the blocked spins of the AFM nanograins, on the other hand (actually, due to the interface structural disorder, both the interactions are expected to be described by distributions of exchange coupling constants [57]). Hence, in all the samples, H_{ex} is maximum at $T = 5\text{ K}$, when the AFM interfacial spins are completely frozen (figure 4.5) and subjected to a high effective anisotropy, allowing them to exert a strong torque

action on the NiFe spins.

The concomitant great enhancement of H_C , with respect to the value measured in the reference NiFe sample ($\sim 20\text{ Oe}$), reveals the presence of AFM spins which can be dragged by the FM spins during the magnetization reversal, probably because they are subjected to a lower local anisotropy within the frozen state. In fact, dragged AFM spins are generally claimed to be the main cause of the magnetic hardening of the FM phase, often observed in FM/AFM systems [24]. In order to account for the different values of H_{ex} at $T = 5\text{ K}$ in the three samples, further considerations are needed. As regards the distributions of exchange coupling constants between the NiFe and the glassy phase and between the glassy phase and bulk AFM, we can assume that they are similar in all the samples: as the 5 nm -thick NiFe layer lies below the IrMn layer in the sample stack, we have no reason to expect that the degree of interface structural disorder is substantially different. Therefore, another element must be taken into account, namely the magnetic anisotropy of the bulk AFM nanograins. In fact, when the sample is cooled down to $T = 5\text{ K}$, if the anisotropy that blocks the spins of bulk AFM nanograins is relatively weak, their orientation may deviate, to some extent, from the anisotropy directions, in order to minimize the exchange energy with the interfacial frozen spins. Therefore, the final configuration of the interfacial frozen spins better fulfils the necessity of minimizing simultaneously also the exchange energy with the NiFe moments, compared with the case in which the bulk AFM spins are firmly blocked along their anisotropy directions. Among the three samples, the case of strong anisotropy of the bulk AFM spins is represented by A6 and A10, having the barrier distribution associated with the AFM nanograins centered at $T > 350\text{ K}$ (figure 4.11). In fact, at $T = 5\text{ K}$, A6 and A10 show approximately the same H_{ex} value ($\sim 640\text{ Oe}$, figure 4.5(a)). The case of weak anisotropy of the bulk AFM spins, leading to a much higher value

of $H_{ex} \sim 1000 \text{ Oe}$ at $T = 5 \text{ K}$, is that of sample A3, whose barrier distribution is centered at a temperature substantially lower ($\sim 230 \text{ K}$).

With increasing T above 5 K , the effective local anisotropy of the frozen interfacial AFM spins gradually reduces, which causes a decrease in both H_{ex} and H_C , in all the samples (figure 4.5). At $T \sim 100 \text{ K}$, the frozen collective state definitely breaks up, but the AFM interfacial spins do not enter the full paramagnetic regime - this event would definitely decouple the FM and AFM layers as regards exchange interaction - because of the polarizing action of the adjacent NiFe and bulk AFM spins. They undergo a sort of re-entrant transition into the AFM nanograins. Regarding the EB effect, it is to be expected that, in this new magnetic regime above 100 K , only the interfacial AFM spins tightly anchored to the spin lattice of the bulk AFM nanograins, i.e. those which re-enter into the AFM state from the frozen regime, contribute effectively to H_{ex} , provided that their spatial orientation still minimizes the exchange energy with the NiFe spins. The remaining fraction of interfacial spins, loosely bound to the spin lattice of the bulk AFM nanograins, will follow the NiFe spins during the magnetization reversal and will neither be involved in the EB effect nor contribute substantially to H_C . Experimental results from Ohldag et al., who performed x-ray magnetic circular dichroism measurements at room temperature on different exchange bias sandwiches [56], appear to be in agreement with our picture as they estimated that only a fraction of the spins at the FM/AFM interface, as small as 4%, was tightly locked to the AFM lattice and caused EB.

So, at $T > 100 \text{ K}$, the EB effect persists as long as some AFM nanograins exist, with a tightly blocked spin lattice, able to anchor some interfacial spins. At the temperature at which all the anisotropy energy barriers associated with the AFM nanograins are so low that the AFM spins are unable to exert the torque action on the NiFe spins, the only hint of the presence of FM/AFM exchange coupling may be

a larger H_C , compared to the value of reference NiFe. Even this last trace disappears when all the AFM energy barriers are overcome by thermal activation, namely when the AFM nanograins enter the superparamagnetic regime. This thermal evolution of H_C is clearly visible in sample A3 (figure 4.5(b)). Between 5 and 100 K, H_C versus T shows a strong decrease associated with the reduction of the effective anisotropy of the interfacial spins within the frozen state, previously discussed. Then, up to 150 K, H_C is larger than 200 Oe and stays approximately constant, whereas H_{ex} decreases (figure 4.5(a)). This last behavior reveals that, with rising T , the fraction of AFM nanograins able to sustain the EB effect progressively reduces, but AFM nanograins exist whose spin lattice is dragged by the NiFe spins during the magnetization reversal, enhancing H_C . At $T = 250$ K, no EB effect is observed, but H_C is still larger than 60 Oe. Only at $T = 300$ K, $H_C \sim 15$ Oe, a value much closer to that of the reference NiFe sample at this temperature (~ 5 Oe). Hence, although the exchange coupling mechanism involves directly the interfacial AFM spins, the bulk AFM nanograins play a crucial role: they sustain the magnetism of the interfacial spins and the thermal decrease of H_{ex} as well as of H_C is closely correlated to the crossing of their anisotropy energy barriers. In particular, at $T = 300$ K, the sample retaining the highest H_{ex} is A10 (figure 4.5(a)), since the main peak of the AFM barrier distribution is centered at higher temperature compared to A3 and A6 (figure 4.11). Moreover, it is worth noticing that, for $100 \text{ K} < T < 300 \text{ K}$, H_C in A6 and A10 is always lower than in A3, whereas H_{ex} is higher (figure 4.5). This indicates that in A6 and A10 the AFM nanograins, having higher anisotropy energy barriers, are more efficiently involved in the EB mechanism, whereas the effect of magnetic hardening of the NiFe phase is depressed.

4.3 Aging and Training Effects in Bilayers

The stability over time of the exchange bias properties in exchange coupled FM/AFM systems is an important issue to be taken into account for practical use, because the electronic devices based on such a system need to be stable and reliable. Despite this need, the dynamical environment in which we find our systems appears to change their exchange bias properties, as suggested to be by the magnetic relaxation measurements reported in section 4.2.2. In fact, we observed that long-term relaxation processes take place even in the absence of an external magnetic field applied, and we referred to this phenomenon as the aging effect, and also that an irreversible decrease in the H_{ex} occurs with the number of hysteresis loops we measure, the so called training effect. Although the origin of both phenomena is still under debate, in literature they have often been reported in systems based on exchange coupling. Both aging and training effects have commonly been ascribed to the same mechanism, dealing with a gradually reconfiguration of AFM entities seeking local energy minima, but different models have been formulated so far [69, 74, 75].

In the following, we report about these phenomena through data collected from magnetics measurements performed on samples both in as prepared condition and after post-deposition treatment solutions that have been proposed to partly limit the changes that we usually observe.

4.3.1 As Prepared Samples

The training effect is generally reported to be distinctive in polycrystalline AFM, but very small when the AFM layer is in form of single crystal [70, 71]. In our case, the IrMn layer is expected to grow polycrystalline with a thin amorphous structure at the interface with the NiFe layer, so the training effect is expected.

In order to investigate both the training and aging effects, we focused our atten-

tion on a sample with $t_{AFM} = 10\text{ nm}$, because the highest values of H_{ex} are measured at room temperature in this case, and that belongs to the B stacking sequence series, namely with composition Si/Cu[5 nm]/IrMn[10 nm]/NiFe[5 nm]. The B stacking sequence is preferred because of the larger sensitivity of MOKE magnetometer when the FM layer is a surface layer, and of the need to be sensitive to variation of a few Oersted in the measurements of H_C and H_{ex} .

So, we performed consecutive magnetic measurements on this sample, and we repeated the analysis at different times to assess whether the effect of aging is present or not. The consecutive magnetization loops ($n = 1, 2, \dots$) were collected at room temperature both with SQUID and MOKE magnetometer. After the measurements, the external field was removed and the sample was left in its remanence state. Then, the sample was measured again after waiting for a time interval equal to t_w , of about 1 week; overall, the procedure was repeated several times, at $t_w = 1\text{ week}, 2\text{ weeks}, \dots$

In figure 4.14(a) the MOKE magnetization loops, normalized to the saturation value, M_S , corresponding to $t_w = 0$ are presented; in figure 4.15, the corresponding H_{ex} dependence on n is displayed (i.e. the training effect) in blue for the case $t_w = 0$, and a decrease of H_{ex} as a function of n is observed, as expected (and similar results are obtained from SQUID magnetometry). Moving from $t_w = 0$ up to $t_w = 3$ weeks, the H_{ex} dependence on n progressively diminishes. In addition, if we focus on the $H_{ex}(n = 24)$ dependence on t_w , namely on the H_{ex} value measured during the last hysteresis loop of the measurement series, when the variation of H_{ex} with n is almost negligible, we observed that $H_{ex}(n = 24)$ increases with t_w and reaches the highest value at $t_w = 3$ (see figure 4.14(b)); this is the experimental evidence of the aging effect. The coercive field H_C is reported to decrease as well in regards of the training effect, as it was observed that it is the descending branch of the hysteresis loop that

shifts to the right with n rather than an overall shift of the hysteresis loop (see figure 4.14(a)). However, H_C appears to be quite the same irrespective of t_w , namely it does not vary significantly with time.

So, NiFe/IrMn systems show relaxation effects on two different time scales, a short one (training) and a long one (aging), and leaving the system in its remanence state, that is nearly equivalent to the saturation state (see figure 4.14(a)), increases the H_{ex} value.

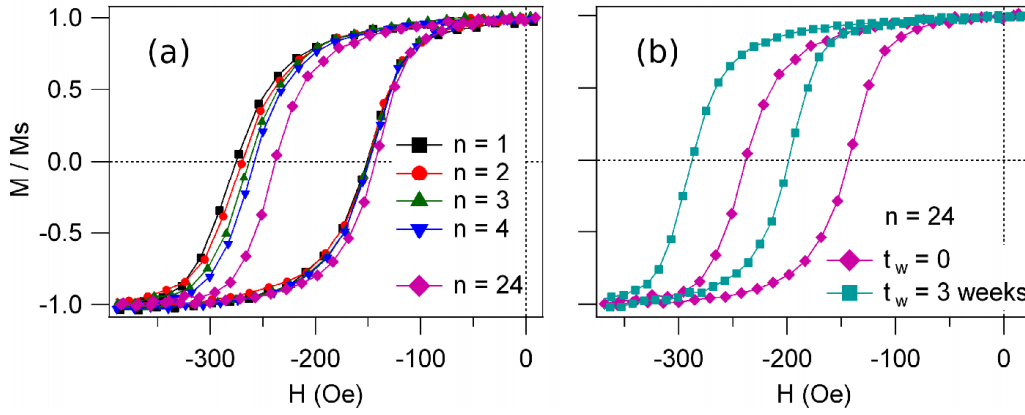


Figure 4.14: (a) Consecutive magnetization loops ($n = 1, 2, 3, 4$ and $n = 24$) recorded for $t_w = 0$ by MOKE magnetometry. (b) $n = 24$ magnetization loops recorded at $t_w = 0$ and $t_w = 3$ weeks.

