Domain walls and exchange-interaction in Permalloy/Gd films

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Abstract. In this work we study the exchange coupling in Permalloy (Py)/gadolinium (Gd) bilayers. The exchange-coupled Py/Gd system is very temperature dependent and moreover the magnetization process in the Py layer is mainly due to domain wall (DW) displacements which are strongly controlled by pinning effects. We propose that this pinning could be caused by magnetostatic and exchange interactions between Py DWs and the magnetostrictive Gd layer. These effects mask the antiferromagnetic coupling between layers and, depending on temperature and Py thicknesses, apparent ferromagnetic coupling occurs. The study has been performed in the 80–300 K temperature range for different Py layer thicknesses and different Py induced anisotropies.

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1. Introduction

The antiferromagnetic coupling between transition magnetic metals (TMM) and heavy rare earths has been widely studied in multilayers such as Fe/gadolinium (Gd) and Co/Gd [1]–[8]. In these systems the magnetization processes are highly controlled by the rotation of magnetization across the interface between Gd and TMM forming a twisted magnetic structure [9]–[11]. The sharpest interfaces are usually obtained in Fe/Gd multilayers, where both experimental and theoretical works show the existence of these twisted magnetic structures [5]. Twisted structures appear in strongly exchange-coupled layers when their thicknesses are lower than the exchange correlation length. In these samples, the magnetization process is mainly due to magnetization rotation. However, for thicker layers and low exchange coupling interaction, the magnetization changes due to twisted structures can be negligible and magnetization processes due to lateral domain wall (DW) displacement can predominate. In particular, in the case of multilayers of hard/soft magnetic materials, when the magnetizing field is applied in the easy axis direction of the soft magnetic material, the magnetization inversion in this layer could be mainly due to lateral DW displacements.

The aim of this work is to show that the above mentioned magnetization process due to lateral DW displacements could influence the exchange-bias field determination and that they could even produce an apparent positive exchange coupling in antiferromagnetic coupled multilayers [12]–[15]. The proposed explanation is to suppose that there is a local interaction between the soft magnetic layer DW and the hard magnetic layer that pins the DW, increasing the magnetic field necessary to reverse the magnetization in the soft layer. This pinning is produced by magnetostatic and exchange interactions. In the case of negative exchange coupling, the DW pinning reduces the apparent exchange bias field effect. In some cases, DW pinning can be even greater than the exchange bias field effect, and an apparent positive exchange coupling appears.

We have selected a system based on two antiferromagnetic exchange-coupled thin films: a hard magnetic layer that produces the bias field and a soft magnetic layer in which we have studied the magnetization process. The Curie temperature of the hard magnetic film is much lower than the soft magnetic film, so the strength of the exchange coupling can be controlled by changing the system temperature. For weak exchange coupling, it is expected that the magnetization process of the soft magnetic film takes place by lateral DW displacements, whereas for stronger exchange coupling, a twisted magnetic structure will appear at the interface and magnetization rotation can be expected. Which magnetization process is more prevalent will also depend on the soft film thickness.

The Permalloy (Ni$_{80}$Fe$_{20}$)/Gd bilayers fulfill the above mentioned requisites [16]–[18]. Permalloy (Py) has high permeability and high Curie temperature (853 K), its anisotropy is easily controlled by growing conditions, and its magnetostriction is nearly zero. Gd has no crystalline anisotropy, but at temperatures below 200 K it has magnetostriction, $\lambda_s \approx 100$–200 ppm (depending on the axis direction) [19]. In the case of polycrystalline Gd, $\lambda_s \approx 13$ ppm [20], with polycrystalline Py $\lambda_s \approx 0$, Ni $\lambda_s \approx -34$ ppm and Fe $\lambda_s \approx -7$ ppm. The internal mechanical stresses produced during growth joint to the Gd magnetostriction result in a high local random anisotropy, as is confirmed by the high magnetic field necessary to saturate it and by its remanence, 0.5 $M_S$ (figure 1: inset). The Curie temperature of Gd is about 290 K and its coercive field widely increases as the temperature decreases due to its magnetostriction temperature behavior [20]. The coercive field of the Py/Gd bilayer, lower than the Gd one, also
Figure 1. Coercive field as a function of the temperature for the bilayer Py(100 nm)/Gd(50 nm) (•) and a Gd layer of 50 nm (■). Inset: (●) magnetic hysteresis loop of a 50 nm Gd single layer at 80 K.

increases as temperature decreases (figure 1), showing an unexpected temperature behavior for antiferromagnetically exchange-coupled bilayers.