SQUID measurements confirmed this trend, and added important information about the aging effect, namely that no significative change in the absolute value of the magnetic moment is observed. This finding rules out the hypothesis of oxidation contributing to the progressively increase of the H_{ex} with time. In fact, in case the degree of oxidation increased, the thickness of the FM would diminish, and an increase of the H_{ex} would be expected as $H_{ex} \propto \frac{1}{t_{FM}}$.

For what concerns training effect, it is mostly admitted that consecutive cycling of the FM triggers the spin configurational relaxation at the interface towards equilibrium. Based on the idea that training is related to irreversible reconfigurations at the FM/AFM interface with each hysteresis cycle, modifying H_C and

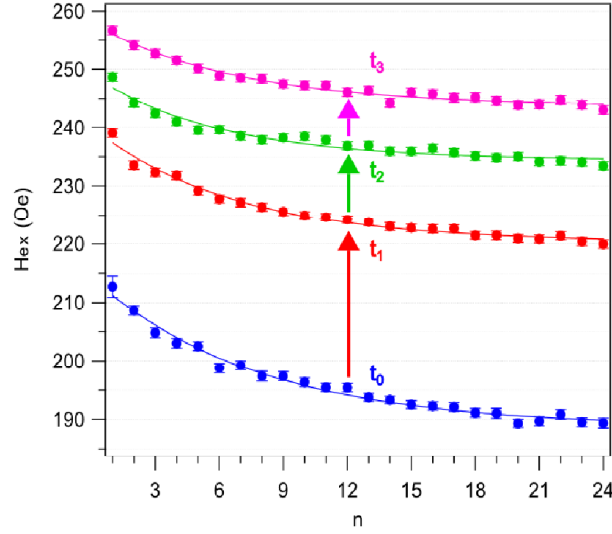


Figure 4.15: H_{ex} dependence on n (the hysteresis loop number) is presented for different measurement time t_w ; data extracted by MOKE magnetometry. A set of vertical arrows displays the H_{ex} aging effect for a fixed n value. Each fit curve is the sum of two exponential curves (see the text for a discussion).

H_{ex} accordingly, different approaches were developed to study this phenomenon. Some considered an implicit sequence equation that determines how the training effect manifests in conventional EB systems in relation to a generic change of the interfacial AFM magnetization [58, 59, 60], while others suggested to consider two particular AFM components that are expected to behave differently, in particular they distinguished between fixed and rotatable AFM interfacial spins [66], similarly to the model we proposed in section 4.2.4. Hence, we decided to describe the data collected in figure 4.15 with the following expression to simulate the relaxation of the exchange bias field H_{ex} as a function of n :

$$H_{ex}(n) = H_{ex}(\infty) + A_f \exp(-n/P_f) + A_r \exp(-n/P_r) \quad (4.2)$$

where $H_{ex}(n)$ is the exchange bias of the n th hysteresis loop, $H_{ex}(\infty) \equiv H_{ex}(n \rightarrow \infty)$ the equilibrium exchange bias value. The A parameters, for both fixed and rotatable AFM interfacial spins, have dimensions of magnetic field (Oersted) while

the P 's are dimensionless parameters and resemble a relaxation time, where the time continuous variable is replaced by a discrete variable, namely, the hysteresis index n .

The parameters obtained using equation 4.2 to fit to the H_{ex} data collected at t_0 were similar to the ones reported in [66], with the fixed component that appears to relax about 10 times slower as compared to the other one. Fit to data collected at subsequent t_w did not give further insight because of the larger uncertainties associated.

4.3.2 Post-Deposition Treatment

We made resort to two different experiments to assess whether the exchange coupling long-term relaxation processes are dependent on temperature, as some reported to be [67]. The first post deposition treatment consisted in keeping a sample at a very low temperature $T \sim 77\text{ K}$; we made resort to liquid nitrogen (LN_2), inside which the sample was kept for a fortnight and replaced there after each magnetic measurement.

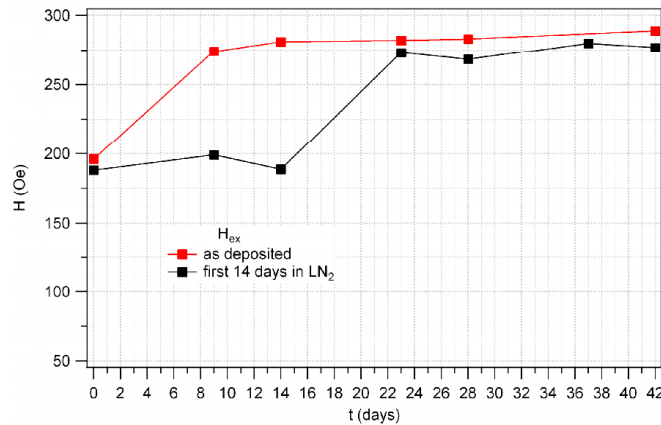


Figure 4.16: Time dependence of the exchange bias field H_{ex} for twin samples with composition Si/Cu[5 nm]/IrMn[10 nm]/NiFe[5 nm] kept either in air at room temperature (red data) or in both LN_2 for 14 days and at room temperature (black data).

We observed no change of magnetic properties during the period of time in which the sample was kept in LN_2 , whereas the usual aging effect began as soon as the sample was removed from the LN_2 , with the same intensity reported for a reference twin sample kept at room temperature (see the comparison between black and red curves in figure 4.16). In figure 4.16 the two curves of H_{ex} in function of t are compared, the H_{ex} values are calculated from the hysteresis loops measured at room temperature, in correspondence of a large value of n , in order to neglect the contribution of the training effect (the latter in fact is still present, with similar intensity, for the sample kept in LN_2).

The temperature $T \sim 77 K$ must be then considered low enough to completely prevent the long-relaxation process to take place, in other terms, the time scale of the rearrangements processes was too large to be appreciated, and the aging effect appears not to be activated at the temperature considered in this first experiment.

This result seems in accordance with the trend of the relaxation curve at similar temperature reported in figure 4.12. In that case it is to be pointed out that the sample AFM layer was $6 nm$ and the sample was kept in presence of a inverse field H_{inv} . For what concerns coercivity we report no differences, being $H_C \sim 120 Oe$ for both the samples considered, namely the one kept in LN_2 and the twin reference sample stored at room temperature.

In a second experiment, a sample with the same composition, after being produced, was annealed at $T = 400 K$, and field cooled down to room temperature with a $3 T$ cooling field and then measured over time at room temperature: no change in H_{ex} against time was observed, corresponding to a significant improvement of the achieved thermal stability. However, when the sample was immersed and stored in a negative external field large enough to provide for the complete reversal of the magnetization, we lost the acquired stability against time: the forced antiparallel

alignment of the NiFe magnetization, with respect to the unidirectional anisotropy direction, guided the re-alignment of interfacial AFM spins, by means of the exchange coupling with the FM spins [67, 68, 69]. This process is responsible for the decrease of H_{ex} (compare the H_{ex} behaviour during both a1 and a2 time intervals in figure 4.17).

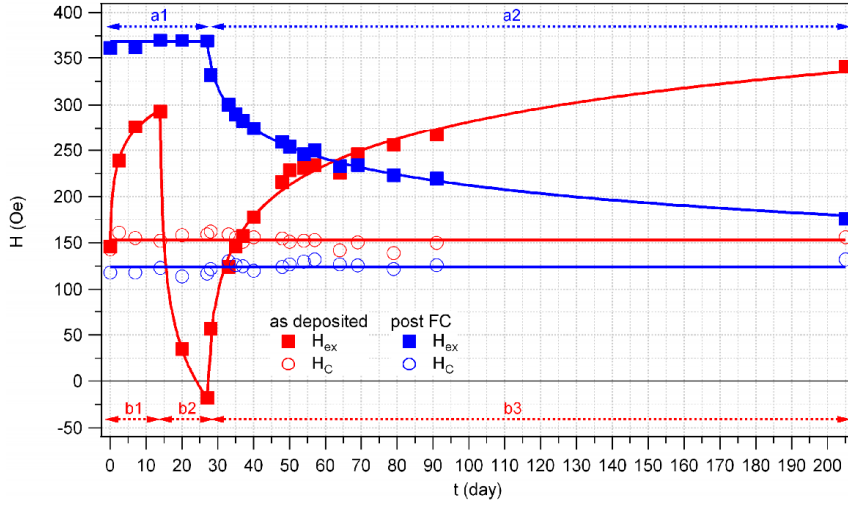


Figure 4.17: Time dependence of the exchange bias field H_{EX} and coercivity H_C for twin samples with composition Si/Cu[5 nm]/IrMn[10 nm]/NiFe[5 nm] both stored in air; red data refer to the as prepared state of the sample, blue data refer to the sample treated with post deposition field cooling. Fit curves are logarithmic curves.

The usual aging effect is observed for the twin sample stored in air in its remanence state (red data in figure 4.17) during time interval b1; then we submitted this sample to the same storage in a negative field and observed again a decrease of H_{ex} , which reduced to negative value (corresponding to shift to the right of the hysteresis loop).

Interestingly, we report a constant difference of $\sim 30 Oe$ in the coercive fields between the sample annealed and its corresponding twin reference sample, this difference is illustrated in figure 4.17 with the help of two horizontal line fitting the H_C values measured for the two samples. Apart from this observed variation, no other change in the shape of the hysteresis loops can be appreciated, as illustrated

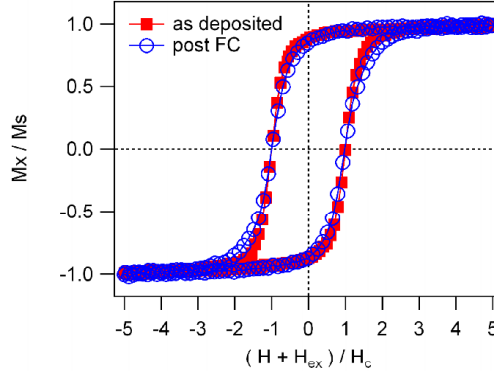


Figure 4.18: Shape comparison between hysteresis loops measured for the as deposited sample (squares) and field cooled one (open circles): the hysteresis loops appear similar in regards of squareness and symmetry properties.

in figure 4.18. The coercitive field decrease, that is obtained with the annealing procedure, is understood in terms of the model proposed, if we consider a reduction in number of interfacial AFM spins that can be dragged by the FM magnetization during its reversal, responsible for the coercivity, and that can establish a link to the AFM stable nanograins thanks to the thermal energy supplied by the annealing procedure.

Making resort to a logarithmic law to best approximate the measured time dependent increase and decrease of H_{ex} , according to what is proposed in references [72, 73, 74], we easily calculated the rate of change of H_{ex} with time. Focusing the attention on the time intervals during which the anti-parallel alignment of the FM magnetization is induced, namely a2 and b2 for the annealed sample and the as prepared twin sample respectively, we observed a significant difference between the two initial rates of change, that is between the two rates of change calculated at the beginning of the two time intervals for the two samples. In details, for the as-deposited sample this value is about ~ 15 times larger than the corresponding initial rate of change of the annealed counterpart, and we ascribed the smaller value of the last to the greater thermal stability achieved via the annealing procedure.

4.4 Modeling the Exchange Bias Interaction with OOMMF

In the following section we will briefly introduce the software we exploited for performing the micromagnetic analysis beside the parameters used in the calculations, and in section 4.4.2 we will illustrate the model system we proposed to account for the exchange bias effect on a simulated ferromagnetic layer.

4.4.1 OOMMF working principles

Micromagnetic simulations were performed using the three-dimensional Object Oriented MicroMagnetic Framework (OOMMF) software, which applies the Landau Lifshitz Gilbert (LLG) equation to simulate the spin configuration and compute the energy and magnetization of nanostructures [76]. It is a finite difference code and it models a magnetic system using a 3D mesh made of rectangular cells; all the cells have the same size and the magnetization within each cell is supposed to be uniform. LLG equation is:

$$\frac{d\vec{M}}{dt} = -\gamma\vec{M} \times \vec{H}_{eff} - \frac{\gamma\alpha}{M_s}\vec{M} \times (\vec{M} \times \vec{H}_{eff})$$

where $\vec{M}$ is the pointwise magnetization (A/m), H_{eff} is the pointwise effective field (A/m) defined as the energy (E) first derivative $\vec{H}_{eff} = \mu_0^{-1} \frac{\partial E}{\partial \vec{M}}$, γ the gyro-magnetic ratio ($m/(As)$), and α the damping coefficient (dimensionless).

In particular, what we are most interested in is the possibility to simulate the reversal of the magnetization under an external magnetic field in AFM/FM nanostructures. To this purpose, we let OOMMF calculate equilibrium spin configuration for each value of the external field applied, by solving LLG equation, after the system

has been defined. To define our system, first of all we defined the x and y axes to be in the plane of the film and the external field to be applied along the x direction. In the x and y directions the size of the cells was of 10 nm , equal to the size of the AFM grains, while in the z direction it was of 5 nm , equal to the NiFe layer thickness. For the NiFe phase, we set the saturation magnetization $M_S = 800\text{ emu/cm}^3$, stiffness constant $A = 13 \cdot 10^{-7}\text{ erg/cm}$, and negligible magnetocrystalline anisotropy; these values are the ones usually exploited in the simulation of NiFe alloy. We imposed that, at the beginning of each calculation, the NiFe magnetization was parallel to the applied magnetic field, which is the condition effectively present in a real loop measurement, when we start measuring with a field large enough to saturate the magnetization along its direction.

For simulation of the IrMn phase problems arise, since the OOMMF code does not allow an antiferromagnetic material to be truly reproduced, so we tried to simulate the presence of the AFM layer interacting with the FM as explained in the following dedicated subsection.

4.4.2 How To Simulate the Antiferromagnet

In the simulated FM/AFM system, the AFM component was simulated by a FM layer to which we assigned very peculiar characteristics. Its saturation magnetization is chosen to be significantly lower than that of the simulated NiFe, in order to make dipolar interaction, that would favor antiparallel alignment between the magnetizations of the two phases, negligible in comparison with the exchange interaction. Its magnetocrystalline anisotropy coefficient K_{AFM} and direction d_{AFM} were chosen on a cell basis. The d_{AFM} axes were in the plane of the layer. The exchange interaction at the AFM/FM interface was assumed to be ferromagnetic and the coupling constant was $A_{ex} = 10^{-7}\text{ erg/cm}$. Anyway, we consider the choice of the exchange

coupling constant A_{ex} to be an oversimplification, since in this way a fixed, positive, value is used instead of a distribution of exchange coupling constants, that is likely to originate from the interface structural disorder [40]. The appearance of H_{ex} was obtained after a variable percentage p of the cells constituting the AFM phase, uniformly distributed within the system, was given a high K_{AFM} value ($2 \cdot 10^7 \text{ erg/cm}^3$) and d_{AFM} was randomly assigned so as to have a positive projection on the x axis. The high K_{AFM} was set so as to forbid the spin rotation under the external field and, hence, to simulate the presence of blocked AFM grains able to sustain the EB mechanism working as pinning centers. A small $K_{AFM} = 2 \cdot 10^4 \text{ erg/cm}^3$ was assigned to the remaining cells in order to simulate the existence of AFM grains whose spin lattice easily follows the NiFe magnetization during its reversal, or ensemble of spins that are not anchored to AFM stable grains and, accordingly, do not contribute to exchange bias. Even if in the previous sections the pinning centers were considered to be single AFM spins at the interface, we consider the model proposed for the simulation to be the best compromise, considered the calculation time required for the simulation.

An example of a portion of the simulated AFM layer, corresponding to a percentage of pinning center $p \sim 11\%$, is illustrated in figure 4.19, where the presence of a negative external field H_{meas} determines the reversal of the FM moments (the neighboring simulated FM layer is not shown) and of all moments with low anisotropy that belong to the AFM phase.

The simulated hysteresis loop that corresponds to the distribution of pinning centers illustrated in figure 4.19, is shown in figure 4.20(a). In figure 4.20(b) H_{ex} values, calculated from the same distribution, but considering different values of the exchange coupling constant A_{ex} , appear to change linearly with A_{ex} .

We designed the OOMMF micromagnetic input format file (also known as mif

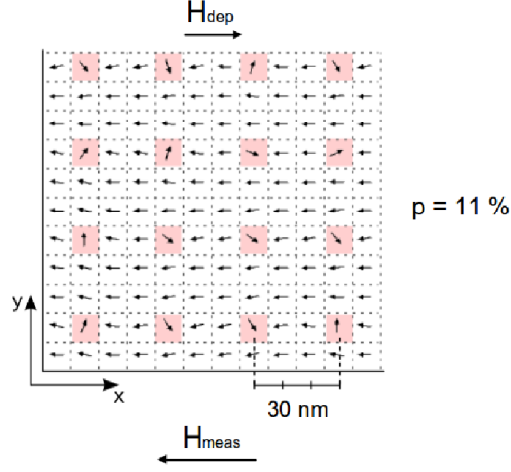


Figure 4.19: Simulated AFM layer: the cells with a strong anisotropy are highlighted in red and their magnetic moments are not affected by the negative field of measure

file) so as to give the users the chance to vary the pinning centers periodicity in square or rectangular lattice, and even the extent of every pinning center can be modified. In the example reported in figure 4.19 the periodicity of the pinning centers is 30 nm , that is 3 micromagnetic cells, along both x and y directions, and the array of pinning centers results in a square lattice. In this case the extent of every pinning center is the lowest possible, namely it corresponds to a single micromagnetic cell.

Inside each pinning center with larger extent, $n \times n\text{ cell}^2$ with $n > 1$, it is also possible to make the moments share a common direction in order to simulate correlated areas inside the AFM layer that may account for modeling a real system at low temperature regime, when the appearance of a correlation length is expected (see subsection 4.2.4).

Other basic examples of square array with equal periodicity but increasing dimension of the pinning centers are reported in figure 4.21.

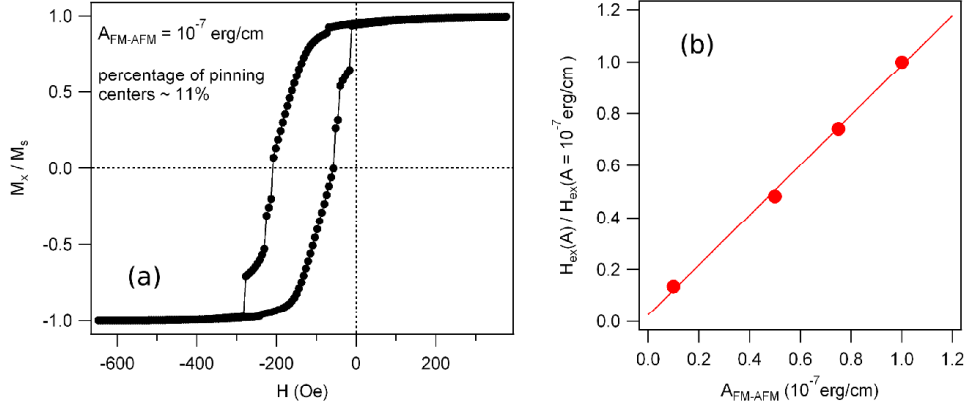


Figure 4.20: Simulated hysteresis loop corresponding to a pinning center distribution with percentage of pinning centers $p \sim 11\%$ (a). The same distribution is exploited to assess the dependence of H_{cx} on the exchange coupling parameter A_{cx} .

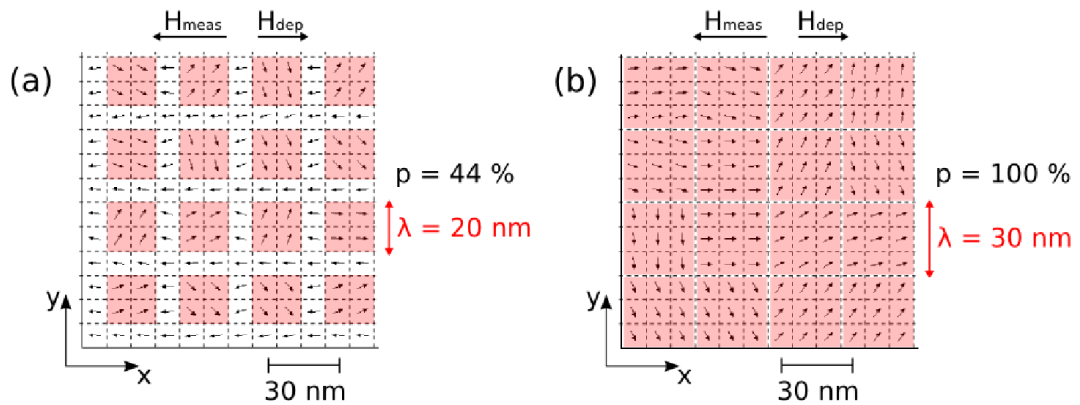


Figure 4.21: Other examples of simulated AFM layer, in which the extension of pinning centers is varied from 2×2 (a) to 3×3 cells² (b).