2. Experimental

Samples were grown in a dc-magnetron sputtering system on corning-glass substrates. The residual pressure in the growth chamber was under $4 \times 10^{-7}$ mbar. Depositions were done at room temperature (RT), with an Ar pressure of $2 \times 10^{-3}$ mbar and a deposition power of 30 W in all cases. A 40 Oe magnetic field was applied in the sample plane during the growth to induce a magnetic anisotropy direction in the Py layer.

It has been reported that multilayers of Co/Gd and CoNi/Gd show Co and Ni diffusion into the Gd layer, creating an interfacial compound that greatly influences the magnetic and electrical properties [2, 6, 21]. In Py/Gd/Py trilayers [22]–[24], we have found Ni diffusion in the Gd/Py interface, but the diffusion is almost negligible in the Py/Gd interface. This result has been used in [22] to obtain Py/Gd bilayers without Ni diffusion in the interface. The samples used in this work were:

1. Py/Gd bilayers with a Gd layer 50 nm thick and Py layers of 100, 50 and 25 nm thickness.
2. Py/Mo/Gd in which a Mo spacer of 20 nm has been inserted between the Py and Gd layers in order to break the Py–Gd exchange interaction.
3. Gd single layer of 50 nm and Py single layer of 75 nm, which have also been grown for further comparison.

In all samples, Mo layers of 10 and 60 nm were used as buffer and capping layers. For lower thickness samples, two Py/Gd bilayers were required in order to enhance the magnetic signal. To that effect, Py/Gd bilayers separated by 20 nm Mo spacer layers, Mo(buffer)/[Py/Gd/Mo]$_2$/Mo(capping), were grown.
Figure 2. Exchange-bias field ($H_{\text{bias}}$) as a function of the temperature. $H_{\text{bias}}$ has been obtained from the hysteresis loops recorded after cooling-down the samples under an applied magnetic field of 1 T (a) Py(200 nm)/Gd(50 nm), (b) Py(100 nm)/Gd(50 nm), (c) [Py(50 nm)/Gd(50 nm)/Mo(20 nm)]$_2$ and (d) [Py(25 nm)/Gd(50 nm)/Mo(20 nm)]$_2$.

High-angle diffraction $\theta$–$2\theta$ and Auger electron spectroscopy (AES) measurements were done to characterize the samples. These measurements revealed the almost null Ni interdiffusion in the studied samples and rule out a partial oxidation of the Gd layer. X-ray diffraction scans show that both Py and Gd were polycrystalline. Py/Gd interface roughness was measured by means of atomic force microscopy in samples with the structure Mo(buffer)/Py(25 nm) deposited under the same growth conditions as the Py/Gd bilayers [22]. The rms value was 0.6 nm, a good roughness for a sputtered film.

The conclusions of this study will be mainly derived from low field measurements, at which the Gd layer is at remanence. Therefore, its magnetization is uniformly distributed in a $2\pi$ solid angle, due to its random uniaxial anisotropy. This effect would reduce the effective exchange interaction. Another source of exchange interaction reduction is the interface roughness and the small Ni diffusion. Figure 2 shows exchange bias field ($H_{\text{bias}}$) obtained by measuring hysteresis loop displacement after cooling the sample under a field of 1 T, in similar way to other authors [25]. The measured bias field at 76 K was $-47$ Oe. From this value and taking into account Gd magnetization distribution at remanence, the net exchange energy per unit of area ($\gamma_{\text{ex}}$) between Gd and Py can be evaluated from $\gamma_{\text{ex}} = 2\mu_0 M_{\text{SPy}} e_{\text{Py}} H_{\text{bias}}$ where $M_{\text{SPy}}$ denotes the Py saturation magnetization, $e_{\text{Py}}$ the thickness of the Py layer. From this expression it is obtained a value of about $4 \times 10^{-4}$ Jm$^{-2}$ for $\gamma_{\text{ex}}$.