Chapter 5

EB effect in nanodots

So far, magnetic properties on thin film bilayers have been studied to provide a reference model in view of the realization of three-dimensional confined structures, in fact a complete understanding of the AFM/FM exchange coupling in nanodots has not been achieved yet [24, 38]. In particular, for the case of IrMn/NiFe bilayers, what we want to achieve is a comprehensive description of the EB phenomenon in nanodots systems, assessing and elucidating the interplay among the effects of magnetic thermal stability of the AFM nanograins, the magnetothermal evolution of the glassy AFM interfacial regions (characterized by the onset of a frozen collective regime below a critical temperature), and the spatial confinement.

Accordingly, we studied the magnetic properties, and the magnetothermal behavior, of arrays of nanodots in form of both square dots, with decreasing lateral size from $1\,\mu\text{m}$ to $300\,\text{nm}$, and circular dots, with a diameter of $140\,\text{nm}$. The present chapter shows the experimental results obtained together with the interpretation given in light of the proposed model.

5.1 Square dots: from $1\ \mu m$ to $300\ nm$

In order to assess the interplay among thermal stability of the AFM nanograins, the magnetothermal evolution of the glassy AFM interfacial regions, and spatial confinement effects, we studied the properties of the IrMn/NiFe nanodots systems in the form of arrays of square dots with different lateral size: $1000\ nm$, $500\ nm$, and $300\ nm$, which have been labeled as A, B, and C, respectively [61]. For each case, interdot distances were chosen to prevent the possible presence of dipolar interactions among the dots, and they are $300\ nm$, for both A and B arrays, and $100\ nm$ for C array. The fabrication of the dot arrays was performed in order to guarantee same processing conditions for all the patterns. In fact, the different dot arrays, each one occupying an area of the order of $(100 \times 100)\ \mu m^2$, were patterned on the very same substrate of naturally oxidized Si(100). Moreover, a single squared structure with size of $500\ \mu m$, named as Q, was left on the substrate to be used as a reference continuous film.

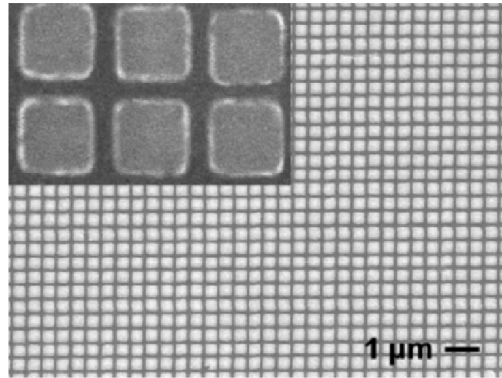


Figure 5.1: SEM image of the IrMn/NiFe square dots. In particular, a portion of the array of dots with size of $300\ nm$ is shown (the inset is an enlarged view).

The fabricated dots were analyzed by scanning electron microscopy (SEM), and a typical image is displayed in figure 5.1, which refers to the C array (we thank Annamaria Gerardino from “Istituto di Fotonica e Nanotecnologie” CNR Rome for

the SEM images provided and presented here and below). The dots appear to be well separated and regularly distributed. Atomic force microscopy observations confirmed the SEM results and indicated that the dots thickness was consistent with the nominal one (20 nm) within an uncertainty of 10%, irrespective of the dot size. Moreover, the atomic force microscopy analysis confirmed the good degree of thickness uniformity of the dots: a small thickness decrease (at most, $2 \div 3\text{ nm}$) was found to affect just a very narrow region at the outer border of some dots (namely, the dot profile did not appear squared but slightly rounded).

The magnetic properties of the dot arrays were investigated using the focused MOKE apparatus equipped with a cryostat, allowing us to span the $10 \div 300\text{ K}$ temperature range.

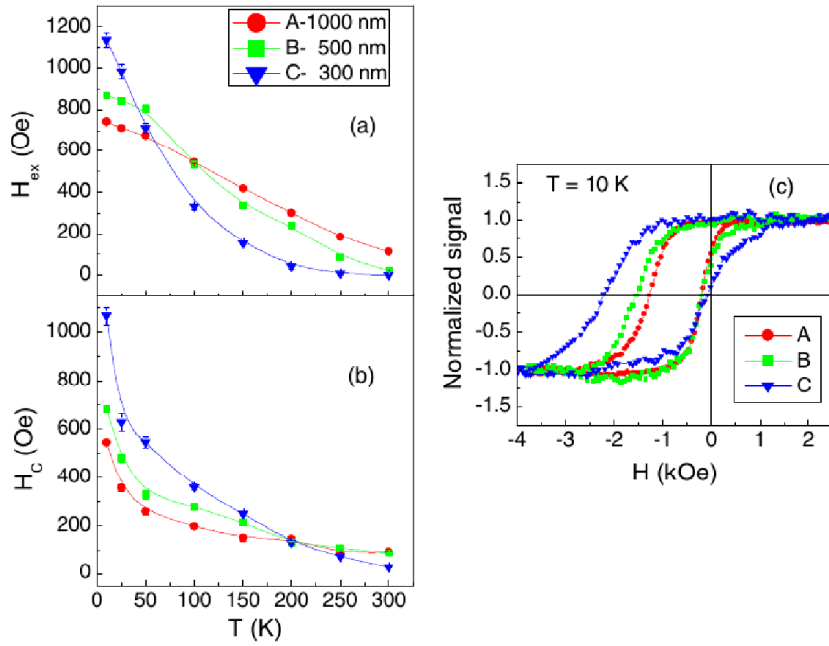


Figure 5.2: H_{ex} (a) and coercivity H_C (b) as functions of T ($10 \div 300\text{ K}$) on dots A, with size 1000 nm (circles), dots B, 500 nm (squares), and dots C, 300 nm (triangles). (c) Hysteresis loops (normalized to the positive saturation value), measured by MOKE at $T = 10\text{ K}$ on the different arrays of dots.

The curves of H_{ex} and H_C vs. T for the dots of type A, B, and C are displayed

in figures 5.2(a) and 5.2(b), while figure 5.2(c) shows the MOKE loops measured on the three arrays of dots at $T = 10\text{ K}$. The behavior of the A dots is not substantially different from that of the reference square Q in the same temperature interval and for this reason, the curves for Q have not been shown.

On the contrary, at $T = 300\text{ K}$, H_{ex} is significantly lower in the B array ($\sim 25\text{ Oe}$) and vanishing in the C one. In all the arrays, H_{ex} increases with decreasing T , but the effect is much more pronounced in the dots with smaller size. Thus, for $T < 100\text{ K}$, H_{ex} in B and C dots overcomes that in the A array and at $T = 5\text{ K}$ the highest H_{ex} is measured in the C dots ($\sim 1140\text{ Oe}$).

To account for these results we resorted to the phenomenological model discussed in section 4.2.4, according to which the interfacial AFM spins responsible for EB were assumed to be homogeneously dispersed throughout the interface area. When a patterned area is considered, we can imagine that the same fraction of pinning AFM spins is distributed in the dots. If they are very diluted in the continuous film, as reported by Ohldag et al. [56], only a few of them will reside in each dot and this number will tend to zero with decreasing dot size. The probability that they are located towards the edge of the dot increases with decreasing dot size (in fact, with decreasing the dot size, the relative spatial extension of the dots border region is enhanced). Picture 5.3 schematically depicts this trend.

However, the strength of their pinning action depends also on the thermal stability of the AFM grains to which they are anchored. The stability can be reduced in AFM grains located at the dot edge. In fact, their volume is likely to be smaller, compared to that of the grains at the center, because of the spatial confinement that hinders the normal expansion of the grains during the growth process. Similar conclusions have also been formulated elsewhere [38].

Moreover, we supposed that bulk AFM grains are essentially exchange decoupled.

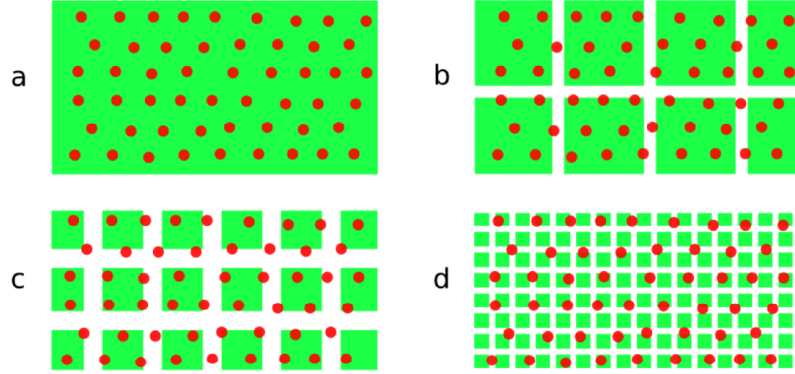


Figure 5.3: The same pinning centers distribution (red dots) is put over a section of continuous film (a) and patterns of square dots with progressively reduced size (b, c, d).

Nevertheless, the existence of a weak partial coupling between adjacent grains may, on one hand, enhance the thermal stability of those located at the center of the dot, on the other hand, compromise the thermal stability of those interior grains that are adjacent to grains at the edge.

In this view, at $T = 300\text{ K}$, the very small value of H_{ex} in the B dots and the substantial absence of EB in the C dots can be accounted for considering that the number of effective AFM pinning centers per dot is too low to cause the insurgence of a strong macroscopic unidirectional anisotropy for the FM magnetization. The enhanced values of H_C ($\sim 90\text{ Oe}$ in A and B and $\sim 30\text{ Oe}$ in C dots), compared to that of the NiFe layer at room temperature, are consistent with the interfacial AFM spins rotating with the FM magnetization, as it happened for the case of the continuous film. With decreasing T from 300 K , the thermal stability of the AFM phase improves more and more and this implies that the polarizing action of the bulk AFM grains on the interfacial spins gets stronger and that the number of interfacial spins able to act as pinning centers for the FM magnetization, as well as their efficiency, gets larger. At the same time, we excluded the possibility that this behavior is due to shape anisotropy, through the comparison with measurements

performed on NiFe reference arrays of nanodots with similar size. Therefore, both H_{ex} and H_C increase in a percentage much larger for the smaller dots (figure 5.2). Moreover, as the temperature $T = 100\text{ K}$ is approached, that being schematically indicated as the temperature that marks the passage to the frozen regime for the interfacial spins, magnetic interactions among interfacial spins must be taken into account, as well.

The effect of the large increase of H_{ex} at low temperature is more clearly visible as the dot size is reduced, suggesting that the formation of the spin glass phase, whose appearance is associated to the appearance of a spin glass correlation length (as discussed in the subsection 4.2.4), is more effective when the interfacial spins are less effectively polarized by nanograins in the smaller dots. Taking into account then the two variables that we considered key features to understand the EB effect at low temperature, such as the dot size and the spin glass correlation length, we suggested that correlated spins exert a stronger pinning action on the FM magnetization as the dot size and the correlation length become comparable.

The hypothesis that the EB effect in nanostructures is governed by a characteristic dimension, corresponding to correlated regions in the AFM layer, has been recently reported also by Laureti et al. [39]. Although the authors state that the AFM grains are exchange decoupled, in order to account for the EB properties in different sets of Co/CoO nanodots they invoke the existence of a correlation length that may vary with the geometrical and structural features of the dots (thickness of the Co layer and oxidation degree). We unambiguously identify this critical length with the correlation length among the AFM spins of the interfacial spin-glass-like region.

It should be noted that, in principle, in a canonical spin glass the correlation length becomes infinite when the fully glassy state is finally established [55]. Thus,

we would expect that at $T = 10\ \text{K}$, H_{ex} tends to the same value in all the arrays. This is not experimentally observed, actually, and can be ascribed to the fact that the interface region is inherently inhomogeneous, both from the structural and magnetic point of view, implying that an infinite correlated glassy state is not established at $T = 10\ \text{K}$. Moreover, during the loop measurement for determining H_{ex} , the application of the magnetic field (up to $5\ \text{kOe}$) may break up the “infinite” glassy state and limit the extent of the correlation length. A hint of this is the high H_C at very low T , see figure 5.2(b), revealing that, despite the high effective anisotropy of the AFM spins in the frozen regime, some of them can still be dragged by the FM moments under a high enough magnetic field. Hence, at the lowest measuring temperature ($T = 10\ \text{K}$), the ratio between the dot size and the correlation length will be much smaller in dots C than in dots B and A, which accounts for the different H_{ex} values measured at that T in the different arrays (figure 5.2(a)).

In order to support the forementioned description, we performed micromagnetic simulations by OOMMF, exploiting the strategy illustrated in section 4.4.2 to simulate the exchange interaction at the interface of FM/AFM systems.

In particular, we were firstly interested in verifying our prediction on the variation of H_{ex} with the percentage p of pinning centers, and for this purpose we simulated different configurations corresponding to different values of p . To guarantee a homogeneous distribution of the pinning centers inside the dot we arranged the pinning centers on a square array, i.e. the distance between pinning centers along x and y directions was fixed to precise values, and according to the adopted scheme, a given distance between the pinning cells resulted in the same p value, irrespective of the size of the dot. For example, for all the simulated dots, a percentage $p \sim 11\%$ corresponds to a center-to-center distance of 3 micromagnetic cells (this is the case illustrated in figure 5.4), a percentage $p \sim 6\%$ to a distance of 4, and so on.

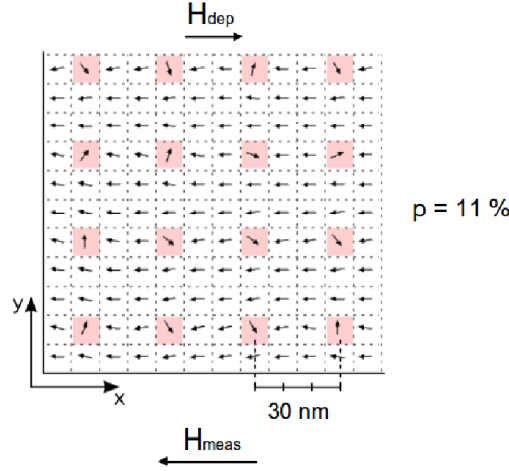


Figure 5.4: Distribution of simulated AFM pinning centers (micromagnetic cells highlighted in red) corresponding to a percentage $p \sim 11\%$

The calculations were performed for a dot with size of 1000 nm (A type) and for a dot of 300 nm (C type) and the final H_{ex} values were the average of those obtained over different replicas of the same simulation. Actually, the replicas performed for a fixed value of p were calculated changing the relative position of the square pinning centers lattice with respect to the dot perimeter. This allowed us to ascertain the absence of any significant dependence of H_{ex} on the specific layout of the pinning centers. In other words, H_{ex} does not depend on the detailed position of the pinning centers, but just on the value of p . The variation of H_{ex} with the percentage of pinning centers for A type and C type dots is shown in figure 5.5: H_{ex} increases with increasing the amount of pinning centers, passing from just $\sim 10 Oe$, when 1% of pinning centers is present, to a final value larger than $600 Oe$. This trend of H_{ex} is very similar in the simulated A and C dots.

Hence, the low H_{ex} measured in the smaller dots (C dots) at $T = 300 \text{ K}$, see figure 5.2(a), can be accounted for if one considers that only a fraction of the pinning centers effectively pin the FM phase, and that this fraction is expected to reduce with decreasing the dot size. This may happen due to the fact that the thermal

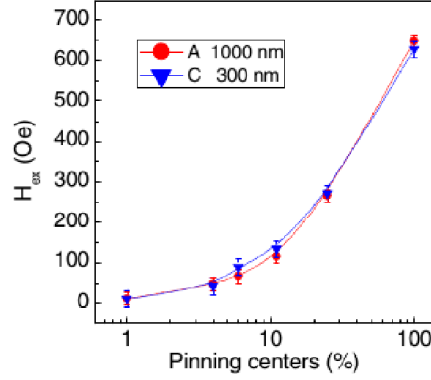


Figure 5.5: Dependence of H_{ex} on the percentage of AFM pinning centers as obtained by micromagnetic calculations simulating the magnetic behavior of a single squared dot with size 1000 nm and 300 nm .

stability of the nanograins located at the border of the dot is reduced, so some pinning centers may become less effective as the dot size is reduced. For instance, based on the simulation, the value of H_{ex} measured at $T = 300\text{ K}$ in the A dots ($\sim 115\text{ Oe}$) is consistent with a fraction of pinning centers around 10%, whereas the vanishing H_{ex} in the C dots indicates the presence of a very low amount of effective pinning centers (less than 1%). It is to be noted that these percentages seem quite reasonable, also in the light of the results presented by Ohldag et al. [56]. In concomitance, the results in figure 5.5 also support our statement that the observed increase in H_{ex} with decreasing T from room temperature reflects the increase in the amount of effective pinning centers.

In order to assess the effect of magnetic correlation among the AFM pinning centers in the low-temperature frozen regime, we increased the size of every pinning center to a specific value λ and considered that the whole simulated AFM layer is pinning, corresponding to the case with a percentage of pinning area $p = 100\%$. Inside each larger new pinning center the micromagnetic cells are given an high anisotropy value and share a common random anisotropy direction d_{AFM} , chosen so as to have a positive projection on the x axis, as illustrated in figure 5.6.

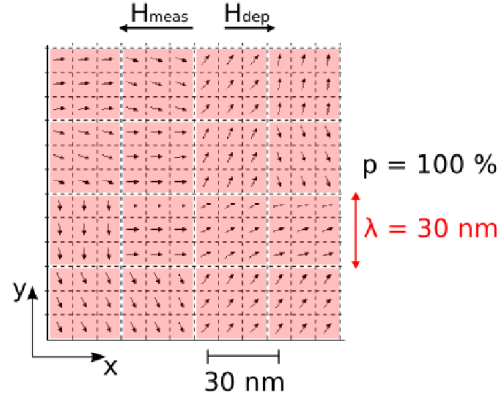


Figure 5.6: Distribution of simulated AFM pinning centers with size λ (every group of 3×3 micromagnetic cells highlighted in red is a large pinning center) corresponding to a pinning surface percentage $p \sim 100\%$.

So, the case of the absence of magnetic correlation is considered when λ equals the size of micromagnetic cells, and this case actually coincides with the simulation for 100% pinning centers reported in figure 5.5. Hence, starting from this case of complete uncorrelation, in order to simulate the appearance of correlation, i.e. the enhancement of λ , the same type of calculation was performed for increasing values of λ : 20, 50, 100, 200, and 300 nm.

In all the calculations we have considered a dot size $D = 1200 \text{ nm}$, slightly larger than that of the A dots, in order to include also the case of correlation length $\lambda = 300 \text{ nm}$. We did not consider larger λ values, closer to D , or smaller dots, despite that these cases would have been very interesting for our study. In fact, when the ratio between λ and D approaches unity, this simulation method does not produce reliable results. Beside the fact that the uncertainty related to H_{ex} becomes very high in these cases, we think that this lack of reliability arises because of the inherent difficulty of modeling a glassy magnetic structure, featuring a collection of spins subjected to an effective local anisotropy (randomly changing in magnitude and direction on a spatial scale comparable or even shorter than the AFM grain

size) and to competing magnetic interactions. In the specific case of the IrMn/NiFe system, the configuration of the frozen interfacial AFM spins must also minimize the exchange interaction energy with the spins of both the NiFe phase and of the AFM grains.

However, problems arise when modeling of the magnetic anisotropy. If λ/D is small (namely, the pinning layer is composed of a large number of cells), the random and local character of the magnetic anisotropy of the glassy interfacial phase can be reproduced by our simulation method to a good extent. On the contrary, as λ approaches D , the pinning layer is partitioned in just a few cells and the random and local character of the anisotropy is progressively and inevitably lost.

The result of the simulation is shown in figure 5.7: H_{ex} increases significantly as the size of the correlated regions grows from 10 to 300 nm (from ~ 650 to $\sim 730\ \text{Oe}$), indicating a trend which is coherent with our expectation (also in this case the H_{ex} values are the average of those obtained over ten replicas of the same simulation).

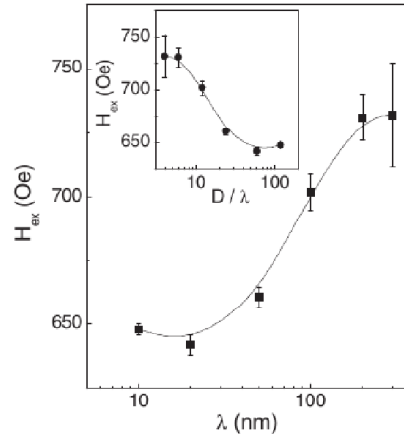


Figure 5.7: Dependence of H_{ex} on the length λ of magnetic correlation among AFM pinning centers as obtained by micromagnetic calculations simulating the magnetic behavior of a squared dot with size $D = 1200\ \text{nm}$. In the inset H_{ex} is plotted as a function of the ratio D/λ .