The magnetic characterization was performed by means of a vibrating sample magnetometer (VSM) for different Py thicknesses and different anisotropy directions. At higher temperatures, the exchange interaction is so weak that the Py film magnetically behaves like a single layer and the magnetization process in the Py easy axis will be mainly due to DW displacements. At lower temperatures, the Py layer is fully exchange coupled to the Gd layer, and the magnetization process will be mainly due to magnetization rotation.
3. Results and discussion

The Py/Gd bilayer hysteresis loops are approximately the superposition of a soft and a hard magnetic loop (Py and Gd loops) (figure 3). The coercive field of the soft layer is directly related to the two large magnetization jumps corresponding to the magnetization inversion of the Py layer. As temperature decreases the Gd remanent magnetization increases and the magnetization jumps are vertically shifted, so the coercive field increases widely by this effect (figure 3). This shift is not directly related to the magnetic coupling between Py and Gd layers. A more direct parameter related with the magnetic coupling is the switching field ($H_{sw}$). $H_{sw}$ is the magnetic field necessary to switch the magnetization of the Py layer. Taking into account that the Gd layer is at remanence, the switching field is the Py coercive field plus the effective exchange bias field. $H_{sw}$ is measured as half the distance in the field axis between the middle points of the magnetization jumps (figure 4 inset). The Gd coercive field is much higher than the Py coercive field. If there were a negative exchange coupling between layers, the switching field, $H_{sw}$, would decrease as temperature decreases and it would even become negative. Figure 4 shows $H_{sw}$ plotted as a function of temperature for Py($x$ nm)/Gd(50 nm) samples ($x = 100$, 50 and 25 nm). $H_{sw}$ increases almost linearly from RT up to 120 K in all samples. Below this temperature, the $H_{sw}$ behavior strongly depends on the Py thickness. For 50 and 100 nm Py samples there is a large increase of the switching field, an increase that, in principle, could be attributed to a positive exchange coupling in this temperature range or to an increasing of DW pinning process. Later on it will be shown that in fact this behavior can be explained by taking into account DW pinning processes. For the 25 nm Py sample, the $H_{sw}$ behavior indicates a negative coupling.

To understand these results we have measured the exchange decoupled samples Py($x$ nm)/Mo(20 nm)/Gd(50 nm) (figure 4), observing that $H_{sw}$ increases almost linearly for the whole temperature range, with behavior similar to the exchange-coupled samples in the RT to 120 K range. As can be observed in figure 5, exchange coupled and decoupled samples with the same Py and Gd thicknesses show almost indistinguishable hysteresis loops in this temperature range and, therefore, magnetization mechanisms must be analogous in both types of samples. In decoupled samples the magnetization processes are only due to Py lateral DW
displacements, then it can be inferred that for the RT to 120 K range all the other samples have similar magnetization processes and the DWs pinning mechanism is also similar.

As the temperature decreases the switching field of decoupled samples linearly increases. However, the switching field is nearly constant in the single Py layer (figure 4). This effect can only be due to a magnetostatic interaction between Py DWs and the Gd layer. The Py DWs stray field produces local changes in the Gd magnetization (figure 6) to close the magnetic flux inside the Gd (a quasi-DW [26]) that pins Py DW. This pinning is produced by the interaction between the Py DW and the non-homogeneous Gd magnetization due to its magnetoelastic energy fluctuations. The pinning field, $H_p$, can be calculated following the Kersten idea [27].

We determine the DW pinning field through the magnetostatic energy variation of the Gd zone magnetized by the DW field dispersion. Assuming a periodic fluctuation of the Gd anisotropy, $K$, due to a random distribution of internal stresses $K = (3/2)\lambda_{sGd} (\sigma) (1 + \cos(2\pi x / \xi))$, with $\lambda_{sGd}$
the Gd saturating magnetostriction constant, $\xi$ the wavelength of anisotropy fluctuation, $M_S$ the Gd saturation magnetization, $Y_{Gd}$ the Gd Poisson coefficient and $\langle \sigma \rangle = (3/2) Y_{Gd} \lambda_{Gd} (M/M_s)^2$ the average internal stresses calculated assuming a non-deformable substrate that avoids the Gd local elongations when Gd becomes ferromagnetic.