The inset of figure 5.7 is the plot of H_{ex} as a function of the ratio between the dot size and the correlation length D/λ : it illustrates at a glance the concept that

H_{ex} increases as D/λ approaches 1. Indeed, we consider the dot of 1200 nm as a model system, where our micromagnetic simulations can be reliably applied to gain unambiguous information on the dependence of H_{ex} on the glassy correlation length. Hence, a trend qualitatively similar of H_{ex} vs. D/λ is expected to be observed also in dots of different size. Finally, we have also calculated the case $\lambda = 1200\text{ nm}$, which provides the highest H_{ex} value (980 Oe). Although this result may appear coherent with our prediction, actually it is factitious and for this reason it has not been reported in figure 5.7. In fact, in this calculation we have forced the anisotropy of the pinning layer to stay parallel to the initial NiFe magnetization direction¹.

5.2 Circular dots with reduced IrMn thickness

In this section we focused on the case of circular dots with a reduced thickness of IrMn, that is $t_{AFM} = 3\text{ nm}$, where the exchange coupling with the FM phase may expected to be due to the disordered AFM region on its own. Moreover, the dots are $\sim 140\text{ nm}$ in size (figure 5.8), and so, even more laterally confined with respect to the square dots considered in the previous section, in order to gain an insight into the exchange coupling mechanism when the disordered IrMn region is further confined in the three spatial dimensions [63]. The thickness of both Cu underlayer and NiFe FM phase is decreased to 3 nm as well, with different purposes: for the former, the purpose is to unfavor the polycrystalline growth of the AFM phase, for the latter to enhance the EB effect as much as possible, since H_{ex} is known to be inversely proportional to the FM layer thickness, so to better investigate its thermal dependence. As we did before, the results are compared with the case of a continuous reference film with the same stacking.

The circular shape of the dots was chosen in relation to the nanofabrication

¹however, if a perpendicular orientation were imposed, no loop shift would be observed

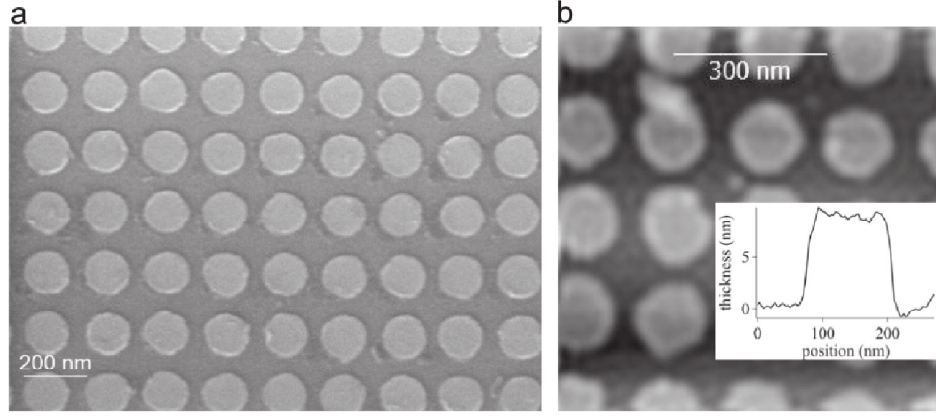


Figure 5.8: (a) SEM image of the IrMn/NiFe dot array. (b) Atomic force microscopy image of the circular dots, obtained in collaboration with Samuele Fin from “Università degli Studi di Ferrara”; in the inset, the height profile of a single dot is displayed.

strategy exploited. As for the case of the square dots, circular AFM/FM dots were prepared by electron beam lithography and sputtering deposition, followed by a lift-off procedure. But here, the fabrication process of the dot array is a bit different, in fact it was carried out exploiting the writing strategy of the e-beam lithography system. Due to the requirements about dot size (around 150 nm) and density (pitch, i.e. center-to-center distance, $\sim 200\text{ nm}$) and the need of a large patterned area ($3 \times 3\text{ mm}^2$, in order to obtain a measurable magnetic moment), we exposed a square Si substrate with 5 mm side, previously spin-coated with a polymeric resist, imposing an electron beam step size (the distance between two consecutive exposed pixel) equal to the desired pitch. This strategy allows one to expose single pixels with a periodicity equal to the beam step size: the dots are exposed as single pixels and it is possible to control the dot size, directly correlated to the beam shape, changing the exposure beam current and the dwelling time, as to say the exposure dose.

Magnetic hysteresis loops of the reference film were measured at increasing values of temperature T via SQUID magnetometry after cooling the sample from room

temperature down to 5 K in a static magnetic field $H_{cool} = 50\text{ kOe}$, applied in the same direction as H_{dep} (field-cooling mode, FC), to ensure the EB to develop with its characteristic uniform unidirectional anisotropy.

The loops were corrected for the diamagnetic contribution of the Si substrate and then analyzed to extract the exchange field H_{ex} and the coercivity H_C . The curves of H_{ex} and H_C vs. T are shown in figure 5.9. The EB effect is absent at room temperature. With decreasing T , it appears at $T \sim 200\text{ K}$. Both H_{ex} and H_C increase with reducing T , especially below $T \sim 100\text{ K}$ and at $T = 5\text{ K}$ they reach 1270 Oe and $\sim 860\text{ Oe}$, respectively.

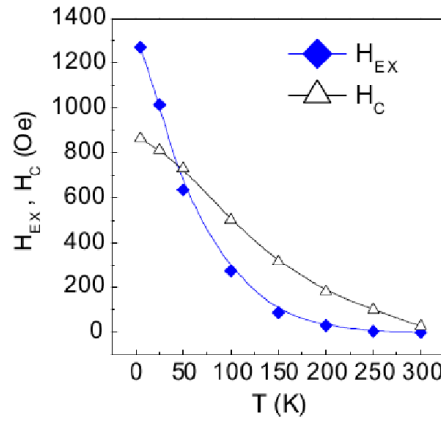


Figure 5.9: H_{ex} (full symbols) and H_C (open symbols) as functions of temperature T , measured in FC mode. The error bars are smaller or comparable to the size of the dots. Solid lines are guides to the eye.

Loop measurements on the dot array were carried out after cooling down from room temperature to $T = 5\text{ K}$ with no field applied (zero-field-cooling mode, ZFC) and in the FC mode ($H_{cool} = 50\text{ kOe}$). The comparison between FC and ZFC measurements is usually exploited in nanogranular system to assess, e. g., whether interactions among the nanograins (the dots) are present. In this case, the hypothesis that each nanograin is a monodomain is frequently used, and the lateral size of 140 nm seems to suggest this possibility for our circular dots [64]. In figure 5.10(a),

the curves of ZFC and FC H_{ex} vs. T are shown: as in the continuous film, no EB effect is observed at room temperature. In the ZFC case, a non-vanishing EB effect appears only at $T < 50 K$ and at $T = 5 K$ $H_{ex} \sim 30 Oe$. As for the FC data, a significant value of H_{ex} is measured at $T = 200 K$; it increases with decreasing T and $H_{ex} \sim 90 Oe$ at $T = 5 K$, a value much smaller than that measured in the continuous film. The FC procedure does not affect noticeably the coercivity (figure 5.10(b)). At $T = 300 K$, $H_C \sim 80 Oe$, a value higher than that measured in the continuous film at the same temperature ($\sim 30 Oe$). Inherent factors, most likely structural defects at the edge of the dots, may be responsible for this slight magnetic hardening. Then, with reducing T , H_C increases, but at $T = 5 K$ it does not exceed $180 Oe$.

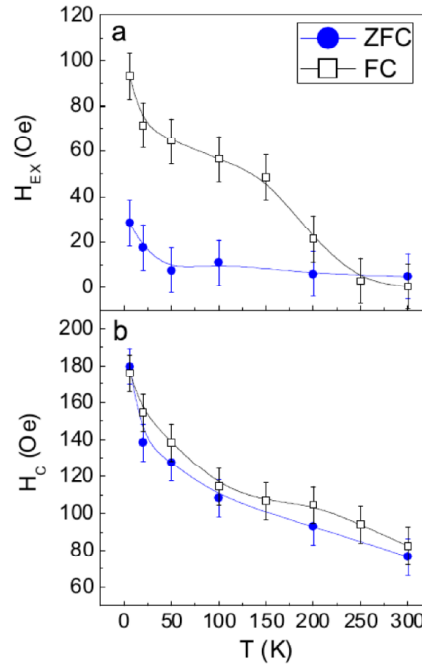


Figure 5.10: Exchange field H_{ex} (a) and coercivity H_C (b) vs. T in the dot array, measured in ZFC (full symbols) and FC mode (open symbols; applied field $H_{cool} = 50 kOe$).

To assess if, in spite of the difference in the values of H_{ex} , the general description

we used to explain the thermal dependence of the EB effect, holds for the circular dots too, we probed the AFM energy barrier distribution of the array. The reference continuous film was grown so to enhance the effect of the disordered AFM phase, so the glassy state at low temperature is expected to be the only effective source of anisotropy. Due to that we expect it to show just the low temperature part of the energy barrier distribution shown in figure 4.11.

For the circular dots, in order to probe the distribution of the effective anisotropy energy barriers of the AFM phase, we exploited a measurement procedure that is different from the one we presented and discussed in section 4.2.1, because it was difficult to keep trace of the temperature dependence of the magnetization of this system due to the low magnetic moment of the array of nanodots. The procedure exploited here consisted in measuring hysteresis loops at $T = 5\text{ K}$ after field cooling (FC) with $H_{cool} = 50\text{ kOe}$, from different temperatures T_i . At the end of each loop measurement, the sample was warmed up to 300 K so as to restore the initial condition unsetting all AFM entities with blocking temperature lower than 300 K . For the case at $T_i = 5\text{ K}$ the measurement corresponds to a zero-field cooling measurement. For each value of T_i , only the AFM spins whose anisotropy is overcome by the supplied thermal energy are able to change their orientation so as to minimize the exchange energy at the interface with the FM spins, aligned to saturation by H_{cool} . Hence only these AFM spins effectively cooperate to the FC value of H_{ex} , measured at $T = 5\text{ K}$, with respect to the ZFC one. On the contrary, the interfacial AFM spins that at T_i are in fixed positions, due to their local effective anisotropy, are expected to be not effectively involved on average in the EB effect. The derivative of H_{ex} with respect to T_i is generally assumed to be a figure of the AFM anisotropy energy barrier distribution [65]. In the latter procedure, the hysteresis loops are always measured at the same temperature and this allowed to derive the

energy barrier distribution independently from any possible thermal dependence of the magnetocrystalline AFM anisotropy K_{AFM} , or the exchange coupling J_{FM-AFM} , characteristic of the interaction between the two magnetic phases at the interface.

The calculated AFM anisotropy energy barrier distribution is displayed in figure 5.11(b): it shows a peak at $T = 40\text{ K}$ that can be associated to the collective freezing of the IrMn spins, in agreement with our model, and no other sharp peak at higher temperature is observed. That is reasonable in case a distribution of nanograins is lacking, namely the presence of nanograins inside the AFM phase can be considered to give a negligible contribution to the magnetic properties. The onset of a low-temperature frozen magnetic regime is also consistent with the marked enhancement of H_{ex} and H_C observed for $T < 100\text{ K}$, both in the ZFC and FC modes (figure 5.10). Thus it appears that, even in the possible absence of AFM nanograins, the exchange coupling mechanism at low temperature is still dominated by the activation of a spin-glass like phase, in analogy with the continuous IrMn/NiFe film.

To account for the much smaller H_{ex} measured in the dot array with respect to the continuous reference film, we consider first the case of the ZFC measurements. It is to be pointed out that, at $T = 300\text{ K}$, the ratio between the remanent magnetization (M_r) and the saturation magnetization (M_S) of the dot array was just ~ 0.23 and it is worth recalling what is often observed in nanogranular exchange-coupled materials. A nanogranular system can be simply described as a collection of soft FM nanoparticles - with typical size $\sim 10\text{ nm}$ (i.e. small enough to be in a single domain magnetic state) and with a random orientation of the anisotropy axis - dispersed in an AFM matrix. Indeed, each nanoparticle is subjected to exchange anisotropy also after ZFC from a temperature higher than the AFM phase blocking temperature (i.e. the temperature above which no loop shift is observed). However, as the nanoparticles are randomly oriented, the exchange anisotropy averages out

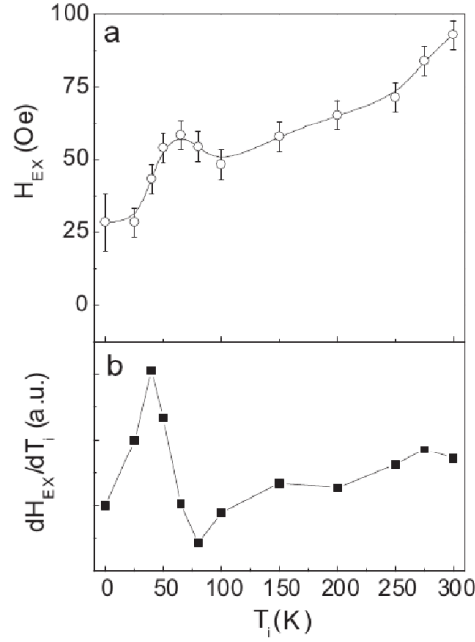


Figure 5.11: (a) Exchange field H_{ex} measured in the dot array at $T = 5 K$ shown as a function of the temperature T_i at which $H_{cool} = 50 kOe$ was applied. (b) Temperature derivative of the curve in (a), providing a figure of the anisotropy energy barrier distribution of the AFM phase.

to a negligible value and no EB effect is produced [62].

A similar explanation may be applied to the dot array measured in the ZFC mode provided that: (i) in the investigated temperature range each dot behaves as a single-domain, namely is in a high-remanence magnetic state; (ii) at $T = 300 K$, i.e. above the blocking temperature, the net magnetization vectors of the dots have different orientations so that, before cooling the sample down to $T = 5 K$, the array as a whole is in an almost un-magnetized state (low-remanence state). In order to assess if these two conditions subsist, we have simulated different scenarios using the OOMMF program. Regarding item (i), we simulated the magnetic configuration of a $3 nm$ thick NiFe disk with diameter of $141 nm$. We used cubic cells with size of $3 nm$ and saturation magnetization and exchange stiffness of $800 emu/cm^3$ and $13 \cdot 10^{-7} erg/cm$, respectively; the contribution of magnetocrystalline anisotropy was

assumed to be negligible. We used a cell size smaller than the NiFe exchange length, $\sim 5\text{ nm}$, to have a better approximation of the circular shape. The simulation of the remanent magnetic state of the dot was done by calculating a loop starting from the positive saturation, to mimic the experimental loop measurement procedure.

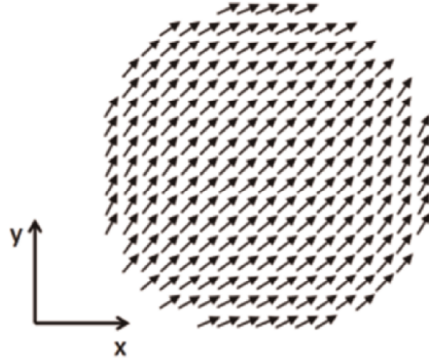


Figure 5.12: Magnetic configuration at remanence for an isolated NiFe dot. The in-plane arrangement of the magnetization vectors of the cells corresponds to the onion state; the magnetization is nearly uniform (for clarity, each vector represents the average over a square of 5×5 cells).

The result, shown in figure 5.12, confirms that the magnetic configuration of the dot is a high-remanence state, being the ratio $M_r/M_S \sim 0.69$. In particular, it is the so-called onion state configuration to take place, among the different possible configurations [64]. Hence, we can exclude that the small M_r/M_S , measured in the array at room temperature, is due to the fact that each dot is in a low-remanence magnetic state (vortex-like configuration).

Since each dot presents a high-remanence magnetic configuration, dipolar interactions must be considered. Due to the small interdistance between two adjacent dots, of about 60 nm , it is expected that dipolar interactions establish a low-remanence magnetic configuration of the whole array, so that condition (ii) would be fulfilled. Therefore, we simulated the magnetic state of an array of 9 NiFe circular dots with the same magnetization, exchange stiffness and anisotropy values used for the single NiFe dot. The dot size was 141 nm and they were arranged on a square

lattice with center-to-center distance of 200 nm.

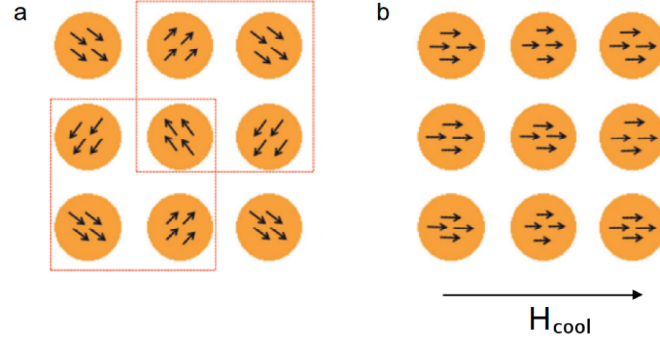


Figure 5.13: (a) Magnetic configuration at remanence for an array of 9 interacting dots; just a few vectors are displayed to indicate the magnetization orientation in the dots. In (b), an external cooling field H_{cool} is applied, and the ideal case of perfect alignment of the magnetic moments with H_{cool} is considered.

The simulation reveals that groups of four dots form, realizing a closure-like configuration as schematically highlighted by the squared red frames in figure 5.13(a). The calculated value of M_r/M_S for the whole array is then ~ 0.24 , much lower than that calculated for the case of single isolated dot and close to the experimental one. This confirms that, at $T = 300\text{ K}$, the net magnetization vectors of the individual dots are aligned along different directions in order to minimize the magnetostatic energy of the array as a whole. Hence, when the low-temperature region is reached in ZFC mode, the exchange anisotropy axes in the dots are non-collinear. Accordingly, H_{ex} in the array is very small even at the lowest temperature.

To address the case of FC measurements of H_{ex} we recall again what happens in the case of a nanogranular material: a loop shift can be observed only by cooling across the blocking temperature in presence of an external field H_{cool} , which magnetizes the nanoparticles assembly. In fact, a net exchange anisotropy is produced in this way and, actually, H_{ex} is seen to increase with increasing H_{cool} , reflecting the enhancement in the alignment degree of the nanoparticle moments [62]. Similarly, in the dot array, at all temperatures the FC H_{ex} values are higher than those mea-

sured in the ZFC mode (figure 5.10(a)), consistent with the improved alignment of the dot magnetization vectors attained through $H_{cool} = 50 \text{ kOe}$, as depicted in case of a perfect alignment in figure 5.13(b). Nevertheless, the measure H_{ex} values are definitely smaller than those reported for the continuous reference film at $T = 5 \text{ K}$, and in the whole temperature range where EB effect is observed.

In order to account for such a small H_{ex} at low temperature, different explanations may be invoked: (I) a decrease in the number of IrMn spins acting as pinning centers, i.e. effectively involved in the EB mechanism, compared to the continuous film (assuming a homogeneous spatial distribution of the pinning centers, we can also speak of a decrease in the effective AFM/FM interface area); (II) a uniform reduction of the exchange anisotropy acting on the NiFe magnetization in the dot compared to the continuous film; (III) a competition between dipolar interactions, that, as shown above, favor a non-collinear arrangement of the dot magnetization vectors, and exchange anisotropy, assumed to be collinear in all the dots. To test these hypotheses again we drew upon the OOMMF software. In order to simulate a set of magnetically interacting dots, easy to be handled from the computational point of view, we represented the system in a different way with respect to that previously described. First, the dots were considered as single domains (i.e. as dipoles) and we used squared cells 50 nm wide and 3 nm thick, so that each dot is composed of (3×3) cells and it is at saturation. Moreover, the effective pinning action on the NiFe magnetization was taken into account by applying a static magnetic field, indicated as the pinning field H_P along x axis (with the same direction of the cooling field H_{cool}). So there is just one layer to consider in the calculation, and we point out that this is quite a different approach from the one exploited in the previous section, and it is preferred now to make the calculations run faster. In each cell where H_P is applied, it can be considered as the measure of the average pinning

effect provided by all the frozen AFM spins inside that cell itself, in other words the related Zeeman energy term, calculated as the dot product between H_P and the magnetic moment of the micromagnetic cell, mimics the effective interaction with an AFM layer.

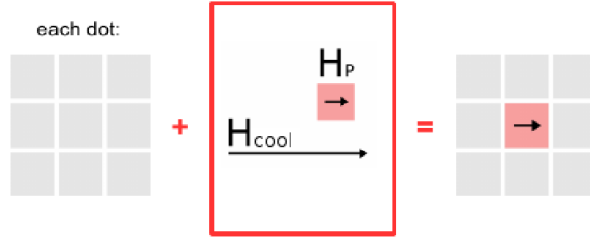


Figure 5.14: A simulated dot consisting of a group of 3×3 micromagnetic cells; a pinning field H_P is assigned to the central cell to perform type A simulation (see the text).

The dots were arranged on a square lattice of (13×13) elements and the center-to-center distance was 200 nm ; to get rid of finite size effects of the array, the hysteresis loops were calculated just for the central sub-lattice of (5×5) elements. We performed two different calculations: type A, in which H_P was applied just to the central cell of each dot (see figure 5.14) to simulate hypothesis (I); type B, in which H_P was applied to all the cells of each dot to simulate hypothesis (II).

The results of the calculation are presented in figure 5.15 (the x-axis scale is logarithmic, this choice permits to better display the results of both calculations). In both cases, H_{ex} increases almost linearly with increasing H_P . In type A simulation, an H_{ex} value close to that measured at $T = 5\text{ K}$ in the dot array ($\sim 90\text{ Oe}$) is obtained for $H_P \sim 850\text{ Oe}$ whereas, in type B, $H_P \sim 90\text{ Oe}$ must be applied.