The magnetostatic energy density of the Gd zone magnetized by Py DW magnetic field dispersion is calculated from $\gamma_M = \frac{1}{2} \mu_{Gd} H_{Gd}^2 \delta (\text{Jm}^{-2})$ where $\mu_{Gd}$ is the Gd permeability, $H_{Gd}$ the field produced by the Py DW poles in the Gd zone near the Py DW, and $\delta$ the Py DW thickness. $H_{Gd}$ is evaluated by considering the Py DW and the Gd system as a magnetic circuit (figure 6) and $\mu_{Gd}$ by assuming reversible magnetization rotation. For $\xi = \delta$, the DW pinning field or magnetostatic pinning field, $H_{pM}$ reaches its maximum value, $H_{pM} = (1/2\mu_0 M_{SPy}) (\partial \gamma_M / \partial x)$ where $M_{SPy}$ is the Py saturation magnetization. For $T \approx 120$ K, $H_{pM} \approx 12$ Oe in good agreement with the experimental $H_{sw}$ results (figure 4).

Below 120 K, the $H_{sw}$ behavior is strongly dependent on Py thicknesses. In samples of 25 nm Py thickness, $H_{sw}$ decreases, showing that the entire Py layer is antiferromagnetically exchange-coupled to the Gd layer. In samples of thicker Py layers, $H_{sw}$ greatly increases. Therefore, we can assume that lateral DW displacement magnetization processes still exist and that another stronger pinning mechanism associated with the exchange coupling appears.

This new pinning mechanism must arise from the antiferromagnetic exchange coupling. The exchange energy by unit of area is dependent on the saturation magnetization of Gd and Py and on the angle, $\theta$, between them. In the studied temperature range, the Py saturation magnetization is almost constant, whereas the Gd saturation magnetization changes from almost zero up to 2.5 T. On the other hand, there is a dispersion of anisotropy in the Gd layer that produces fluctuations in the angle between magnetizations in both layers, fluctuations that in turn reduce the effective exchange coupling in magnetic domains and also produce changes in Py DW energy. These Gd magnetization fluctuations are widely reduced below the Py DW zone due to the strong dispersion magnetic field above mentioned. Hence, the DW net exchange energy fluctuation is

$$\gamma_{wex} = \frac{C}{2} \left( 1 + \cos \left( \frac{2\pi x}{\xi \alpha} \right) \right),$$

where $C$ is the maximum value of $\gamma_{wex}$ and $2\pi \alpha$ is the maximum dispersion angle between the Gd magnetization and the dispersion field produced by Py DW in the Gd layer.

$H_{pex}$ is the pinning field related to the exchange coupling and is determined from $H_{pex} = (1/2\mu_0 M_{SPy}) \gamma_{wex} / \partial x$, obtaining $2\pi \alpha \sin(2\pi \alpha) = (2\mu_0 M_{SPy} H_{pex} \delta / C)$. Assuming
Figure 7. A bilayer of Py(100 nm)/Gd(50 nm) has been measured at 80 K. (a) A first minor hysteresis loop has been measured after a magnetic field of $10^4$ Oe has been applied in the Py easy axis direction (◼). Then, the sample is rotated $\pi/2$ and a second hysteresis loop is recorded with the applied magnetic field in the hard axis direction (○). Inset: schematic representation of the measurement configuration. (b) In this case, a first minor hysteresis loop has been measured after a magnetic field of $10^4$ Oe has been applied in the Py hard axis direction (◼). Then, the sample is rotated $\pi/2$ and a second hysteresis loop is recorded with the applied magnetic field in the easy axis direction (○). Inset: schematic representation of the measurement configuration.

The thickness of the wall $\delta \approx 100$ nm, taking the experimental value of $H_{\text{per}} \approx 30$ Oe, and the above obtained $\gamma_{\text{ex}} = 4.10^{-4}$ J m$^{-2}$, the maximum dispersion angle obtained is: $2\pi\alpha \approx 0.9$ rad. This result, 0.14 (2$\pi$), shows the high reduction of Gd magnetization fluctuations due to the Py DW dispersion field.