In order to verify hypothesis (III), type A and B simulations were performed also for a single dot consisting of 9 cells, so it is deprived of neighbouring dots in order not to have dipolar interactions. Curves of H_{ex} vs. H_P were obtained similar to those shown in figure 5.15 for the 13×13 lattice. This result indicates that interdot

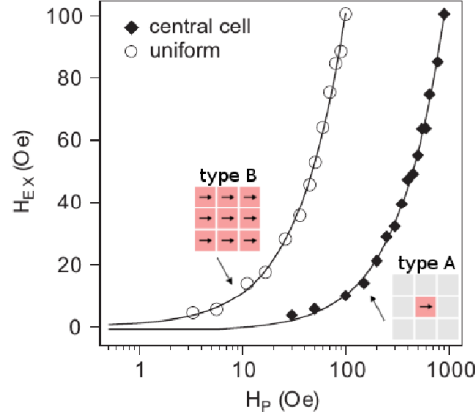


Figure 5.15: Exchange field H_{ex} as a function of the pinning field H_P , calculated for an array of 13×13 dots, each dot consisting of 9 cells. Two types of simulations were carried out: H_P applied only to the central cell of each dot (type A, solid symbols) and applied to all the cells of each dot (type B, open symbols). The lines are guides to the eye.

dipolar interactions do not affect H_{ex} , namely exchange anisotropy dominates on magnetostatic effects. Hence, hypothesis (III) can be disregarded. Coming again to 5.15, it is to be noted that, for the same calculated value of H_{ex} , the ratio between the H_P value used in type A calculation and that used in type B is nearly $\frac{1}{9}$. This means that the final macroscopic effect of the AFM pinning action, namely the loop shift, is roughly proportional to the product between H_P and the total area where H_P is applied, namely between the strength of the pinning exerted by a single cell and the number of pinning cells. Therefore, the simulations do not allow us to ascertain whether the weak EB effect is due to a low H_P acting uniformly on all the dots or to a decrease in the effective AFM/FM interface area, namely it is not possible to discriminate between hypotheses (I) and (II).

Hypothesis (I) substantially means that, even at the lowest temperature when the frozen collective regime of the AFM spins is established, only those located at the center of the dots exert a pinning action strong enough to give rise to the EB effect. This may occur if the thickness of the IrMn layer is vanishingly small in the

outer region of the dots, due for instance to shadowing effect during the sputtering deposition process. However, the atomic force microscopy analysis we reported in figure (see the profile measurement in the inset of figure 5.8(b)) indicates that the dots have a quite squared and sharp profile.

Hypothesis (II) means that a uniform decrease in the strength of the pinning action exerted by the frozen AFM spins on the FM ones occurs, with respect to the case of the continuous film. A possible explanation to account for this effect is the following. Considering the small dot size and the ultrafine IrMn layer thickness, we are actually addressing a case in which the length of magnetic correlation among the IrMn spins may be spatially limited by confinement effects along all the three spatial dimensions. In other words, the volume of the IrMn glassy region is so small that the collective pinning action of the frozen IrMn spins is much weaker than in the continuous film, where they are free to magnetically correlate over an arbitrary length at least along two dimensions. We indicate with D the physical size of the AFM/FM system and with λ_0 the correlation length of the AFM spins in the absence of spatial constraints. Thus, going from the continuous film to the dots, one actually moves from the condition $\frac{D}{\lambda_0} > 1$ to $\frac{D}{\lambda_0} < 1$. So we propose that, since in the case of circular dots the magnetic correlation among the IrMn spins is spatially delimited, the spins themselves are thermally less stable than they are in the continuous film. This scenario is also contrary to what happened in the case of the larger square dots reported in the previous section, for which the proposed $\frac{D}{\lambda_0} > 1$ regime accounted for larger H_{ex} values with respect to the corresponding reference film.

In conclusion, it is also possible to guess that the worse thermal stability of the interfacial IrMn spins of circular dots should especially affect those located at the border of the dots. Therefore, this explanation actually combines together hypotheses (I) and (II), namely the reason of the weak EB effect in the dots is an

overall reduction of the pinning strength of the IrMn spins, especially of the outer ones, due to spatial confinement effects.

Chapter 6

Conclusions

The work carried out during this thesis concerned both the preparation and the study of the properties of bilayer nanostructures grown by dc-magnetron sputtering. They consist of two thin layers of $\text{Ni}_{80}\text{Fe}_{20}$ (NiFe) and $\text{Ir}_{25}\text{Mn}_{75}$ (IrMn), the FM and AFM phases respectively, exchange coupled at their common interface, in form of both continuous films and arrays of nanodots.

Based on both the experimental study of the magnetothermal behavior of the EB effect in our samples and their microstructural investigation, we proposed a model for the magnetic structure of the AFM layer including different magnetic entities. The region of the AFM layer close to the interface with the FM layer was observed to have a disordered structure, both from the structural and from the magnetic point of view. Differently, the rest of the AFM layer, namely the bulk of the AFM phase, was made of AFM nanograins that were supposed to be independent or weakly interacting. The disordered magnetic region at the interface is supposed to consist of AFM spins which mediate the exchange interaction between the bulk of the AFM phase and the FM phase, and so their presence is considered to be essential for the appearance of the EB effect. As mediators of the interaction with the FM phase,

interfacial spins can be polarized by the AFM nanograins and, at the same time, if temperature is properly decreased (below $\sim 100\text{ K}$), form a spin glass state with its own anisotropy.

This picture was developed thanks to the analysis, presented in chapter 4, of the experimental data collected on the bilayer samples in form of continuous films. In particular, we observed that the values of H_{ex} and H_C fields increased with decreasing T , and this was particularly visible when the temperature is decreased below $\sim 100\text{ K}$. Accordingly, we have distinguished two magnetic regimes, the so called high and low temperature regimes, depending on whether the temperature is above or below $T \sim 100\text{ K}$, respectively. We focused our attention on the samples with stacking $\text{NiFe}[5\text{ nm}]/\text{IrMn}[t_{AFM}]$, namely with IrMn grown after NiFe, the ones for which we observed the more remarkable difference between the high and low temperature regimes. Those two regimes were further investigated exploiting an original protocol that allowed us to derive the energy barriers distribution of the AFM phase. Looking at the energy barriers distribution of three samples with $t_{AFM} = 10, 6$ and 3 nm , we observed two main peaks for each sample, one at high ($T > 100\text{ K}$) and one at low ($T < 100\text{ K}$) temperature. As we observed that the high temperature peak shifts towards lower energies with the decrease of the AFM thickness, we associated it to the volume distribution of nanograins inside the AFM phase; the highest values of H_{ex} at room temperature were observed for $t_{AFM} = 6$ and 10 nm , where the corresponding high peaks are found to be above room temperature, namely the nanograins with larger volumes are still blocked thanks to their larger anisotropy energy term. We supposed that blocked nanograins are able to polarize the interfacial spins that, in turn, sustain the exchange interaction with the NiFe phase, giving raise to EB effect. We further assumed that only a percentage of all interfacial spins are able to effectively pin the NiFe phase, the ones

we called pinning centers, and that they are homogeneously dispersed throughout the interface area.

For what concerns the peak observed at low temperature, we associated it to the appearance of a frozen collective state of interfacial AFM spins, namely the spin glass state, and it is such a state that is expected to sustain the EB effect thanks to its high effective anisotropy, allowing interfacial spins to exert a collective stronger torque action on the NiFe spins.

When the case of patterned area is considered, namely the arrays of dots with both square and circular shape studied in chapter 5, in the assumption that the same percentage of pinning AFM spins is distributed in the dots as it was in the continuous film, the probability that they are located towards the edge of the dot increases with decreasing dot size. We then suggested that in nanostructures, the stability of the AFM grains located at the edge can be compromised, and as a consequence the pinning action reduced at the edge. In fact, we observed that, at room temperature, the EB effect becomes vanishingly small with the reduction of the dot size, namely as the contribution to the EB of the edge region, with its less effective AFM pinning spins, should be expected to be more considerable. With decreasing T from 300 K , in the high temperature regime, the thermal stability of the AFM nanograins improves and this implies that the polarizing action of the bulk AFM grains on the interfacial spins gets stronger and also that the number of pinning centers may increase. According to this picture, we experimentally observed a general increment of EB effect with decreasing temperature.

However, as the temperature $T = 100\text{ K}$ is approached, our model predicts that the interfacial AFM spins start to correlate, interacting each other to form a spin glass state. We supposed that the passage to the frozen regime, as temperature is decreased below 100 K , in the low temperature regime, is marked with the increasing

of a correlation length λ among the AFM interfacial disordered spins. In continuous film, λ might be expected to arbitrarily increase as the spin-glass is fully established; this is the behavior reported for a canonical spin glass. But, due to the fact that the real interface region may be inhomogeneous both from structural and magnetic point of view, we expected a finite value for the correlation length, λ_0 , even in absence of lateral constraints, namely for the case of a continuous system. As the lateral confinement is taken into account for the study of square dots with size D ranging from $1\ \mu m$ to $300\ nm$, we observed that H_{ex} and H_C increase as the size of the dot, D , is reduced and formulated the idea that correlated spins exert a stronger pinning action on the FM magnetization as D is decreased. We supposed that the onset of the spin glass phase is more effective as D is decreased, because the interfacial AFM spins are less effectively polarized by the nanograins at the dot edge. To better understand how D and the spin glass phase, with characteristic correlation length λ_0 , contribute to determine the EB, we performed micromagnetic calculations considering system with different $\frac{D}{\lambda_0}$ values, and the results obtained helped us to conclude that the large enhancement of EB effect should be expected in the case $\frac{D}{\lambda_0} > 1$, namely as the size of the dot D is decreased towards λ_0 .

On the contrary, considering the case of circular dots with size $140\ nm$, and thinner AFM layer as well, so that the lowest polarizing action should be expected by the AFM nanograins, we observed small EB effect, even at the lowest temperature. We then suggested that we are actually addressing a case in which the length of magnetic correlation among the IrMn spins is spatially limited by confinement effects, namely the case in which $\frac{D}{\lambda_0} < 1$, and different hypotheses have been considered and tested through micromagnetic simulations to account for the weak EB effect measured. In other words, we proposed that the volume of the spin glass state is unavoidably lower and the spins themselves are thermally less stable (the

reduced volume of the spin glass phase that implies a decrease for the corresponding anisotropy energy term), with respect to either the continuous film or the square dots ($\frac{D}{\lambda_0} \gtrsim 1$ and presence of nanograins), where they were more free to magnetically correlate on a large length along the two surface dimensions.

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