It is noteworthy that the bias field is related to the Py–Gd exchange coupling whereas the switching field is related to the effective magnetic field necessary to invert the Py magnetization. Therefore, the switching field is the Py coercive field and the exchange bias field addition.

$$H_{\text{sw}} = H_c + H_{\text{bias}}.$$  

In antiferromagnetically coupled systems, when temperature decreases, both $H_c$ and the absolute value of $H_{\text{bias}}$ increase, leading to a temperature-dependent increase or reduction in $H_{\text{sw}}$. The pinning field is low dependent on the Py layer thickness whereas the bias field effect on the Py layer decreases when the Py layer thickness increases. Both effects explain the $H_{\text{sw}}$ behavior.

These assumptions can be verified by measuring Py lateral DW displacements in two situations: (i) Py DW parallel to Gd magnetization in such a way that $H_c$ and $H_{\text{bias}}$ affect DW displacements and (ii) Py DW perpendicular to Gd magnetization in such a way that only $H_c$ affects DW displacements because $H_{\text{bias}}$ is perpendicular to DW. To perform this experiment, we have obtained minor hysteresis loops, at 80 K, in the thicker (100 nm) Py/Gd bilayers, with the Py domain magnetization parallel (figure 7(a)) and perpendicular (figure 7(b)) to Gd magnetization, with the bias field first parallel and then perpendicular to the magnetizing field. Figure 7(a) shows the low field zone of the magnetization curve obtained after saturating the sample at $10^4$ Oe in the Py easy axis and then reducing the field down to $-50$ Oe to observe the Py magnetization inversion. Then another minor hysteresis loop is measured, applying the magnetizing field perpendicular to Gd magnetization and later to the Py easy axis. Thus,
the magnetization process will be mainly due to $90^\circ$ magnetization rotation. Given the Gd coercive field is higher than $|-50|$ Oe, we can suppose that the Gd magnetization is almost at remanence. Since in this measurement configuration the negative exchange coupling takes place in the Py easy axis direction, the effect of the bias field is to reduce the hysteresis loop slope, with an increase in the apparent anisotropy field up to 50 Oe (in the Py single layer the anisotropy field is about 15 Oe). In the same way, figure 7(b) shows the low field zone of the magnetization curve obtained after saturating the sample at $10^4$ Oe in the Py hard axis, and then reducing the field down to $-50$ Oe. Afterwards, a second minor hysteresis loop is measured, applying the magnetizing field in a direction perpendicular to Gd magnetization and thus parallel to the Py easy axis. The magnetization process is mainly due to Py DW displacements and both Py DW and Py domain magnetization are perpendicular to Gd magnetization as can be observed in figure 7(b). Then, the negative exchange coupling effect on domain energy is the same in all magnetic domains. So, the unique pressure on DW is produced by the applied magnetic field and the high coercivity of the Py/Gd bilayer can only be due to a DW pinning. The coercive field of the second minor hysteresis loop is in the same order as the $H_{sw}$ measured in figure 3. This confirms that the large increase of $H_{sw}$ below 120 K is due to the DW pinning produced by exchange coupling.

4. Conclusions

In summary, in Py–Gd bilayers the interface exchange interaction is weak at a temperature in the 300–120 K range, and the magnetostatic interaction between Py DW and Gd magnetization can explain the switching field behavior.

Below 120 K the Py magnetization process depends widely on Py thickness:

1. For thickness below the exchange correlation length, i.e. for the sample of the 25 nm Py layer, the Py magnetization process is due to an in-plane magnetization rotation and so the exchange interaction produces a negative bias field that reduces the switching field.

2. In samples with a thicker Py layer, the magnetization process seems to be mainly due to DW displacements. The exchange interaction results in two effects: (i) an exchange coupling between Py DWs and Gd magnetization and (ii) a negative exchange coupling of Gd with the Py domain magnetizations that can be considered as a bias field. The first one produces a strong pinning in DW displacements due to the high Gd magnetostriction and to the local Gd anisotropy fluctuations, a pinning that increases the switching field. The second one reduces the switching field. In spite of the negative exchange coupling between Py and Gd layers, for certain temperatures and Py thicknesses the first effect is greater than the second one and an apparent positive exchange interaction appears between Py and Gd.

In principle, these effects are not restricted to multilayers with antiferromagnetic coupling and they could also appear in systems with ferromagnetic coupling. Therefore, they must be taken into account in research on multilayers with DW magnetization processes. The exchange coupling produces a bias field and also DW pinning.

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